

BIOPROTA

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

Se-79 in the Soil-Plant System

Phase 2: Approaches to Modelling

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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.org.

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 updated consideration of those processes considered key to the understanding the behaviour of selenium-79 (Se-79) in soils and uptake into plants and discusses conventional and novel methods for addressing these processes within biosphere assessment models. Results from the application of the models to a hypothetical scenario description are also presented. Technical inputs were provided by CIEMAT (Spain), ANDRA (France), Posiva (Finland), Mike Thorne and Associates Ltd (UK) and IRSN (France) and financial support of ANDRA, NUMO and POSIVA is gratefully acknowledged.

This 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 user's risk.

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1. INTRODUCTION

In various studies relating to the deep geological disposal of solid radioactive wastes, Se-79 has been identified as a radionuclide of potential radiological significance. Furthermore, there are a number of uncertainties associated with biosphere modelling of Se-79, not least in relation to the redox-related behaviour of selenium in soils and consequences for plant uptake.

In response to these acknowledged uncertainties, a selenium working group was established in 2008, beginning with a focused workshop, hosted by NAGRA at Wettingen, Switzerland, to identify key processes for the modelling of selenium in soils and uptake into plants [Smith, 2008].

Subsequent to the workshop, a Se-79 working group report on 'Modelling the Abundance of Se-79 in Soils and Plants for Safety Assessments of the Underground Disposal of Radioactive Waste' [Smith et al, 2009] was published. The report aimed to present up to date information that would help to develop a better understanding of the processes affecting Se-79 accumulation in soils and uptake into plants, which could be used as a basis for improving models of the long-term behaviour of Se-79. Both an analysis of the important features, events and processes (FEPs) relevant to selenium environmental behaviour and an interaction matrix, which demonstrated linkages between FEPs and effectively represents a generic conceptual model, were produced.

Since this report was published, further information has become available; for example, a Se-79 biosphere review has been published by the Nuclear Decommissioning Authority – Radioactive Waste Management Directorate (NDA RWMD) [Limer and Thorne, 2010], and new modelling approaches have been developed, e.g. Coppin et al [2011] and Perez-Sanchez et al [2012]. A phase 2 programme of work, presented here, was therefore established to take account of these developments as a means of further improving understanding of key processes and approaches to modelling.

1.1 OBJECTIVES

The overall objectives of the Phase 2 Se-79 project were to address some of the remaining uncertainties associated with modelling the behaviour of Se-79 in soils and subsequent uptake from soils to plants and to consider the application of different modelling approaches in addressing the key environmental factors of relevance to selenium. By exploring the significance of different models and data assumptions at the conceptual and quantitative levels, it is intended that the level of complexity required in models, for example, the need to include more mechanistic processes, can be elucidated in different assessment contexts. Such consideration is aimed ultimately at increasing confidence in the long-term biosphere modelling of Se-79 for safety assessments in support of radioactive waste disposal facilities.

In meeting this objective, new sources of information have been reviewed and relevant information used to augment the information on processes considered in Smith et al [2009] to be key to understanding and modelling the behaviour of Se-79 in soils and uptake into plants. Models, both conventional and bespoke, applied to a hypothetical scenario are presented and results discussed in relation to whether key uncertainties in Se-79 environmental behaviour are addressed.

1.2 REPORT STRUCTURE

Section 2 outlines the key processes in the soil-plant system. Conventional approaches for representing these processes are outlined in Section 3. Section 4 then presents recent model developments that provide alternative approaches to Se-79 biosphere modelling. Results from the application of models to a hypothetical scenario are presented in Section 5. An overall summary and conclusions are provided in Section 6 and Section 7 lists the references used.

2. KEY PROCESSES FOR SE-79 IN THE SOIL-PLANT SYSTEM

Selenium-79 (Se-79) is one of the key radionuclides assessed as contributing to human dose following release of radioactivity from disposal facilities to the biosphere. The key factors affecting dose are the behaviour in soils and subsequent uptake into, and distribution within plants that may then be consumed by humans or animals.

