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Investigation of Cl-36 Behaviour in Soils and Uptake into Crops

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BIOPROTA

Key Issues in Biosphere Aspects of Assessment of the Long-term Impact of Contaminant Releases Associated with Radioactive Waste Management

Investigation of Cl-36 Behaviour in Soils and Uptake into Crops

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PREFACE

BIOPROTA is an international collaboration forum which seeks to address key uncertainties in the assessment of radiation doses in the long term arising from release of radionuclides as a result of radioactive waste management practices. It is understood that there are radio-ecological and other data and information issues that are common to specific assessments required in many countries. The mutual support within a commonly focused project is intended to make more efficient use of skills and resources, and to provide a transparent and traceable basis for the choices of parameter values, as well as for the wider interpretation of information used in assessments. A list of sponsors of BIOPROTA and other information is available at www.bioprota.com.

The general objectives of BIOPROTA are to make available the best sources of information to justify modelling assumptions made within radiological assessments of radioactive waste management. Particular emphasis is to be placed on key data required for the assessment of long-lived radionuclide migration and accumulation in the biosphere, and the associated radiological impact, following discharge to the environment or release from solid waste disposal facilities. The programme of activities is driven by assessment needs identified from previous and on-going assessment projects. Where common needs are identified within different assessment projects in different countries, a common effort can be applied to finding solutions.

This report describes work undertaken to compare and contrast different assessment models for the accumulation of Cl-36 in soils and its uptake into crops and the food chain. It was started following a workshop on Cl-36 behaviour in the biosphere held within the BIOPROTA framework and hosted by ANDRA in September 2006. The study was carried out by staff from ANDRA, EDF, IRSN, JGC Corporation, NWMO, SCM, Quintessa and GMS Abingdon. The study was financially supported by ANDRA, JGC Corporation, the NDA and NUMO.

The report is presented as working material for information. The content may not be taken to represent the official position of the organisations involved. All material is made available entirely at the users' risk.

Version History

Version 1.0: Work-plan and scenario description prepared by Enviros following review by Working Group participants and distributed to 19 April 2007.

Version 1.1: Included some model descriptions, incorporating some information from the May 2007 BIOPROTA meeting in Prague and distributed to Working Group participants, 13 July 2007.

Version 1.2: Updated by GMS Abingdon Ltd and distributed to Working Group participants, 4 January 2008.

Version 1.3: Updated by GMS Abingdon after a Working Group meeting at ANDRA 22 January 2007, and distributed to Working Group participants, 24 January 2008.

Version 2.0: Updated by Quintessa to include most model descriptions, incorporating some of the results from illustrative calculations before 12 April 2008, and distributed to Working Group participants, 30 April 2008.

Version 2.1: Updated by Quintessa following a workshop held by the Working Group participants as part of the May 2008 BIOPROTA meeting at Nagra, incorporating new analyses of the illustrative calculations and conclusions, and distributed to Working Group participants, 2 June 2008.

Version 3.0: Updated by Quintessa and GMS Abingdon in the light of comments from participants. 2 July 2008.

Version 3.1: Updated by Quintessa in the light of comments from participants. 22 July 2008.

Version 3.2: Updated by Quintessa in the light of comments from participants. 3 September 2008.

EXECUTIVE SUMMARY

This report describes a study undertaken following a workshop on Cl-36 behaviour in the biosphere hosted by ANDRA in September 2006 within the BIOPROTA international cooperation framework. Some initial investigations and discussions were reported in the 2007 Annual BIOPROTA workshop report, after which a detailed project scope and plan was developed.

The question the study sought to address was the typical Performance Assessment question, i.e. what is the annual individual dose to an average member of a potential exposure group (PEG). However, this study stopped at the assessment of Cl-36 concentrations in the edible parts of the harvested crops which could be consumed by the PEG or by animals forming part of the human food chain.

The objectives of the study were:

- to compare the scientific basis supporting alternative approaches used to assess the contamination of soil by Cl-36 and its uptake by crops,
- to make relevant illustrative calculations using models based on possibly alternative assumptions, and
- to make recommendations concerning dose assessment for long-term post-closure assessment of Cl-36 releases from waste repositories.

There is a general need for the assessment process to be informed of how much uncertainty is acceptable, within the context of a particular assessment. This requirement applies to all the relevant radionuclides and work to develop an understanding of how to determine the level of acceptable uncertainty in a specific context would be useful across the board.

The different assessment contexts include issues such as whether climate and environmental change should be used, or only the site's present day condition. This will have a material impact on the relevance and need for site specific data, and/or data from other sites which could be analogues today for the future condition of the site under investigation.

A significant range of model types and alternative data assumptions has been described and compared, demonstrating the degree of understanding of the likely behaviour of Cl-36 soil and plant systems. Results have been presented for Cl-36 concentrations in crops based on unit activity concentration in upwelling groundwater and irrigation water, both deterministic and probabilistic. A wide range of sensitivity calculations has also been presented

Despite the variations in the models, the results for concentrations in crops are typically within about an order of magnitude of each other for the main scenario considered. Furthermore, the spread of results obtained in probabilistic calculations does not indicate much greater variation for a given set of site conditions. This implies that knowledge of stable chlorine levels is critical for the specific activity models, and for other models, that the soil and plant systems must be sufficiently well characterised to allow usefully confident choices of distribution coefficients and root uptake factors, or to provide the other more detailed data. This is a truism, but something not to be forgotten. The model input data requirements in turn help to define site investigation requirements.

The range of results presented here, in combination with the model explanations, can support consideration of whether particular model assumptions can be considered as overly-cautious, cautious or more likely to represent the likely outcome in particular contexts.

There is a general need for the assessment process to be informed of how much uncertainty is acceptable, within the context of a particular assessment. This requirement applies to all the relevant radionuclides and work to develop an understanding of how to determine the level of acceptable uncertainty in a specific context could be useful across the board.

The application of the models used here could be extended to the calculation of doses to potentially exposed groups. Different approaches are employed to define these groups and that comparison would be instructive, both for Cl-36 and generally. The purpose would not be to identify a best approach, since that will be context dependent, but to investigate how different assumptions affect the results and also to investigate whether the variation in dose results is dominated by variant assumptions in the calculation of concentrations in crops, or in those for human behaviour.

In combination, these activities could then inform requirements for future investigations at the site specific level.

The study was carried out by staff from ANDRA, EDF, IRSN, JGC Corporation, NWMO, SCM, Quintessa and GMS Abingdon. The study was financially supported by ANDRA, JGC Corporation, the NDA and NUMO.

CONTENTS

1 INTRODUCTION

1.1 BACKGROUND

Cl-36 has been identified as one of the radionuclides dominating estimates of annual individual doses to potential exposure groups in post-closure performance assessments of both high and intermediate level radioactive waste repositories. In addition, there are notable uncertainties in some of the data employed in making those dose estimates, see for example discussion in BIORPOTA (2006). While in general, interception by growing crops of contaminated irrigation water is of interest (see report of interception models in BIOPROTA (2005a), for Cl-36, the largest uncertainties are associated with assumptions for how it would accumulate in soil given a long-term release from the geosphere, and how that Cl-36 might then be taken up by plants. These plants might then be consumed by humans or enter the human food chain via animal products consumed by humans. Transfer through animals, however, was not considered.

Given these soil accumulation and plant uptake uncertainties, a cautious approach to dose estimation has generally been adopted in dose assessments, with the intention that any estimated doses are unlikely to have been significantly under-estimated. However, this may result in significant over-estimation of radiological impact. Although these estimated doses may themselves not be significant compared to other sources of, say, natural radiation exposure, there is good sense in removing unnecessary pessimism within performance assessments since that may result in misallocation of research and other resources. At the same time, conventional models routinely applied to all radionuclides may in some circumstances lead to dose underestimation, due to omission of processes which are specifically relevant to special radionuclides such as Cl-36, which have unique characteristics. More broadly, there is a general desire to develop as good an understanding of the system under assessment as possible within wider resource constraints, so as to generate broad confidence in the overall assessment conclusions (BIOPROTA, 2007).

1.2 OBJECTIVES

The specific objectives of this study were:

- to compare the scientific basis supporting alternative approaches used to assess the uptake of Cl-36 from soils to crops,
- to make relevant illustrative calculations using models based on the alternative assumptions, and
- to make recommendations concerning dose estimation methods and/or requirements for improved data for long-term post-closure assessment of Cl-36 releases from waste repositories.

1.3 SCOPE

The scope is limited to the long-term release of repository derived Cl-36 in groundwater from the geosphere to the biosphere, the behaviour of that Cl-36 in agricultural soils, and the uptake of that Cl-

36 into farm crops which may contribute to various ingestion pathways for potential exposure groups. The interception of Cl-36 by the aboveground plant has been disregarded; a decision justified using the known dominance of root to plant transfer. Two source terms were assumed by which the Cl-36 in groundwater reaches the rooting zone of agricultural soil:

1. as a result of irrigation, and

2. as a result of capillary action and any/all other transfers from below

The models used in the inter-comparison approach can be distinguished on the basis of the soil and the soil to plant sub models. Four types of model Cl-36 behaviour in soil and root uptake were considered.

a) Conventional (radionuclide generic) models which assume: (1) a radionuclide residence time and accumulation in root-zone soil with consideration of infiltration into deeper soil by water; (2) an instantaneous equilibrium soil-water distribution coefficient to determine the mobile fraction; and (3) a fixed plant to soil concentration ratio, through which the radionuclide concentration in the plant is determined as a result of root uptake. Other processes are included in some applications, such as erosional losses, mixing to deeper soil by biota and losses from the root-zone due to uptake and cropping. (See a review and summary of variant applications in BIOPROTA (2005).) The crops may also become contaminated on their surface due to soil splash. Although some of the model parameter values are specific to chlorine or Cl-36, this approach does not take explicit account of processes specific to chlorine behaviour in soils and is structurally the same as that applied to a wide range of radionuclides.

b) Specific activity models in which it is assumed that Cl-36 reaches equilibrium in some components of the system in the same proportions as stable chlorine. Dynamic treatment of movement between other components is still needed; the decision on which components are at equilibrium is dependent upon many factors and still under discussion.

c) Compartmental models which consider the behaviour of Cl-36 in soil in a similar way to the conventional generic models but use an isotope ratio (like that of the specific activity models) for the plant uptake of Cl-36 from the soil.

d) More detailed models which include further consideration of the dynamics of water flows and stable chlorine fluxes as well as chemical forms and greater spatial and temporal discrimination. Transpiration is one of the modes for plant uptake of Cl-36 considered in such models.

1.4 STRUCTURE OF REPORT

After this introduction, Section 2 provides details of the different types of models which have been applied or recently developed to address the dose assessment issues under investigation in this study (see Section 1.3 above). Section 3 describes an assessment situation, or “scenario”, which may be considered a typical requirement in the evaluation process for long-term repository performance assessments. Section 4 sets out the specific data applied to the scenarios for the separate models described in Section 2. Section 5 contains results and discussion. Section 6 provides conclusions and recommendations.

2 CONCEPTUAL MODEL DESCRIPTIONS AND MATHEMATICAL REPRESENTATIONS

There is a variety of broad classes of model that have been used for this inter-comparison, distinguished on the basis of the soil and the soil to plant sub models (Table 2-1). Table 2-2 offers a summary of the models involved in this comparison exercise, with the abbreviations used in the results. Details of these models are given in Sections 2.1 to 2.4, and a summary of the model similarities and differences is given in Section 2.5.

Table 2-1: Soil and soil-to-plant sub models used for the model inter-comparison

Section	Soil sub model	Soil-to-plant sub model	General model
2.1	Complete budget for water and Cl including losses via percolation	Transfer factor	EPRI, BIOMASS, JGC, Aquabios, SAMM-TR
2.2	Budgeting Cl-36 and stable Cl input	Isotope ratio	AquaCl36, NWMO
2.3	Complete budget for water and Cl including losses via percolation	Isotope ratio	EDF, SAMM-IR
2.4	Complete budget for water and Cl including losses via percolation	Water uptake via transpiration × Cl-36 activity[a]	IRSN, NDA-RWMD

[a] In the NDA RWMD model the following processes are also considered: active uptake (using Michaelis-Menton kinetics), root exudation

Table 2-2: Summary of models used in this study

Model type	Participant	Model name	Code used for model implementation
Conventional Radionuclide Generic	EPRI	EPRI	AMBER
	BIOMASS	BIOMASS (ERB2A is used for the well scenario; ERB2B is used for the groundwater scenario)	AMBER
	JGC	JGC	AMBER
	ANDRA	Aquabios	MoM
	SCM	SAMM-TR	Matlab
Specific Activity (SA)	ANDRA	AquaCl36	MoM
Compartmental with isotope ratio for plant uptake	SCM	SAMM-IR	Matlab
	EDF	EDF	ECOLEGO
Complex	IRSN	^{36}Cl SP236Cl	GoldSim / SYMBIOSE
	NDA RWMD	MTA	ModelMaker 4.0

2.1 CONVENTIONAL MODELS

Conventional models assume a complete budget for water and radionuclides in the soil, be it for one layer of soil or multiple layers of soil. In general, the rate of change in the amount of a given radionuclide in a model compartment is

$$\frac{dN_i}{dt} = \left(\sum_{j \neq i} \lambda_{ji} N_j + \lambda_M M_i + S_i(t) \right) - \left(\sum_{j \neq i} \lambda_{ij} N_i + \lambda_N N_i \right) \quad (1)$$

where:

N_i is the amount of the radionuclide in compartment i, mol

M_i is the amount of the precursor radionuclide in compartment i, mol (M decays to N)

$S_i(t)$ is the external source of N to compartment i, mol y^{-1}

λ_M, λ_N are the decay constants for M and N respectively

λ_{ji} are the transfers from compartments j ($\neq i$) into compartment i, y^{-1}

λ_{ij} are the transfers from compartment i into compartments j ($\neq i$), including external losses, y^{-1}

In this study, there is only a single transfer of Cl-36 into the soil compartment, S_i, that of either irrigation or groundwater depending upon the scenario under consideration. All of the models discussed below consider percolation as a means of transferring Cl-36 from the topsoil to deeper soil layers, or out of the system altogether. Within the soil compartments an instantaneous equilibrium soil-water distribution coefficient to determine the mobile fraction of Cl-36 in the soil.

In conventional models, concentrations of radionuclides in the flora, fauna and atmosphere are assumed to be in equilibrium with the dynamically modelled concentrations in the donor environment, i.e. soil, from which they receive their radionuclides, so that these biosphere receptors do not need to be dynamically modelled as well. Thus, in order to determine the radionuclide concentration in the plant as a result of root uptake a fixed plant to soil concentration ratio is used.

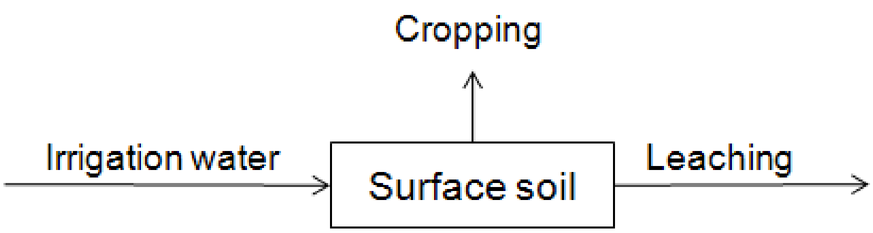
2.1.1 APPROACH BASED ON EPRI MODEL

Introduction

This model was developed to model the migration and accumulation of radionuclides in soil following introduction to the soil via irrigation water applied to agricultural and garden soil in a near desert climate at Amargosa Valley, Nevada, as part of a Total System Performance Assessment for HLW disposal at Yucca Mountain (EPRI, 2002). It is the same as the EPRI model considered in an earlier inter-comparison exercise (BIOPROTA, 2005b), but using Cl-36 specific parameter values.

The model assumes a single soil compartment, with transfers of radionuclide in and out of the soil as illustrated below (Figure 2-1). At the site for which this model was developed, the water table is so far from the surface that up-welling is not considered in the model.

Figure 2-1: Schematic of the EPRI model



Inputs and losses of contaminants to the system

The radioactive source term to the soil is a function of concentration in the irrigation water and the amount of water applied per unit area per year. Contaminated water is assumed only to reach the soil via spray irrigation. The amount of irrigation water applied per year is dependent upon climate, number of crops per year and type of crop. The credible range of irrigation is from 0.1 m/y by gardeners who might chose to irrigate at a low level up to 1.2 m/y by alfalfa growers who produce multiple crops in a year. Since the soil could be used for different purposes over the potentially extended period of contaminated irrigation, a single value for all crops is used and a long term average contamination level is then determined.

Losses of contamination from the soil which are taken into account in the model include migration in infiltrating groundwater and losses in cropping. The equations used to describe these transfers are given in Table 2-3.

Uptake of contaminant by the crop

The radionuclide concentration in the crop, C_{crop} (Bq kg^{-1}, fresh weight of crop), is given by

$$C_{crop} = \frac{\left(F_{p2} CF_{crop} + F_{p1} S_{crop}\right) C_s}{\left(1-\theta_t\right)\rho} + I_{crop} V_{irr} C_w \left(\frac{\left(1-F_{abs}\right) e^{-TW} F_{p3}}{Y} + \frac{F_{abs} F_{p2} F_{trans}}{Y} \right) \qquad (2)$$

where:

C_s is the radionuclide concentration in the soil compartment, Bq m^{-3}

I_{crop} is the fraction of radionuclide in spray irrigation water that is initially deposited on standing biomass,

S_{crop} is the soil contamination on the crop, kg (dry weight soil) kg^{-1} (fresh weight of crop),

F_{trans} is the fraction of absorbed activity that is translocated to the edible portion of the plant by time of harvest (translocation fraction),

F_{abs} is the fraction of intercepted radionuclide initially deposited on the plant surface that is absorbed from external surfaces into plant tissues,

F_{p1} is the fraction of external soil contamination on the edible part of the crop retained after processing,

F_{p2} is the fraction of the internal contamination associated with the edible art of the plant at harvest that is retained after food processing has occurred,

F_{p3} is the fraction of external contamination from interception that is retained on the edible part of the crop after food processing,

W is the removal rate of radionuclide deposited on the plant surface by irrigation by weathering processes (weathering rate) including mechanical weathering, wash-off and leaf fall, y^{-1},

T is the interval between irrigation and harvest, y

and other parameters are defined in Table 2.3.

The first term of this equation represents crop contamination due to both internal uptake from the surface soil compartment via the roots and external contamination from the deposition of re-suspended sediment from the surface soil compartment. The second term corresponds to plant uptake via irrigation. However, in this study, the focus is on accumulation in soil and root uptake, so that here the interception of irrigation water by standing biomass is disregarded, i.e. I_{crop} is set to zero.

Table 2-3: Functions used for transfers of interest in the EPRI model

Transfer	Function	Parameter definitions
Irrigation	$V_{irr} C_w$	V_{irr} - irrigation rate applied to the surface soil compartment, $m^3\ y^{-1}$ C_w - radionuclide concentration in the water, $Bq\ m^{-3}$
Leaching	$\frac{I}{R\theta d}$ where R is given by $1+\frac{(1-\theta_t)\rho}{\theta}K_d$	I- annual infiltration/recharge rate, $m\ y^{-1}$ R - retardation coefficient for the compartment from which infiltration/recharge is coming θ - water filled porosity of the compartment from which infiltration/recharge is coming d - depth of the compartment from which infiltration/recharge is coming, m θ_t - total porosity of the sediment or soil compartment, ρ - grain density of the sediment or soil compartment, $kg\ m^{-3}$, K_d - sorption coefficient of the sediment or soil compartment, $m^3\ kg^{-1}$
Cropping	$\frac{(CF_{crop}+S_{crop})Y_{crop}}{(1-\theta)\rho}$	$CFcrop$ - concentration factor from root uptake for the crop, $Bq\ kg^{-1}$ (fresh weight of crop)/$Bq\ kg^{-1}$ (dry weight of soil) $Scrop$ - soil contamination on the crop, kg (dry weight soil) kg^{-1} (fresh weight of crop) $Ycrop$ - wet weight biomass of the crop, $kg\ m^{-2}\ y^{-1}$, obtained at harvest from the unit area irrigated

2.1.2 APPROACH IN BIOMASS REFERENCE BIOSPHERES REPORT

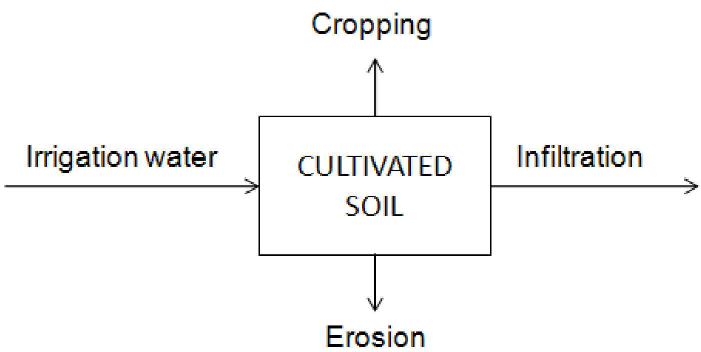
Introduction

The IAEA Programme on BIOsphere Modelling and ASSessment (BIOMASS) was launched in Vienna in 1996 (IAEA 1996). The programme was concerned with developing and improving capabilities to predict the transfer of radionuclides in the environment. As part of this programme the concept of a reference biosphere for application to the assessment of the long-term safety of geological repositories for radioactive waste was developed. Compartmental flow models were devised, considering a range of ecosystems and geosphere-biosphere interfaces. Exposure pathways to the human community were considered in great depth, but are beyond the scope of this study. The following description of the BIOMASS model has been derived from BIOMASS (2003).

The setup used for the two scenarios differ in that they consider either a single habitat or a series of interacting habitats for the 'irrigation water source' and 'natural groundwater source' scenarios respectively.

The model used for the 'irrigation source' scenario is able to track contaminant movement from the irrigation water via soil, atmosphere, crops and animals to humans, with excreted material (from both animals and humans) being recycled. However, the processes of primary interest involve contamination of the soil and crop. Crops will become contaminated due to direct deposition of irrigation water. A fraction can be retained on the plant surface and another fraction transferred within the plant, particularly to edible parts. Weathering of plant surfaces results in transfer of intercepted radionuclides to the soil. Crops may then become contaminated by root uptake, and soil splash may result in further crop contamination. Although seasonal factors can substantially influence details of what could happen in any one year, the above processes can be modelled on the basis of equilibrium between the concentration in the irrigation water and the crops, or the soil and the crops (BIOMOVS II, 1996; see Figure 2-2).

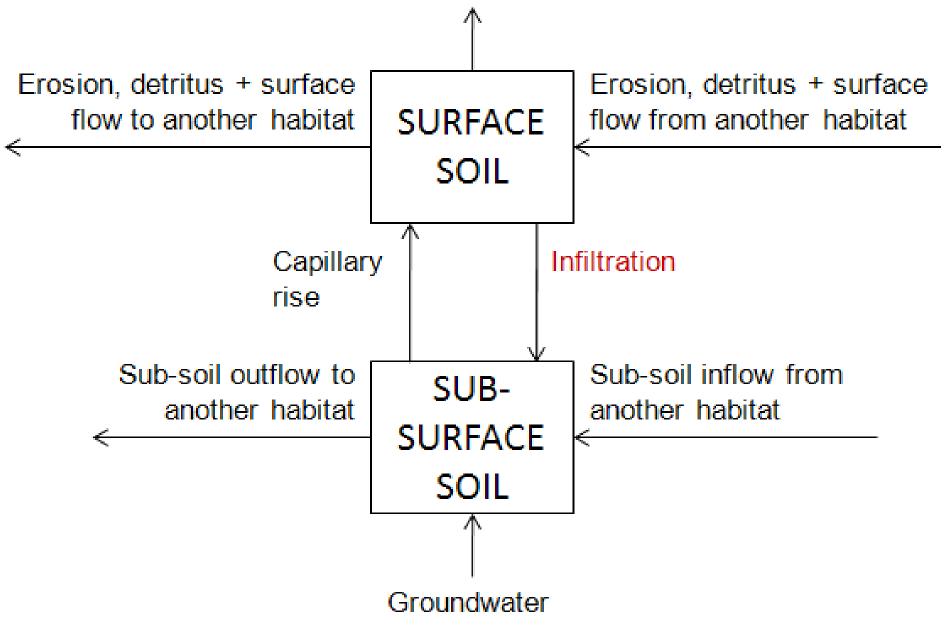
Figure 2-2: Simplified BIOMASS model for the BIOPROTA Cl-36 study – 'irrigation water source' scenario



For the 'natural groundwater source' scenario, a range of habitats were considered (woodland, pasture, arable land, shrub land, marsh, river and lake), with additional assumptions as to the spatial connectivity of these habitats. Within each of these habitats transfers between the following compartments are considered - aquifer, soils, atmosphere, plants, animals and sinks – with the dynamics of contaminants within the soil being of key interest (Figure 2-3). As with the irrigation scenario model, only the dynamics of the soil compartment need be simulated when considering the contaminant uptake by the crop; the crop uptake is discussed below. The habitat of relevance for this

study is the arable land. It is considered that some of each habitat will be adjacent to a river and some adjacent to a lake.
In addition to interactions within the habitats, interactions between the habitats (hydrological flows, atmospheric volatilisation and deposition, soil movement, exchange of animal/plant material) are also considered. However, these are beyond the scope of this study.

Figure 2-3: Simplified BIOMASS model for the BIOPROTA Cl-36 study – 'natural groundwater source' scenario. Transfers highlighted in red are only considered for the root and green vegetables.



<u>**Inputs and losses of contaminant to the system**</u>

For the 'irrigation water source' scenario, the contaminant is assumed to enter the system as the irrigation water (Figure 2-2). For the 'natural groundwater source' scenario contaminated water can enter the system either directly from below or as a result of hydrological inflows from a neighbouring habitat (Figure 2-3).

The ways in which contaminant can leave the system depend upon the scenario being considered. For the 'irrigation water source' scenario it is assumed that contaminant leaves the system via infiltration, erosion and cropping (Table 2-4). However, erosion losses can be disregarded (BIOMASS 2003) since the amount lost is not significant compared to the other transfers. For the 'natural groundwater source' scenario it is assumed that contaminant leaves the system via either the movement of water to the sub- or surface soil of a neighbouring habitat, detritus movement to another habitat and loss to crops (Table 2-5).

With respect to the crop, it is assumed that contamination can be lost via food preparation (F_{p1}, F_{p2} and F_{p3}). The time between irrigation and cropping can affect final concentrations in the crop, bearing in mind the time for uptake from soil and the rate of weathering loss from contamination on plant surfaces arising from interception of irrigation water. The latter is not considered in this study, and the former is assumed to occur in this model as if all processes are occurring through the year at the annual average rate.

Transfers of contaminant within the system and uptake by the plant

The algebraic form used for each dynamic transfer process in the 'irrigation water source' scenario is shown in Table 2-4, and those for the 'natural groundwater source' scenario are shown in Table 2-5.

The radionuclide concentration in the edible part of the crop, Bq kg^{-1} (fresh weight of crop) has the same form for both scenarios, and is given by the same equation as used in the EPRI model (Equation 2). However, there is a difference in where the soil Cl-36 concentration comes from.
For the irrigation scenario (ERB2A) there is only one habitat, and thus only one topsoil compartment whose Cl-36 concentration to be considered for the calculation of plant contamination. However, in the groundwater scenario (ERB2B) it is assumed that there will be an area of relevant habitat (arable land) adjacent to a river and another area of this land adjacent to a lake. The soil Cl-36 concentration used in determining plant concentration of Cl-36 is the arithmetic mean of the soil Cl-36 concentration in these two arable habitats.

Table 2-4: Transfers between compartments in the BIOMASS 'irrigation water source' scenario model

Transfer	Equation	Parameter definitions
Irrigation source term to soil, S	$V_{irr}C_w$	V_{irr} – irrigation water application rate, m^3 y^{-1} C_w – radionuclide concentration in well, Bq m^{-3}
Infiltration and recharge, λ_{II}	$\frac{I}{R\theta d}$ where R is given by $1+\frac{(1-\theta_t)\rho}{\theta}K_d$	I - annual infiltration/recharge rate, m y^{-1} R - retardation coefficient of cultivated soil θ - water filled porosity of cultivated soil d - depth of cultivated soil, m θ_t - total porosity of the cultivated soil compartment, ρ - grain density of the cultivated soil compartment, kg m^{-3}, K_d - sorption coefficient of the cultivated soil compartment, m^3 kg^{-1}
Erosion, λ_{IE}	$\frac{E}{d}$	E - erosion rate of the soil compartment, m^3 y^{-1}
Cropping, λ_{IC} [transfer from soil to plant]	$\frac{\sum_{crop}\frac{1}{4}(CF_{crop}+S_{crop})Y_{crop}}{(1-\theta_t)\rho d}$	CF_{crop} - concentration factor from root uptake for crop, Bq kg^{-1} (Fresh weight of crop) / Bq kg^{-1} (dry weight soil) S_{crop} - soil contamination on the crop, kg (dry weight soil) kg^{-1} (fresh weight of crop) Y_{crop} - wet biomass of the crop, kg y^{-1}, obtained at harvest from the unit area irrigated

Table 2-5: Transfer between compartments, and between habitats, in the BIOMASS 'natural groundwater source' scenario

Transfer	Equation	Parameter definitions
Groundwater source term to sub-soil compartment, W_i	G_iC_g	G_i - volume of groundwater source term to the sub-soil of area i, m^3 y^{-1} C_g - radionuclide concentration in the groundwater, Bq m^{-3}
Capillary rise of soil water from the sub-soil to surface soil, λ_{Ui}	$\frac{U_i}{R_{ia}\theta_{ia}V_{ia}}$ where $R = 1 + \frac{(1-\theta_{tia})\rho_{ia}}{\theta_{ia}} K_{dia}$ $V_{ia} = Area_i d_{ia}$	U_i - volume of capillary rise from sub-soil to the surface soil compartment of area i, m^3 y^{-1} R_{ia} - retardation coefficient for the sub-soil compartment of area i θ_{ia} -water filled porosity of the sub-soil compartment of area i V_{ia} - volume of the sub-soil compartment of area i, m^3 θ_{tia} - total porosity of the sub-soil compartment of area i ρ_{ia}- grain density of the sub-soil compartment of area i, kg m^{-3} K_{dia} - sorption coefficient of the sub-soil compartment of area i, m^3 kg^{-1} $Area_i$ - area of sub-soil compartment of area i, m^2 d_{ia} - thickness of the sub-soil compartment of area i, m
Recharge of soil water to the sub-soil from the surface soil, λ_{Di}	$\frac{D_i}{R_{ib}\theta_{ib}V_{ib}}$	D_i - volume of recharge from the surface soil compartment of area i, m^3 y^{-1} R_{ib} - retardation coefficient for the surface soil compartment of area i θ_{ib} -water filled porosity of the surface soil compartment of area i V_{ib} - volume of the surface soil compartment of area i, m^3
Loss of surface soil due to erosion, λ_{Ei}	E_i/V_{ib}	E_i - transfer of surface soil from area i to another habitat, m^3 y^{-1}

Table 2-5 continued: Transfer between compartments, and between habitats, in the BIOMASS 'natural groundwater source' scenario

Transfer	Equation	Parameter definitions
Flow from sub-soil of area i to the sub-soil of another area, k, λ_{Aik}	$\frac{A_{ik}^{S}}{R_{ia}\theta_{ia}V_{ia}}$	A_{ik}^{S} - volume of sub-horizontal flow from the sub-soil compartment of area i to the sub-soil compartment of area k during the summer, $m^3\ y^{-1}$
Flow from sub-soil of area i to the surface soil of area k, λ_{Aik}	$\frac{A_{ij}^{W}}{R_{ia}\theta_{ia}V_{ia}}$	A_{ik}^{W} - volume of sub-horizontal flow from the sub-soil of compartment of area i to the surface soil compartment of area k during the winter, $m^3\ y^{-1}$
Flow from surface soil of area i to the surface soil of area, k, λ_{Aik}	$\frac{I_{ik}}{R_{ib}\theta_{ib}V_{ib}}$	I_{ik} - volume of sub-horizontal flow from the surface soil compartment of area i to the surface soil compartment of area k, $m^3\ y^{-1}$
Loss of detritus from surface soil of area i to the surface soil of another area, k, λ_{Pik}	$\frac{P_{ik}\left(CF_{crop}+Soil_{crop}\right)Y_{crop}Area_{i}}{(1-\theta_{tib})\rho_{ib}V_{ib}}$	P_{ik} - fraction of primary productivity in area I lost as detritus, $Soil_{crop}$ - soil contamination on the crop, kg (dw soil) kg^{-1} (fw crop) θ_{tib} - total porosity of the surface soil compartment of area i ρ_{ib}- grain density of the surface soil compartment of area i, kg m^{-3}

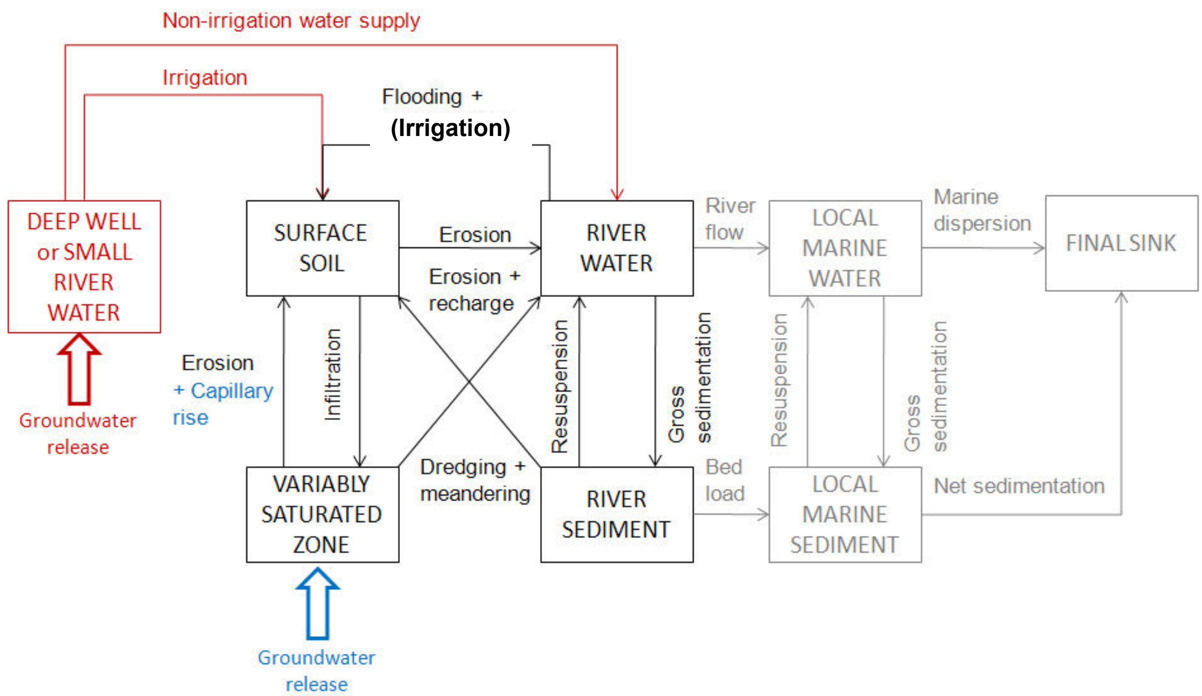
2.1.3 APPROACH ADOPTED BY JGC

Introduction

This model was developed to consider a biosphere receptor as part of a performance assessment for the potential geological disposal of high level radioactive waste (HLW) in Japan (JNC, 1999; Lawson & Smith, 1985). The model was designed to be as generic as possible, considering different combinations of location (inland, coastal, island), topography (mountainous, hilly and plain) and geology (crystalline, sedimentary). A variety of potential receptors were considered: soil, surface freshwater, well and marine (Little 2002; Little et al., 2002 a, b, c and d). These receptors were chosen to be indicative of the potential receptors that could be associated with the various properties of any potential geological repository.