The Se-79 working group has previously produced a report [Smith et al, 2009] that presented information aimed at developing a better understanding of the processes affecting Se-79 accumulation in soils and uptake into plants. Both an analysis of the important features, events and processes (FEPs) relevant to selenium environmental behaviour and an interaction matrix, which demonstrated linkages between FEPs were produced. A number of FEPs were identified as a result of this work programme for inclusion within biosphere models for Se-79 (Table 1), some of which were considered to be new to Se-79 specific modelling. Newly identified FEPs are largely phenomena that affect processes and pathways through the environment.

Selenium is recognised as having a complex environmental behaviour with multiple chemical species potentially co-existing at any one time within a given soil. Although the range of FEPs detailed in Table 1 are all relevant for Se-79 biosphere modelling, the different selenium species in soils and their specific properties were considered in Smith et al [2009] to be of particular importance for Se-79 biosphere models, since chemical speciation governs bioavailability and thus entry into the food chain. Volatilisation was also identified as a potentially key loss mechanism for selenium both from soils and plants. The degree of volatilisation is also thought to be dependent on the chemical species present. Such processes are therefore included in the focus of the current report.

Table 1 Features, Events and Processes relevant to Se-79 in the soil/plant system (from Smith et al [2009])^a

FEP	New for Se-79 ^b	Potential for alteration of current FEP
Bioturbation		
Capillary rise		✓
Colloidal selenium	✓	
Cropping loss (plants & animals)	✓	
Decomposition	✓	
Degassing	✓	
Discharge from below		✓
Effect of plants on Se speciation	✓	
Environmental change	✓	
Erosion		
Fertiliser addition	✓	
Foliar uptake	✓	
Infiltration		
Ingestion		
Interception		✓
Isotopic dilution	✓	
Micro-organisms & fungi	✓	
Organics, complexes and binding mechanisms	✓	
Percolation		✓
pH	✓	
Precipitation	✓	
Recalcitrant Se	✓	
Redox potential	✓	
Root uptake		✓
Soil hydrology variation	✓	
Sorption		✓
Stable Se concentration	✓	
Translocation		✓
Volatilisation	✓	
Weathering		✓

^a For definitions of FEPs, the reader is referred to Smith et al [2009].

^b A FEP is considered new if it is not routinely considered in previously existing Se-79 models, although it is recognised that it may be taken into account in some models.

2.1 SELENIUM IN SOILS

Selenium, an essential micronutrient for humans and mammals, occurs naturally in the environment. Concentrations in soils are largely determined by the availability of selenium in sedimentary rock. In regions with low selenium concentrations in the bedrock, selenium deficiency can occur. Agricultural practise in such areas may require selenium supplementation to animal feed and/or to soils (agronomic biofortification). Irrigation practice may also contribute to anthropogenic sources of selenium in soils [Winkel et al, 2012], particularly where groundwater is abstracted from regions with high concentrations of selenium in sedimentary rocks or from surface waters rich in selenium. Where agronomic biofortification occurs, it is estimated that between 5 and 20% of the annual selenium application is taken up by plants; the remainder being lost from the system by volatilisation processes and leaching [Winkel et al, 2012]. Where leaching occurs in combination with lateral water movement (e.g. surface run-off), this may result in increased concentrations of selenium in surface waters, potentially increasing selenium concentrations in aquatic plants and fish: accumulation in aquatic biota has been observed in the US following the use of selenium fertilisers [Winkel et al, 2012]. Alternatively, selenium-rich surface water may be used as a source of irrigation water thus cycling selenium back into agricultural soils.