The model assumes two soil compartments, with transfers in and out as illustrated below (Figure 2-4). The grey boxes represent processes and compartments included in the model which are not relevant for the scenarios under consideration. In addition, where compartments or transfers are considered only for one of the specific source terms they are highlighted appropriately. Specifically, red is used to highlight processes included in the irrigation scenario only and blue is used to highlight processes in the scenario of the source coming from depth.

Figure 2-4: Schematic of the JGC model (modified from JNC 1999)



Inputs and losses of contaminants to the system

Two possible pathways of the contaminant entering the system are considered: a groundwater release entering a well or small river whose water is used for irrigation, and a groundwater release where the contaminant enters the surface soil environment through capillary rise and erosion.

The contaminant is lost from the compartments of the system of interest to this study (black boxes in Figure 2-4) in two ways: via river flow and bed load.

$$\text{River flow} = \frac{Q_w}{V_w} \tag{3}$$

$$\text{Bed load} = \frac{K_{drsed} B_l}{\left(\theta_{rsedw} + K_{drsed}\left(1-\theta_{rsed}\right)\rho_{grsed}\right)V_{rsed}} \tag{4}$$

where:

Q_w is the discharge rate of water from the water compartment to the downstream water compartment, $m^3\ y^{-1}$,

V_w is the volume of the water compartment from which the water is discharged, m^3,

K_{drsed} is the sorption coefficient of the river bed sediment compartment, $m^3\ kg^{-1}$,

B_l is the bed load of the river, $kg\ y^{-1}$,

θ_{rsedw} is the water filled porosity of the river bed sediment compartment,

θ_{rsed} is the total porosity of the river bed sediment compartment,

ρ_{grsed} is the grain density of the river bed sediment compartment, $kg\ m^{-3}$,

V_{rsed} is the volume of the river bed sediment compartment, m^3

With respect to the crop, it is assumed that contamination can be lost via food preparation (F_{crop}) and weathering ($e^{-TW_{crop}}$). However, the timeframe under consideration in this study is up to time of harvest without consideration of food preparation, meaning that F_{crop} can be set to zero.

Transfers of contaminant within the system and plant uptake

The algebraic form used for each dynamic transfer process in the conceptual model is shown in Table 2-6.

Table 2-6: Functions used for the transfers of interest in the JGC model

Transfer	Function	Parameter definitions
Non-irrigation water supply	V_{nirr}/V_{dw}	V_{nirr} - annual volume of non-irrigation water abstracted from well water, m^3 y^{-1} V_{dw} - volume of the well water compartment from which non-irrigation water is taken, m^3
Irrigation	V_{irr}/V_{dw}	V_{irr} - irrigation rate applied to the surface soil compartment, m^3 y^{-1}
Infiltration and recharge	$\frac{d_d}{R_{sed}\theta_{sedw}d_{sed}}$ where R_{sed} is given by $1+\frac{(1-\theta_{sed})\rho_{gsed}}{\theta_{sedw}}K_{dsed}$	d_d - annual infiltration/recharge rate, m y^{-1} R_{sed} - retardation coefficient for the compartment from which infiltration/recharge is coming θ_{sedw} - water filled porosity of the compartment from which infiltration/recharge is coming d_{sed} - depth of the compartment from which infiltration/recharge is coming, m θ_{sed} - total porosity of the sediment or soil compartment, ρ_{gsed} - grain density of the sediment or soil compartment, kg m^{-3}, K_{dsed} - sorption coefficient of the sediment or soil compartment, m^3 kg^{-1}
Flooding and sedimentation	V_{fw}/V_{rw}	V_{fw} - annual volume of flood water which infiltrates the surface soil compartment, m^3 y^{-1} V_{rw} - volume of the river water compartment from which flooding occurs, m^3
Capillary rise	$\frac{d_{cap}}{R_{vsed}\theta_{vsedw}d_{vsed}}$	d_{cap} - annual depth of capillary rise from the variably saturated zone compartment, m y^{-1} [a] R_{vsed} - retardation coefficient for the variably saturated zone compartment θ_{vsedw} - water filled porosity of the variably saturated zone compartment d_{vsed} - depth of the variably saturated zone compartment, m

[a] Note that d_{cap} represents the annual depth of capillary rise, it does not represent the velocity of capillary rise.

Table 2-6 continued: Functions used for the transfers of interest in the JGC model

Transfer	Function	Parameter definitions
Erosion [a]	E_{sed} / d_{sed}	E_{sed} - erosion rate for the sediment or soil compartment, m y^{-1} d_{sed} - depth of sediment or soil compartment, m
Dredging and meandering	$\frac{V_{dm}}{V_{rsed}}$	V_{dm} - volume of the sediment deposited on the surface soil compartment due to dredging/meandering, m^3 y^{-1} V_{rsed} - volume of the river bed sediment compartment, m^3
Re-suspension [b]	$\frac{r_{sed}}{d_{rsed}}$	r_{rsed} - rate of re-suspension from the river bed sediment compartment, m y^{-1} d_{rsed} - depth of the river bed sediment compartment, m
Gross sedimentation	$\frac{K_{dsed} S_g (1-\theta_{sed}) \rho_{gsed} A_w}{(1+K_{dsed}\alpha_w) V_w}$	K_{dsed} - sorption coefficient of the suspended sediment in the water compartment, m^3 kg^{-1} S_g - gross sedimentation rate from the water compartment to the associated bed sediment compartment, m y^{-1} θ_{sed} - total porosity of the bed sediment compartment A_w - area of the water compartment, m^2 ρ_{gsed} - grain density of the bed sediment compartment, kg m^{-3} α_w - sediment load in the water compartment, kg m^{-3} V_w - volume of the water compartment, m^3

[a] The same function is used to describe the transfers due to erosion from the surface soil compartment to the river water compartment; from the variably saturated zone compartment to the surface soil compartment; and from the variably saturated zone compartment to the river water compartment.

[b] A different function is used to describe the transfer due to the re-suspension of a marine bed.

In addition to the transfers described in Table 2-6, of particular interest is the radionuclide concentration in the crop, C_{crop} (Bq kg^{-1}, fresh weight of crop), which is given by

$$C_{crop} = \frac{\left(CF_{crop} + \left(1 - F_{crop}\right)S_{crop}\right)C_{ss}}{\left(1-\theta_{ss}\right)\rho_{gss}} + \mu_{crop} d_{icrop} C_{rw} \frac{\left(1-F_{trans}\right)\left(1-F_{crop}\right)e^{-TW_{crop}} + F_{trans}}{Y_{crop}} \qquad (5)$$

where:

CF_{crop} is the concentration factor for the crop, Bq kg^{-1} (fresh weight of crop)/Bq kg^{-1} (dry weight of soil),
F_{crop} is the fraction of external contamination on the crop lost due to food processing,
S_{crop} is the soil contamination on the crop, kg (dry weight soil) kg^{-1} (fresh weight of crop),
C_{ss} is the radionuclide concentration in the surface soil compartment, Bq m^{-3},
θ_{ss} is the total porosity of the surface soil compartment,
ρ_{gss} is the grain density of the surface soil compartment, kg m^{-3},
μ_{crop} is the interception fraction for irrigation water on the crop,
d_{icrop} is the depth of irrigation water applied to the crop, m y^{-1},
C_{rw} is the radionuclide concentration in the river water compartment, Bq m^{-3},
F_{trans} is the fraction of activity transferred from external to internal plant surfaces (translocation fraction),
Y_{crop} is the yield of the crop, kg m^{-2},
W_{crop} is the removal rate of irrigation water from the crop by weathering processes (weathering rate), y^{-1},
T is the interval between irrigation and harvest, y.

The first term of this equation represents crop contamination due to both internal uptake from the surface soil compartment via the roots and external contamination from the deposition of re-suspended sediment from the surface soil compartment. The second term corresponds to plant uptake via irrigation. However, in this study, we are not interested in assessing the concentration in crops due to irrigation, so μ_{crop} is set to zero, i.e. the second term is disregarded.

2.1.4 APPROACH ADOPTED BY ANDRA

Introduction

The transfer factor (TR) approach looks explicitly at the dynamic behaviour of water and Cl-36 in the soil column using the water budget to estimate losses and the solid/liquid distribution coefficient (K_d) for retention. Transfer to plants is modelled on the basis of empirical transfer factors.

The transfer approach has classically been used for radionuclides of radioecological significance. It treats the soil as a dynamic system considering accumulation, losses and an equilibrium value based on the individual radionuclide behaviour. Transfers from the soil to all other compartments, including plants are modelled using empirical transfer coefficients. The extent of the dynamic part of radionuclide accumulation in soil depends on the system losses (the higher the losses the more quickly equilibrium is achieved). In case of a significant dynamic increase in the soil all associated compartments, up to the final dose received by man, will also show these dynamics.

The equations and associated databases are grouped in the model called *Aquabios* (Albrecht, 2007) which is managed by the Andra model management code MoM.

Inputs and losses of contaminants to the system

Irrigation by contaminated well water is the only modelled source of contamination (Figure 2-5). Irrigation depends on the water budget of the soils. Local information of monthly precipitation (T_{pluie}) and evapotranspiration (ETP) are used to assess the yearly need of irrigation ($T_{eaucult}$). This procedure is illustrated based on meteorological data from the inland and coastal sites used in this study (Météo France, 2008). Precipitation is directly measured, evapotranspiration either measured or modelled; the relationship between both can be constructed on the basis of meteorological data (5 years in the case of this study). Once constructed for a specific site (Figure 2-6) this relationship can be used to quantify irrigation on the basis of any available data for precipitation (i.e. during a probabilistic model approach).

Figure 2-5: Schematic of Aquabios model

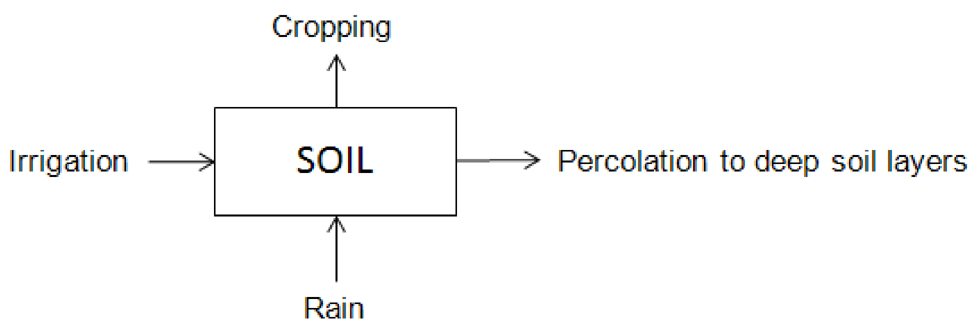
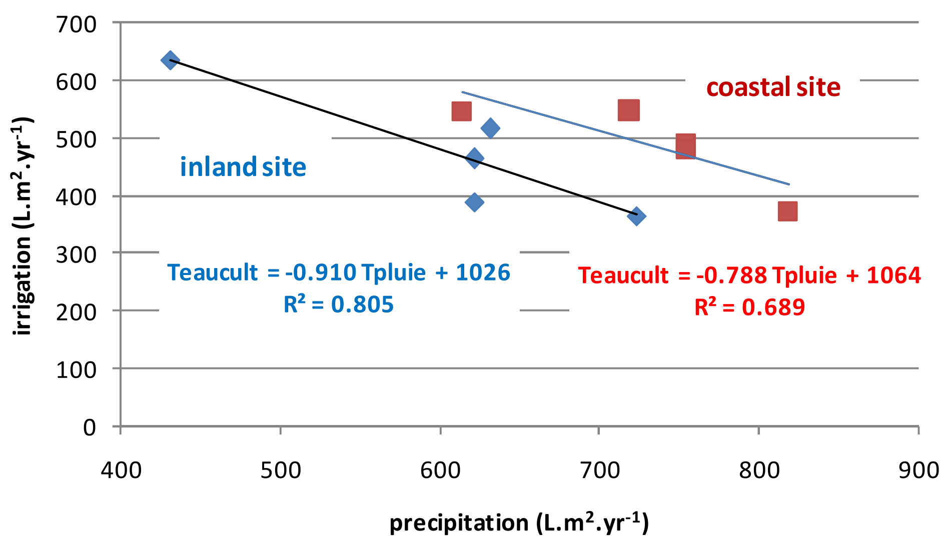


Figure 2-6: Correlation between yearly precipitation and irrigation for the coastal and inland sites based on monthly data for five years



Fluxes of Cl-36 (Q, Bq m^{-2} y^{-1}) are modelled using yearly averaged irrigation ($T_{eaucult}$, L m^{-2} y^{-1}) and the concentration in the irrigation or well water ($C_{eaucult}$, Bq L^{-1}):

$$Q = T_{eaucult} \cdot C_{eaucult} \tag{6}$$

Under humid conditions, particularly during the colder season, where precipitation exceeds evapotranspiration, yearly excess water in the system exits via percolation. This excess water is set equal to the hydraulic budget and calculated using the following equation:

$$Bihy_{cult} = T_{pluie} + T_{eaucult} - ETP \tag{7}$$

This percolating water has the ability to transport dissolved radionuclides into deeper sections of the soil, where they become unavailable for root uptake. Related losses (λ_{lix}, y^{-1}) can be estimated using the hydraulic budget, soil physical data (soil water content, *tensolceau* (-); soil porosity, *poro_cult* (-); depth of soil homogenisation and root growth, *profcult*, (m) and the retention factor (R_s, -)

$$\lambda_{lix} = \frac{Bihy_{cult}}{tensolceau \cdot poro\_cult \cdot Prof_{cult} \cdot R_s} \tag{8}$$

The latter is defined by

$$R_s = 1 + \frac{Kdsol \cdot (1 - poro\_cult) \cdot Rho_{part}}{tensolceau \cdot poro\_cult} \tag{9}$$

where *Kdsol* (kg/L) stands for the solid/solution distribution coefficient and Rho_{part} for the soil density (kg m^{-3}).

An additional loss term is related to the export of Cl-36 at harvest (λ_{exp}). It can be modelled based on the yield of a blend of cultivated plants (*Rendcult*, kg$_{fresh}$ m^{-2} y^{-1}), the soil to plant transfer coefficient for this mix of plants (*FTsolplant*, kg$_{dry}$/kg$_{fresh}$) and the soil physical parameters depth of homogenisation ($Prof_{cult}$) and soil density (*Rho_cult*, in kg/m^3):

$$\lambda_{\exp} = \frac{Rendcult + FTsolplant}{Rho\_cult \cdot Prof_{cult}} \tag{10}$$

Transfers of contaminant within the system and uptake by plants

The evolution of Cl-36 concentration in the soil can be modelled using a mass balance approach:

$$\frac{dC_{soil}}{dt} = \frac{Q}{Rho\_cult \cdot Prof_{cult}} - (\lambda_{lix} + \lambda_{\exp} + \lambda_R)C_{soil} \tag{11}$$

This differential equation has an analytical solution, which is integrated into Aquabios:

$$C_{sol}(t_j) = \frac{\frac{Q}{Rho\_cult \cdot Prof_{cult}}}{\lambda_{lix} + \lambda_{\exp} + \lambda_R} + \left[\left(C_{sol}(t_i) - \frac{\frac{Q}{Rho\_cult \cdot Prof_{cult}}}{\lambda_{lix} + \lambda_{\exp} + \lambda_R} \right) e^{-(\lambda_{lix} + \lambda_{\exp} + \lambda_R)(t_j - t_i)}\right] \tag{12}$$

For radionuclides, such as Cl-36 with a high mobility (low K_d, high λ_{lix}) and significant plant uptake (λ_{exp}), the exponential part of the equation vanishes and the soil concentration quickly reaches equilibrium. Losses related to radioactive decay (λ_R) are insignificant.

To obtain the concentration of Cl-36 in fresh plant tissue, a simple transfer approach is applied, which is based on empirically determined transfer factors (*FTsolplant*). An example of the transfer of Cl-36 from soil to cereals is given for illustration:

$$Ccer\_sol = Csolcult \cdot FTsolcer \tag{13}$$

Interception of Cl-36 and transfer from leaves to the edible portion is considered in Aquabios, but has been turned off for this study. This has little impact on the results for cereals, as root uptake clearly dominates cereal activity. However, for root and leafy green vegetables, as much as a third of plant uptake of Cl-36 may be attributable to interception.

2.1.5 APPROACH ADOPTED BY SCM

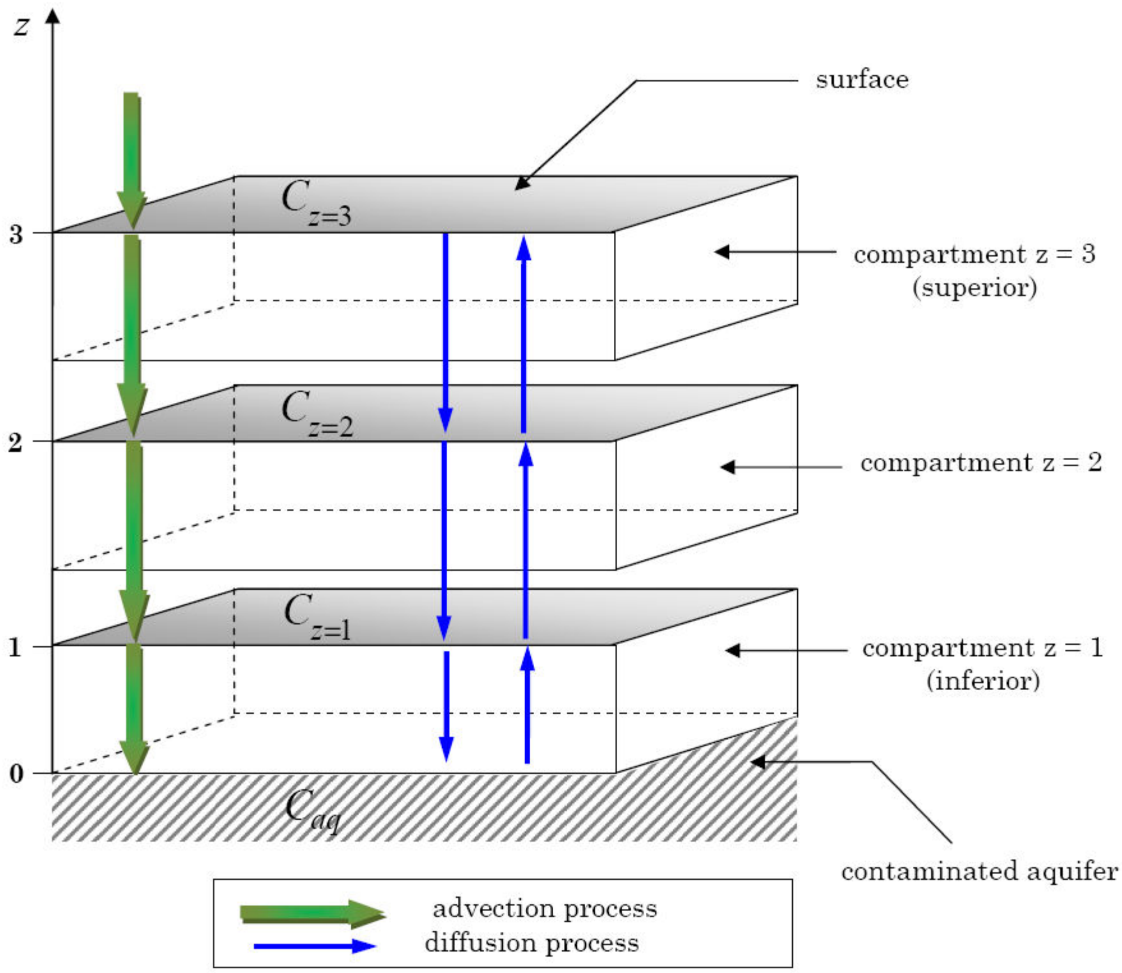
Introduction

The aim of the SCM – ANDRA Multicompartment Model (SAMM; Miquel, 2008) is to simulate the propagation of radionuclides in soils of the biosphere. This model, integrating the processes of advection, diffusion and radioactive decay, is a synthesis of the two models available at ANDRA (Albrecht, 2007 and Klos, 2007). Contamination can take place via irrigation from the surface of the soil and advectively moving to the aquifer or to move upwards via diffusion from the contaminated aquifer to the surface.

The soil is described by a vertical column consisting of a freely chosen number of compartments. The concentration of Cl-36 can be computed for each compartment including the rooted top compartment.

To illustrate transfer processes and mathematical treatment, let's take an example with three compartments. For radionuclides, such as Cl-36 with a high mobility and long half-life period, the influence of radioactive decay is insignificant. The diagram of the discretised vertical soil column, with advection and diffusion processes indicated, is shown in Figure 2-7.

Figure 2-7: SAMM: Discretised vertical soil column with three compartments



The SAMM software computes the concentration in Bq/kg of Cl-36, and the concentration of stable chlorine in g/kg, (for the compartment z at the time t) resolving numerically the Advection, Diffusion and radioactive Decay differential Equation (ADDE) which is given by the formula:

$$\frac{\partial C(z,t)}{\partial t} = \left(v(z,t)\frac{\partial C(z,t)}{\partial z} + C(z,t)\frac{\partial v(z,t)}{\partial z} \right) + \left(D_a(z,t)\frac{\partial^2 C(z,t)}{\partial z^2} + \frac{\partial D_a(z,t)}{\partial z}\frac{\partial C(z,t)}{\partial z} \right) + \left(-\lambda C(z,t) + \lambda_p C_p(z,t) \right) \quad (14)$$

where:

- $C(z,t)$ is the concentration in the soil for the position z at the time t
- $v(z,t)$ is the speed of propagation of radionuclides in the soil for the position z at the time t
- $D_a(z,t)$ is the coefficient of apparent diffusion for the position z at the time t
- λ is the constant of radioactive decay
- $C_p(z,t)$ is the concentration in the soil of an eventual radionuclide parent
- λ_p is the constant of radioactive decay of an eventual radionuclide parent

The three bracketed terms correspond to advection, diffusion and decay respectively. The speed of propagation of radionuclides in the soil (v) and the coefficient of apparent diffusion (D_a) depend on various soil factors:

$$v = \frac{d_v}{\left(t_e\,\omega + (1-\omega)\,\rho_p\,Kd_{sol}\right)S} \quad (15)$$

$$D_a = \frac{D_0}{\left(t_e\,\omega + (1-\omega)\,\rho_p\,Kd_{sol}\right)\left(1 + \mu_s\,\frac{(1-\omega)}{\omega}\right)} \quad (16)$$

where:

- t_e is the water content in the soil (-)
- ω is the soil porosity (-)
- ρ_p is the soil density (kg m^{-3})
- Kd_{sol} is the solid / solution distribution coefficient (m^3 kg^{-1})
- μ_S is the tortuosity parameter (-)
- S is the section of the soil column (m^2)
- d_v is the water volumetric debit in the soil column (m^3 y^{-1})
- D_0 is the coefficient of diffusion in water (m^2 y^{-1})

The step-width used for temporal discretisation used to represent time can be automatically or freely chosen, thus daily, monthly or yearly calculations are possible.

SAMM computes the concentration of Cl-36 in a non-cultivated soil. The loss term related to the export of Cl-36 at harvest isn't taken into account. Thus, if accumulation of the radionuclide in the soil is possible, then the calculated concentration of the contaminant in the soil is higher in SAMM than in model which considers a cultivated soil (e.g. Aquabios).

Contaminant uptake by the plant

The transfer coefficient (TR) approach used for example by Andra in the *Aquabios* model uses the empirically determined transfer factors (FT_{sol_cer}) and is also applied to SAMM. The model which uses this transfer factor approach is known as SAMM-TR. For example, the concentration in Bq/kg_{fresh} of Cl-36 in cereals grown in the surface compartment (z = Z) can be calculated for any given time t using the following equation:

$$C_{cer}(Z,t) = C_{sol}(Z,t) \times FT_{sol\_cer} \tag{17}$$

2.2 SPECIFIC ACTIVITY MODELS

Specific activity models assume that, for each compartment of the environment, the ratio between Cl-36 and stable chlorine is the same as those of the other compartments. For example, for plants, the following relationship can be written:

$$\frac{^{36}\mathrm{Cl}_{\mathrm{plant}}}{\mathrm{stable\_Cl}_{\mathrm{plant}}} = \frac{^{36}\mathrm{Cl}_{\mathrm{env\_plant}}}{\mathrm{stable\_Cl}_{\mathrm{env\_plant}}} \tag{18}$$

where $^{36}Cl_{plant}$ and $stable\_Cl_{plant}$ are the concentrations of Cl-36 and stable chlorine in the plant respectively (in $Bq.kg^{-1}$ and $mg.l^{-1}$); $^{36}Cl_{env\_plant}$ and $stable\_Cl_{env\_plant}$ are the concentrations of ^{36}Cl and stable chlorine in the environment of the plant respectively (in $Bq.kg^{-1}$ and $mg.l^{-1}$). Thus, in order to calculate the concentration of Cl-36 in the plant, the plant environment needs to be defined first. Typically this is the topsoil.

2.2.1 APPROACH ADOPTED BY ANDRA

Introduction

Originally developed for carbon-14 in the Canadian waste disposal safety calculations (Davis et al., 1993), the IR approach was adapted for chlorine-36 by Sheppard (2001) and applied for the ANDRA high level waste Dossier 2005 (Andra, 2005) and the ANDRA graphite (FAVL, low activity long lived) waste assessment (Leclerc-Cessac, 2004). The principal hypothesis of the IR approach is that stable and radioactive chlorine rapidly achieve equilibrium in all connected compartments and that the specific Cl-36 / stable Cl (Cl-s) ratio can be considered identical. As stable chlorine is readily measurable in all compartments, it is sufficient to know the ratio of Cl-36 over Cl-s in the upstream compartment to calculate the unknown Cl-36 activity in the downstream compartment.

The equations and associated databases are grouped in the model called *AquaCl36* which is managed by the ANDRA model management code MoM.

Inputs and losses of contaminants to the system

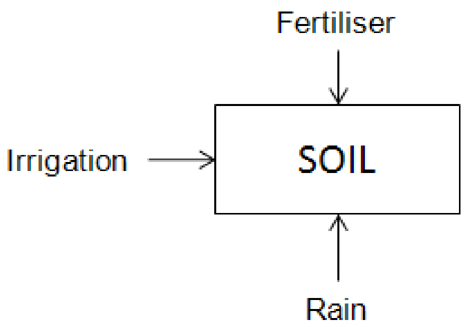
As with ANDRA's conventional model, Aquabios, irrigation by contaminated well water is the main modelled source of contamination. The inputs of water to the soil are determined identically in Aquabios and AquaCl36.

Different types of soils can be considered by Andra's models; the one relevant for this current study is the cultivated soil (*solcult*). The activity of Cl-36 (in Bq/kg_{dry}) and the concentration of stable chlorine (mg/kg_{dry}) can be estimated based on fluxes into the soil and the concentrations of stable and radioactive chlorine. This allows computation of the isotopic ratio (RI in Bq/mg):

$$RI_{solcult} = \frac{T_{eaucult} \cdot C_{eaucult}}{T_{eaucult} \cdot C_{eaucult\_S} + T_{pluie} \cdot C_{pluie\_S} + T_{engrais\_cult} \cdot Engrais\_s} \qquad (19)$$

The fluxes of water are given in L m^{-2} y^{-1} (T) with *eaucult* indicating irrigation and *pluie* precipitation (Figure 2-8); activities of Cl-36 ($C_{eaucult}$) are in Bq/L and concentrations of stable chlorine ($C_{eaucult\_S}$, C_{pluie_S}) in mg/L. Fertiliser addition is given in kg m^{-2} y^{-1} ($T_{engrais\_cult}$), with stable chlorine concentration (*Engrais_s*) in g/kg.

Figure 2-8: Schematic of AquaCl36



Contaminant uptake by the plant

Isotopic ratios in plants are assumed to be identical to the ratios in the soil:

$$\left(\frac{Cl-36}{Cl-s}\right)_{plant} = \left(\frac{Cl-36}{Cl-s}\right)_{soil} \qquad (20)$$

As the stable chlorine content of the plant (*Cl-s*, in mg/kg_{fresh}) can be measured, the Cl-36 in the plant in Bq/kg_{fresh} can be easily calculated:

$$\left(Cl-36\right)_{plant} = \left(\frac{Cl-36}{Cl-s}\right)_{soil} \left(Cl-s\right)_{plant} \tag{21}$$

In *AquaCl36* three equations are defined for the three types of plants considered:

$$\begin{aligned} C_{cer} &= RI_{solcult} \cdot Cl\_s\_cer \\ C_{rac} &= RI_{solcult} \cdot Cl\_s\_rac \\ C_{feuil} &= RI_{solcult} \cdot Cl\_s\_feuil \end{aligned} \tag{22}$$

For each plant (*Cl_s_...* in mg/kg_{fresh}) stable chlorine information is required with *cer* standing for cereals, *rac* for root vegetables and *feuil* for leafy vegetable. *C…* indicates the activity of Cl-36 (in Bq/kg_{fresh}) in each plant.

2.2.2 APPROACH ADOPTED BY NWMO

The NWMO modelling approach was developed by Zach et al. (1996), and is described as follows from Gierszewski et al. (2004).

A total internal dose for Cl-36 is first estimated by the internal dose pathway equations. These equations account for internal dose from ingestion of various food types and their associated pathways as well as internal dose from ingestion of drinking water, soil and from inhalation. This estimated dose is compared with the maximum possible internal dose, which is set by assuming all chlorine in soft tissues is at the same ratio of Cl-36 to stable chlorine as in the background groundwater. As such, the NWMO model is the simplest, most conservative of the models considered in this study. The minimum of the two doses is taken to be the internal dose:

$$Dit^{Cl36}(\text{pathways model}) = \sum_{k,i} De^{Cl36,k,i} + Dw^{Cl36} + Ds^{Cl36} + Di^{Cl36} \tag{23}$$

and

$$Dit^{Cl36} = \min\begin{pmatrix} Dit^{Cl36}(\text{pathways model}) \\ DF^{Cl36} \cdot \left[Ca_{gw}^{Cl36} \big/ \left(C_{gw}^{Cl} + Ca_{gw}^{Cl36} \cdot gb^{Cl36}\right)\right] \cdot B_{Cl} / B_m \end{pmatrix} \tag{24}$$

where

Dit^{Cl36} is the human total internal dose from Cl-36 (Sv y^{-1})

$De^{Cl36,k,i}$ is the internal dose from Cl-36 due to ingestion of food type j along pathway k (Sv y^{-1})

Dw^{Cl36} is the internal dose from Cl-36 due to ingestion of drinking water (Sv y^{-1})

Ds^{Cl36} is the internal dose from Cl-36 due to ingestion of soil (Sv y^{-1})

Di^{Cl36} is the internal dose from Cl-36 due to inhalation (Sv y^{-1})

DF^{Cl36} is the internal dose conversion factor for Cl-36 [(Sv y^{-1})/(Bq kg^{-1})]

gb^{Cl36} is the mass/radioactivity conversion factor for nuclide i (kg Bq^{-1})

Ca_{gw}^{Cl36} is the Cl-36 activity in the groundwater (Bq m^{-3} $_{water}$)

C_{gw}^{Cl} is the stable chlorine concentration in the groundwater (kg m^{-3} $_{water}$)

B_{Cl} is the total chlorine content of soft tissues in a human (kg)

B_m is the total soft tissue mass in a human (kg).

Since this model does not consider directly the contamination of a food source (e.g. plants) via uptake from soil, this model has not been used further in this study. However, the description and approach illustrates how one can assess doses directly from the concentrations of Cl-36 in water.

2.3 COMPARTMENTAL MODELS WITH AN ISOTOPE RATIO APPROACH TO PLANT UPTAKE OF CONTAMINANT

2.3.1 APPROACH ADOPTED BY SCM

Introduction

The model setup used is almost identical to the one used for the conventional model, considering either irrigation water or groundwater as a contamination source (see Section 2.1.5). The difference in the approach taken for the specific activity model, as compared to the conventional model, is the way in which contaminant uptake by the plant is modelled. SAMM models the Cl-36 and the stable chlorine concentration based on known fluxes and concentrations in the rain and irrigation water and in the fertiliser added to the soil. Both Cl forms are treated identically, with the exception that stable chlorine does not decay. Equations are identical to those used in Aquabios (see Section 2.1.4), with the exception, that the differential equation is solved analytically and that an extra term is used to consider stable Cl input via fertiliser addition. The modelled concentrations of stable and radioactive chlorine are then used to estimate plant uptake using the isotope ration approach. This model is known as SAMM-IR (IR stands for isotope ratio).

Contaminant uptake by the plant

SAMM-IR can be used to compute the concentration of any stable element, including stable chlorine in the non-cultivated soil (in g/kg). The stable chlorine originates from stable chorine present in irrigation and rainfall water and in fertilizers.

Isotopic ratios between Cl-36 and stable chlorine are assumed to be identical in all connected environmental compartments. With knowledge of the concentration of stable chlorine in plants ($C_{plant}(Cl_{stable})$ in g/kg), the concentration of Cl-36 in plants ($C_{plant}(Cl\text{-}36)$ in Bq/kg) can be obtained :

$$C_{plant}\left(Cl-36\right)=\left(\frac{Cl_{soil}\left(Cl-36\right)}{Cl_{soil}\left(Cl_{stable}\right)}\right)C_{plant}\left(Cl_{stable}\right) \tag{25}$$

2.3.2 APPROACH ADOPTED BY EDF

Introduction

The approach taken by EDF was to define the environment of the plant and then model chlorine (stable and Cl-36) dynamics in that environment, so that the Cl-36 concentration in the plant would follow automatically. Since Cl-36 is a mobile element, it may be considered that transfer to plants occurs through the uptake of the contaminant as a consequence of transpiration of water taken up from the rooting zone of the soil. Thus, it may be written:

$$\frac{{}^{36}Cl_{plant}}{stable\_Cl_{plant}}=\frac{{}^{36}Cl_{soil\_water}}{stable\_Cl_{soil\_water}} \tag{26}$$

where ${}^{36}Cl_{soil\_water}$ and $stable\_Cl_{soil\_water}$ are the concentrations of Cl-36 and stable chlorine in water that is taken up from the soil by plants for supplying their growth needs respectively (in Bq kg^{-1} and mg l^{-1} respectively). According to Equation 26, the general structure of the model that is described below aims at estimating the Cl-36 and stable chlorine concentrations in water that is taken up by plants for transpiration.

In the EDF model, the soil compartment is split into four layers. Certain processes for the movement of water (and associated Cl-36 and stable chlorine) are independent of the scenario being considered:

- the topsoil directly receives water inputs from rainfall
- water (and associated Cl-36 and stable chlorine) can be transferred to subsoil layers (downward direction) by percolation if the water content of a given soil layer is higher than some critical level (the field capacity)
- evaporation (loss from topsoil directly to the atmosphere), that is calculated at each month according to atmospheric temperature and solar radiation
- transpiration (loss from topsoil to the atmosphere via plants).

Building upon these fundamental model assumptions, EDF developed two models, one for each scenario under consideration: irrigation (Figure 2-9) or groundwater (Figure 2-10). The differences are:

- ***Irrigation scenario.*** The topsoil receives water inputs from irrigation. All the water transfers that are calculated by the model finally aim at providing a relevant evaluation of the transpiration fluxes from the different soil layers to the plants.
  For root and leafy green vegetables (respectively potatoes and lettuce), the irrigation amount is constant during a chosen period of the year. However, for maize culture, the irrigation amount is calculated on a monthly time-step (during the cultivation period) in order to ensure that the water content of the soil satisfies the requirements of the plants, i.e. doesn't fall below

some critical level. Thus, for cereal, irrigation occurs only when a deficit in the topsoil water budget is observed (see below for further details).

- ***Groundwater scenario.*** Water can be transferred from subsoil layers (upward direction) by capillarity rise. It is assumed that a water flow from subsoil arises when the water budget of the soil is under the wilting point. This mechanism appears only for climate when evapotranspiration is important compared to the precipitations. This capillarity depends on the type of soil considered. Indeed the mechanism increases with the porosity of the soil.

Figure 2-9: Schematic of the EDF model: irrigation scenario

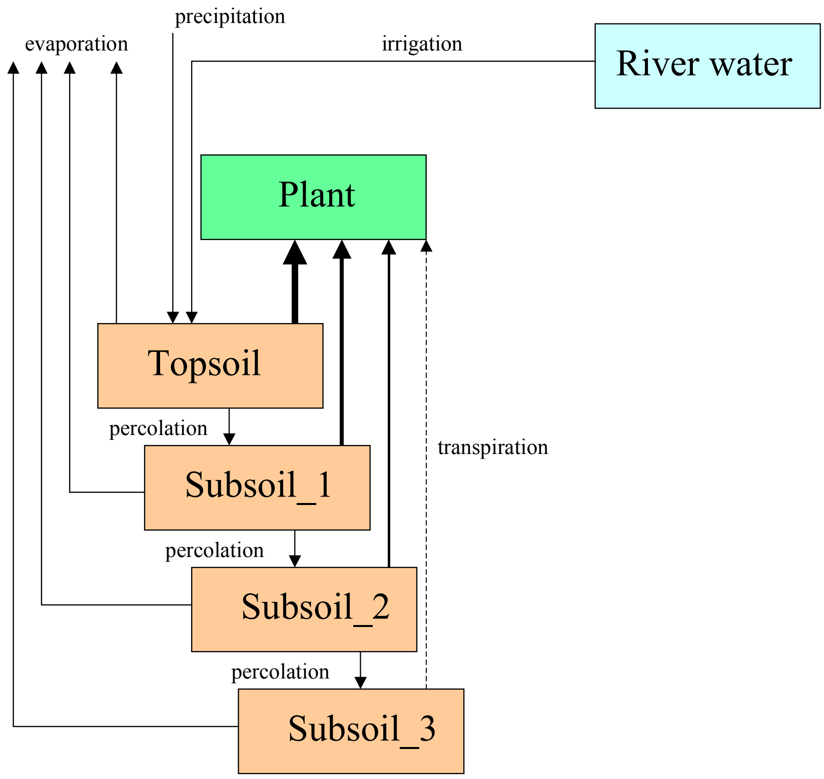
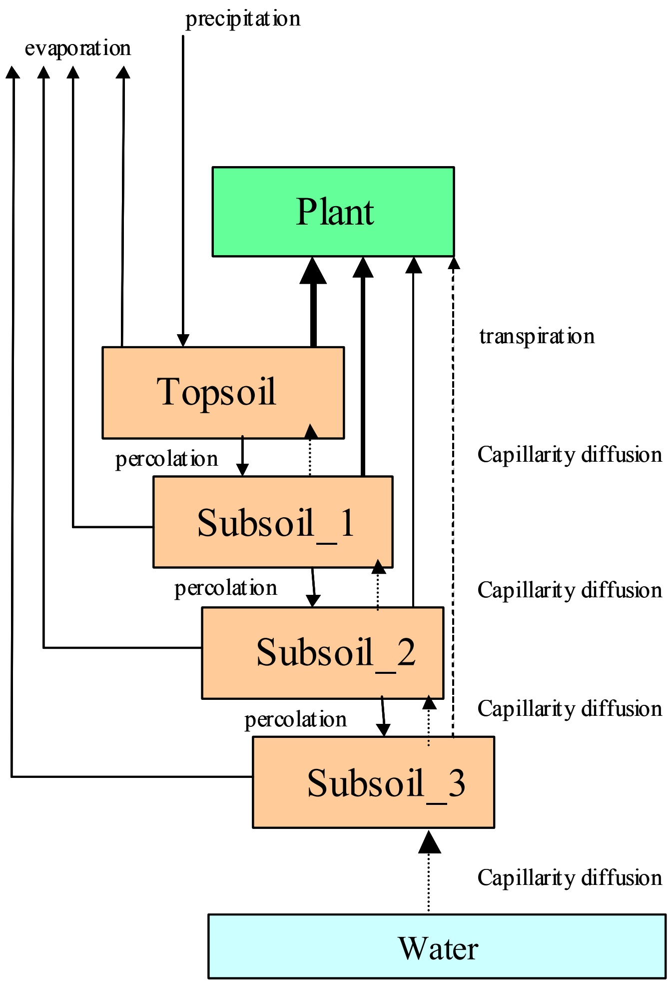


Figure 2-10: Schematic of the EDF model: groundwater scenario



<u>Inputs and outputs of water to the system</u>

First the input and output processes which are common to both scenarios are considered; the equations which describe these processes are given in Table 2-7.

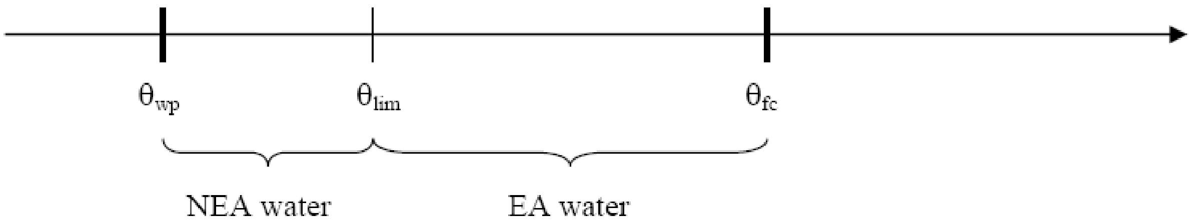
The monthly precipitation data the model uses are site specific (e.g. Météo France 2008). Evapotranspiration is defined as the water flux from soil to the atmosphere resulting from both direct evaporation from topsoil and transpiration by plants. The model calculates both potential evapotranspiration (evapotranspiration if sufficient water were available; Turc 1954, 1961) and actual evapotranspiration (calculated using the FAO model (cited in Maraux et al, 1998)), which accounts for factors that moderate evapotranspiration. These factors are:

- the plant canopy and its stage of development. The actual evapotranspiration may be estimated by taking into account the potential evapotranspiration and a time-dependent cultural coefficient that may be lower or higher to unity according to the stage of development of the plant;

- the water content of the soil. When soil contains enough water, there is no potential resistance for root uptake and evapotranspiration. On the contrary, when soil water content decreases, evapotranspiration is limited because of the strength necessary for taking up water.

Thus, we distinguished two different pools in water contained in topsoil: the ‘Easily available’ (EA) and ‘Not-easily available’ (NEA) soil water. The boundary between EA and NEA soil water is defined by a limiting water content, θ_{lim}, situated between the wilting point water content, θ_{wp}, and the field capacity water content, θ_{fc} (Figure 2-11).

Figure 2-11: Pools of water in the topsoil (EDF model)



The partitioning of actual evaporation between soil evaporation and plant transpiration is governed by the solar radiation that can actually reach the soil and that can be spent as latent energy to evaporate water from soil surface. The fraction of solar radiation which can reach the soil decreases with the increase of leaf area. According to some crop models (e.g. Sau et al, 2004), the plant transpiration can be estimated from actual evapotranspiration as a function of LAI (leaf area index) and an extinction factor. For calculating LAI_n, it is assumed that aerial plant biomass linearly increases with time until harvest (Soltner, 1990).

Table 2-7: EDF model – scenario independent inputs and outputs of water to the system

Process	Equation	Parameters
Potential evapotranspiration	$ET_{p,n} = 0.4\frac{T_n}{T_n + 15}(Ig_n + 50)$ where, if Ig_n isn't given in the site specific data it can be calculated using $Ig_n = Ig_A.\left(0.18 + 0.62\frac{h_n}{H_n}\right)$	$ET_{p,n}$ -potential evapotranspiration at month n, mm.month^{-1} T_n - mean atmospheric temperature at month n, °C Ig_n - global solar radiation at month n, cal.cm^{-2}.j^{-1} Ig_A - maximum radiation (without atmosphere)[a], cal.cm^{-2}.j^{-1} H_n - monthly daylight duration[a], h h_n - monthly sunshine duration, h
Actual evapotranspiration	if $\theta_{lim} < \theta_{topsoil,n} < \theta_{fc}$, then $ET_{a,n} = K_c.ET_{p,n}$ if $\theta_{wp} < \theta_{topsoil,n} < \theta_{lim}$, then $ET_{a,n} = \frac{\theta_{topsoil,n}}{\theta_{lim}} K_c.ET_{p,n}$	K_c - cultural coefficient at time n $\theta_{topsoil,n}$ - water content in topsoil at time n
Transpiration	$Trans_n = ET_{a,n}.[1 - \exp(-\alpha_{extinction}.LAI_n)]$ If $t_{germ} < n < t_{harv}$, $LAI_n = LAI_{harv}.\left(1 - \frac{t_{harv} - t_n}{t_{harv} - t_{germ}}\right)$	$Trans_n$ - plant transpiration at time step n, mm.month^{-1} $\alpha_{extinction}$ - extinction factor, LAI_n: leaf area index at time n (LAI_{harv} is leaf area index at harvest) t_{harv} - harvest date; t_{germ} - germination date

[a] Ig_A and H_n are given according to the latitude in tables available in the literature (Soltner, 2004)

In the irrigation scenario, irrigation is assumed to occur only when no 'easily available' water is present in topsoil and thus when water needs of plants are not satisfied. Consequently, the water content in the topsoil must be determined prior to the irrigation rate. Within this model this is done for each time step.

At the end of time step n, water content in topsoil depends on inputs by rainfall and irrigation and on outputs from evapotranspiration. Soil water content in the topsoil is further constrained by two limits, the wilting point (θ_{wp}) and the field capacity (θ_{fc}), so that at any time n, $\theta_{wp} < \theta_n < \theta_{fc}$. It is assumed that inputs and outputs are uniformly distributed over the overall height of the topsoil layer. According to these assumptions, water content at the end of time step time n can be written as:

$$\theta_{topsoil,n} = \min\left\{\theta_{fc}\,;\max\left[\theta_{wp}\,;\theta_{topsoil,n-1} + \frac{PP_n + Qirr_n - ET_{a,n} - v_{perc,n}}{h_{topsoil}}.\Delta t\right]\right\} \tag{27}$$

where:

- $\theta_{\mathrm{topsoil,n}}$: water content in topsoil at the end of time step time n (m^3 water.m^{-3} soil);
- θ_{fc} : water content at field capacity (m^3 water m^{-3} soil);
- θ_{wp} : water content at wilting point (m^3 eau.m^{-3} soil);
- $\mathrm{PP(t)}$: rainfall at time t (m^3 water m^{-2} soil d^{-1});
- $\mathrm{Qirr(t)}$: irrigation rate at time t (m^3 water m^{-2} soil d^{-1});
- $\mathrm{ET_{a,n}}$: actual evapotranspiration at time step n (m d^{-1}) ;
- $v_{perc,n}$: percolation velocity in topsoil at time step n (m.j^{-1}) ;
- $\mathrm{h_{topsoil}}$: height of the topsoil layer (m) ;
- $\Delta\mathrm{t}$: duration of time step (d).

Obviously, irrigation does not occur when no plant is present on the field. Thus, independently of the water content in topsoil:

$$\mathrm{Qirr}_n\text{=0 if } n < t_{germ} \text{ or } n > t_{harv} \tag{28}$$

where are t_{germ} and t_{harv} are the dates of germination and harvest of the plant of interest.

During the cultivation period, irrigation is governed by two main conditions:

- irrigation occurs only if no 'easily available' water is present in topsoil, i.e. if $\theta_n < \theta_{\lim}$
- as classically conducted in agriculture, each irrigation operation provides a constant and pre-determined quantity of water that must allow water content to exceed water content at field capacity.

As a consequence, irrigation rates are calculated as follows:

If $t_{germ} < n < t_{harv}$:

$$\text{if } \theta_{\text{topsoil,n}} > \theta_{\text{lim}} \text{, then } \text{Qirr}_{\text{n}} = 0 \quad (29)$$

$$\text{if } \theta_{\text{topsoil,n}} < \theta_{\text{lim}} \text{, then } Qirr_n = \frac{Qirr_{event}}{\Delta t}.\left(1 + ENT\left|\frac{h_{topsoil}.(\theta_{fc} - \theta_n)}{Qirr_{event}}\right|\right) \quad (30)$$

where :

- Qirr_{n} : irrigation rate at time step n (m d^{-1}) ;
- $\text{Qirr}_{\text{event}}$: quantity of water distributed to the field for one 'irrigation event' (m).

In the groundwater scenario, water enters the soil via capillary diffusion when the water content of the topsoil is below the wilting point, i.e. $\theta_{wp} > \theta_{topsoil}$. When this condition is satisfied then the water that is acquired from the water flow from subsoil layers can be estimated: water capillarity velocity at time n is given by the following relationship:

$$v_{capill,n} = \frac{\max\left\{0; \left[\theta_{wp} - (\theta_{topsoil} + \frac{PP_n + Qirr_n - ET_{a,n}}{h_{topsoil}}.\Delta t\right].h_{topsoil}\right\}}{\Delta t} \quad (31)$$

Capillarity loss rate at time step n is in each soil layer is given by the following relationship:

$$\lambda_{capill,soil\_layer\_i,n} = \frac{v_{capill,n}}{h_{soil\_layer\_i-1}.\theta_{topsoil,n}} \times \varphi \quad (32)$$

with φ the soil porosity.

<u>Transfers of contaminant within the soil system</u>

For both scenarios, stable chlorine and Cl-36 enters the soil layers via rainfall, while outputs are led by physical percolation processes. Indeed, if water content exceeds maximum water content (i.e. water content at field capacity), some water (and associated chlorine) is lost from one soil layer to the deeper one by percolation. At each time step, the percolation loss rate of each soil layer need to be determined before the chlorine budget of the soil layers. An additional output pathway is that of plant transpiration. This output depends on the transpiration rate (see equation (7)), the retardation coefficient and the relative root density (see equation (15)), which allows to estimate the part of transpiration coming from the soil layer. These equations are given in Table 2-3.

For the individual scenarios, stable chlorine and Cl-36 enter the system via irrigation water or the water entering the soil via capillary diffusion. The amount of chlorine entering via these pathways depends upon the rate of water flow. The budget equations for stable chlorine and Cl-36 in the topsoil are given in Equations 35 and 36.

Note that these equations contain terms for inputs via both irrigation and capillary rise. For the irrigation scenario, $\lambda_{capill\_soil\_layer\_i,n}$ is set equal to zero, and for the groundwater scenario $Qirr$ is set to zero.

The budget equations for stable chlorine and Cl-36 in the deeper soil layers are similar (see Equations 37 and 38). Again, $\lambda_{capill\_soil\_layer\_i,n}$ is set equal to zero for the irrigation scenario.

Uptake of contaminant by the plant

The calculation of the specific activity in plants is calculated as:

$$\frac{{}^{36}\mathrm{Cl}_{\mathrm{plant}}}{\mathrm{stable\_Cl}_{\mathrm{plant}}} = \frac{{}^{36}\mathrm{Cl}_{\mathrm{transpiration\_water}}}{\mathrm{stable\_Cl}_{\mathrm{transpiration\_water}}} \tag{33}$$

Stable chlorine and ^{36}Cl in transpiration water can be estimated from concentrations in the soils layers where water is extracted for transpiration purposes. Then:

$$\frac{{}^{36}\mathrm{Cl}_{\mathrm{plant,n}}}{\mathrm{stable\_Cl}_{\mathrm{plant,n}}} = \sum_{\mathrm{soil\_layers}} \mathrm{RRD}_{\mathrm{soil\_layer\_i}} \cdot \frac{{}^{36}\mathrm{Cl}_{\mathrm{soil\_layer\_i,n}}}{\mathrm{stable\_Cl}_{\mathrm{soil\_layer\_i,n}}} \tag{34}$$

Table 2-8: EDF model: processes driving movement of stable chlorine and Cl-36 through the soil

Process	Equation	Parameters
Percolation velocity of the topsoil	$v_{perc,n} = \dfrac{\max\left\{0;\left[\theta_{topsoil,n-1} + \dfrac{PP_n + Qirr_n - ET_{a,n}}{h_{topsoil}}.\Delta t - \theta_{wp}\right].h_{topsoil}\right\}}{\Delta t}$	$v_{perc,n}$ - water percolation velocity in topsoil at time n, m y^{-1}
Percolation loss rate in soil layers	$\lambda_{perc,soil\_layer\_i,n} = \dfrac{v_{perc,n}}{h_{soil\_layer\_i}.\theta_{topsoil,n}}$	$\lambda_{perc,soil\_layer\_i,n}$ - percolation loss rate at time step n in each soil layer i, y^{-1} $\theta_{topsoil,n}$ - water content in topsoil at the end of time step time n, m^3 water m^{-3} soil $h_{soil\_layer\_i}$ - height of soil layer i, m
Retardation coefficient	$Rs = 1 + \dfrac{(1-\omega)}{\theta\omega}\rho K_d$	ω - soil porosity ρ - soil density K_d - 36Cl
Distribution of roots according to depth	$RRD_{soil\_layer\_i} = \dfrac{2.h_{soil\_layer\_i}}{\delta_{max}}.\left(1 - \dfrac{\delta_{soil\_layer\_i}}{\delta_{max}}\right)$	$RRD_{soil\_layer\_i}$ - relative root density in soil layer i (-) (i.e. the root density in soil layer i divided by the total root density in soil) δ_{max} - maximum rooting depth, m $\delta_{soil\_layer\_i}$ - mean depth of soil layer i, m

$$Cl_{topsoil}(t+dt) = Cl_{topsoil}(t) + Cl_{fertilizer} + \frac{PP(t).Cl_{rainfall}.dt}{h_{topsoil}} + \frac{Qirr(t).Cl_{irr}.dt(t)}{h_{topsoil}} - \lambda_{perc,topsoil}(t).R_s.Cl_{topsoil}(t).dt - \frac{RRD_{topsoil}.Trans(t).R_s.Cl_{topsoil}(t)}{h_{topsoil}.\theta_{topsoil}(t)} + \lambda_{capill\_soil\_layer\_i,n}.R_s.Cl_{subsoil1} \tag{35}$$

$$^{36}Cl_{topsoil}(t+dt) = {}^{36}Cl_{topsoil}(t) + \frac{PP(t).{}^{36}Cl_{rainfall}.dt}{h_{topsoil}} + \frac{Qirr(t).{}^{36}Cl_{irr}.dt(t)}{h_{topsoil}} - \lambda_{perc,topsoil}(t).R_s.{}^{36}Cl_{topsoil}(t).dt - \frac{RRD_{topsoil}.Trans(t).R_s.{}^{36}Cl_{topsoil}(t)}{h_{topsoil}.\theta_{topsoil}(t)} + \lambda_{capill\_soil\_layer\_i,n}.R_s.{}^{36}Cl_{subsoil1} \tag{36}$$

$$Cl_{soil\_layer\_i}(t+dt) = Cl_{soil\_layer\_i}(t) + \lambda_{perc,soil\_layer\_i-1}(t).Cl_{soil\_layer\_i-1}(t).dt - \lambda_{perc,soil\_layer\_i}(t).R_s.Cl_{soil\_layer\_i}(t).dt - \frac{RRD_{soil\_layer\_i}.Trans(t).R_s.Cl_{soil\_layer\_i}(t)}{h_{soil\_layer\_i}.\theta_{soil\_layer\_i}(t)} + \lambda_{capill\_soil\_layer\_i,n}.R_s.Cl_{soi\_layer\_i-1} \tag{37}$$

$$^{36}Cl_{soil\_layer\_i}(t+dt) = {}^{36}Cl_{soil\_layer\_i}(t) + \lambda_{perc,soil\_layer\_i-1}(t).{}^{36}Cl_{soil\_layer\_i-1}(t).dt - \lambda_{perc,soil\_layer\_i}(t).R_s.{}^{36}Cl_{soil\_layer\_i}(t).dt - \frac{RRD_{soil\_layer\_i}.Trans(t).R_s.{}^{36}Cl_{soil\_layer\_i}(t)}{h_{soil\_layer\_i}.\theta_{soil\_layer\_i}(t)} + \lambda_{capill\_soil\_layer\_i,n}.R_s.{}^{36}Cl_{soi\_layer\_i-1} \tag{38}$$

2.4 MORE COMPLEX FLOW AND FLUX MODELS

2.4.1 APPROACH ADOPTED BY IRSN

Introduction

The model for Cl-36 developed by IRSN, for use in the SYMBIOSE simulation and modelling platform for environmental pollutant risk assessments, is described in Tamponnet (2006, 2007). This study only refers to the part of the model dedicated to the Plant-Soil system.

This model considers that the driving force controlling the movements of stable chloride and Cl-36 is the circulation of water in the system. Contamination may occur via rain (or wet deposition) or irrigation. The case of direct contamination of soil from groundwater was not considered basically in this model.

The equation to describe the Cl-36 concentration in the plant, in mol kg^{-1} fresh weight, is given by,

$$\frac{\partial}{\partial t}[^{36}Cl]_{P.Org} = \delta_{Cropping=1}(TCl36_P^{InPTra} + TCl36_P^{UpTra} - TCl36_P^{Bio} - TCl36_{P.Org}^{Rad}) - \delta_{P=AnnualCrop}(\delta(t-t^{Ger+}) \times [^{36}Cl]_{P.Org}) \quad (39)$$

where

$\delta_{Cropping=1}$ is set to 1 if the plant is in the ground and is zero otherwise

$TCl36_P^{InPTra}$ is the concentration of Cl-36 in the irrigation water intercepted by the aboveground plant (mol kg^{-1} f.w. s^{-1})

$TCl36_P^{UpTra}$ is the concentration of Cl-36 in the contaminated water taken up by the roots (mol kg^{-1} f.w. s^{-1})

$TCl36_P^{Bio}$ is the concentration of Cl-36 in water eliminated from the plant, e.g. via transpiration (mol kg^{-1} f.w. s^{-1})

$TCl36_{P.Org}^{Rad}$ is the loss of Cl-36 through radioactive decay (mol kg^{-1} f.w. s^{-1})

$\delta(t-t^{Ger+})$ is used to reset the concentration at the time of germination, but only if the plant under consideration is an annual crop

Finally, the organic chloride pool was not considered in this model at its current stage of development. The results presented below assume that 50% of the water taken in as evapotranspiration goes as transpiration, with the other 50% being evaporated into the atmosphere.

2.4.2 APPROACH ADOPTED BY NDA RWMD

Introduction

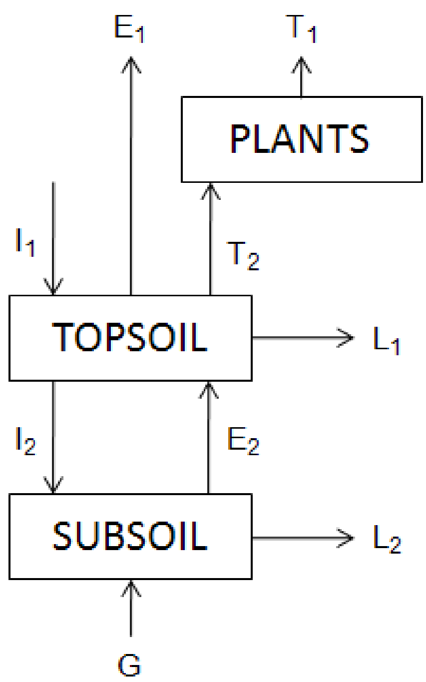
The model for Cl-36 developed for United Kingdom Nirex Ltd for use in support of future generic performance assessment studies is described in Thorne (2007a). This model takes account of the observation that Cl-36 uptake by plants is affected both by hydrological conditions and by concentrations of stable chloride in soils. The effects of competition with other chemical species, such as nitrate, are not represented explicitly, but the model could readily be extended to include such competition. Furthermore, because of the high mobility of chloride in soils, it was considered sufficient to use a two-layer soil model. Such a model is the standard level of complexity used by Nirex in its biosphere assessment calculations (Thorne, 2007b).

The basic model structure relating only to chloride is shown in Figure 2-12. The soil zone is distinguished into topsoil and subsoil compartments. Groundwater discharges occur to the subsoil compartment, whereas precipitation and irrigation waters enter the topsoil compartment.

The model can broadly be considered to have four components:

- A representation of water flow in soils and uptake by plants:
- Stable chloride transport in soils and plants;
- Organic chlorine transport in soils and plants;
- Cl-36 transport in soils and plants.

Figure 2-12: Structure of the NDA RWMD Cl-36 Model showing water flows



Water flow and chloride transport

The water contents of the compartments are taken to be time invariant, so the water flows are considered to be in balance. Note that the system is assumed to be maintained such that there is no surface runoff of meteoric water. However, as such surface runoff plays no part in the transport of chloride; the model can still be used in circumstances in which it occurs. A small water loss through harvesting plants and, therefore, removing their water content has been neglected. This loss is trivial compared with the throughput of water in the transpiration stream. The water balance formulation is very similar to that adopted in standard Nirex assessment calculations (Thorne, 2007b).