2.1.1 SPECIATION

Selenium can be present in soils in multiple states, depending upon conditions such as pH and redox potential. These include inorganic forms such as selenide (-2), elemental Se (0), selenite (+4) and selenate (+6) as well as organic forms such as methylated selenium compounds, selenoamino acids and selenoproteins. Each selenium species behaves differently in the environment; speciation is therefore of key importance to understanding and evaluating plant uptake.

The selenium species present can largely be determined in relation to soil conditions [Neal and Sposito, 1989; Winkel et al, 2012]:

- Selenate – alkaline to neutral and oxic conditions;
- Selenite – neutral to acidic soils and less oxic conditions;
- Elemental selenium – anoxic conditions;
- Selenide – highly reducing conditions.

Limer and Thorne [2010] identify selenate as the likely form for Se-79 to be transported to the biosphere. However, the form in soils will vary greatly depending upon soil redox conditions. Factors affecting soil redox conditions (e.g. changes in soil hydrology, the addition of nitrogenous fertilisers etc) will therefore have an effect on selenium speciation.

2.1.2 PARTITIONING BETWEEN SOLID AND LIQUID PHASES

The partitioning of selenium between liquid and solid phases varies between the different selenium species such that varying soil redox conditions have the potential to affect soil solid-liquid partitioning (K_d) and volatilisation. Solubility ultimately affects mobility within the environment and bioavailability and is therefore of high importance with regard to the modelling of long-term radiological impacts. As such, the need to explore further the dynamics of, and key processes governing, sorption and desorption of selenium species was identified as a key requirement in Smith et al [2009].

Selenate is regarded as the least sorbed species and will dominate in oxic conditions. As conditions become more reducing (e.g. as a result of higher soil saturation) selenate will progressively reduce to selenite, through elemental selenium to selenide, with solubility and, hence, mobility decreasing [Limer and Thorne, 2010]. Elemental selenium is not soluble in water [Winkel et al, 2012].

Although selenate is not considered to be a highly sorbed species, it does form easily reversible surface complexes on clay minerals which render it less available although sulphate, an analogue of selenate, will compete if present for available binding sites thus potentially increasing selenate mobility [Winkel et al, 2012].

The speciation of selenium is further complicated through the complex interplay between soil pH, mineralogical and chemical composition of the soil, the nature (ionic composition) of absorbing surfaces and microbial interactions [Neal, 1995]. For example, under neutral and alkaline conditions, selenium is largely present as highly mobile selenate whereas in acidic conditions, selenium is usually found as selenite bound to mineral soil components [Smith et al, 2009]. Selenite may be subject to irreversible sorption to iron oxides, clay minerals and soil organic matter [Winkel et al, 2012]. The following environmental behaviours have been noted in relation to selenium in the review by Limer and Thorne [2010] that result in either more or less selenium in soil solution:

- Increasing pH results in a lower adsorption of selenite;
- Phosphate and arsenate ions compete with selenite for binding sites on, for example, clay particles thus increasing the amount of selenium in soil solution and thus bioavailability; similarly, sulphate and nitrate compete with selenate, again increasing bioavailability; and,
- Sorption capacity is increased in organic soils in which the majority of selenium is associated with organic matter.

An increased sorption of selenium in organic soils as compared with mineral soils was also observed by Keskinen et al [2009]. In Coppin et al [2009] the sorption on organic matter was found to be due to sorption on mineral phases linked with organic matter. This sorption to organic matter is particularly associated with the 50 - 200 µm particle size fraction [Smith et al, 2009]. Soil organic matter content can also influence soil redox conditions; reducing conditions being associated with high organic matter content, potentially resulting in the formation of elemental selenium or metal selenides thus reducing selenium concentrations in the aqueous phase [Winkel et al, 2012].

2.1.3 DISTRIBUTION IN SOILS

A further potentially important factor to consider is the distribution of selenium within the soil column, particularly within the soil rooting zone. It was noted in Smith et al [2009] that this has received relatively little attention.