The stable chloride content of the two soil compartments is related to the concentration in water in those compartments. Thus:

$$Q_{Topsoil} = \left(\theta_{Topsoil} C_{Topsoil} + \rho_{Topsoil} C^{\#}_{Topsoil}\right) d_{Topsoil} \tag{40}$$

$$Q_{Subsoil} = \left(\theta_{Subsoil} C_{Subsoil} + \rho_{Subsoil} C^{\#}_{Subsoil}\right) d_{Subsoil} \tag{41}$$

where

Q_i is the chloride content of compartment i (mol m^{-2})

C_i is the chloride concentration in compartment i (mol m^{-3})

θ_i is the water-filled porosity of compartment i (-)

ρ_i is the dry bulk density of compartment i (kg m^{-3})

$C^{\#}_i$ is the chloride concentration on soil solids in compartment i (mol kg^{-1})

d_i is the depth of compartment i (m)

For convenience and as a suitably general relationship, a Freundlich isotherm (Freundlich, 1907), in which the concentration in the solid phase is taken to be proportional to some positive power of the concentration in the liquid phase, is adopted for the sorption of chloride onto solid particles in the soil, i.e.

$$C^{\#}_i = K C_i^{\,N} \tag{42}$$

In principle, the two calibration coefficients K and N could be dependent upon whether topsoil or subsoil is being considered. However, in practice, it seems likely that the same values would be used in both cases. It should be noted that chlorine as chloride is not considered to be redox-sensitive, so no dependence of K and N on water content is included.

Chloride contents of plants vary with species, locality and distance from the sea. However, a representative concentration is about 5000 µg g^{-1} (dry weight) (Coughtrey et al., 1983). Furthermore, for any particular plant type adapted to local conditions, this concentration is likely to vary within a fairly narrow range. For the purposes of the model, the stable chloride concentration in plants is assumed to be constant. Thus:

$$Q_{Plants} = BC_{Plants} \tag{43}$$

where

Q_{Plants} is the chloride content of plants (mol m^{-2})

B is the standing biomass of plants (including roots) (kg d.w. m^{-2})

C_{Plants} is the stable chloride concentration in plants (mol kg^{-1} d.w.)

In principle, B can be time-dependent, but, in the first instance, it is considered a useful simplification to treat harvesting or grazing as a continuous process and to make B time-independent. Although this approach may appear somewhat artificial, it is a reasonable representation of the actual time-varying process, since the underlying growth-dilution effect is continuous. Furthermore, although the rate of growth varies on a seasonal basis, to a first approximation processes of uptake and metabolism will vary in a similar way. Nevertheless, it could be useful to extend the model in future to explore explicitly the effects of seasonality on Cl-36 transport. In the approach adopted, as both B and C_{Plants} are held constant, Q_{Plants} is constant.

Stable chloride enters the model domain in upwelling groundwater (G) and in precipitation plus irrigation water (I_1). In each case, the chloride concentrations need to be specified by the user. As chloride is not lost in the evaporation and transpiration streams, zero fluxes are associated with water flows E_1 and T_1.

Chloride fluxes in water flows between the topsoil and subsoil compartments and in losses from those compartments are determined by the chloride contents in soil water in those compartments. These are the computed quantities $C_{Topsoil}$ and $C_{Subsoil}$. Thus, the four fluxes (expressed in units of mol m^{-2} y^{-1}) are given by:

$$I_2C_{Topsoil}\ L_1\ C_{Topsoil} \qquad E_2C_{Subsoil}L_2C_{Subsoil}$$

Uptake to plants is considered to occur by both active and passive processes. As stable chloride concentrations in plants are far higher than in the associated soils (e.g. Coughtrey *et al.*, 1983), uptake by plants is inferred to be mainly by an active process and saturable Michaelis-Menten kinetics seems an appropriate representation, as this form is widely used for other ions (Epstein, 1966; Nye and Tinker, 1977; Barber, 1984). Furthermore, as this is an active process, the governing factor should be the concentration of chloride in water in topsoil and not the flux of water entering the plant. However, that flux of water will also carry chloride passively into the plant. Thus, a reasonable overall parameterisation is:

$$T_2 D_{Plants} C_{Topsoil} + F_{Plants} C_{Topsoil} / \left(R + C_{Topsoil}\right) \tag{44}$$

where the two terms represent passive and active uptake, respectively. In the first term, a discrimination factor, D_{Plants}, has been introduced to allow for the possibility that chloride may be discriminated against relative to water molecules at the root membrane. Thus, D_{Plants} should be defined on the range [0, 1]. Values of >1.0 are not appropriate, as this would indicate an active process. In the second term F_{Plants} (mol m^{-2} y^{-1}) is the maximum chloride flux that can be taken up by plants and R (mol m^{-3}) is a calibration coefficient.

Chloride can be lost from plants by two processes, cropping and exudation. In a model formulation in which B is taken to be constant, cropping/harvesting is treated as a continuous process, with rate constant Y/B, where Y (kg d.w. m^{-2} y^{-1}) is the crop yield. Thus, the rate of loss by cropping is $Q_{Plants}\ Y/B$ (mol m^{-2} y^{-1}). However, as $Q_{Plants} = C_{Plants}B$, this can be written as $C_{Plants}Y$. Note that this formulation does not imply that all parts of the plant are cropped. However, it does imply that chloride is well-mixed in crops. Observationally, this is found to be the case (see Coughtrey *et al.*, 1983). That part of the crop that remains in the ground is taken to return chloride to soil by exudation, as discussed below.

Chloride is present in plants at a higher concentration than in the soil. Therefore, there is expected to be a diffusive flux out of the root system back into the soil. Indeed, studies in which plants were grown in nutrient solution have demonstrated that between 24% and 38% of the total plant chloride content could be lost by root exudation (Coughtrey *et al.*, 1983). This exudation flux will depend on the concentration difference between the plants and the soil. Note that this diffusive flux competes with active uptake processes to define the overall equilibrium concentration of chloride in plants. In the above formulation, C_{Plants} is expressed on a dry weight basis. However, most of the chloride will be present in plant water. Defining the dry weight to wet weight ratio of the plants to be φ, the concentration of chloride in plant water can be approximated as:

$$\rho_{Water} C_{Plants} \varphi / (1-\varphi) \qquad (45)$$

where ρ_{Water} is the density of water (kg m^{-3})

Thus, an appropriate formulation for loss by diffusion across the root boundary back into soil is:

$$\lambda \left[\rho_{Water} C_{Plants} \varphi / (1-\varphi) - C_{Topsoil} \right] \qquad (46)$$

where λ (m y^{-1}) is a calibration coefficient that can be thought of as being of the form D_{diff}/L_{diff} in which D_{diff} (m^2 y^{-1}) is a diffusion coefficient and L_{diff} (m) is a diffusion length.

Inclusion of Organic Chlorine

To extend the model to include organic chlorine, three additional compartments are required. These represent organic chlorine in plants, topsoil and subsoil, respectively. The organic chlorine content of plants, $Cl_{org\text{-}pl}$ (mol Cl_{org} m^{-2}), is calculated from:

$$Cl_{org-pl} = \xi B \qquad (47)$$

where

ξ is the concentration of organic chlorine in plants (mol Cl_{org} kg^{-1} (d.w.))

B is the standing biomass, as defined previously (kg (d.w.) m^{-2})

It would be expected that organic chlorine concentrations would be higher in topsoil than subsoil. Thus, it is appropriate to distinguish the two layers and use:

$$Cl_{org-Topsoil} = \omega_{Topsoil} d_{Topsoil} \quad (48)$$

$$Cl_{org-Subsoil} = \omega_{Subsoil} d_{Subsoil} \quad (49)$$

where

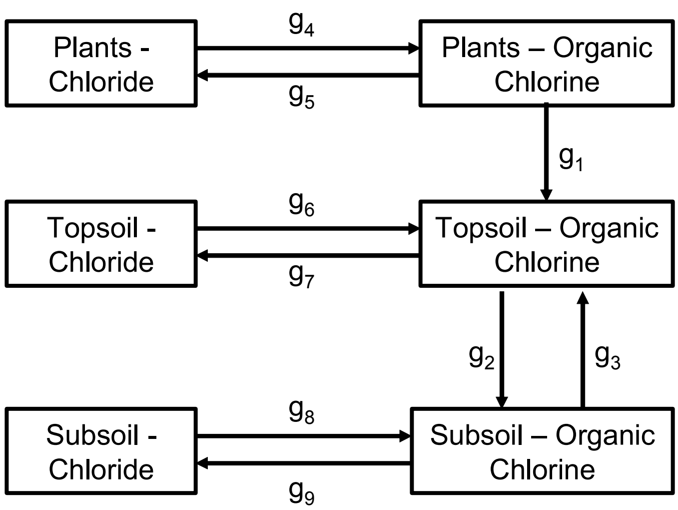
$Cl_{org\text{-}Topsoil}$ and $Cl_{org\text{-}Subsoil}$ are the organic chlorine contents of topsoil and subsoil (mol Cl_{org} m^{-2})

$\omega_{Topsoil}$ and $\omega_{Subsoil}$ are the concentrations of organic chlorine in topsoil and subsoil (mol Cl_{org} m^{-3})

$d_{Topsoil}$ and $d_{Subsoil}$ are the depths of topsoil and subsoil (m)

Transfers of organic chlorine in the soil-plant system and exchanges between organic chlorine and chloride are represented as shown in Figure 2-13.

Figure 2-13: Representation of transfers of Organic Chlorine and Exchanges with Chloride



The rate coefficient g_1 represents transfer of organic chlorine to soil by the senescence and death of plants. As in the case of the chloride component:

$$g_1 = Y / B \tag{50}$$

Transfers between topsoil and subsoil represent the physical mixing of organic matter and leaching processes. As these processes are expected to be slow, g_2 is taken to have a reference value of 0.01 y^{-1}. This is based on a dry mass density of 1325 kg m^{-3}, a topsoil depth of 0.3 m and a rate of turnover of soil due to bioturbation of 1 kg y^{-1} across the 0.3 m depth boundary. The value of g_3 is estimated to preserve a mass balance of soil in the model. Thus:

$$g_3 = g_2 \frac{d_{Topsoil} \rho_{Topsoil}}{d_{Subsoil} \rho_{Subsoil}} \tag{51}$$

The terms g_4 and g_5 represent internal transfers within plants. A wide variety of plants are known to produce chlorometabolites, but only a few studies have addressed the quantification of such compounds (Öberg et al., 2005). However, based on typical rates of metabolic reactions, it seems reasonable to suppose that incorporation of chloride in organic compounds occurs on a timescale of hours to days. Thus, g_4 is set to a reference value of 300 y^{-1}. The value of g_5 is set to achieve an appropriate balance between chloride and organic chlorine concentrations in plants. As discussed above, the concentration of organic chlorine in plants is only 1% of the stable chloride concentration. On this basis, g_5 = 100 g_4.

Organic chlorine in soils is taken to be incorporated in physically or chemically stabilised organic matter. Thus, rates of release as chloride are likely to be slow. Using a degradation half life of 1000 years for physically and chemically stabilised organic matter (Jenkinson and Rayner, 1977), g_7 and g_9 are each estimated as 7 10^{-4} y^{-1}.

An alternative way of estimating g_7 and g_9 is to assume that there is negligible conversion of chloride to organic chlorine in soils and, therefore, to set g_6 and g_8 to zero. Taking a biomass yield of 0.25 kg (d.w.) m^{-2} y^{-1} and an organic chlorine concentration in plants of 0.05 mg Cl_{org} g^{-1} (d.w.), corresponding to 0.05 g Cl_{org} kg^{-1} (d.w.), the Cl_{org} input rate to soil is 0.25×0.05 = 0.0125 g Cl m^{-2} y^{-1}. With a Cl_{org} content of soils of 12 g m^{-2}, the chloride release rate from this organic content to achieve balance is 1.04 10^{-3} y^{-1}. The similarity of this value with that estimated on the basis of organic matter degradation is encouraging. Overall, a reference value of 1 10^{-3} y^{-1} is estimated for g_7 and g_9, to be used with a reference value for g_6 and g_8 of zero.

However, conversion of chloride in soils to organic chlorine may occur. Although the processes involved are not well understood, the degree of such conversion may be considerable (Öberg et al., 2005). Indeed, for a forested catchment, Öberg et al. (2005) have used mass balance arguments to estimate a rate of transformation of 0.2 g m^{-2} y^{-1}. Adopting this value for topsoil with a chloride content in soil solution of 0.2 mol m^{-3} or 7 g m^{-3}, a porosity of 0.5 and a depth of 2.0 m, gives a rate coefficient of 2.86 10^{-2} y^{-1}.

With this degree of conversion, the rate of transfer from organic to inorganic forms must be augmented to 0.2 + 0.0125 = 0.2125 g Cl m^{-2} y^{-1} in order to achieve mass balance. Again adopting a Cl_{org} content of soils of 12 g m^{-2}, the chloride release rate from this organic content to achieve balance is 1.77 10^{-2} y^{-1}. Thus, a variant reference basis is defined in which g_7 and g_9 are taken as 1.77 10^{-2} y^{-1} and g_6 and g_8 are taken as 2.86 10^{-2} y^{-1}.

Inclusion of Cl-36 (and uptake of Cl-36 by plants)

Having defined the system in terms of stable chloride fluxes, calculations for Cl-36 follow directly. This is shown in the following equations, in which C^* is the activity concentration of Cl-36 expressed as Bq m^{-3} or Bq kg^{-1} (d.w.), as appropriate. Radioactive decay of Cl-36 has been neglected for simplicity, as the radionuclide is extremely long-lived, and constant concentrations of Cl-36 are assumed in the input water supplies. Thus, the relevant differential equation for topsoil is

$$d_{Topsoil}\left(\theta_{Topsoil} + \rho_{Topsoil}Kd_{Topsoil}\right)\frac{dC^*_{Sopsoil}}{dt} = E_2C^*_{Subsoil} + I_1C^*_{PI} + \left(\frac{\lambda C^*_{Plants}}{C_{Plants}}\right)\left(\frac{\rho_{water}C_{Plants}\varphi}{1-\varphi} - C_{Topsoil}\right) - \left(I_2 + L_1\right)C^*_{Topsoil} - T_2D_{Plants}C^*_{Topsoil} - \frac{F_{Plants}C^*_{Topsoil}}{R + C_{Topsoil}} \qquad (52)$$

Similarly, for subsoil the relevant differential equation is

$$d_{Subsoil}\left(\theta_{Subsoil} + \rho_{Subsoil}Kd_{Subsoil}\right)\frac{dC^*_{Subsoil}}{dt} = I_2C^*_{Topsoil} + GC^*_G - \left(E_2 + L_2\right)C^*_{Subsoil} \qquad (53)$$

Finally, for plants

$$B\frac{dC^*_{Plants}}{dt} = T_2D_{Plants}C^*_{Topsoil} + \frac{F_{plants}C^*_{Topsoil}}{R + C_{Topsoil}} - C^*_{Plants}Y - \left(\frac{\lambda C^*_{Plants}}{C_{Plants}}\right)\left(\frac{\rho_{water}C_{Plants}\varphi}{1-\varphi} - C_{Topsoil}\right) \qquad (54)$$

In the equation for the uptake of Cl-36 to the plant, none of the parameters are crop specific, so that identical concentrations of Cl-36 would be calculated for each crop for a given scenario.

In these equations, the concept of Kd has been introduced. However, it should be recalled that a Freundlich isotherm approach is being used. Thus, the relevant term in the equations is $KC_i^N(C^*_i/C_i)$ and this has been replaced by KdC^*_i. Thus, equating these two forms, it is readily seen that the Kd values used in these equations for Cl-36 should be calculated using $Kd = KC_i^{(N-1)}$.

2.5 SUMMARY OF MODEL SIMILARITIES AND DIFFERENCES AND KEY DATA REQUIREMENTS

2.5.1 MODEL SIMILARITIES AND DIFFERENCES

The processes considered by the models for the irrigation water source and natural groundwater source scenarios are given in Table 2-9 and Table 2-10 respectively.

Within the conventional models, all of them consider losses via percolation to lower soil layers, whereas only four of the five models consider the process of cropping. Since the SAMM-TR model does not consider losses of Cl-36 from cropping, the concentration of Cl-36 in the soil, and therefore the plant, is expected to be higher than in the models where cultivated soils are simulated (Miquel and Albrecht, 2008).

Irrespective of the number of soil layers considered within each model, the majority of them take into account only the topsoil layer in calculations of root uptake of Cl-36 (and, if appropriate, stable Cl). The exception to this is the EDF model (Section 2.3.1); in the EDF model plant uptake is assumed to occur in all layers where roots are present, with the proportion of the uptake from each layer being determined by the root density in that layer (see Equation 34 and Table 2-8). As a consequence, it would be expected that the EDF will predict lower concentrations of Cl-36 in the crops than SAMM-IR.

Table 2-9: Processes considered by the models – irrigation water source scenario

Model Type	Conventional					Specific Activity	Compartmental with isotope ratio approach for plant uptake		More Complex	
Model Name	**EPRI**	**BIOMASS (ERB2A)**	**JGC (Well)**	**Aquabios**	**SAMM -TR**	**AquaCl36**	**SAMM-IR**	**EDF**	***IRSN***	***NDA RWMD***
Rainfall				X	X	X	X	X	X	X
Evaporation								X	X	X
Transpiration								X	X	X
Evapotranspiration				X	X		X	X	X	
Sorption of Cl-36 to soil	X	X	X	X	X		X	X		X[a]
Percolation / Leaching / Infiltration to lower soil layers	X	X	X	X	X		X	X	X	X
Erosion		X	X							
Fertiliser						X	X	X		
Cropping	X	X	X	X					X	X
Irrigation	X	X	X	X	X	X	X	X	X	X

[a] Although the NDA RWMD model considers sorption processes, in the best estimate calculations (Sections 5.1.1 and 5.2.1) this is switched off.

Table 2-10: Processes considered by the models – natural groundwater source scenario

Model Type	**Conventional**			**Compartmental with isotope ratio approach for plant uptake**		**More Complex**
Model Name	**BIOMAS (ERB2B)**	**JGC (Soil)**	**SAMM-TR**	**SAMM-IR**	**EDF**	**NDA RWMD**
Rainfall			X	X	X	X
Evaporation					X	X
Transpiration					X	X
Sorption of Cl-36 to soil	X	X	X	X	X	X[a]
Percolation / Leaching / Infiltration to lower soil layers	X	X	X	X	X	X
Water loss to other parts of the system	X					X
Flooding & sedimentation		X				
Dredging & meandering		X				
Re-suspension		X				
Gross sedimentation		X				
Erosion	X	X				
Fertiliser				X	X	
Cropping	X	X				X
Capillary rise	X	X	X[b]	X[a]	X	X

[a] Although the NDA RWMD model considers sorption processes, in the best estimate calculations (Sections 5.1.1 and 5.2.1) this is switched off.

[b] In SAMM-TR and SAMM-IR the capillary rise is via diffusion.

2.5.2 Model Data Requirements

For a given scenario, the parameters used in each model fall into two types, those which are site independent and those which are site specific. For those parameters which are site independent, model default values are typically used, so that no further consideration is required before these parameters can be used in implementations of the models. However, it should not be assumed that because some parameters are pre-defined (i.e. assigned 'default' values) that these parameters are known with total certainty.

Site specific climate data are required by each model independent of its type in order to determine the water balance (Section 4.1). For other processes, the number, and nature, of required site specific parameters often depends upon the model type; these are given in Table 2-11 and are discussed in greater detail in Sections 4.2, 4.3 and 4.4.

Table 2-11: Site specific model type dependent data requirements

Model Type	Data requirements
Conventional	Sorption coefficient of Cl to soil, K_d Concentration factors for the uptake of Cl-36 from the soil by the plant roots Soil physical parameters
Specific Activity	Stable Cl content of the plants Stable Cl content in the rain, in the irrigation water and in the fertiliser And potentially, stable Cl content of the soil, depending upon which chlorine pools are assumed to be in equilibrium
Compartmental with isotope ratio for plant uptake	Sorption coefficient of Cl to soil, K_d Soil physical parameters Stable Cl content of the soil (this is modelled based on: Stable Cl content in the rain, in the irrigation water and in the fertiliser) Stable Cl content of the plants
Complex	Stable Cl content of the soil Stable Cl content of the plants Plant transpiration (IRSN)

2.5.3 Model Comparisons

Many of the participants in this study carried out two forms of calculations with their models: a single run using the 'best estimate' of the model parameters, and a batch of runs taking uncertainty in the true value of some of the input parameters into account. For the sensitivity calculations, traditional methods for analysing the results from such simulations include the Pearson correlation coefficient (Box et al. 1978), which is an indicator of linear correlations between individual input parameters and an output variable.

An alternative approach to this is to use a Gaussian process model (Oakley 2000, Kennedy and O'Hagan 2001), such as that encoded within GEM-SA[a] (Gaussian Emulation Machine for Sensitivity Analysis; Kennedy 2004). This statistical software uses a Gaussian process prior probability distribution to describe beliefs about an unknown code output, as a function of the code inputs. Heuristically, a Gaussian process model can be viewed as a probabilistic non-parametric modelling approach for black-box identification of non-linear dynamic systems (Likar and Kocijan 2007). Using such a method it is possible to ascertain the dependency of the output on both the individual parameters and the combined effects of the parameters acting together. Gaussian process models are used to analyse complex problems in many disciplines, e.g. the manufacturing of specialised steel (Bailer-Jones et al. 1999), the cost-effectiveness of medical treatments (Stevenson et al. 2004), and ecosystem modelling (Kennedy et al. 2008).

[a] http://www.tonyohagan.co.uk/academic/GEM/index.html

3 SCENARIO DESCRIPTION

Consistent with the scope described above, two source terms were assumed by which the Cl-36 in groundwater reaches the rooting zone of agricultural soil:

1. as a result of irrigation, and
2. as a result of capillary action and any/all other transfers from below

For case 1), it was assumed that the concentration of activity in the irrigation water is 1 Bq/l and the irrigation water entirely reaches the surface of the root-zone soil. The amounts of irrigating water and how and when in the season will depend on matters described below. For 2) the same Cl-36 concentration in groundwater was assumed (1 Bq/l) and that the groundwater table was 2 m below the relevant ground surface. The stable chlorine concentration in irrigation water was not specified. (excluding any contribution from the assumed Cl-36). Rainwater is assumed to be 10 mg/l for a coastal site and 2.5 mg/l for an inland site[a]. The intention was to avoid arbitrary alternative assumptions for the geosphere-biosphere interface. Four publications of Kashparov were considered a valid data source, at least for the inland site (Kashparov et al 2005a,b; Kashparov et al. 2007a,b).

The question of interest for this study is the typical Performance Assessment question, i.e. what is the annual individual dose to an average member of a potential exposure group (PEG)? However, in this study the focus is on the assessment of Cl-36 concentrations in the edible parts of the harvested crops which are to be consumed by the PEG or by animals to be part of the PEG exposure network[b].

In carrying out a model comparison, there is a delicate balance to strike between over- or under-constraining the scenarios under consideration. If all the input parameters for the models were to be provided, this would prejudice the lessons that can be learnt from the results comparisons. If the definition of the scenarios were to be left too loose, too many options are left open and there may be no points of comparison between the models.

The following compromise was therefore adopted, i.e. each user sets their own constraints based on their own assessment context, in this case, primarily information on site characteristics, but also factors concerned with relevant assessment endpoints. This way it is possible to learn from:-

- the experience of sharing information on included and excluded processes, bearing in mind the differences between different site generic contexts and between different site specific contexts;
- seeing what difference the different types of sites *appear* to have on results;
- the degree to which the differences actually arise because of the different models applied or data available.

For both scenarios, modern agronomic methods were assumed, including the use of fertiliser and soil and farming features as described in Kasparov et al. (2005a,b). Monthly climate data were provided for a coastal and inland site (Météo France, 2008), though primary focus was given to the inland site

[a] The effect of chlorine deposition on Chernobyl Cs-137 mobility Norway in coastal and inland areas is interestingly recorded in Steinnes et. al. (2007).
[b] It is outside the scope of this study to consider and compare assumptions for PEG behaviours.

(Sections 5.1 and 5.3.1). It was then left to the project participants to use or interpret this data as appropriate to their model requirements and contexts; e.g. as annual averages. All four Kashparov publication were given as primary sources (Kashparov et al 2005a,b; Kashparov et al. 2007a,b), from which ANDRA derived data (ranges) for soil type, soil physical properties and other data (such as concentration ratios, stable chlorine concentration in the plants). This data was subsequently used by many of the other participants (e.g. EDF, SCM).

Given these constraints, calculations were made of the concentration of Cl-36 in the following crops: root vegetables, leafy green vegetables and cereal. These three were chosen because the edible parts are very different, i.e. for root vegetables, the edible part is below ground, and lettuce, the edible part is above ground; while for grain it is only the seed body which is eaten. It was proposed to calculate first the best estimate of concentration of Cl-36 in the crops at time of harvest in Bq/kg fresh weight (fw), using the first year of the climate data (Météo France, 2008), together with estimates of the potential range of concentrations given uncertainties associated within an average year. If concentrations were thought to be potentially higher in subsequent years, a continuous sequence of five years of climate was suggested (Météo France, 2008), repeated if more than 5 years was needed.

In a second phase of the calculations, those parameters which were considered pertinent but uncertain, a series of stochastic calculations was carried out to quantify the impact of their uncertainties on model output, and to find relationships between those parameters and the final result.

4 MODEL PARAMETERS AND DATA

The data requirements for each model can be split into three categories: (1) climate and hydrology, (2) soil and fertilizer, and (3) plant. The data used in these categories are discussed in Sections 4.1 to 4.3, highlighting which parameters have been used as constants in deterministic analyses and those which have been regarded variable in probabilistic approaches. However, it should not be assumed that because some parameters are pre-defined (i.e. assigned 'default' values) that these parameters are known with total certainty. More realistically it is the case that the uncertainty in these parameters is not relevant to the given study.

These specific 'uncertainties' of the site specific parameters are stemming from either an incomplete understanding of the site and its management (e.g. soil properties, the cropping rate) or uncertainty in the empirical quantification of that parameter (e.g. the sorption of Cl-36 to the soil). These parameters, and the distributions which have been assumed for these parameters, are given in many of the tables below. Samples from the distributions of the site-specific 'uncertain' parameters were then used in the stochastic calculations of the scenarios to examine the model sensitivities (Sections 5.1.2 and 5.2.2).

In addition to the processes which are parameterised in all of the models, parameters associated with processes considered in specific models only are discussed in Section 4.4. The only data used by the models which can be considered truly independent of either the site or scenario, and to some extent model, is given in Table 4-1.

Table 4-1: Site and scenario independent fixed parameters

Parameter	Units	Value	Participant using this data
Concentration of Cl-36 in source water	Bq l^{-1}	1.00E+00	All
Half life of Cl-36	y	3.01E+05	EPRI, BIOMASS, JGC
		3.02E+05	ANDRA, SCM

4.1 CLIMATE AND HYDROLOGY PARAMETERS

The site specific climate and hydrology data used in simulations of the inland and coastal sites are given in Table 4-2 and Table 4-3 respectively. With the exception of the conventional models, additional information on the stable chlorine concentration of the precipitation and source water was required, and is given in Table 4-4 and Table 4-5 for the inland and coastal sites respectively.

Monthly climate data for two sites in France were provided for this model inter-comparison (Météo France, 2008), one site being inland (St. Dezier) and the other coastal (Lille). The precipitation data (m $month^{-1}$) spanned a five year period, whereas the other three climate variables - temperature (^{o}C), monthly daylight duration (h) and monthly sunshine duration (h) – came from one year but were repeated five times. From the 5 year precipitation data, ANDRA and SCM derived an annual average irrigation rate (Figure 2-6) which was then also used by EPRI, BIOMASS and JGC in their calculations. The EDF and IRSN models operate on a monthly and daily time step respectively (see

Sections 2.3.2 and 2.4.1), so that for these models irrigation rates were calculated during a model run[a]. The NDA RWMD model assumed a total input of water from aboveground to be 0.8 m y^{-1}, with 0.75 m y^{-1} as precipitation and the remainder as irrigation water.

From the climate data the potential evapotranspiration was calculated by EDF using Turc's relationship (Turc 1954, 1961) and used directly in their submodel for soil water budget determination (Section 2.2). Andra and SCM used these evapotranspiration data in their estimation of irrigation needs (Section 2.3). Although evapotranspiration was also used the IRSN model (Section 2.4.1), one of the more complex models, it was calculated on a crop-by-crop basis using an alternative method (Kramer 1959; Tamponnet 2007).

Table 4-2: Climate and hydrology data – inland site

Parameter	Best estimate	Distribution	Low	High	Participant using this data
Amount of precipitation (m/y) [b]	5.94E-01	Uniform (U)	4.31E-01	7.23E-01	ANDRA, SCM, EDF, IRSN
Relation rain-irrigation: slope (see Figure 2-6)	-9.10E-01				ANDRA
Relation rain-irrigation: intercept (see Figure 2-6)	1.026E+03				ANDRA
Irrigation rate (m/y)	4.85E-01	U	3.68E-01	6.34E-01	ANDRA [c], BIOMASS, EPRI, JGC
	3.02E-01	U	1.83E-01	4.24E-01	SCM
	3.60E-01				EDF [d]
Evapotranspiration (m/y)	7.12E-01				ANDRA, SCM, EDF
	5.50E-01				NDA RWMD[e]

[a] IRSN divided the monthly precipitation rates by 30 to approximate daily precipitation rates for use in their model.
[b] Météo France (2008)
[c] This was calculated by ANDRA using the relationship between precipitation and irrigation rate (Figure 2-6).
[d] This is the amount distributed per irrigation event (based on 3.0E-02 m^3 water.m^{-2} soil.$month^{-1}$)
[e] This is the default value from Thorne (2007b).

Table 4-3: Climate and hydrology data – coastal site

Parameter	Best estimate	Distribution	Low	High	Participant using this data
Amount of precipitation (m/y)	7.42E-01	U	6.14E-01	8.18E-01	ANDRA, SCM, EDF, IRSN
Relation rain-irrigation: slope (see Figure 2-6)	-7.88E-01				ANDRA
Relation rain-irrigation: intercept (see Figure 2-6)	1.064E+03				ANDRA
Irrigation rate (m/y)	4.79E-01	U	4.19E-01	5.80E-01	ANDRA, BIOMASS, EPRI, JGC
	2.79E-01	U	1.86E-01	3.64E-01	SCM
	3.60E-01				EDF [a]
Evapotranspiration (m/y)	7.21E-01				ANDRA, SCM, EDF
	5.50E-01				NDA RWMD

Table 4-4: Stable chlorine concentration of water inputs - inland site

Parameter	Best estimate	Distribution	Low	High	Participant using this data
Stable chlorine concentration in precipitation (g/L)	7.00E-05	Triangular (T)	7.00E-05	5.60E-03	ANDRA, SCM
	2.50E-03				EDF, NDA RWMD
Stable chlorine concentration of source water (g/L)	2.00E-03	U	2.00E-03	8.00E-02	ANDRA, SCM
	1.60E-02				EDF
	3.00E-02				NDA RWMD

[a] This is the amount distributed per irrigation event (based on 3.0E-02 m^3 water.m^{-2} soil.$month^{-1}$)

Table 4-5: Stable chlorine concentration of water inputs - coastal site

Parameter	Best estimate	Distribution	Low	High	Participant using this data
Stable chlorine concentration in precipitation (g/L)	9.55E-03	T	7.00E-05	2.15E-02	ANDRA, SCM
	1.00E-02				EDF,NDA RWMD
Stable chlorine concentration of source water (g/L)	4.50E-02	T	2.00E-03	5.00E-01	ANDRA, SCM
	4.80E-02				EDF
	3.00E-02				NDA RWMD

Table 4-6: Cl-36 concentration of water inputs

Parameter	Best estimate	Participant using this data
Cl-36 in rainfall (Bq m^{-3} rainfall)	0 or 6.25E+01 [a]	EDF, NDA RWMD
Cl-36 in groundwater (Bq m^{-3})	1.0 10^3 or 0.0	NDA RWMD

4.2 SOIL AND FERTILIZER PARAMETERS

Neither the soil nor fertilizer parameters were site specific, so are presented here in Table 4-7 and Table 4-8 respectively.

Table 4-7: Soil parameters

Parameter	Best estimate	Distribution	Low	High	Participant using this data
K_d (m^3 kg^{-1})	5.0E-05	T [b]	0	5.8E-03	EPRI, JGC, SCM, ANDRA[c], BIOMASS
	1.0E-03	U	1.0E-04	1.0E-02	EDF

[a] 6.25E+01 is used for the irrigation scenario by the NDA RWMD model only.
[b] Andra HAVL Dossier (2005), Andra Centre Manche and Pelletier et al 2007, Kashparov et al. (2005 a,b; 2007 a,b)
[c] ANDRA consider this parameter for their conventional model, Aquabios, only.