Hydrology will vary throughout the soil profile which in turn is likely to affect the species of selenium present due to changing redox conditions with soil saturation. It was therefore recommended [Smith et al, 2009] that further consideration be given to modelling the vertical differentiation of soil layers, specifically within the soil rooting zone.

2.2 UPTAKE FROM SOILS TO PLANTS

Although selenium is an essential micronutrient in humans and other mammals, it is not believed to have an essential role in plant nutrition, yet plants are an important source of selenium for those with essential nutrition requirements.

Accumulation of selenium in plants can vary. For example, Winkel et al [2012] note that accumulation can vary by more than two orders of magnitude between different plant taxa grown in soil with a given selenium concentration. White et al [2004] note that most plants contain up to 25 $\mu\text{g Se g}^{-1}$ d.w. and are unable to tolerate high levels of selenium in the environment. Nonetheless, some plants (e.g. from the genera *Astragalus*, *Neptunia* and *Stanleya*) are able to hyper-accumulate selenium to concentrations up to 15 mg Se g^{-1} d.w. [Sors et al, 2005].

The concentration of selenium in soils will influence the plant species that can grow, depending upon their tolerance. In general, food crops are considered to have a low tolerance to selenium in soils [Plant et al, 2004] with yields reducing as soil concentrations increase [Bisbjerg and Gissel-Nielsen, 1969]. Crops grown in low-selenium soils show little difference in selenium uptake; changing the type of crop grown therefore largely has little impact on the selenium concentration in food crops [Plant et al, 2004].

The primary route of entry of selenium into plants is from soils via roots although some foliar absorption (up to 30% of plant uptake) may occur [Limer and Thorne, 2010]. Foliar uptake has been demonstrated to be greater for selenate than selenite when applied via fine spray to leaves [Kapolna et al, 2009]. Although, as noted in Smith et al [2009], foliar uptake is likely to be a minor pathway for soil-plant systems, the importance of this route of uptake may be increased where competition for root uptake sites is high (e.g. in the presence of high soil sulphate and/or phosphate concentrations). In addition, the possibility of absorption associated with volatilisation from the soil cannot be precluded and may lead to a lower loss from the system than currently anticipated from volatilisation alone. Information on the uptake of volatilised forms is however required in order to assess the potential impact of this route of uptake.

2.2.1 BIOAVAILABILITY

Since root uptake is the dominant mechanism for selenium entering plants, selenium behaviour in soils (i.e. speciation) is considered key to fully understanding bioavailability [Smith et al, 2009].

The predominant forms of selenium available to plants are selenate and selenite although organic forms are also readily taken up [Zayed et al, 1998; Sors et al, 2005]. Uptake of selenate by plants is reported to be up to eight times greater than that of selenite [Bisbjerg and Gissel-Nielsen, 1969]. In soils, selenium can also become bound to organic matter rendering it less bioavailable than that in solution. Selenite in particular has an affinity for binding to soil organic matter [Winkel et al, 2012]. Mineralisation of organic matter can then release selenium back to the bioavailable pool and may therefore be a continued source of selenium over time.

Microbial activity can also influence the form of selenium in soils [Smith et al, 2009]. Microbial activity is moderated by various factors, including nutrient availability, thus nutrient-deficient soils may have a slower rate of microbially-induced speciation than nutrient-rich soils.

Soil conditions affecting selenium speciation are therefore inherently linked to the uptake of selenium by plants.

2.2.2 ISOTOPIC DILUTION

The presence of selenium in soils, both natural and as modified by additives and other human actions, will have an effect on the uptake of Se-79 as a result of isotopic dilution.

Natural selenium concentrations can be highly variable in soils (and may be artificially amended through agronomic biofortification). Any activity that increases the stable selenium concentration (or bioavailability) in soils would serve to dilute Se-79 with resultant effects on transfer of Se-79 from soils to plants.