Table 4-7 continued: Soil parameters

Parameter	Best estimate	Distribution	Low	High	Participant using this data
Topsoil thickness (m)	2.0E-01	T	1.0E-01	3.0E-01	ANDRA, SCM[a]
	3.0E-01				EPRI, BIOMASS, JGC, NDA RWMD [b]
	5.50E-02[c]	U	1.0E-03	1.0E-02	EDF[d]
	1.5E-01 (Vegetables) 2.5E-01 (Cereal)				IRSN
Soil Porosity (-)	4.00E-01				BIOMASS, JGC
	4.50E-01				ANDRA, SCM, EDF
	5.00E-01				EPRI
Soil water content (m^3 water m^{-3} soil)	2.5E-01	T	2.0E-01	5.0E-01	ANDRA[e]
	3.50E-01				BIOMASS
	4.00E-01				IRSN
	5.00E-01				NDA RWMD
Soil grain density (kg m^{-3})	2.65E+03				EPRI, BIOMASS
	2.60E+03				JGC, ANDRA, SCM, EDF
	1.40E+03				IRSN
	1.325E+03				NDA RWMD[f]
Soil stable chlorine concentration	Inland: 2.0E-03 (g kg^{-1}) [g] Coastal: 1.0E-02(g kg^{-1})				ANDRA, SCM, EDF
	5.6 10^{-3} mol.kg^{-1} (d.w.)				IRSN

[a] SCM used only the best estimate value as this model was not used for any probabilistic calculations.

[b] In the NDA RWMD model the subsoil is assumed to be 1.7m thick.

[c] Ciffroy et al. (2005)

[d] EDF considers an additional three soil layers, each 2.00E-01m thick.

[e] ANDRA consider this parameter for their conventional model, Aquabios, only.

[f] This density is used for the subsoil as well.

[g] Not used for calculations by ANDRA or SCM.

Table 4-8: Fertilizer parameters

Parameter	Best estimate	Distribution	Low	High	Participant using this data
Fertiliser application [kg m^{-2} y^{-1}]	2.50E-02	T	8.33E-03	7.50E-02	ANDRA, SCM
Stable chlorine concentration in the fertilizer	1.90E+02 [g/kg] [a]				ANDRA, SCM
	1.00E+00 [g m^{-2} soil] [b]				EDF

4.3 PARAMETERS RELATING TO ROOT UPTAKE

The parameters of interest in the conventional models for the uptake of Cl-36 into the plants are given in Table 4-9; the same distributions were used for both the inland and coastal sites.

For the other models, the parameter(s) of interest for the root uptake of Cl-36 was the stable chlorine content of each crop. In the NDA RWMD model this was fixed at 1.40E-01 mol kg^{-1}, independent of crop or site under consideration. The stable chlorine concentrations in the crops assumed for the inland and coastal sites of the remaining models are given in Table 4-10.

Some of the models (EPRI, BIOMASS, JGC, Aquabios, IRSN, NDA RWMD) consider cropping as a means of losing chlorine from the system. The parameters used for these cropping rates were assumed to be both scenario and site independent and are given in Table 4-12.

Table 4-9: Plant specific uptake parameters - conventional models

Parameter	Best estimate	Distribution	Low	High	Participant using this data
Concentration ratio, root vegetables (-) [c]	1.50E+01	T	8.50E+00	1.31E+02	EPRI, JGC, SCM, ANDRA, BIOMASS
Concentration ratio, leafy green vegetables (-) [a]	3.00E+01	T	8.50E+00	1.49E+02	EPRI, JGC, SCM, ANDRA, BIOMASS
Concentration ratio, cereal (-) [a]	1.34E+02	T	1.20E+01	2.46E+02	EPRI, JGC, SCM, ANDRA, BIOMASS

[a] Kashparov et al. 2007b
[b] Kashparov 2006
[c] Andra HAVL Dossier (2005), Andra Centre Manche and Pelletier et al 2007, Kashparov et al. (2005 a,b; 2007 a,b)

Table 4-10: Plant specific uptake parameters – stable chlorine concentrations [g kg^{-1} fw] for the inland site

Crop	Best estimate	Distribution	Low	High	Participant using this data
Independent	4.97E-01				IRSN
	1.40E-01				NDA RWMD [a]
Root vegetables	3.02E-01	T	5.20E-01[b]	8.10E-01	ANDRA, SCM, EDF[c]
Leafy green vegetables	2.45E-01	T	1.75E-01	3.50E-01	ANDRA, SCM
	2.45E-01				EDF
Cereal	5.04E+00	T	3.30E-01	8.58E+00	ANDRA, SCM, EDF[c]

Table 4-11: Plant specific uptake parameters – stable chlorine concentrations [g kg-1 fw] for the coastal site

Crop	Best estimate	Distribution	Low	High	Participant using this data
Independent	4.97E-01				IRSN
Root vegetables	6.04E-01	T	5.20E-01	1.62E+00	ANDRA, SCM
	1.21E+00				EDF
Leafy green vegetables	4.90E-01	T	1.75E-01	7.00E-01	ANDRA, SCM
	9.80E-01				EDF
Cereal	1.01E+01	T	3.30E-01	1.72E+01	ANDRA, SCM
	2.02E+01				EDF

[a] mol kg^{-1} (d.w.) rather than g kg^{-1} (f.w.)
[b] The minimum value was by mistake set higher than the best estimate; as this has only a minor effect on the probabilistic results, but no effect on the deterministic results, no revision was necessary.
[c] EDF used the same upper and lower limits for the probabilistic simulations, but with a uniform distribution.

Table 4-12: Harvest yield data used (kg fw m^{-2} y^{-1})

Crop	Best estimate	Distribution	Low	High	Participant using this data
Crop independent	2.0E+00	T	0.0E+00	4.0E+00	ANDRA
	3.00E+00				EPRI
	2.50E+00				NDA RWMD
Root vegetables	3.00E+00				BIOMASS
	3.15E+00				JGC
	3.37E+00				IRSN
Leafy green vegetables	3.00E+00				BIOMASS
	1.5E+00				JGC
	2.12E+00				IRSN
Cereal	4.00E-01				BIOMASS, JGC
	2.30E+00[a]				IRSN

[a] This is based on maize.

4.4 ADDITIONAL MODEL PARAMETERS

4.4.1 CONVENTIONAL MODELS

The additional default parameters used by some of the conventional models, BIOMASS (ERB2A), and JGC are given in Table 4-13 and Table 4-14 respectively.

Table 4-13: Additional user defined parameters in BIOMASS model - irrigation water source scenario, ERB2A

Parameter	Symbol	Units	Value
Infiltration rate	I	$m\ y^{-1}$	7.0E-01
Retardation coefficient	R	-	1
Erosion rate	E	$m^3\ y^{-1}$	0
Soil contamination on the crop	S_{crop}	kg (dry weight soil) kg^{-1} (fresh weight of crop)	0
Fraction of the internal contamination associated with the edible art of the plant at harvest that is retained after food processing has occurred	F_{p2}	-	1
Fraction of external soil contamination on the edible part of the crop retained after processing	F_{p1}	-	N/A

Table 4-14: User defined parameters in JGC model

Transfer	Parameter	Symbol	Units	Value
Water supply	Annual volume of water abstracted from well water compartment for purpose other than irrigation	V_{nirr}	$m^3\ y^{-1}$	0 [a]
	Volume of the well	V_{dw}	m^3	1.00E+06 [a]
Infiltration and recharge	Annual infiltration/recharge rate	d_d	$m\ y^{-1}$	0.7
	Retardation coefficient for the compartment from which infiltration/recharge is coming	R_{sed}	-	1

[a] This parameter is used only in the irrigation water source scenario

Table 4-14 continued: User defined parameters in JGC model

Transfer	Parameter	Symbol	Units	Value
Erosion	Erosion rate of the soil compartment	E_{sed}	m y^{-1}	1.00E-04
Crop concentration	Fraction of external contamination on the crop lost due to food processing	F_{crop}	-	N/A
	Soil contamination on the crop	S_{crop}	kg (d.w. soil) kg^{-1} (f.w. crop)	0
	Interception fraction for irrigation water on the crop	μ_{crop}	-	0
	Depth of irrigation water applied to the crop	d_{icrop}	m y^{-1}	N/A

4.4.2 COMPARTMENTAL MODELS WITH AN ISOTOPE RATIO APPROACH TO PLANT UPTAKE OF CONTAMINANT

The additional user defined parameters of the EDF model have been split into crop independent (Table 4-15) and crop dependent (Table 4-16).

Table 4-15: User defined parameters of the EDF model

Parameter	Symbol	Units	Value
Wilting point water content [a]	θ_{wp}	m^3 water m^{-3} soil	1.65E-01
Water content at field capacity [b]	θ_{fc}	m^3 water m^{-3} soil	3.40E-01
Limit water content [b]	θ_{lim}	m^3 water m^{-3} soil	1.83E-01
Quantity of water distributed to the field for one 'irrigation event'	$Qirr_{event}$	m^3 water m^{-2} soil $month^{-1}$	3.00E-02
Cultural coefficient [c]	K_c	-	1.01E+00
Leaf area index at harvest [d]	LAI_{harv}	-	4.00E+00
Extinction factor [c]	$\alpha_{extinction}$	-	4.00E-01

Table 4-16: Crop specific user defined parameters in the EDF model

Parameter	Crop		
	Root vegetables	Leafy Green Vegetables	Cereal
Maximum rooting depth (m) [e]	0.20E-01	1.00E-01	1.00E+00
Date of harvest of the plant (day of the year)	270	150,210,270	270
Date of germination of the plant (day of the year)	105	90,151,211	90
Maximum stable chlorine concentration in plant (units)	3.02E-01	2.45E-01	5.04E+00

[a] Baes and Sharp (1983)
[b] Maruax et al. (1998)
[c] Sau et al. (2004)
[d] IAEA (1994)
[e] Soltner (2004)

4.4.3 More Complex Flow and Flux Models

Additional scenario independent, user defined parameters in the IRSN and NDA RWMD models are given in Table 4-17 and Table 4-18.

Table 4-17: User defined parameters in the IRSN model

Parameter	Units	Value
Infiltration rate	m^3 water m^{-2} s^{-1}	1.20E-08

Table 4-18: User defined parameters in the NDA RWMD model

Transfer type	Parameter	Units	Value
Water flows	Precipitation plus irrigation inputs to topsoil	$m\ y^{-1}$	8.00E-01
	Evaporative losses from topsoil	$m\ y^{-1}$	5.00E-02
	Losses from plants in transpiration	$m\ y^{-1}$	5.00E-01
	Infiltration from topsoil to subsoil	$m\ y^{-1}$	2.50E-01
	Capillary driven upward movement: subsoil to topsoil	$m\ y^{-1}$	3.50E-01[a]
	Groundwater discharge to subsoil	$m\ y^{-1}$	1.00E-01
Soil properties	Density of water	$kg\ m^{-3}$	1.00E+03
Chloride sorption to soils	K value for topsoil. Used in Freudlich isotherm.	Depends on N	0
	K value for subsoil. Used in Freudlich isotherm.		0
	N value for topsoil. Used in Freudlich isotherm.	-	1.00E+00
	N value for subsoil. Used in Freudlich isotherm.	-	1.00E+00
Plant characteristics	Standing biomass density	kg (d.w.) m^{-2}	2.50E-01
	Michaelis-Menten maximum uptake rate	mol m^{-2} y^{-1}	1.00E+00
	Michaelis-Menten concentration coefficient	mol m^{-3}	1.00E+00
	Plant discrimination factor for passive uptake	-	1.00E+00
	Dry weight to wet weight ratio of plants	-	1.00E-01

[a] Adjusted to give $L_2 = 0$

Table 4-18 continued: User defined parameters in the NDA RWMD model

Transfer type	Parameter	Units	Value
Organic chlorine information	Rate of bioturbation from topsoil to subsoil	y^{-1}	1.00E-02
	Conversion of plant chloride to plant organic chlorine	y^{-1}	3.00E+02
	Conversion of chloride to organic chlorine in soil[a]	y^{-1}	0
	Conversion of organic chlorine to chloride	y^{-1}	1.00E-03

[a] The same parameter value is used for both soil layers.

5 RESULTS AND DISCUSSION

The results from calculations of Cl-36 concentrations in crops for both scenarios are presented in this chapter. In the first two sections the irrigation scenario is considered in detail, with greater focus given to the calculations for the inland site (Section 5.1) rather than the coastal site (Section 5.2). In the third section (Section 5.3), the groundwater release scenario is considered, though fewer results were contributed than for the irrigation scenario. In the final section (Section 5.4), the effect of altering assumptions about the stable chlorine concentration in the plants is considered, together with the stable chlorine concentration in the source water.

For the irrigation scenario, the effect of sorption of Cl-36 to the soil, and the impact of the consideration of cropping losses on calculated Cl-36 soil and crop concentrations are discussed (Section 5.1.3). For the groundwater scenario, the effect of altering the defined stable chlorine concentration in the plant is discussed (Section 5.4). For both scenarios, the effect of altering the rate of inflow of contaminated source water (site contamination) is considered (Sections 5.1.4 and 5.3.3).

5.1 IRRIGATION WATER SOURCE SCENARIO – INLAND SITE

The models from the following organisations were applied to the irrigation water source scenario at the inland site: EPRI, BIOMASS, JGC, ANDRA, SCM-ANDRA, EDF, IRSN and NDA RWMD. In the first part of this section, focus is given to the concentration of Cl-36 in the crops at harvest based on best estimates for the parameter values in each model. In the second part, focus is given to the sensitivity of some of the models to the parameter values each participant used. The third part of this section contains a more detailed examination of the interaction between sorption of Cl to the soil and cropping losses. The fourth part of this section considers the sensitivity of the model predictions of Cl-36 concentration in the plants to irrigation rates.

5.1.1 DETERMINISTIC CALCULATIONS

The concentration of Cl-36 in the various crops after five years are shown in Figure 5-1, Figure 5-2 and Figure 5-3. Square symbols are used to denote the conventional models, a triangle for the specific activity model, diamonds for the compartmental models that use an isotope ratio approach for the plant uptake of Cl-36, and circles for the more complex models.

The results show agreement in model predictions within a factor of 10 for the root vegetables (Figure 5-1) and a factor of 6 for the leafy green vegetables (Figure 5-2). Within the conventional models the agreement is even better (within a factor of 3).

There is a greater degree of variability in the predictions for the Cl-36 concentration in the cereal, with agreement only to a factor of two orders of magnitude if all the models are considered (Figure 5-3; note the log scale on the y-axis). However, the agreement within the models is greatly increased by excluding AquaCl36 (the specific activity model) and the NDA RWMD model (one of the more complex models), giving results that agree within a factor of 15. As with the other two crops, within the conventional models the agreement is within a factor of 3. The models which used an isotopic ratio approach to plant uptake used parameter values for the uptake based upon both the grain and straw of the cereal, whereas the conventional models considered uptake only into the grain. Kashparov et al (2007b) reported concentration ratios for the grain and straw components of both wheat and oats. There was at least a factor of 6 difference in the reported concentration ratio, dependent upon the plant component under consideration, with the highest uptake occurring in the straw.

Although the difference is most marked for the crops, AquaCl36 consistently predicted the highest concentration of Cl-36 in all of the crops. AquaCl36 uses the simplest mathematical approach evaluating the Cl-36 activity in the plant on the basis of the Cl_36/Cl_s ratio in the soil (plant environment) and the stable chlorine content of the plant. The Cl-36 activities (Bq/kg dry) modelled by AquaCl36 (0.17) are comparable with those obtained by others (Table 5-1). In principle, therefore, the Cl-36 concentrations calculated by AquaCl36 for the plants should be comparable to those of SAMM-IR and EDF since all three models utilise the same stable chlorine concentrations for the plants (Table 4-10). However, the Cl-36 to stable chlorine ratio in the soil calculated by AquaCl36 (84 Bq g^{-1}) is over 50% greater than calculated by either SAMM-IR or EDF[a] (56 Bq g^{-1}).

The majority of the models use crop specific parameters for the uptake of chlorine into the plant, the exception being the NDA RWMD model. Since all of the conventional models used the same crop specific concentration ratios (see Table 4-9) to model the uptake, it is not surprising that their calculated Cl-36 concentration in cereal was nine times greater than the Cl-36 concentration in root vegetables since the CR is nine times higher. Similarly, those models which used an isotope ratio approach to plant uptake of Cl-36 (including the specific activity model, AquaCl36) used the same crop specific parameters (Table 4-10), with the factor of 16 difference between cereal and root vegetable uptake parameters reflected in the calculated crop concentrations of Cl-36. The IRSN model also used crop specific parameters for plant uptake of Cl-36, though the increase in Cl-36 is less than for the models previously discussed. In particular, when modelling the uptake of Cl-36 in cereal, only the grain of the plant was considered by IRSN; it has been shown that the straw component, considered in the other models, has a higher uptake of Cl-36 (Kashparov et al, 2007b).

Since the NDA RWMD model does not use crop specific parameters a higher uptake of Cl-36 by the cereal, as compared to the other crops, is not observed for this model. It is likely that this therefore explains why the calculated concentration of Cl-36 in cereal is much lower than the other models (a factor of 6 compared to the next lowest prediction, the EPRI model).

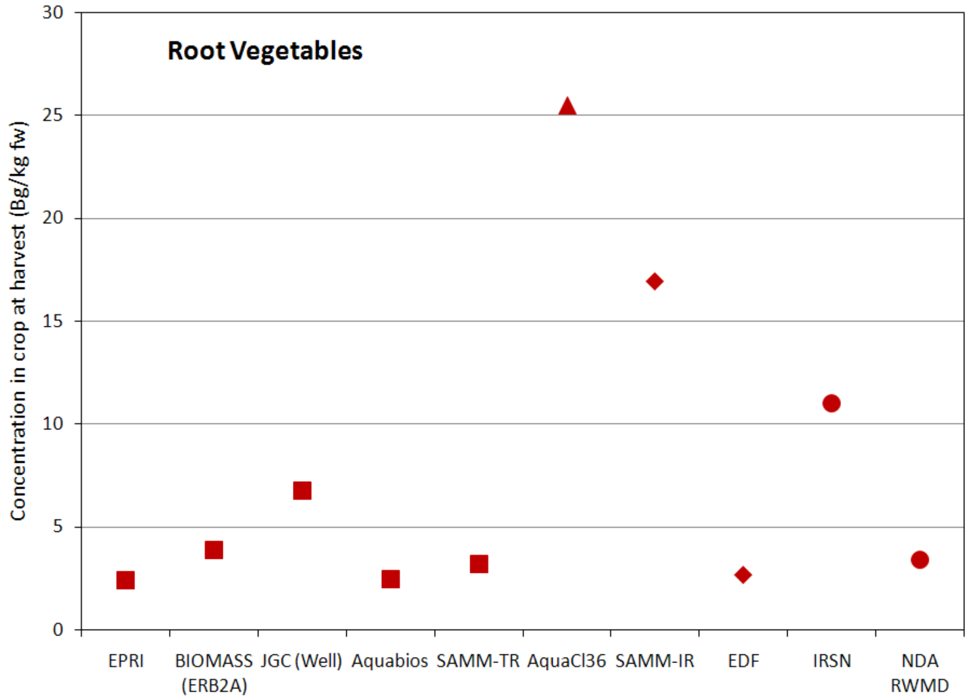
Aquabios and SAMM-TR employ the same differential equation, the first using an analytical, the second a numerical approach, with consistency checked (Miquel 2008). Differences in calculated soil Cl-36 concentrations (Table 5-1), and thus crop Cl-36 concentrations, are solely based on consideration of cropping losses, considered in Aquabios (2 kg m^{-2} y^{-1}), not considered in SAMM-TR. The effect of cropping is discussed further in Section 5.1.3.

The EDF model calculated some of the highest Cl-36 concentrations in the soil (Table 5-1), yet the calculated concentration of Cl-36 in the root and leafy green vegetables are amongst the lowest values. The EDF model uses a more complex approach which models water and chlorine input and uptake as a function of precipitation and evapotranspiration and as a function of depth as well as root depth distribution, so that the ratio Cl-36/Cl_s drops with depth. A fraction of the roots thus sees these lower ratios and the overall modelled Cl-36 activity of the plant drops, compared with if only the topsoil layer was considered.

[a] For the EDF this value applies to the topsoil layer only. The Cl-36 to stable chlorine ratios were lower in the subsoil layers.

Although accumulation in soils is included in many of the models[a], the accumulation of Cl-36 in the soil is so small in the majority of the models that there is effectively no difference in the Cl-36 concentration in the soil, or plant, between 1, 2 and 3 years or beyond (Figure 5-4). One exception to this is the NDA RWMD model, which takes up to 20 years to reach some form of equilibrium. The effects of higher soil accumulation rates of Cl-36, together with the impact of cropping, are discussed in more detail in Section 5.1.3. The other exception is the JGC model with a higher K_d (Nakai and Miyauchi, 2008).

Figure 5-1: Irrigation scenario [inland site]. Deterministic calculations of Cl-36 in root vegetables (Bq/kg fw)



[a] AquaCl36 and the IRSN model are the exceptions.

Figure 5-2: Irrigation scenario [inland site]. Deterministic calculations of Cl-36 in green vegetables (Bq/kg fw)

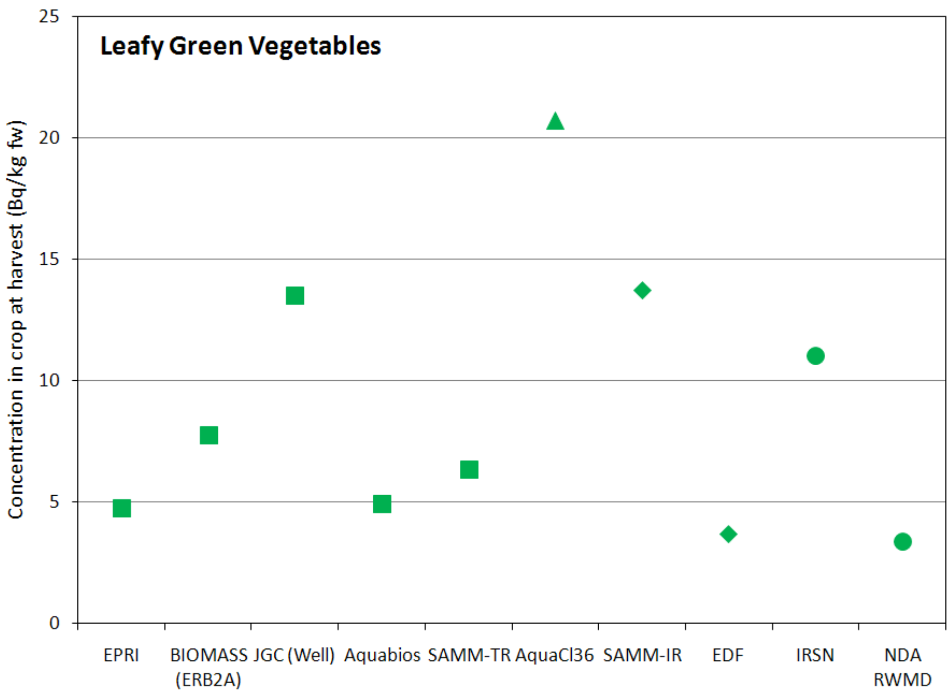


Figure 5-3: Irrigation scenario [inland site]. Deterministic calculations of Cl-36 in cereal (Bq/kg fw)

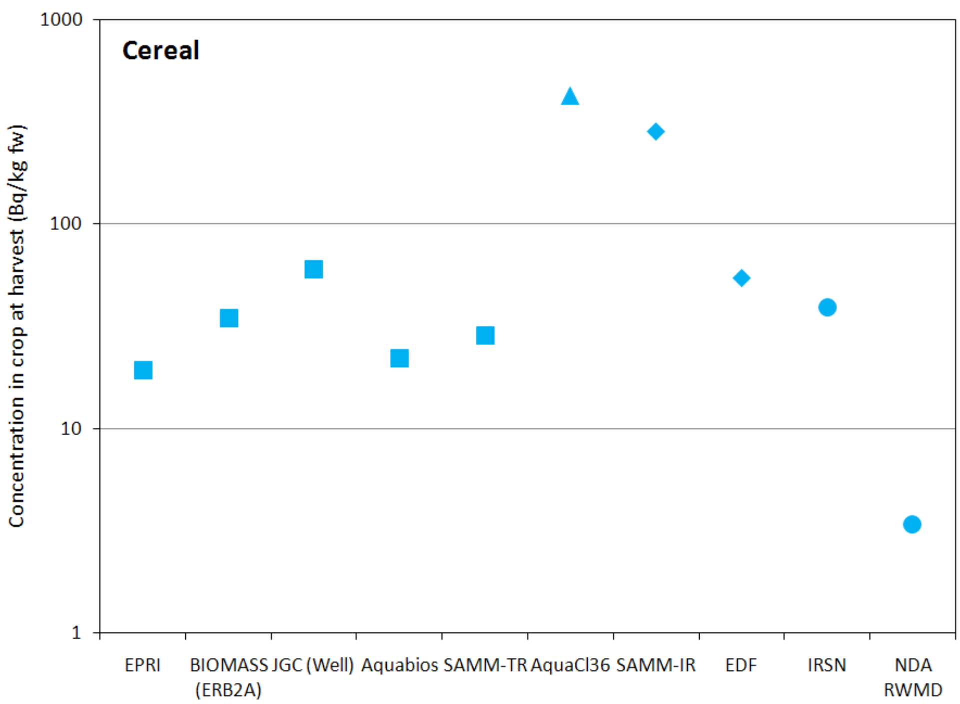
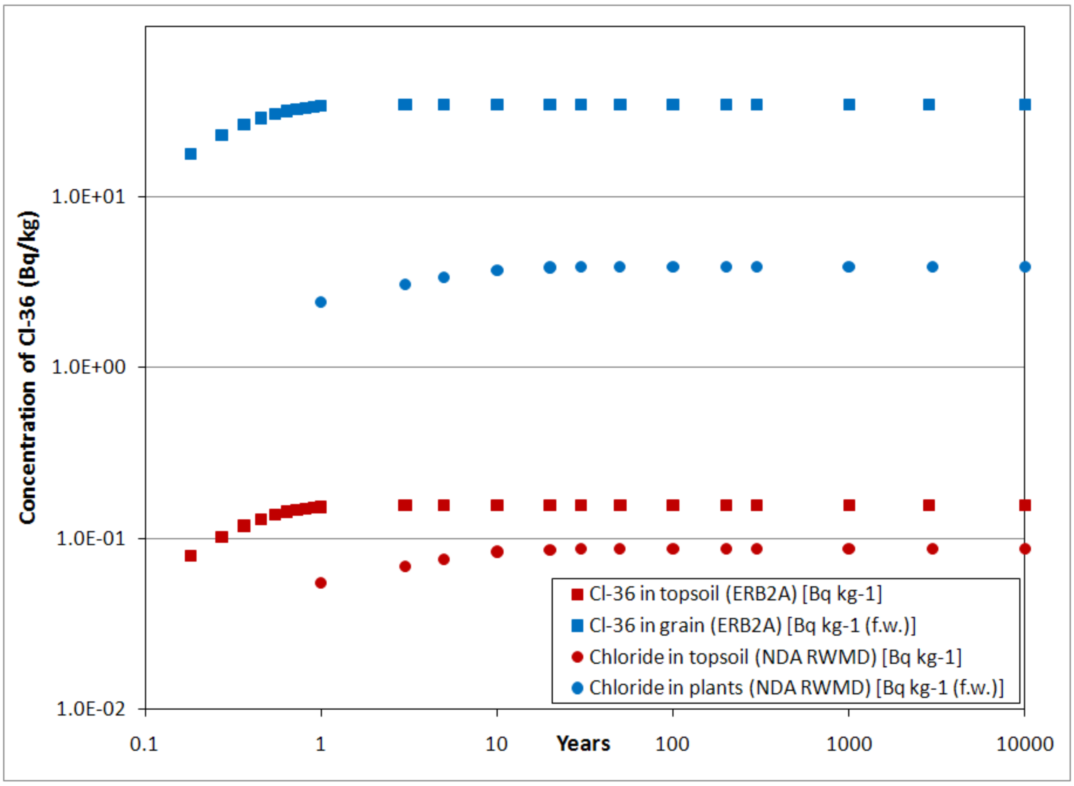


Table 5-1: Concentration of Cl-36 in the soil after 5 years of simulation [inland site]

Model Type	Model	Topsoil concentration of Cl-36 (Bq/kg_{dry})
Conventional	EPRI [a]	Root vegetables: 1.60E-01 Leafy green vegetables: 1.58E-01 Cereal: 1.43E-01
	BIOMASS [b]	2.58E-01
	JGC	4.49E-01
	Aquabios	1.60E-01
	SAMM-TR[c]	2.10E-01
Specific Activity	AquaCl36	1.70E-01
Compartmental with isotope ratio approach for plant uptake	SAMM-IR	2.10E-01
	EDF [c, d]	Root vegetables: 1.40E+00 Leafy green vegetables: 7.00E-01 Cereal: 5.81E-01
More complex	IRSN	1.30E-1
	NDA RWMD [e]	1.51E-01

[a] A soil porosity of 0.5 and a soil density of 2650 kg m^{-3} were used to convert the Cl-36 from Bq m^{-3}.
[b] A soil porosity of 0.4 and a soil density of 2650 kg m^{-3} were used to convert the Cl-36 from Bq m^{-3}.
[c] A soil porosity of 0.45 and a soil density of 2600 kg m^{-3} were used to convert the Cl-36 from Bq m^{-3}.
[d] There is a difference between the crops because there is a different irrigation. This concentration are those in the topsoil (55×10^{-3} m). The concentrations in the sub-soil layers are lower than those in the topsoil.
[e] A soil porosity of 0.5 and a soil density of 1325 kg m^{-3} were used to convert the Cl-36 from Bq m^{-3}.

Figure 5-4: Time series of Cl-36 concentration (Bq kg^{-1}) in soil and plant in the BIOMASS (ERB2A) and NDA RWMD model [irrigation scenario, inland site]



5.1.2 Probabilistic Calculations

Uncertainty analysis

EPRI, BIOMASS, JGC, ANDRA, and EDF carried out sensitivity analyses for the crop concentrations of Cl-36 based on the uncertainty of a specific number of model parameters[a]. The arithmetic mean simulated crop concentrations of Cl-36 together with the 5% and 95% percentiles of the CDF (cumulative density function) of results (box), and the minimum and maximum (the vertical lines) from these simulations are shown in Figure 5-5, Figure 5-6 and Figure 5-7. In addition, the results from the deterministic calculations are overlaid onto the sensitivity simulations. Due to the large variation in calculated concentrations, the y-axis in each graph is on a log-scale. The parameters active in the probabilistic assessments are summarised in Table 5-2.

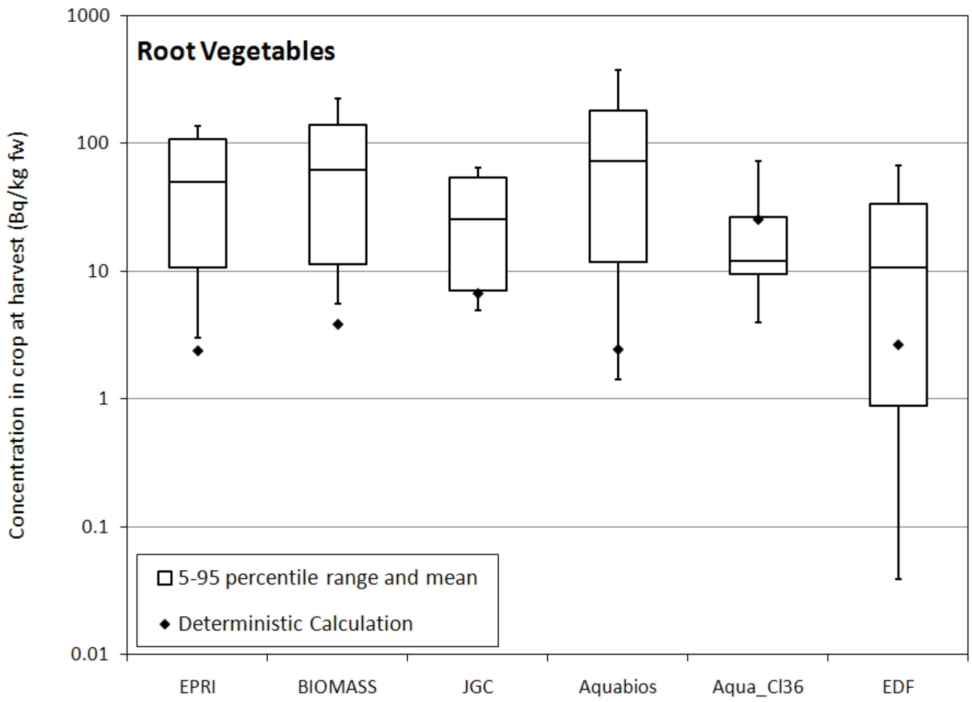
The distributions used in these probabilistic assessments are such that the best estimate value is typically at the lower end of each input distribution. It is therefore to be expected that the value from the deterministic calculation may be lower than the mean value from the probabilistic calculations[b]. Further, the complex interactions between the parameters are such that the deterministic results may lie outside of the range of plant concentrations calculated in the probabilistic assessment.

[a] EDF did not carry out stochastic calculations for the leafy green vegetables.

[b] In other probabilistic assessments, best estimate calculated concentrations have been greater than the range of the probabilistic calculations (Smith, 2008).

From these figures it is clear that the variability within a given model due to parameter uncertainty (typically ranging from a factor of 13 to a factor of 250[a]) is greater than the variability between the models in the deterministic calculations (ranging from a factor of 3 to a factor of 15[b]).

Figure 5-5: Stochastic calculations - root vegetables [inland site]. The 5- and 95-percentiles with the mean is indicated by the two boxes. The tails indicate the extremes of the results from the simulations. [c]



[a] In the stochastic calculations, the concentration of Cl-36 in the root vegetables varied by a factor of 1700 in the EDF model (based upon minimum and maximum predicted concentrations).
[b] This is only valid if AquaCl36 and the NDA RWMD model are excluded from the analysis in the cereals. If all models are considered, the deterministic calculations varied by up to a factor of 126.
[c] The crop concentrations from the deterministic calculations (black diamond's) are shown for reference.

Figure 5-6: Stochastic calculations - leafy green vegetables [inland site].

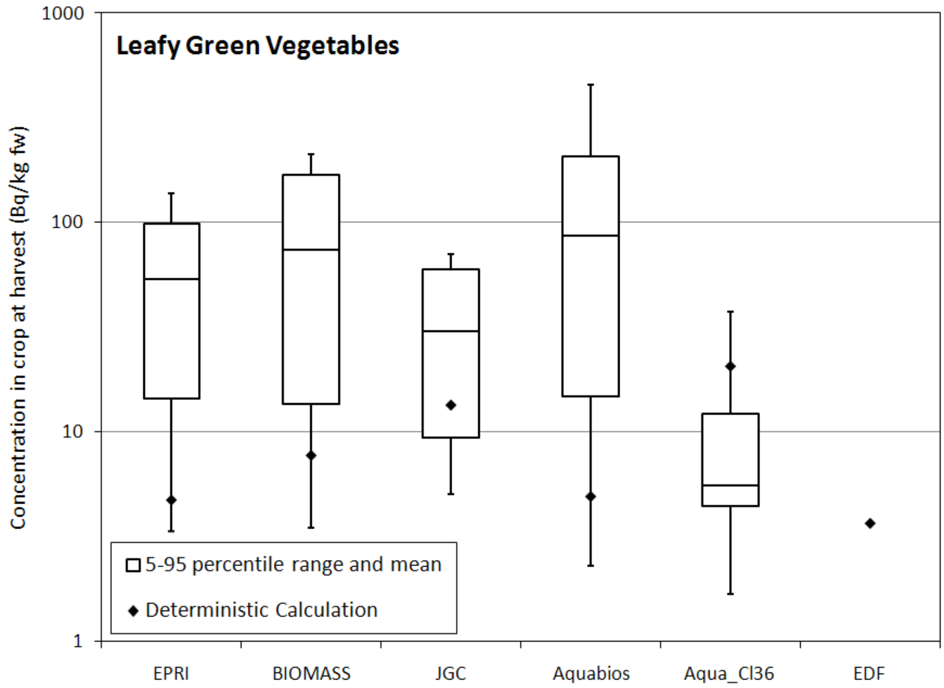


Figure 5-7: Stochastic calculations - cereal [inland site].

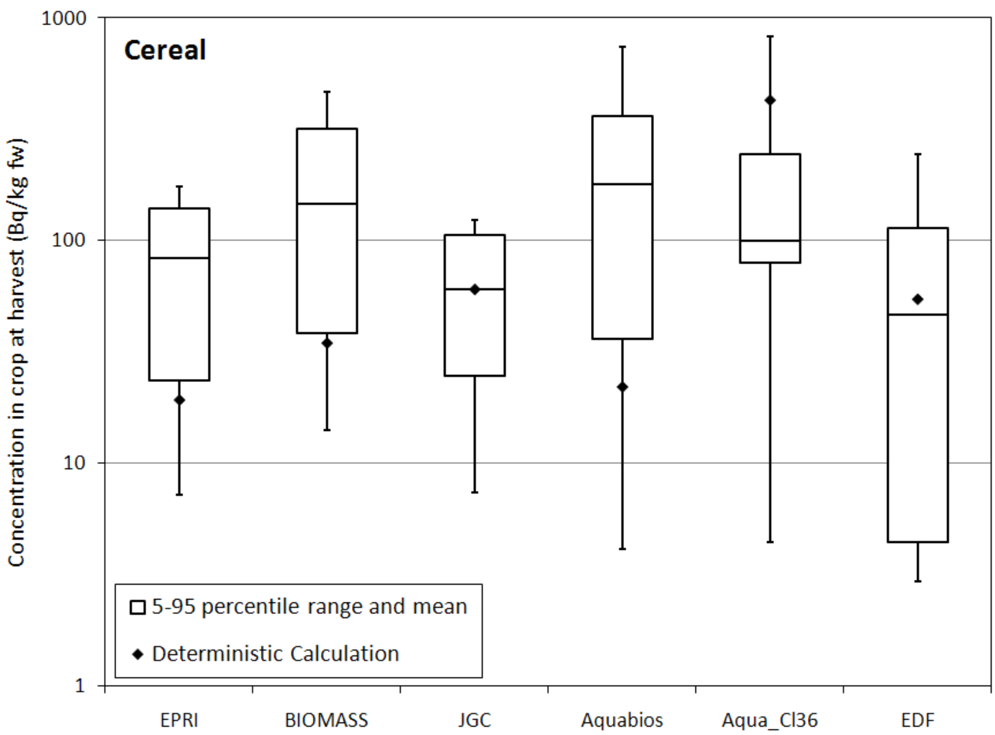


Table 5-2: Parameter varied during the stochastic calculations of the irrigation scenario (inland site)

Model	Parameters varied
EPRI	Sorption of Cl to soil (K_d) Concentration ratio for each crop (CR) Irrigation rate (V_{irr})
BIOMASS	Sorption of Cl to soil (K_d) Concentration ratio for each crop (CR) Irrigation rate (V_{irr})
JGC	Sorption of Cl to soil (K_d) Concentration ratio for each crop (CR) Irrigation rate (V_{irr})
Aquabios	Sorption of Cl to soil (K_d) Concentration ratio for each crop (CR) Precipitation rate [a] Crop yield Crop rooting depth Soil water content
AquaCl36	Stable chlorine content of the plants Stable chlorine content of the well water Stable chlorine content of precipitation Rate of fertilizer application Rate of stable chlorine application to the cultivated soil Precipitation rate [a]
EDF	Sorption of Cl to soil (K_d) Stable chlorine content of the plants Depth of the soil layers

[a] From this the irrigation rate was calculated (Figure 2-6).

Sensitivity analysis

Although the data presented in Figure 5-5, Figure 5-6 and Figure 5-7 offer a perspective as to the overall sensitivity of the models to a sub-set of their parameters, it is useful to examine more closely how individual parameters affect the model predictions, and how these parameters interact.

As seen in the probabilistic results obtained from AquaCl36, the calculated Cl-36 crop concentrations correlate negatively with the amount of precipitation. This can be explained both by the dilution of radioactive Cl with stable chlorine present in the rain or by the reduced need of irrigation with increasing precipitation. Similar arguments can be developed to explain the negative correlations between plant contamination and stable chlorine concentration of both the irrigation water or the extent of fertiliser application. All of these parameters cause an increase in the stable chlorine concentration of the soil, thereby lowering the Cl-36 to stable chlorine ratio.

For each individual crop, the concentration of Cl-36 was positively correlated to the stable chlorine concentration of the plant. This is because more stable chlorine in the plant would indicate more Cl-36 in the plant also. The assumed stable chlorine concentration in plants was found to have a positive effect on the calculated crop concentration of Cl-36 in the EDF model as well.

The sorption coefficient of Cl-36 to the soil and the depth of the topsoil layer were both also found to have a positive effect on the calculated crop concentration; the nature of these interactions is discussed further in Sections 5.1.3 and 5.1.4.

The following discussion now focuses upon the conventional models, since more than one model of this class participated in the sensitivity studies. Considering the parameters separately, the concentration ratios used were strongly positively correlated to the simulated concentration in the crops at harvest (Table 5-3), as were the sorption coefficient and the rate of irrigation[a]. Given the largest amount of variation occurred in the calculation of Cl-36 concentration in cereal, Figure 5-8 and Figure 5-9 contains scatter plots of Cl-36 concentration in cereal against K_d and the concentration ratio of cereal respectively. Table 5-4 demonstrates how much variation in the calculated concentration of Cl-36 in the cereal depended upon the parameters and their interactions. In all cases, it is the concentration ratio of Cl-36 uptake in the cereals which has the greatest impact upon the predicted Cl-36 concentration in the crop at harvest. In most of the models the interaction between sorption and the concentration ratio explained more than 1% of the variance in the results, the JGC model being the exception.

[a] For Aquabios, this correlation has been inferred given a measured significant negative correlation between precipitation and crop concentration of Cl-36 and the assumed negative relationship between precipitation and irrigation (Figure 2-6)

Table 5-3: Conventional modelling of the irrigation scenario - Correlation of Cl-36 concentration in crop at harvest with model parameters. Here + and – symbols have been used to denote positive and negative correlations respectively. +/- represents p < 0.1; ++/-- represents p < 0.05 and +++/--- represents p < 0.01.

Crop	Model	K_d	Concentration ratio (root vegetables)	Concentration ratio (leafy green vegetables)	Concentration ratio (cereal)	Irrigation rate
Root Vegetables	EPRI	+++	+++			+++
	BIOMASS	+++	+++	-		+++
	JGC		+++			+++
	Aquabios	+++	+++		---	+++
Leafy Green Vegetables	EPRI	+++		+++		+++
	BIOMASS	+++		+++		+++
	JGC	+	-	+++		+++
	Aquabios	+++		+++	---	+++
Cereal	EPRI	+++		--	+++	+++
	BIOMASS	+++			+++	+++
	JGC				+++	+++
	Aquabios	+++			+++	+++

Figure 5-8: Concentration of Cl-36 in cereal at harvest (Bq kg-[1] f.w.) against sorption coefficient (K_d)

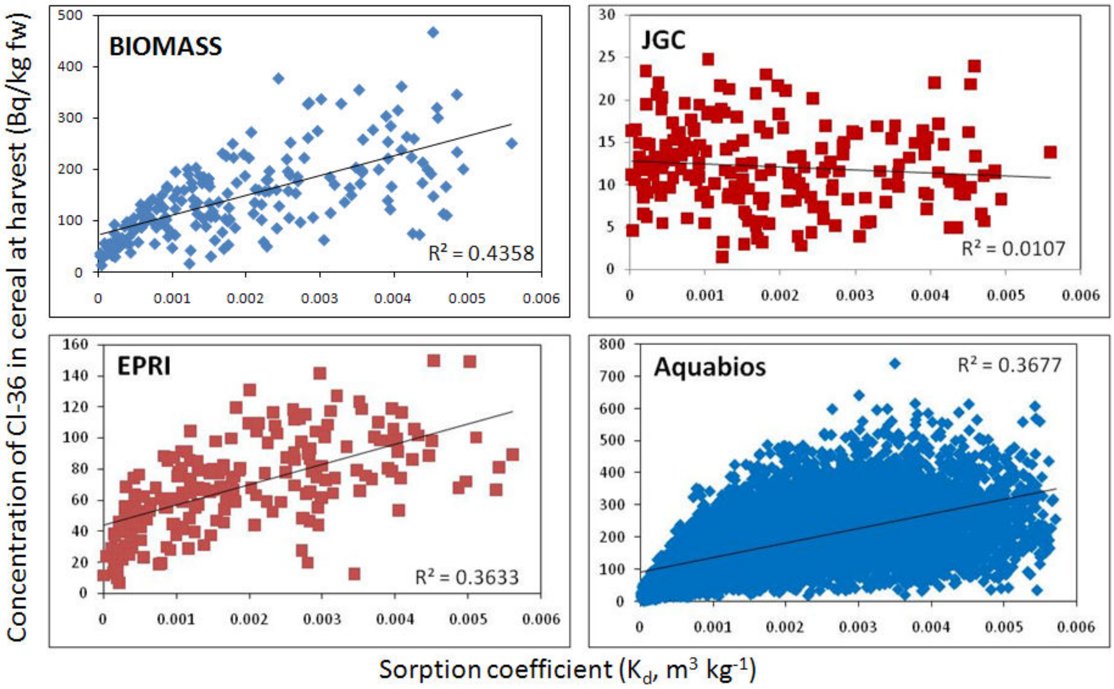


Figure 5-9: Concentration of Cl-36 in cereal at harvest (Bq kg^{-1} f.w.) against concentration ratio (CR) for cereal (-)

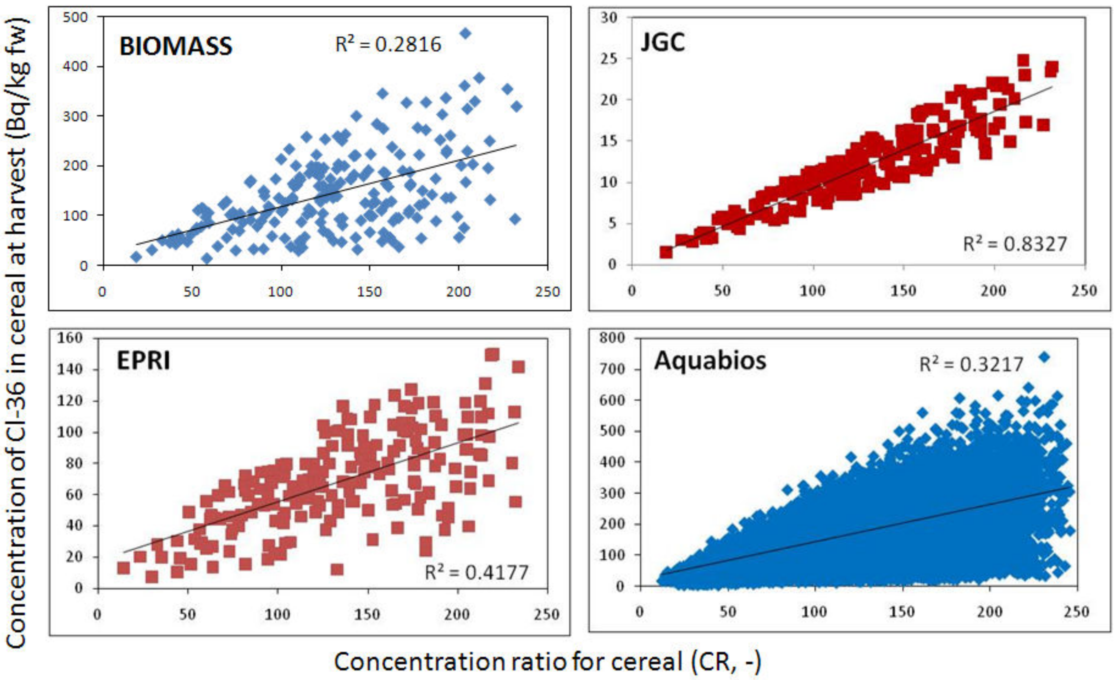


Table 5-4: Variation in calculated Cl-36 concentration in cereal as explained by model input parameters

Parameter	Model			
	EPRI	BIOMASS	JGC	Aquabios[a]
K_d	28.13	35.16	0.04	16.86
CR (Cereal)	53.95	54.86	90.44	43.19
Irrigation rate	13.48	1.71	8.41	0.56
K_d . CR (Cereal)	1.47	3.19	0.06	4.45
K_d . Irrigation rate	0.76	0.47	0.05	1.48
CR (Cereal) . Irrigation Rate	0.42	0.20	0.65	0.49
Other parameters & pair-wise interactions	0.28	3.27	0.09	12.67
Total	98.83	98.86	99.74	75.17

5.1.3 Interaction between sorption and cropping

The following discussion relates to those models which considered both a non-zero K_d and cropping; thus the conventional generic models EPRI, BIOMASS, JGC, Aquabios) and also the NDA RWMD model. Although the EDF and SAMM-TR models consider a non-zero K_d, cropping is disregarded.

Consideration of soil losses based on cropping is necessary in the case of Cl-36 only if accumulation in the soil is significant. For very low K_d values there is no difference between the first and second year Cl-36 concentration, as chlorine is essentially washed out of the system. Only in cases where chlorine Kd is larger a critical value (Nakai and Miyauchi, 2008) does the consideration of cropping impact the plant activity.

The experiment, carried out with the JGC model to increase the sorption coefficient of Cl-36 to the soil, showed that it requires more time for Cl-36 to reach equilibrium, both in the soil and in the plant and the equilibrium value attained will be higher.

The cropping rate has the inverse effect to reduce the time to reach equilibrium and to decrease the ultimate equilibrium concentration of Cl-36 in the soil and the crop (compare Figure 5-10 to Figure 5-11). This effect of cropping rate upon the ultimate equilibrium concentration of Cl-36 in the crops is also observed in Aquabios (Figure 5-12).

The relationship between sorption, the cropping rate, the concentration ratio and the modelled concentration of Cl-36 in the crop is exponential, as can be seen in Figure 5-13 and Figure 5-14. In these figures both root vegetables (with a concentration ratio of 15, solid symbols) and cereals (with a

[a] This analysis is based on a limited subset of the probabilistic results from this model. As a consequence the parameter space will not be as well covered as for the other three models.

concentration of 134, open symbols) are shown. For a low sorption coefficient (Figure 5-13) the relationship between cropping rate and the concentration of Cl-36 in the two crops is effectively log-linear, the overall higher concentrations in the cereals resulting from the higher concentration ratio used for that crop. With a high sorption coefficient (Figure 5-14) if the concentration ratio is low enough then the relationship between cropping rate and the concentration of Cl-36 in the crop is still effectively log-linear. However, if the concentration ratio for uptake of Cl-36 by a crop is high then this no longer holds. This behaviour is not observed in the NDA RWMD model (Figure 5-15).

Figure 5-10: Effect of increasing sorption of Cl-36 to soil in the JGC model - root vegetables with no cropping

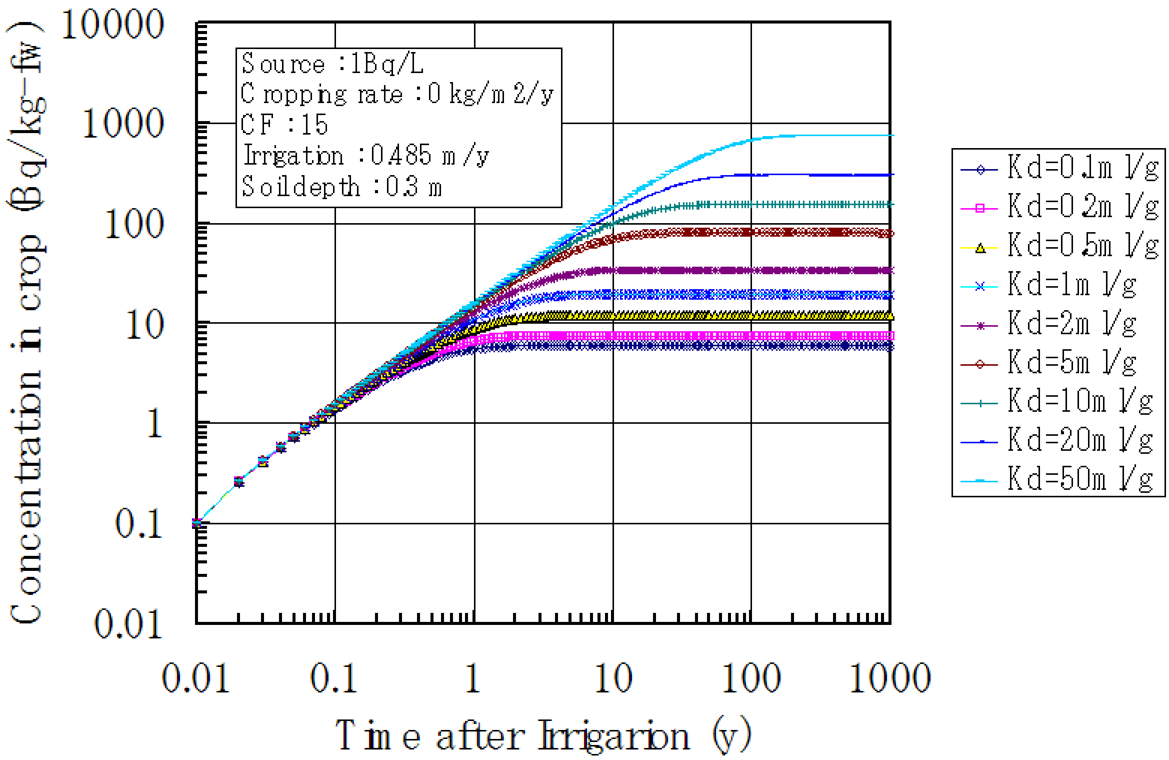


Figure 5-11: Effect of increasing sorption of Cl-36 to soil in the JGC model - root vegetables with cropping at a rate of 5 kg/m^2/y

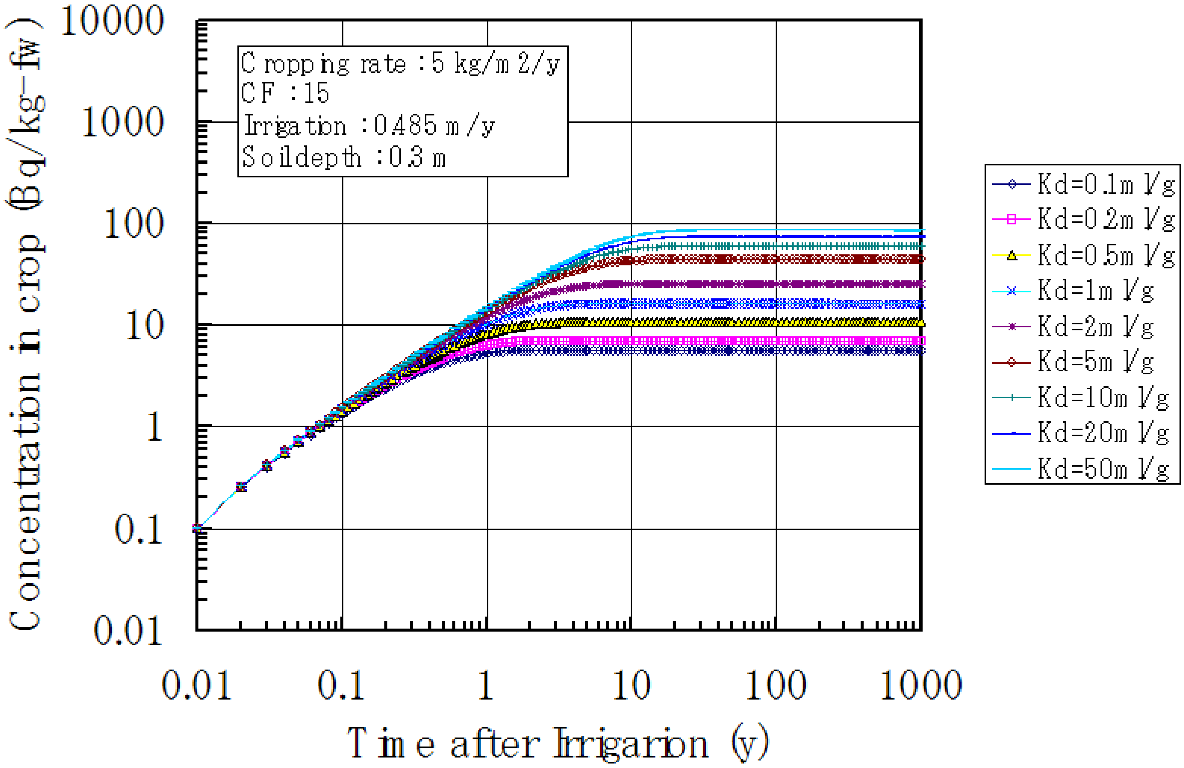


Figure 5-12: Effect of cropping in Aquabios - sorption coefficient of 5.8 l/kg.

(a) No cropping. (b) Cropping at a rate of 2 kg/m^2/y.

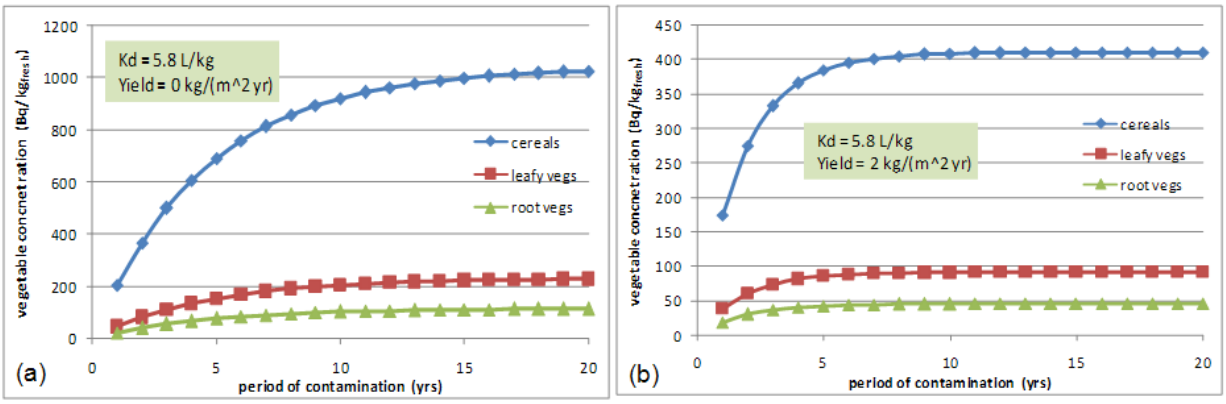


Figure 5-13: Interaction between the cropping rate and concentration ratio in the JGC model (K_d = 0.5 ml g^{-1})

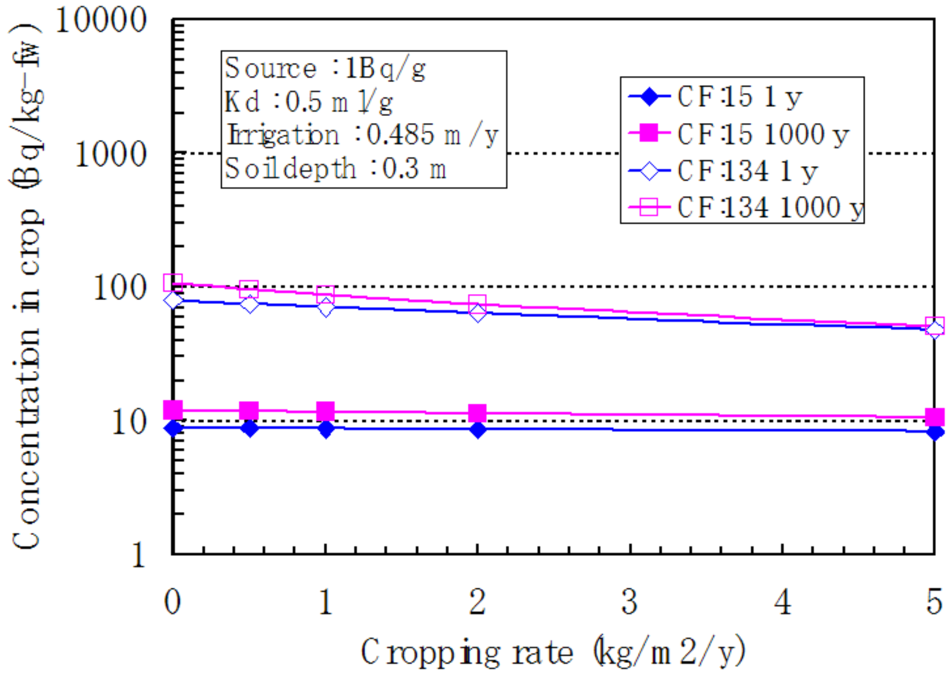


Figure 5-14: Interaction between the cropping rate and concentration ratio in the JGC model (K_d = 10 ml g^{-1})

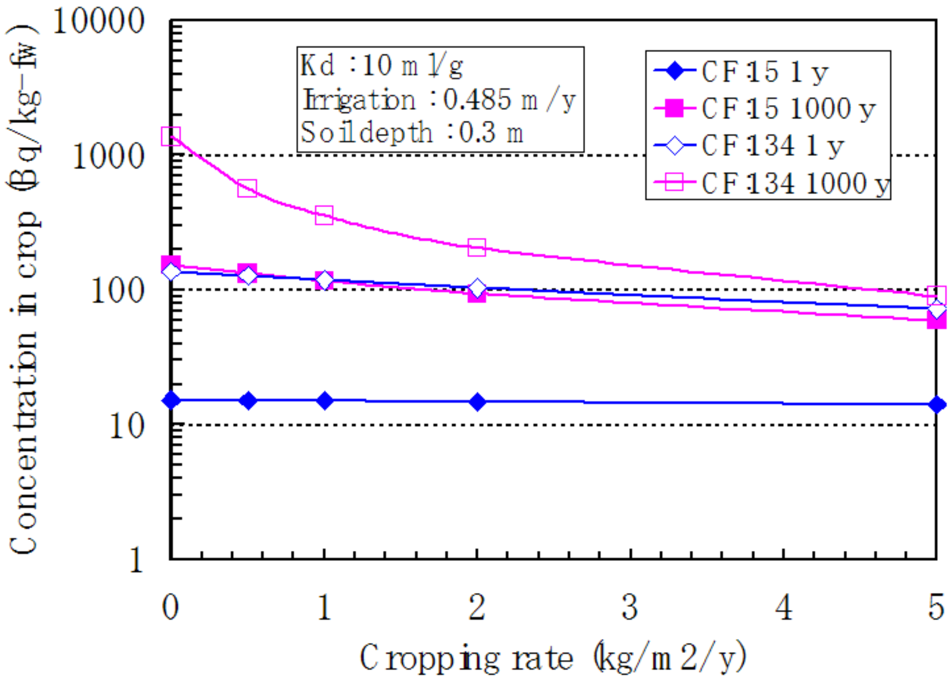
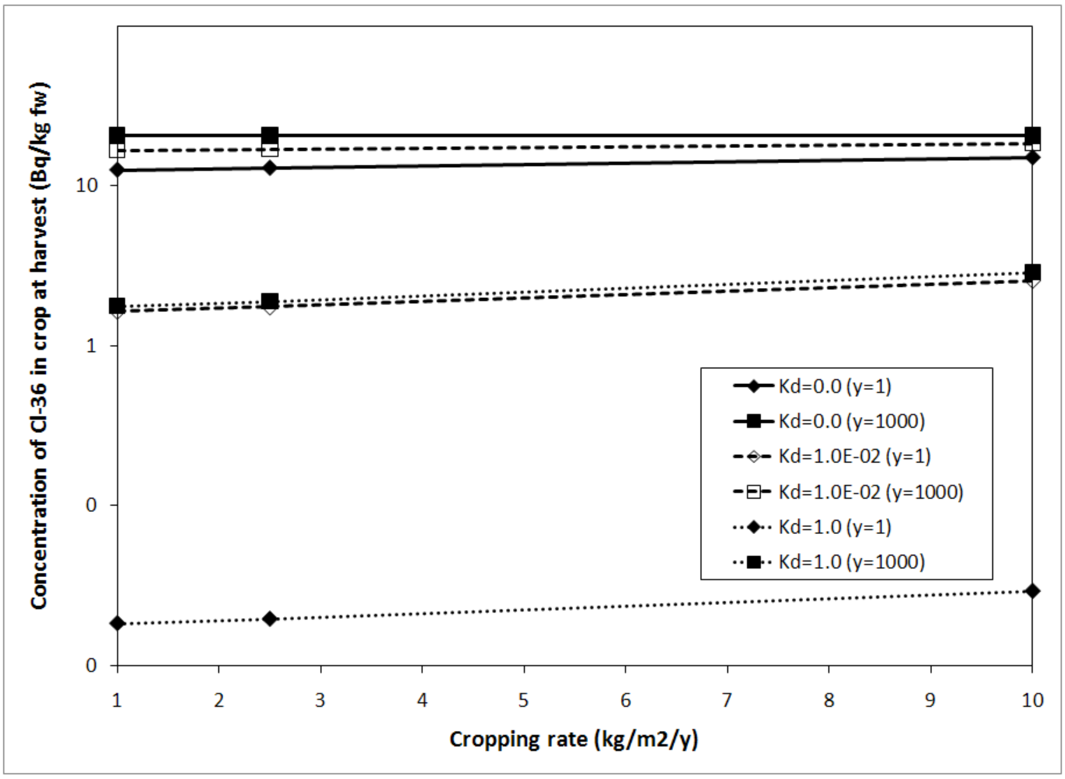


Figure 5-15: Sorption and cropping rate interaction with concentration of Cl-36 in crop at harvest in the NDA RWMD model



5.1.4 SENSITIVITY TO IRRIGATION AMOUNT

Irrigation alone does not explain the entire variability in calculated crop concentration of Cl-36 (Table 5-4 shows its impact for cereal). However, in all models, the calculated concentration of Cl-36 in the crops must necessarily be correlated to the irrigation amount (see Table 5-3 and Figure 5-16). This correlation between irrigation rate and the calculated crop Cl-36 concentration was found to be significantly positive both in conventional models as well as in the specific activity models (i.e. AquaCl36).