2.2.3 ANALOGUE BEHAVIOUR

Selenium is an analogue of sulphur and can readily enter plant roots via an active transport process associated with sulphate transporters that are located within root cell membranes. It is largely selenate that is taken up via such transporters. Research has shown that by increasing sulphate in the plant rooting zone, the uptake of selenate is increasingly inhibited whereas uptake of other forms of selenium (selenite and organic selenium) is not affected [Zayed et al, 1998]. Up to a 60% reduction in selenium uptake has been reported in plants grown on soils to which sulphur has been added [Smith et al, 2009]. Brassicas in particular are unable to distinguish selenium from sulphur resulting in selenium accumulation when readily available in soils [Plant et al, 2004].

There is also evidence to suggest that phosphate transporters play a role in the active transport of selenium, in the form of selenite, into plants [Li et al, 2008]. However, a greater diffusive uptake element is associated with the uptake of selenite when compared with selenate and organic selenium [Zayed et al, 1998].

Concentrations of sulphate and phosphate in soils will therefore influence the uptake of selenium by plants as a result of competition for sulphate and phosphate transport sites, with selenate and selenite uptake being inhibited in the presence of sulphate and phosphate, respectively. The addition of soil improvers is therefore likely to reduce the rate of uptake of selenium into plants; whereas, in soils deficient in sulphate and/or phosphate, enhanced uptake of selenium may occur.

2.2.4 CLIMATE EFFECTS

Climate can have an influence on the behaviour of selenium in soils. In particular, increased precipitation can lead to a shift from oxic to more anoxic soil redox conditions, leading to the reduction of selenate to less bioavailable forms such as selenite, elemental selenium and selenide. Increased precipitation can also lead to increased soil concentrations through the deposition of selenium^a through rainfall [Winkel et al, 2012]. Furthermore, increased precipitation may result in leaching of mobile species that may also reduce the amount of selenium available to plants.

^a Sources of atmospheric selenium include coal and petroleum fuel combustion.

2.3 DISTRIBUTION IN PLANTS

2.3.1 UPTAKE FROM SOIL

The distribution of selenium throughout plants is variable and is greatly influenced by the species of selenium present: selenate is translocated from roots to shoots more readily than selenite or organic Se compounds, as demonstrated in Table 2 [Zayed et al, 1998]. Evidence suggests that the mobilisation of selenate throughout plant tissues is strongly linked to high-affinity sulphate transporters [Sors et al, 2005]. Selenite is readily converted to organic Se in roots where it is largely retained [Zayed et al, 1998; Li et al, 2008].

Table 2 Ratio of shoot to root selenium content as influenced by the chemical form of selenium supplied in hydroponic growth solution (from Zayed et al [1998])

Selenium form	Shoot to root ratio			
	Broccoli	Indian Mustard	Sugarbeet	Rice
Selenate	9.55	17.2	6.12	1.44
Selenite	0.47	0.3	0.24	0.4
Selenomethionine	0.91	0.98	-	0.58

Selenium concentrations in seeds and grain are generally greater than for the roots or shoots of a plant [Bisbjerg and Gissel-Nielsen, 1969]. The accumulation of selenium in the grain of cereal crops suggests that selenium may associate with proteins (see presentation by Sheppard in Smith, [2008].) Further review information on uptake following soil applications of native selenium, selenite and selenate is provided in Bitterli et al [2010].

2.3.2 FOLIAR UPTAKE

Where selenium enters a plant as a result of foliar absorption, it is expected that the majority will remain associated with the above-ground plant; less than 20% is translocated to roots [Limer and Thorne, 2010]. Of the above ground burden, up to 70% may be associated with grain at harvest [Limer and Thorne, 2010].

Limer and Thorne [2010] investigated the implications of foliar absorption uptake on model output for the NDA RWMD Biosphere Spreadsheet Model [Thorne, 2007] concluding that incorporation of this process was of limited consequence for dose calculations since root uptake was the dominant transfer pathway.

2.4 VOLATILISATION

Volatilisation of selenium from soils and plants has been identified as a key process by which Se-79 may be lost from the system; however, many models currently conservatively disregard volatilisation from soils and plants [Smith et al, 2009].

Data relating to both plant and soil volatilisation are presented below. However, it is important to note that the majority of available data on selenium volatilisation are derived from short-term studies with some evidence that volatilisation rates reduce over time [Sheppard and Sheppard, 2008]. Further data derived from long-term field studies would therefore be of benefit in determining the extent to

which volatilisation is reduced over time thus enabling the impact of this process to be evaluated in relation to long-term biosphere modelling.

2.4.1 PLANTS

Certain plants (particularly selenium-accumulating plants) have the capacity to remove selenium from soils by metabolising it into an organic methyl-selenide form that is readily volatilised [Zayed et al 1998; Limer and Thorne, 2010]. The rate of volatilisation will vary with plant type. For example, Terry et al [1992] investigated rates of volatilisation for a number of different plant species with the following conclusions:

- Rice, broccoli and cabbage volatilised selenium at the fastest rates (200 to 350 $\mu\text{g Se per m}^2$ leaf area per day or 1500–2500 $\mu\text{g Se kg}^{-1}$ plant dry wt. d^{-1});
- Carrot, barley, alfalfa, tomato, cucumber, cotton, eggplant and maize had intermediate rates of 30 to 100 $\mu\text{g Se m}^{-2} \text{d}^{-1}$ (300–750 $\mu\text{g Se kg}^{-1} \text{d}^{-1}$); and,
- Sugar beet, bean, lettuce and onion had the lowest rates at less than 15 $\mu\text{g Se m}^{-2} \text{d}^{-1}$ (<250 $\mu\text{g Se kg}^{-1} \text{d}^{-1}$).

In comparing all plant species, selenium volatilisation rate was found to be highly correlated with the concentration in plant tissues, suggesting that the ability of plants to absorb selenium may be an important factor contributing to high rates of volatilisation [Terry et al, 1992]. However, de Souza et al [1998] reported selenium volatilisation rates 2- to 3-fold higher from plants supplied with selenite compared with selenate (considered the most bioavailable form), which was attributed to the conversion of selenite to selenomethionine. Similar conclusions were reached by Sheppard and Sheppard [2008] and Zayed et al [1998]. Pilon-Smits et al [1998] reported that the volatilisation rates were 230-fold higher for selenomethionine compared with selenite, and 1.5-fold higher for selenite than for selenate. The chemical form of selenium present in the root substrate is therefore likely play an important role in governing plant volatilisation rates, with selenate reduction being a rate-limiting step.

2.4.2 SOILS

An extensive review by Sheppard and Sheppard [2008] focused on selenium volatilisation rates reported under field conditions for both sediments and soils and concluded that volatilisation represents an important loss mechanism for selenium.

Volatilisation of selenium from soils occurs as a result of biomethylation by microbes and can be affected by a number of factors, including speciation, agricultural practices such as ploughing, temperature, soil type and soil moisture [Smith et al, 2009]. Sheppard and Sheppard [2008] reported a high degree of variability in volatilisation rates, largely attributable to variability in environmental and experimental conditions (e.g. soil type, exposure time). Evidence suggests that the presence of organic matter within soils serves to reduce volatilisation [Zayed et al, 1998; Sheppard and Sheppard, 2008], but that organic forms of selenium are volatilised by microbial activity to a much greater extent than inorganic forms [Reamer and Zoller, 1980]. However, research undertaken by IRSN has indicated that the addition of organic matter to soils serves to increase volatilisation rates, with differences being observed in effect with soil type and selenium concentration added [Colinon-Dupuich et al, 2011].