Within the EDF model, the irrigation rate is sensitive to the thickness of the topsoil layer, as irrigation events will occur only when the water content of the topsoil layer drops below a critical level, θ_{lim}. The thicker the topsoil layer, the more difficult it is for the water in the topsoil to reach θ_{lim}, and so more irrigation is applied to the system, resulting in an increase in the calculated Cl-36 concentration in the crop (Figure 5-17). However, the plant takes up chlorine from all the soil layers, dampening the effect an increase in topsoil Cl-36 concentration may have on the plants.

The sensitivity of calculated Cl-36 concentration in crops to irrigation rate was not considered for this scenario by the NDA RWMD model. However, sensitivity of crop concentrations to source influx for the groundwater scenario was considered (Section 5.3.3) and it is likely that the same observations would have been made for the irrigation scenario.

Figure 5-16: Effect of irrigation rate on the calculated Cl-36 concentration in cereal (Bq/kg f.w.) after 5 years

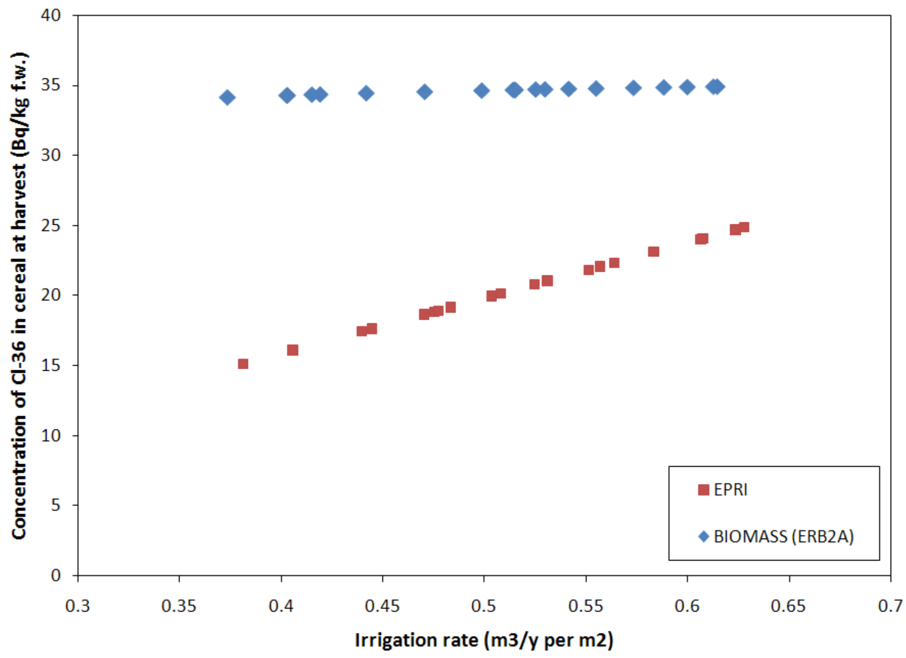
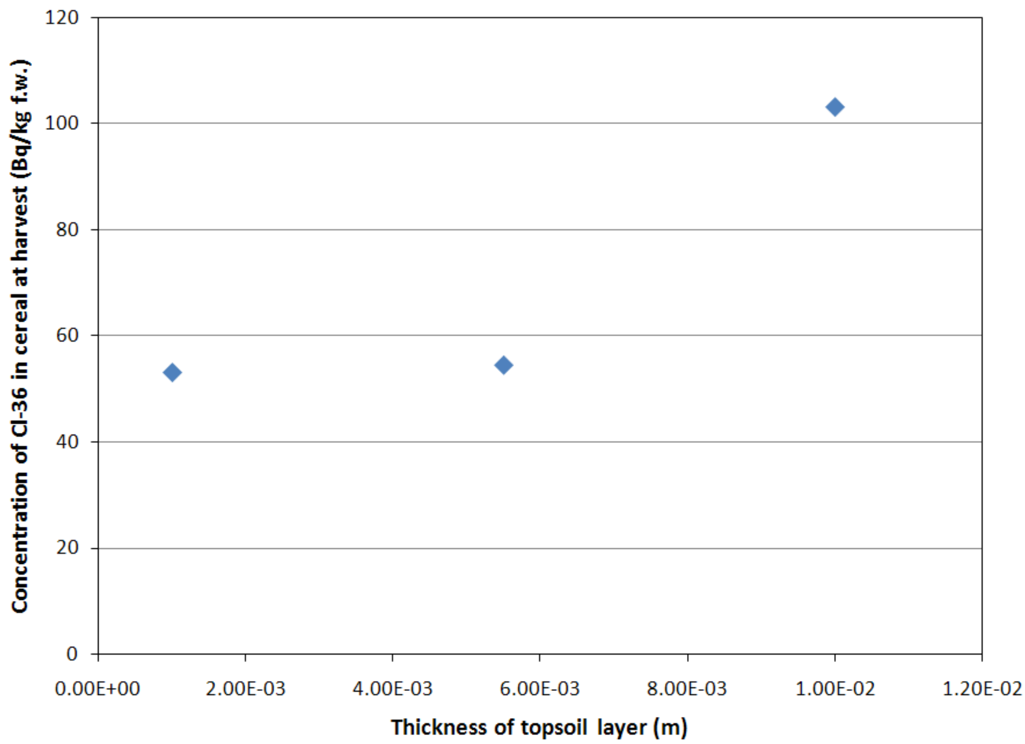


Figure 5-17: Effect of topsoil depth on calculated Cl-36 concentration in cereal using the EDF model



5.2 IRRIGATION WATER SOURCE SCENARIO – COASTAL SITE

EPRI, BIOMASS, JGC, ANDRA, SCM-ANDRA, EDF and NDA RWMD participated in the calculation exercises for the irrigation water source scenario at the coastal site. The only differences between the inland and coastal site are the amount of precipitation with a higher precipitation rate at the coastal site and the stable chlorine content of the precipitation and the well water, (Figure 2-6), and a greater stable chlorine concentration near the ocean (Section 3). The increase in precipitation at the coastal site served to reduce the irrigation rates used by most of the models (every model except the NDA RWMD model), though the change in best estimate of irrigation rate was less than 2% as compared with the inland site.

5.2.1 DETERMINISTIC CALCULATIONS

Within the conventional models, the reduction in irrigation rate at the coastal site led to a reduction in the soil Cl-36 concentration (Table 5-5) and thus a reduction in the plant Cl-36 concentration also. The increase in precipitation served to further decrease the soil Cl-36 concentration in Aquabios and SAMM-TR, which both explicitly consider precipitation for the water balance of the topsoil, so that the drop in calculated crop Cl-36 concentration is more appreciable in these models (cf leafy green vegetable concentrations at the different sites for these two models, Figure 5-2 and Figure 5-19).

Overall, with the exception of the EDF model, the predicted concentration of Cl-36 in any given crop was lower at the coastal site than at the inland site, with the coastal concentration typically anywhere from one third of the inland concentration to barely any difference (see Figure 5-18, Figure 5-19, Figure 5-20 and Table 5-6).

In the EDF model, the stable chlorine concentration assumed for both the plants and the irrigation water at the coastal site was four times higher than the stable chlorine content used for the inland site. Combining this with the increased stable chlorine content of the precipitation then explains the approximate reduction by a factor of two of the calculated crop Cl-36 concentrations at the coastal site compared to the inland site (Table 5-6).

No data was given for stable Cl at the coastal site. ANDRA assumed a stable chlorine concentration in coastal plants about twice that of the inland plants for their AquaCl36 and SAMM-IR models, but a substantially higher increase in the stable Cl concentration of in the irrigation and the rain water (22 times for irrigation). This explains the substantial reduction in calculated plant Cl-36 concentrations observed for these two models (Table 5-6).

The effect of altering the stable chlorine concentration in the plants has been further examined using the NDA RWMD model, for the groundwater scenario (Section 5.4).

Figure 5-18: Irrigation scenario [coastal site]. Deterministic calculations of Cl-36 in root vegetables (Bq kg^{-1} fw)

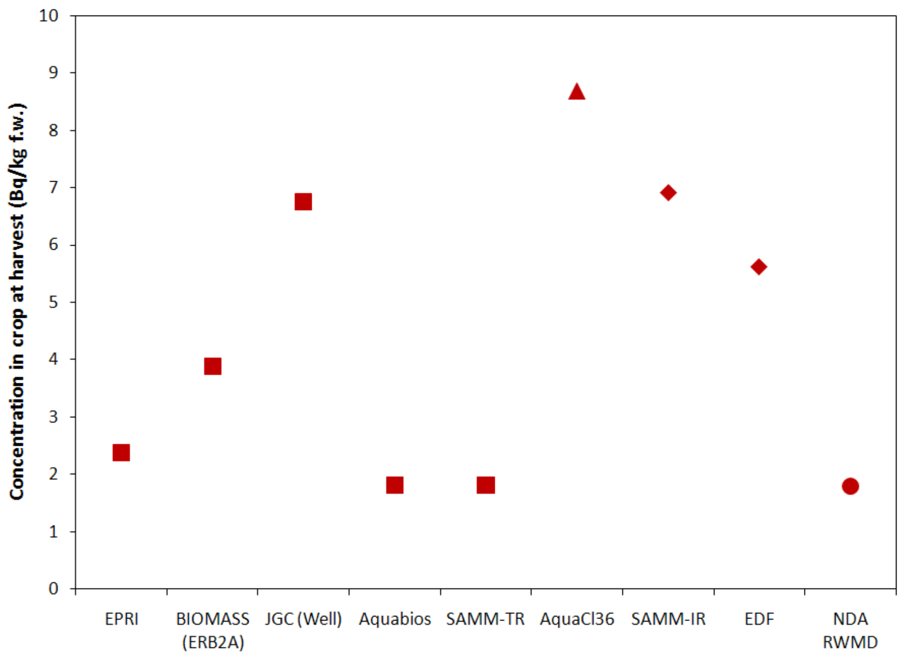


Figure 5-19: Irrigation scenario [coastal site]. Deterministic calculations of Cl-36 in leafy green vegetables (Bq kg^{-1} fw)

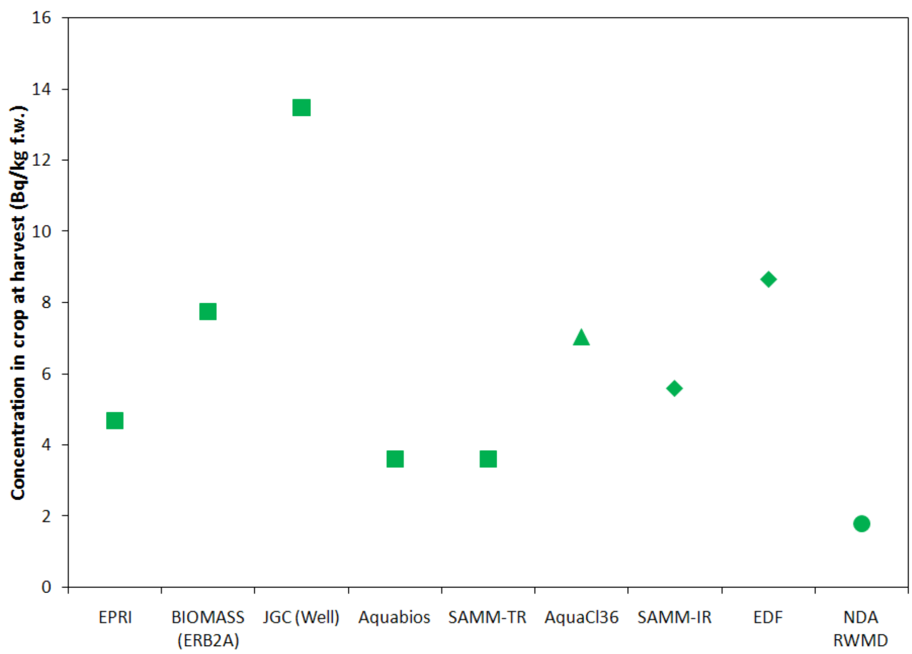


Figure 5-20: Irrigation scenario [coastal site]. Deterministic calculations of Cl-36 in cereal (Bq/kg fw)

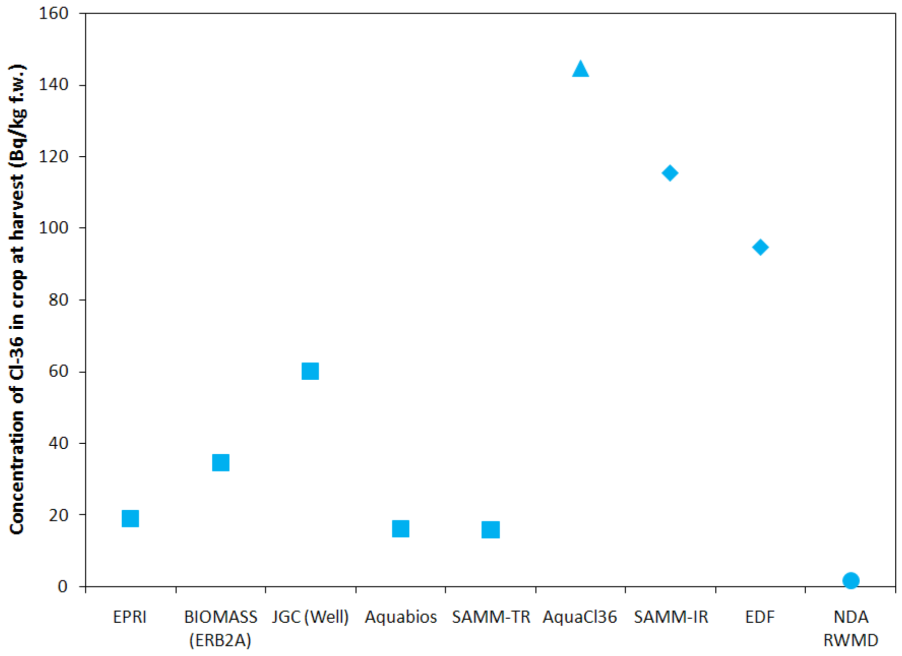


Table 5-5: Concentration of Cl-36 in the soil after 5 years of simulation [coastal site]

Model Type	Model	Topsoil concentration of Cl-36 (Bq/kg_{dry})	Ratio of topsoil Cl-36 concentration: coastal / inland
Conventional	EPRI [a]	Root vegetables: 1.58E-01 Leafy green vegetables: 1.56E-01 Cereal: 1.41E-01	Root vegetables: 9.88E-01 Leafy green vegetables: 9.87E-01 Cereal: 9.86E-01
	BIOMASS [b]	2.58E-01	9.99E-01
	JGC	4.49E-01	1.00E+00
	Aquabios	1.20E-01	7.50E-01
	SAMM-TR [c]	1.20E-01	5.71E-01
Specific Activity	AquaCl36	1.40E-01	8.24E-01
Compartmental with isotope ratio approach for plant uptake	SAMM-IR	1.20E-01	5.71E-01
	EDF [c, d]	Root vegetables: 1.40E+00 Leafy green vegetables: 7.03E-01 Cereal: 6.44E-01	Root vegetables: 1.00E+00 Leafy green vegetables: 1.00E+00 Cereal: 1.11E+00
More complex	NDA RWMD [e]	2.37E+01	1.57E+00

[a] A soil porosity of 0.5 and a soil density of 2650 kg m^{-3} were used to convert the Cl-36 from Bq m^{-3}.
[b] A soil porosity of 0.4 and a soil density of 2600 kg m^{-3} were used to convert the Cl-36 from Bq m^{-3}.
[c] A soil porosity of 0.45 and a soil density of 2600 kg m^{-3} were used to convert the Cl-36 from Bq m^{-3}.
[d] There is a difference between the crops because there is a different irrigation. This concentration are those in the topsoil (55×10^{-3} m). The concentrations in the sub-soil layers are lower than those in the topsoil.
[e] A soil porosity of 0.5 and a soil density of 1325 kg m^{-3} was used to convert the Cl-36 from Bq m^{-3}.

Table 5-6: Ratio of concentration of Cl-36 in crop at harvest after 5 years, coastal/inland (Bq/kg f.w.)

Model Type	Model	Root Vegetables	Leafy Green Vegetables	Cereal
Conventional	EPRI	9.87E-01	9.87E-01	9.87E-01
	BIOMASS	9.99E-01	9.99E-01	9.99E-01
	JGC	9.99E-01	9.99E-01	9.99E-01
	Aquabios	7.33E-01	7.33E-01	7.33E-01
	SAMM-TR	5.67E-01	5.67E-01	5.67E-01
Specific Activity	AquaCl36	3.40E-01	3.40E-01	3.40E-01
Compartmental with isotope ratio approach for plant uptake	SAMM-IR	4.09E-01	4.09E-01	4.09E-01
	EDF	2.10E+00	2.36E+00	1.74E+00
More complex	NDA RWMD	5.27E-01	5.27E-01	5.27E-01

5.2.2 PROBABILISTIC CALCULATIONS

EPRI, BIOMASS, JGC and ANDRA carried out a sensitivity analysis of the crop concentration of Cl-36 with respect to uncertainty in a range of model parameters. The arithmetic mean simulated crop concentrations of Cl-36 together with the 5% and 95% percentiles of the CDF of results (box), and the minimum and maximum (the vertical lines) from these simulations are shown in Figure 5-21, Figure 5-22 and Figure 5-23. In addition, the results from the deterministic calculations are overlaid onto the sensitivity simulations. Due to the large variation in calculated concentrations, the y-axis in each graph is on a log-scale. The parameters varied in each model were identical to those for the inland site (Table 5-2). As was observed for the inland site, the variability within a given model due to parameter uncertainty (typically ranging from a factor of 13 to a factor of 270[a]) is greater than the variability between the models in the deterministic calculations (ranging from a factor of 5 to a factor of 90[b]).

In general, the model results are sensitive to the same parameters as for the inland site. The one exception is the amount of irrigation water, which, apart from in AquaCl36, no longer has a significant effect on the calculated crop Cl-36 concentrations.

[a] In the stochastic calculations, the concentration of Cl-36 in the cereal varied by a factor of 660 in AquaCl36 (based upon minimum and maximum predicted concentrations).

[b] If AquaCl36 and the NDA RWMD model are excluded from the analysis in the cereals, the deterministic calculations varied by up to a factor of 7.

Figure 5-21: Stochastic calculations - root vegetables [coastal site].

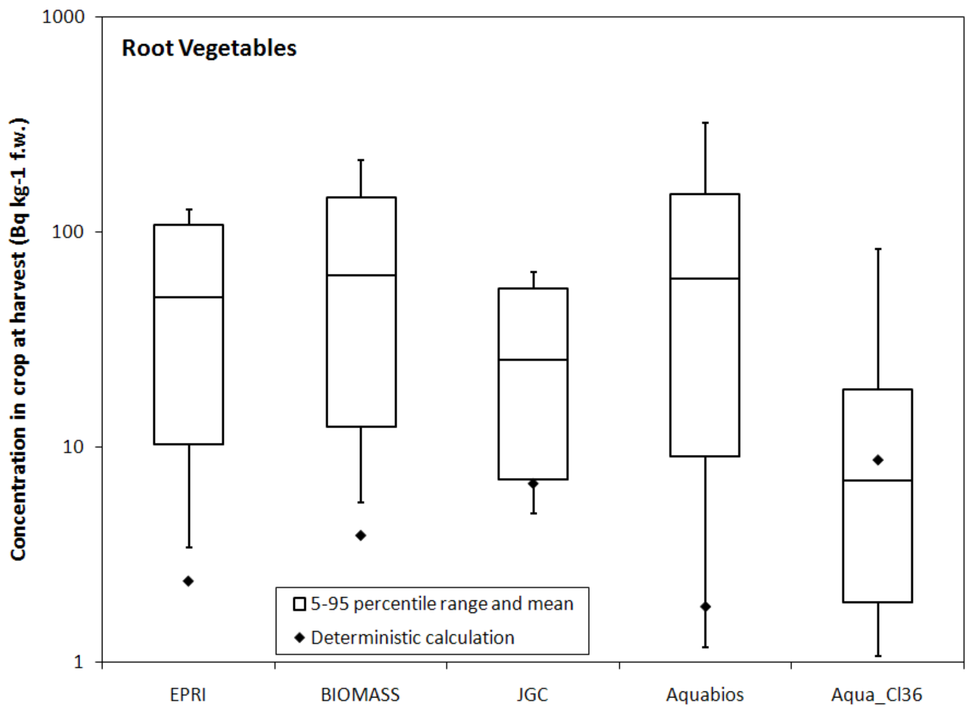


Figure 5-22: Stochastic calculations – leafy green vegetables [coastal site].

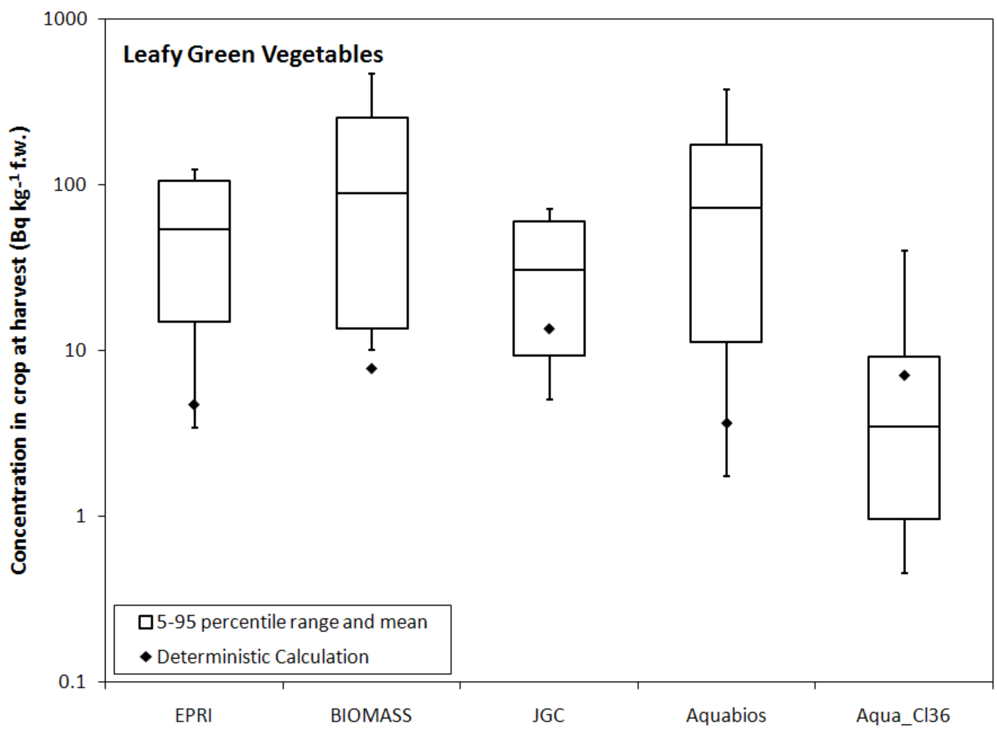
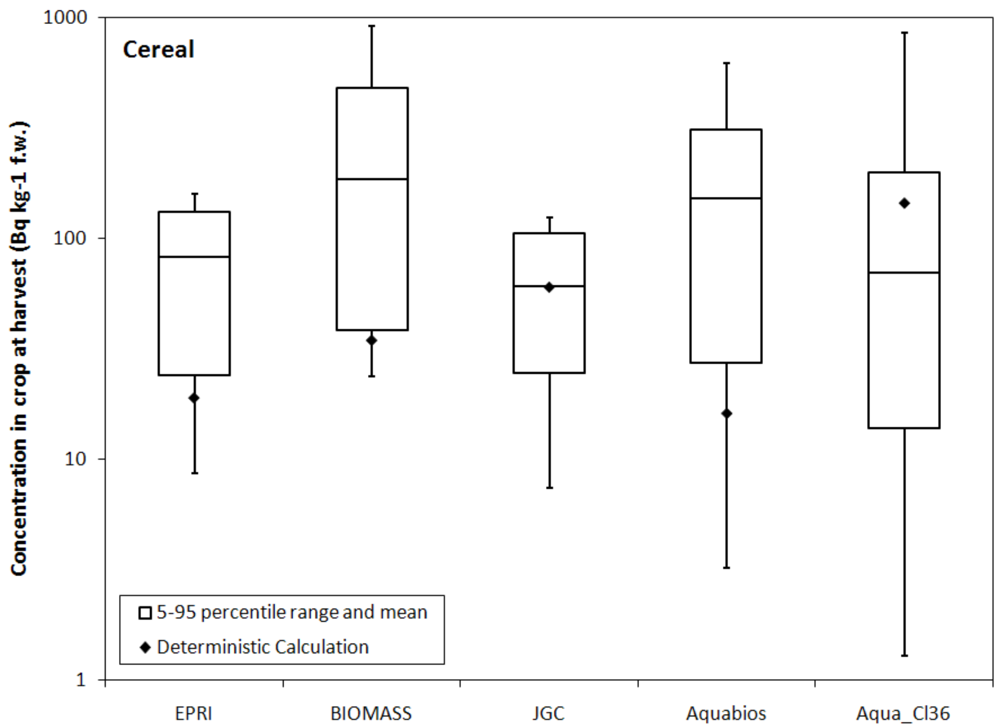


Figure 5-23: Stochastic calculations – cereal [coastal site].



5.3 NATURAL GROUNDWATER SOURCE SCENARIO

BIOMASS, JGC, SCM-ANDRA, EDF and NDA RWMD participated in the calculation exercises for the groundwater scenario. However, given the climate data provided for the two sites, neither SCM-ANDRA nor EDF experienced capillary rise in their models, so no results from these two participants can be presented.

In the first two parts of this section, results from simulations of the two sites are discussed, including comparisons with equivalent results from the irrigation scenario. In the third part of this section, the NDA RWMD model is used to examine how changes in the rate of groundwater influx to the soil would impact upon plant Cl-36 concentrations. In the fourth part of this section, the NDA RWMD model is used to examine the effect of altering assumptions about the stable chlorine concentration of the plant.

5.3.1 INLAND SITE

BIOMASS, JGC and NDA RWMD participated in simulations of the inland site, with the crop concentrations of Cl-36 after five years given in Table 5-7.

Within the conventional models, as compared with the irrigation scenario there is reduction in calculated crop Cl-36 concentration of over 78% (Table 5-8). The combined effect of a low intake rate of contaminated groundwater into the subsoil combined with a low rate of capillary rise leads to the low Cl-36 concentration in the topsoil. Thus, for example, in the BIOMASS model there is a reduction in the calculated soil Cl-36 concentration of nearly two orders of magnitude between the scenarios (Figure 5-24).

The crop Cl-36 concentrations as calculated by the NDA RWMD model are 50% higher than those given for the irrigation scenario, which can be explained in part by the increase in Cl-36 concentration of the topsoil (Figure 5-25). As with the irrigation scenario, it took longer than 5 years for the NDA RWMD model to reach equilibrium (cf. Figure 5-4).

Table 5-7: Groundwater scenario [inland site]. Concentration of Cl-36 in crops (Bq kg^{-1} f.w.) after five years

		Crop concentration of Cl-36 after five years (Bq kg^{-1} f.w.)		
Model Type	**Model**	**Root Vegetables**	**Leafy Green Vegetables**	**Cereal**
Conventional	BIOMASS	4.96E-02	9.93E-02	4.44E-01
	JGC	1.44E+00	2.89E+00	1.29E+01
More Complex	NDA RWMD	5.28E+00	5.28E+00	5.28E+00

Table 5-8: Ratio of crop concentration of Cl-36 for the inland site dependent upon contamination scenario: groundwater / irrigation

		Ratio of crop concentration of Cl-36 after five years (Bq kg^{-1} f.w.) Groundwater / Irrigation		
Model Type	**Model**	**Root Vegetables**	**Leafy Green Vegetables**	**Cereal**
Conventional	BIOMASS	1.28E-02	1.28E-02	1.28E-02
	JGC	2.14E-01	2.14E-01	2.14E-01
More Complex	NDA RWMD	1.56E+00	1.56E+00	1.56E+00

Figure 5-24: Difference between the scenarios - concentration of Cl-36 in the soil in the BIOMASS models. Note the logarithic vertical axis.

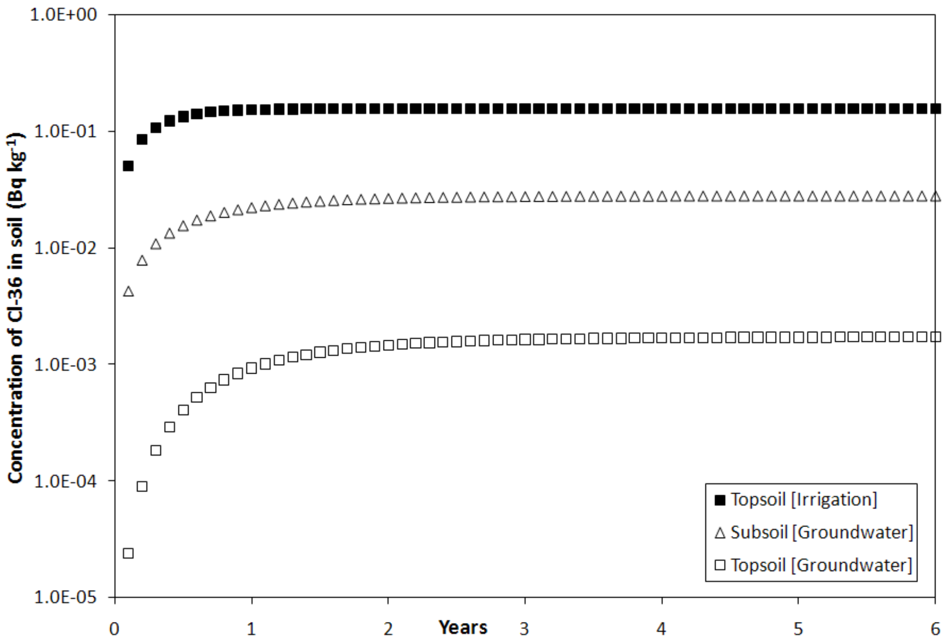
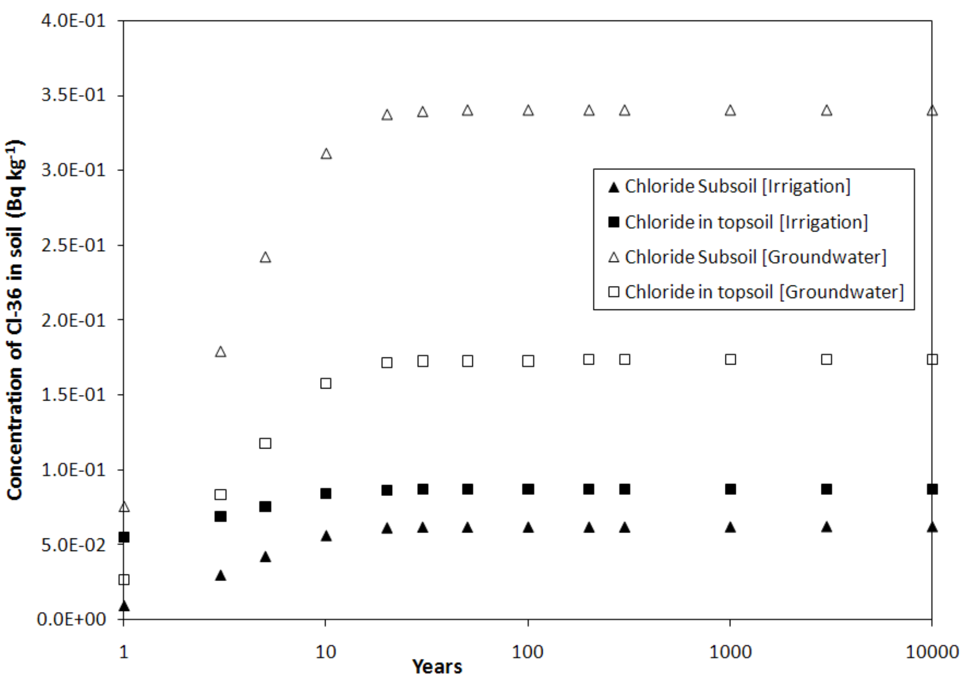


Figure 5-25: Difference between the scenarios - concentration of Cl-36 in the soil (Bq kg^{-1}) in the NDA RWMD model



5.3.2 COASTAL SITE

BIOMASS, JGC and NDA RWMD participated in simulations of the coastal site, with the crop concentrations of Cl-36 after five years given in Table 5-9. As neither the BIOMASS nor JGC models take precipitation explicitly into account the calculated crop calculations are identical to those given for the inland site. As was observed for the simulations of the irrigation scenario, the calculated crop concentration of Cl-36 in the NDA RWMD model is lower for the coastal site than the inland site, with a coastal/inland ratio identical to the irrigation scenario (Table 5-6).

Table 5-9: Groundwater scenario [coastal site]. Concentration of Cl-36 in crops (Bq kg-1 f.w.) after five years

		Crop concentration of Cl-36 after five years (Bq kg^{-1} f.w.)		
Model Type	**Model**	**Root Vegetables**	**Leafy Green Vegetables**	**Cereal**
Conventional	BIOMASS	4.96E-02	9.93E-02	4.44E-01
	JGC	1.44E+00	2.89E+00	1.29E+01
More Complex	NDA RWMD	2.78E+00	2.78E+00	2.78E+00

5.3.3 EFFECT OF ALTERING RATE OF GROUNDWATER INFLOW

In addition to the deterministic calculations presented above, NDA RWMD examined the effect of altering the groundwater up-flow rate, having previously noted that the amount of Cl-36 entering the subsoil was directly proportional to this rate (Thorne, 2007b). For reference, the basic model structure representing the flow of chloride and water is repeated here (Figure 5-26).

The effect of reducing the groundwater up-flow rate (G) by a factor of ten was investigated. To ensure that the loss from the subsoil (L_2) remained at zero the capillary driven upward movement from the subsoil to the topsoil (E_2) was reduced from 0.35 to 0.26 m y^{-1}. This calculation was carried out for the coastal site only. Results were about one order of magnitude lower than in the original simulations, as illustrated in Figure 5-27 and by the ratios given in Table 5-10. These results are reflect similar findings from the irrigation scenario, whereby a reduction in the inflow rate of contaminated water lead to a reduction in the Cl-36 concentration in the plants of the same order of magnitude.

Figure 5-26: Structure of the NDA RWMD Cl-36 Model showing water flows

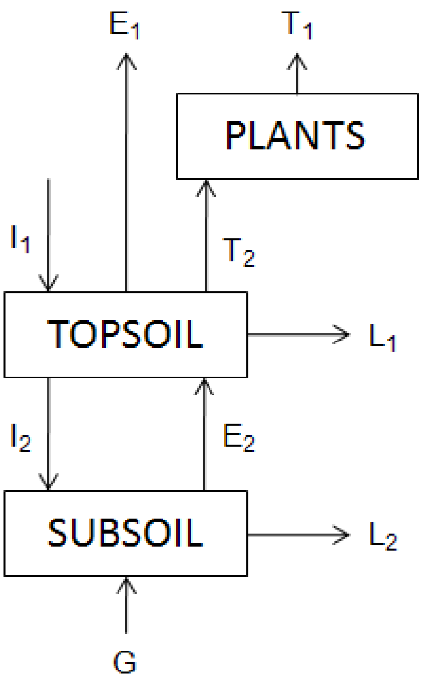


Figure 5-27: Concentration of Cl-36 in crop (Bq kg^{-1} f.w.) dependent upon groundwater up-flow rate

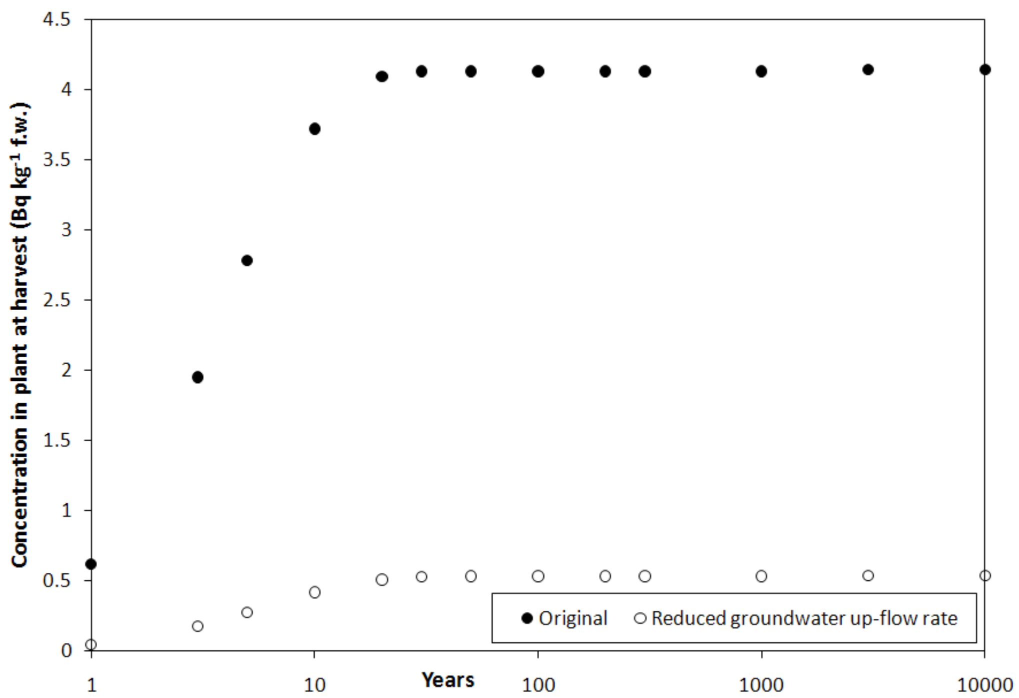


Table 5-10: Ratio of concentrations of Cl-36 in the three compartments of the NDA RWMD model as a result of reducing groundwater up-flow rate

Time (y)	Chloride Plants	Chloride Topsoil	Chloride Subsoil
1	8.38E-02	8.49E-02	1.04E-01
3	9.33E-02	9.43E-02	1.11E-01
5	9.96E-02	1.01E-01	1.18E-01
10	1.12E-01	1.13E-01	1.32E-01
20	1.24E-01	1.26E-01	1.45E-01
30	1.28E-01	1.29E-01	1.49E-01
50	1.29E-01	1.30E-01	1.50E-01
100	1.29E-01	1.30E-01	1.50E-01
1000	1.29E-01	1.30E-01	1.50E-01
10000	1.29E-01	1.30E-01	1.50E-01

5.4 EFFECT OF ALTERING THE STABLE CHLORINE CONCENTRATION IN PLANTS

In the irrigation scenario, the effect of altering the stable chlorine concentration of the plants to reflect differences between the inland and coastal sites was considered. However, the effects observed in the irrigation scenarios were compounded by additional site specific model assumptions. In particular, the stable chlorine concentration of the irrigation water was assumed to differ between the sites.

It was found that quadrupling the stable chlorine concentration of the plants (and irrigation water) in the EDF model to reflect background chlorine levels at the coastal site led to an increase in calculated Cl-36 concentrations. In contrast, a doubling of the stable chlorine concentration in the AquaCl36 and SAMM-IR models led to a large reduction in the calculated crop Cl-36 concentrations, though the increase in stable chlorine concentration of the plants may have been negated by the increase in the stable concentration in the irrigation water of a factor of 22. However, the three models concerned here also have other substantial differences which could contribute to the differences,

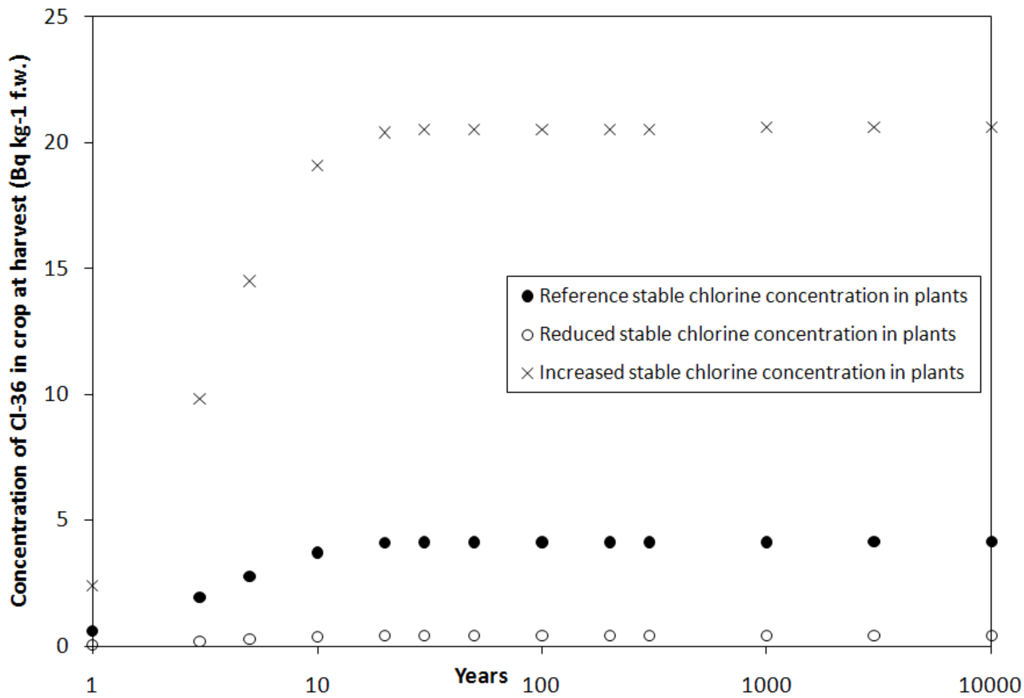
The NDA RWMD model was then used to investigate the effect of stable chlorine concentration in the plant by carrying out some variant simulations of the groundwater scenario at the coastal site. The stable chlorine concentration in any source water was not altered. The results from three cases are presented here (Figure 5-28):

- The reference case, stable chlorine concentration in the plants of 1.40E-01 mol kg^{-1} (d.w.)
- The reduced case, stable chlorine concentration in the plants of 1.40E-02 mol kg^{-1} (d.w.)

- The increased case, stable chlorine concentration in the plants of 7.00E-01 mol kg^{-1} (d.w.)

Reducing the stable chlorine concentration in the plant leads to a reduction in the calculated Cl-36 concentration in the plant. Similarly, an increase in the assumed stable chlorine concentration in the plant results a higher plant Cl-36 concentration. The significance of these changes in plant concentrations of stable chlorine on the calculated plant Cl-36 concentrations is most easily seen by taking ratios of the variant cases with the reference case (0.1 for the reduced case and 5 for the increased case).

Figure 5-28: Concentration of Cl-36 in crop (Bq kg-1 f.w.) dependent upon stable chlorine concentration in the crop. [Simulations carried out using the NDA RWMD model.]



6 CONCLUSIONS AND RECOMMENDATIONS

6.1 OVERVIEW OF THE IMPLICATIONS OF THE RESULTS OBTAINED USING DIFFERENT MODELS

There is a generic question about how much uncertainty is acceptable within long term dose assessments and many causes of uncertainty in assessment results. These apply to Cl-36 releases as much as any others. However the following discussion is limited to Cl-36 behaviour in soils and uptake into crops.

The different assessment contexts include issues such as whether climate and environmental change should be used, or only the site's present day condition. This will have a material impact on the relevance and need for site specific data, and/or data from other sites which could be analogues today for the future condition of the site under investigation.

A significant range of model types and alternative data assumptions has been described and compared, demonstrating the degree of understanding of the likely behaviour of Cl-36 soil and plant systems. Results have been presented for Cl-36 concentrations in crops based on unit activity concentration in upwelling groundwater and irrigation water, both deterministic and probabilistic, and at defined inland and coastal locations. A wide range of sensitivity calculations has also been presented.

Notwithstanding the different level of model complexity and the variations in the processes included, and the data assumption differences, the results for concentrations in crops are typically within about an order of magnitude of each other for the main scenario considered[a]. Furthermore, the spread of results obtained in probabilistic calculations does not indicate much greater variation for a given set of site conditions. This implies that knowledge of stable Cl levels is critical for the specific activity models, and for other models, that the soil and plant systems must be sufficiently well characterised to allow usefully confident choices of distribution coefficients and root uptake factors, or to provide the other more detailed data. This is a truism, but something not to be forgotten. The model input data requirements in turn help to define the site investigation requirements.

The results presented here offer some insight into how both model and parameter assumptions can affect the results, demonstrating that some best estimate calculations lie outside the range of the probabilistic calculations. This can be viewed as an illustration of the need for further research to better understand these conceptual uncertainties.

The range of results presented here, in combination with the model explanations, can support consideration of whether particular model assumptions can be considered as overly-cautious, cautious or more likely to represent the likely outcome in particular contexts.

[a] The exception to this is cereals, where the difference was up to three orders of magnitude. However, this can be explained by not all of the models using crop specific parameters for the uptake of Cl-36 and, if appropriate, stable chlorine.

6.2 SCIENTIFIC APPROACH TO CL-36 UPTAKE FROM SOILS TO CROPS

Section 2 of this report discussed the various scientific approaches that are used to assess the uptake of Cl-36 from soils to crops, highlighting the similarities and differences between the models (Section 2.5). The specific activity models (Section 2.2), were arguably the simplest models, with the compartmental model developed by EDF (Section 2.3.2) and the NDA RWMD model (Section 2.4.2) at the opposite end of the model complexity spectrum.

6.2.1 Processes considered

The loss of Cl-36, and, if appropriate, stable chlorine also, to deeper soil layers was included as a model process in the majority of the models used in this study, the exception being the specific activity models. The significance of the exclusion or inclusion of this process for Cl-36 is best discussed in light of the illustrative calculations which were carried out by the project participants (Section 5). Almost without exception, the highest calculated crop Cl-36 concentrations were from the specific activity model AquaCl36, irrespective of the crop, site or scenario under consideration[a].

There is a need to be clear as to which compartments of a system are assumed to be in equilibrium with each other, with respect both to modelling and experiments. In addition, when empirical data is provided for the input parameters to any model, the representativeness of those measurements to the temporal and spatial assumptions within the model must be understood and correspond. This is of particular importance for the stable chlorine to Cl-36 ratio used in specific activity, and isotope ratio uptake, models. Field measurements of stable chlorine and Cl-36 depend upon recent climatic conditions, such that field conditions may not be at equilibrium at the time the measurements were taken.

It is also important to clearly specify which part of the plant is being treated (e.g. whole plant, grain, stem). For example, Kashparov et al (2007b) reported concentration ratios of Cl-36 for the grain and straw components of both wheat and oats. There was at least a factor of 6 difference in the reported concentration ratio, dependent upon the plant component under consideration, with the highest uptake occurring in the straw.

One of the parameters found to have most impact upon the calculated crop Cl-36 concentrations was the sorption coefficient of Cl-36 to the soil (Section 5.1.3). For radionuclides which can accumulate in the soil the parameters used to describe this process will impact upon both the (peak) equilibrium concentration of radionuclide in the soil and plant and also the length of time it takes for equilibrium to be reached (e.g. Figure 5-10).

It was then shown that if Cl-36 is thought to accumulate in the soil then it becomes increasing important to have an understanding of the agricultural practice at the site under consideration. It is generally thought that an increase in harvest yield of crops will reduce the Cl-36 concentration in the soil, and therefore the concentration of Cl-36 in the crops (Figure 5-13 and Figure 5-14), although there is some evidence to the contrary (Figure 5-15).

[a] The one exception was leafy green vegetables at the coastal site within the irrigation scenario (Section 5.2, Figure 5-19).

6.2.2 UTILITY OF COMPLEX MODELS

The confidence limits presented in the probabilistic results (Sections 5.1.2 and 5.2.2) reflect for each model the range of uncertainty assuming each model is conceptually correct. The spread in results across models reflects the range in results including conceptual uncertainty as well as parameter uncertainty. Given that the variability between the models for the deterministic calculations was less than the variability within a model in the probabilistic results, it is not possible to conclude that the more complex models reduce uncertainties. Thus, the results from this study do not lead to the conclusion that chlorine specific processes need to be included in a model of its uptake into plants from the soil. However, more detailed measurements of the uptake of Cl-36 are required in order to increase confidence in the model outputs. It is this which will help to constrain the range in the results, making site specific data a key factor in reducing uncertainty in the results.

Any model used for assessing radionuclide uptake from soil into plants needs to address the level of conservatism used for processes for which the data are limited. This applies to the inclusion of processes as well as parameter values. If the radionuclide is known to accumulate in soil, then both the inclusion of this accumulation and also of the harvesting processes will increase the realism of the model, and take into account the interaction between these parameters.

In order to determine the concentration of Cl-36 in a specific crop the rate of uptake needs more precise contextual description in order to allow more accurate parameterisation. However, this study cannot be used to conclude whether the concentration ratio or isotope ratio is best. The way to resolve this is via further experimentation and measurements, both in the laboratory and the field.

6.3 RECOMMENDATIONS FOR THE FUTURE

Focussing on the dynamics of Cl-36 in soil, some knowledge gaps remain. Although some assumptions were made about the dynamics of organic chlorine and the inorganic-organic chlorine conversion (Section 2.4.2), empirical data to justify including such processes in a conceptual model are currently limited. In comparing the two sites, different assumptions about the (background) levels of stable chlorine in both the soil and precipitation lead to substantial variation in modelled Cl-36 concentrations in the plants. Further empirical research in these areas would increase our understanding of such processes and therefore improve our conceptual models.

More generally, there is a need for the assessment process to be informed of how much uncertainty is acceptable, within the context of a particular assessment. This requirement applies to all the relevant radionuclides and work to develop an understanding of how to determine the level of acceptable uncertainty in a specific context could be useful across the board.

The application of the models used here could be extended to the calculation of doses to potentially exposed groups. Different approaches are employed to definition of these groups and that comparison would be instructive, both for Cl-36 and generally. The purpose would not be to identify a best approach but to investigate how different assumptions affect the results and also to investigate whether the variation in results is dominated by variant assumptions in the calculation of concentrations in crops, or in those for human behaviour. In combination, these activities could then inform requirements for future investigations at the site specific level.

7 REFERENCES

Abbatt M L and Rood A S (1994). COMIDA: a radionuclide food chain model for acute fallout deposition. Health Physics, 66 (1), 17-29

Albrecht A (2004) Intégration dans le code de calcul Aquabios d'un modèle du comportement de Cl-36 dans la biosphère basée sur le rapport isotopique et analyse de sensibilité; Rapport Andra n° C.NT.ASTR.04.007.

Albrecht A (2007) Les équations du modèle de transfert des radionucléides dans la biosphère (Aquabios) et leur intégration dans le code " Modèle Management, MoM "; Rapport Andra n° C.NT.ASTR.06.0036.B.

Andra (2005) Dossier 2005 - Référentiel "Comportement des radionucléides et des toxiques chimiques d'un stockage dans le Callovo-Oxfordien jusqu'à l'homme" - Site de Meuse/Haute Marne; Rapport Andra n° C.RP.ASTR.04.0032.B.

Baes C F and Sharp R D (1983). A proposal for estimation of soil leaching and leaching constants for use in assessment models. Journal of Environment Quality, 12 (1), pp. 17-28.

Bailer-Jones C A L, Bhadeshia H K D H and MacKay D J C (1999). Gaussian process modelling of austinite formation in steel. Materials Science and Technology 15:287-294.

Barber S A (1984). Soil Nutrient Bioavailability: A Mechanistic Approach, John Wiley and Sons, New York.

BIOMASS (2003). “Reference Biospheres” for solid radioactive waste disposal. Report of BIOMASS Theme 1 of the BIOsphere Modelling and ASSessment (BIOMASS) Programme. IAEA-BIOMASS-6. International Atomic Energy Agency, Vienna.

BIOPROTA (2005a). Model Review and Comparison for Spray irrigation Pathway. A report prepared within the international collaborative project BIOPROTA: Key Issues in Biosphere Aspects of Assessment of the Long-term Impact of Contaminant Releases Associated with Radioactive Waste Management. Main Contributors: U Bergstrom, A Albrecht, B Kanyar, G Smith, M Thorne, H Yoshida and M Wasiolek. Published on behalf of the BIOPROTA Steering Committee by SKB (Swedish Nuclear Fuel and Radioactive Waste Management Company), Stockholm.

BIOPROTA (2005b). Model Intercomparison with Focus on Accumulation in Soil. A report prepared within the international collaborative project BIOPROTA: Key Issues in Biosphere Aspects of Assessment of the Long-term Impact of Contaminant Releases Associated with Radioactive Waste Management. Main Contributors: A Albrecht, C Damois, E Kerrigan, R Klos, G Smith, M Thorne, M Willans and H Yoshida. Published on behalf of the BIOPROTA Steering Committee by ANDRA (Agence nationale pour la gestion des déchets radioactifs), Châtenay-Malabry,_France.

BIOPROTA (2006). Report of an International Forum on Cl-36 in the Biosphere Châtenay-Malabry, France, 27-28 September 2006. Published on behalf of the BIOPROTA Sponsoring Committee by ANDRA (Agence nationale pour la gestion des déchets radioactifs), Châtenay-Malabry,_France.

Box G E P, Hunter W G and Hunter J S (1978). Statistics for experimenters; An introduction to design, data analysis, and model building John Wiley & Son, New York.

Ciffroy P, Siclet F, Damois C, Luck M and Duboudin C (2005) A dynamic model for assessing radiological consequences of routine release in the Loire river: parameterisation and uncertainty: sensitivity analysis. Journal of Environmental Radioactivity, 83, 9-48

Coughtrey (1983). Radionuclides distribution and transport in terrestrial and aquatic ecosystems. AA Balkema, Rotterdam, Netherlands

Davis P A, Zach R, Stephens M E, Amiro B D, Bird G A, Reid J A K, Sheppard M I , Sheppard S C, and Stephenson M (1993) The disposal of Canada's nuclear fuel waste: The biosphere model, BIOTRAC, for postclosure assessment; Rapport Whiteshell Laboratories n° AECL-10720, COG-93-10.

EPRI (2002) Evaluation of the Proposed High-Level Radioactive Waste Repository at Yucca Mountain UsingTotal System Performance Assessment: Phase 6, EPRI, Palo Alto, CA: 2002. 1003031.

Epstein E (1966). Dual patterns of ion adsorption by plant cells and by plants, Nature, **212**, 1324-1327.

Freundlich H (1907) Adsorption Isotherms. Physikalische Chemie, 57(A) pp. 385-470.

Gierszewski et al. (2004). SYVAC3-CC4 THEORY (Version CC4.05). Ontario Power Generation, Nuclear Waste Management Division Report 06819-REP-01300-10072-R00.

IAEA (1994). Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments, IAEA Technical Reports Series No. 364, International Atomic Energy Agency, Vienna.

IAEA (1996) The IAEA Programme on Biosphere Modelling and Assessment Methods: Thems for a New Co-ordinated Research Programme on Environmental Model Testing and Improvement, Theme 1: Radioactive Waste Disposal, Theme 2: Environmental Releases, Theme 3: Biosphere Processes, BIOMASS/G/WD01.

Jenkinson D S and Rayner J H (1977). The turnover of soil organic matter in some of the Rothamsted classical experiments, Soil Science, 123, 298-305.

JNC (1999). H12 : Project to Establish the Scientific and Technical Basis for HLW Disposal in Japan. JNC TN1400 99-023.

Lawson G and Smith G M (1985). BIOS: A Model to Predict Radionuclide Transfer and Doses to Man Following Releases from Geological Repositories. National Radiological Protection Board, NRPB-R169, HMSO, London.

Kashparov V, Colle C, Zvaricha S, Yoschenko V, Levchuk S and Lundin S (2005a). Soil-to-plant halogens transfer studies 1. Root uptake of radioiodine by plants. Journal of Environmental Radioactivity 79 (2005a) 187–204.

Kashparov V, Colle C, Zvaricha S, Yoschenko V, Levchuk S and Lundin S (2005b). Soil-to-plant halogens transfer studies 2. Root uptake of radiochlorine by plants. Journal of Environmental Radioactivity 79 (2005) 233-253.

Kashparov V, Colle C, Levchuk S, Yoschenko V and Svydynuk N (2007a). Transfer of chlorine from the environment to agricultural foodstuffs. Journal of Environmental Radioactivity 94 (2007) 1-15.

Kashparov V, Colle C, Levchuk S, Yoschenko V and Zvarich S (2007b). Radiochlorine concentration ratios for agricultural plants in various soil conditions. Journal of Environmental Radioactivity 95 (2007) 10-22.

Kennedy M C (2004). Description of the Gaussian process model used in GEM-SA. University of Sheffield.

Kennedy M C and O'Hagan A (2001). Bayesian calibration of computer models. Journal of the Royal Statistical Society B 63: pp425-464.

Kennedy M C,O'Hagan A, Anderson C W, Lomas M, Woodward F I, Heinemeyer A and Gosling J P (2008). Quantifying uncertainty in the biospheric carbon flux for England and Wales. Journal of the Royal Statistical Society. Series A, 171: pp 109-135.

Klos R (2007). *A multicompartment soil model in MoM*, Rapport Aleksandria Sciences ASN 07-02, 2007.

Kramer P J (1959). Transpiration and the water economy of plants. In *Plant physiology v2* (ed.) F C Steward (New York: Academic Press) pp 607–726

Leclerc-Cessac E (2004) Projet de stockage en subsurface de Dechets FAVL graphites. Dossier d'Allocation de Performance. Calculs préliminaires des facteurs de conversion biosphère du chlore 36; Rapport Andra n° F NT ASTR 02-073.

Likar B, and Kocijan J (2007). Predictive control of a gas-liquid separation plant based on a Gaussian process model. Computers & Chemical Engineering 31:142-152.

Little R H (2002). The Identification and Justification of an Assessment Biosphere for a Soil Receptor. Quintessa Report QRS-1111A-9 v1.0, Quintessa, Henley-on-Thames, UK.

Little R H, Watkins B M, Egan M J, Walke R C and Smith G M (2002a). Identification of Generic Concepts and Associated Geosphere-Biosphere Interfaces. Quintessa Report QRS-1111A-1 v1.0, Quintessa, Henley-on-Thames, UK.

Little R H, Watkins B M, Walke R C and Smith G M (2002b). The Identification and Justification of an Assessment Biosphere for a Surface Freshwater Receptor. Quintessa Report QRS-1111A-2 v1.0, Quintessa, Henley-on-Thames, UK.

Little R H, Watkins B M, Walke R C and Smith G M (2002c). The Identification and Justification of an Assessment Biosphere for a Well Receptor. Quintessa Report QRS-1111A-3 v1.0, Quintessa, Henley-on-Thames, UK.

Little R H, Watkins B M, Walke R C and Smith G M (2002d). The Identification and Justification of an Assessment Biosphere for a Marine Receptor. Quintessa Report QRS-1111A-4 v1.0, Quintessa, Henley-on-Thames, UK.

Météo France (2008). Five year weather data provided to BIOPROTA by EDF for St. Dezier and Lille. Email 23 January 2008.

Maraux F, Lafolie F and Bruckler L (1998). Comparison between mechanistic and functional models for estimating soil water balance: Deterministic and stochastic approaches. *Agricultural Water Management* 38 (1), pp. 1-20.

Miquel S (2008). Développement et intégration d'un modèle de transfert sol multicouche dans le logiciel Matlab; Rapport Société de Calcul Mathématique, S.A. n° C.RP.FSTR.08.0009.A.

Miquel S and Beauzamy B (2007). *Analyse critique des modes d'échantillonnage, de la réalisation et de l'interprétation des calculs probabilistes réalisés pour les évaluations de transfert en biosphère dans le cadre du dossier HAVL 2005,* Rapport Société de Calcul Mathématique, S.A. n°C.RP.FSTR.07.0004.A.

Miquel S and Albrecht A (2008). SCM – ANDRA Multicompartment Model applied to Cl-36. Rapport Société de Calcul Mathématique.

Müller and Pröhl (1993) Ecosys-87: A dynamic model for assessing radiological consequences of nuclear accidents. *Health Physics* 64 (3), pp. 232-252.

Nakai K and Miyauchi Y (2008). Cl-36 model for LLW disposal in Japan, Oral Presentation, BIOPROTA X Meeting, Wettingen, Switzerland, 7-9 May 2008.

Nye P H and Tinker P B (1977). Solute Movement in the Soil-Root System, Blackwell Scientific Publications, Oxford.

Oakley J E (2000). Bayesian uncertainty analysis for complex computer codes. University of Sheffield, Sheffield.

Öberg G, Holm M, Sandén P, Svensson T and Parikka M (2005). The role of organic-matter-bound chlorine in the chlorine cycle: a case study of the Stubbetorp catchment, Sweden, Biogeochemistry, 75, 241-269.

Sau F, Boote K J, Bostick W M, Jones, J W and Mínguez M I (2004). Testing and improving evapotranspiration and soil water balance of the DSSAT crop models. *Agronomy Journal* 96 (5), pp. 1243-1257.

Sheppard M (2001). Transfer of Chlorine 36 in the biosphere: bibliography and modelling; Rapport Andra n° C RP 0ECO 2000/04/A.

Simmonds J R, Lawson G and Mayall, A (1995). Methodology for assessing the radiological consequences of routine release of radionuclides to the environment. Report EUR 15760-DG Environment, Nuclear Safety and Civil protection

Soltner (1990), Les grandes productions végétales. Collection sciences et techniques agricoles.

Soltner (1994), Les bases de la production végétale: Tome 1 le sol. Collection sciences et techniques.

Stevenson M D, Oakley J E and Chilcott J B (2004). Gaussian process modeling in conjunction with individual patient simulation modeling: A case study describing the calculation of cost-effectiveness ratios for the treatment of established osteoporosis. Medical Decision Making 24:89-100.

Stiennes E, Gjelsvik R, Bjerk T, Gaare E and Varskog P (2007). Large-scale studies of Cs-137 deposition in Norway from the Chernobyl accident. Proc. 9th International Conference on Biogeochemistry of Trace Elements: Environmental Protection, Remediation and Human Health, Beijing, Tsinghua University Press.

Tamponnet C. (2006) Modélisation des transferts environnementaux du Chlore 36. Rapport IRSN/DEI/ SECRE 2006-19, 48pp.

Tamponnet C. (2007) Fichiers de paramètres pour la modélisation des transferts du Chlore 36. Rapport IRSN/DEI./SECRE 2007-20, 37pp.

Thorne M C (2007a). A Point-scale Model for Cl-36 Transport in Soils and Plants, Mike Thorne and Associates Limited Report to United Kingdom Nirex Limited, MTA/P0011b/2006-1: Issue 2, January 2007.

Thorne M C (2007b). A Guide to the Spreadsheet Model used for Groundwater and Well Calculations for Generic Performance Assessments, Mike Thorne and Associates Limited Report to United Kingdom Nirex Limited, MTA/P0011c/2006-4: Issue 2, March 2007.

Turc L (1954) Water balance of soils: relationship between precipitation, evapotranspiration and runoff (in French). Ann Agron 549-595 and 6:5-13.1

Turc L (1961). Estimation of irrigation water requirements, potential evapotranspiration: A simple climatic formula evolved up to date. Ann. Agron. 12 (1), pp. 13-49.

Whicker and Kirchner (1987). Pathway: A dynamic food-chain model to predict radionuclide ingestion after fallout deposition. *Health Physics* 52 (6), pp. 717-737.

Zach R, Amiro B D, Bird G A, Macdonald C R, Sheppard M I, Sheppard S C and Szekely J G (1996). The Disposal of Canada's Nuclear Fuel Waste: A Study of Postclosure Safety of In-Room Emplacement of Used CANDU Fuel in Copper Containers in Permeable Plutonic Rock. Volume 4: Biosphere Model. AECL.