The availability of essential nutrients for microbial activity will also be a key factor affecting the rate of volatilisation [Smith et al, 2009]; hence it may be reasonable to assume that in nutrient-poor soils the rate of volatilisation will be reduced. Work by IRSN has identified a good correlation between the amount of selenium volatilised (normalised by the number of bacteria present in soil) and CaCl₂ extractable (bioavailable) selenium [Colinon-Dupuich et al, 2011].

An important knowledge gap noted in the review by Sheppard and Sheppard [2008] was the lack of data relating to selenium volatilisation rates over the long-term, with field studies focusing on relatively short time frames. From the available data, Sheppard and Sheppard [2008] noted that there was some evidence that volatilisation rates may reduce over time, suggesting that selenium may become incorporated into non-exchangeable forms in the longer term. Such a process could have important implications for the modelling of long-term behaviour of Se-79 in the biosphere.

From the currently available data base, Sheppard and Sheppard [2008] recommended the following volatilisation rates:

- Soils - 3% per annum.
- Sediments – 6% per annum.

Research undertaken by IRSN and reported in Smith et al [2009] has investigated how selenium volatilisation rates vary over time and in response to changing organic matter and selenium soil concentrations. Of the selenium added to soils that was subsequently monitored over a 1.5 year period, only 0.2% was lost through volatilisation. The rate of loss was greatest in organically enriched soils and reduced over time, which is consistent with the findings of Sheppard and Sheppard [2008]. Large variations in volatilisation rates were observed between soil types and evidence suggests that the bioavailability of selenium is an important factor.

Results of the review by Sheppard and Sheppard [2008] indicate that volatilisation of Se-79 in the short-term may substantially reduce the importance of this radionuclide due to loss from the soil-plant system. However, results also indicate that rates of volatilisation reduce over time. Volatilisation rates in the long-term therefore remain a key uncertainty in relation to biosphere aspects of Se-79 dose assessment modelling.

As noted in Perez-Sanchez et al [2012], little information is available on the potential for volatile selenium, released from soils, to subsequently be absorbed by foliage.

2.5 SUMMARY OF KEY FEATURES, EVENTS AND PROCESSES FOR SE-79 IN THE SOIL-PLANT SYSTEM

This section has served to identify those aspects of selenium behaviour in soils that are considered key to improved modelling of the transfer of Se-79 from soils to plants following release from engineered disposal facilities for long-lived radioactive waste. These are summarised in **Error! Reference source not found.** The aspects of behaviour detailed are by no means the only ones relevant to models of selenium behaviour, but are considered most important for predicting uptake into the terrestrial foodchain. Additional aspects are detailed in Smith et al [2009]. Of those FEPs outlined, the common denominator in driving selenium behaviour is speciation in soils. This is therefore considered the single most important feature for understanding the behaviour of selenium in the soil-plant system and for modelling the transport of Se-79 into the foodchain.

Table 3 Key aspects of selenium behaviour in soils and plants

FEP	Drivers
Speciation	Soil redox conditions pH Microbial activity Soil hydrology (incl. Climatic effects)
Solid-liquid partitioning	Strongly linked to speciation pH Soil hydrology Soil composition (e.g. organic matter/clay content)
Bioavailability	Strongly linked to speciation
Plant uptake	Presence of selenate, selenite, selenide and organic selenium Distribution of Se-79 in rooting zone Stable selenium (isotopic dilution) Competing chemical species (sulphate/phosphate) Volatilisation from soil and foliar uptake
Distribution in plants	Form of selenium taken up (speciation) Plant type
Losses from the system	
Cropping	Form of selenium Crop type
Volatilisation (Soil)	Microbial activity Selenium speciation Soil nutrient status Soil type Soil hydrology
Volatilisation (plant)	Speciation Plant type

3. CONVENTIONAL APPROACHES TO MODELLING SE-79 IN THE BIOSPHERE

The conventional approach to modelling the behaviour of selenium in the biosphere in support of performance assessments for radioactive waste repositories is described in this section. Potential limitations in the approach are also identified.

Overall, selenium behaviour in soils and plants is represented by relatively simplistic processes that relate to total selenium in soils (Figure 1); thus speciation and the factors affecting speciation are not considered.

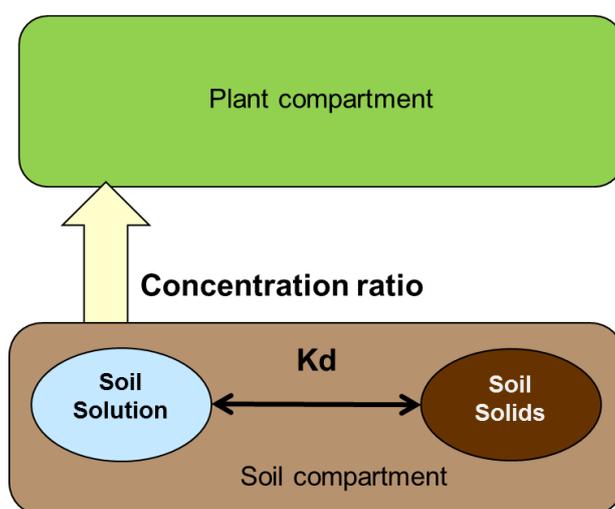


Figure 1 Conventional model approach for Se-79 in soils and plants

3.1 BEHAVIOUR IN SOILS

The standard approach to modelling the partitioning between solid and liquid phases is the use of the partition coefficient, K_d , which represents an instantaneous reversible equilibrium process representative of total selenium in soils (i.e. different selenium species are not specifically addressed).

K_d is largely determined from laboratory batch or column experiments that provide a short-term measure of partitioning between the liquid and solid phases and do not take account of seasonal or longer-term environmental change. Field measurements of K_d can also be made, including analysis of stable elements.

In the case of Se-79, a large variation in K_d is observed as demonstrated by the range in values over approximately three orders of magnitude within the recent IAEA publication on parameter values for radionuclide transfer in terrestrial ecosystems [IAEA, 2009]. IAEA [2009] provides the following recommendations on K_d for different soil types (Table 4).

Table 4 Recommended Kd (L/kg) by soil type for selenium [from IAEA, 2009]

Soil type	Number of observations	Mean	GSD	Minimum	Maximum
All	172	2E+2	3.3	4E+0	2.1E+3
Sand	15	5.6E+1	5.2	4E+0	1.6E+3
Loam and Clay	134	2.2E+2	3	1.2E+1	2.1E+3
Organic	2	1E+3	-	2.3E+2	1.8E+3

The appropriateness of using a single Kd to represent Se-79 behaviour may be questionable on the basis of the different pools of selenium that can occur in any given soil.

3.2 UPTAKE INTO PLANTS

Models representing the uptake of elements from soils to plants often rely upon a simple equilibrium concentration ratio (CR) parameter. It is generally accepted that CR and Kd are linked in a broadly inverse fashion [Smith et al, 2009]. Nonetheless, it is acknowledged that although CR and Kd are inversely linked, the variation in CR is somewhat lower than that observed for Kd [Limer and Thorne, 2010].

It is reasonable to assume that those environmental factors affecting Kd, e.g. chemical speciation, will also influence CR: as noted previously, selenate, the most soluble and mobile of the selenium species, is also the most bioavailable. Any factor leading to an increase of selenium in the liquid phase therefore has the potential to result in an increased uptake into plants. The presence of competing elements, e.g. sulphur, will be an additional factor affecting CR [Smith et al, 2009]. Where sulphur concentrations are enhanced (e.g. as a result soil fertilisation), uptake of selenium by plants is likely to be reduced. Since selenium and ions such as sulphate compete for uptake sites, the sulphur requirements of a plant may also influence uptake [Limer and Thorne, 2010].

Overall there is around an order of magnitude difference between the CR for selenate and selenite [Bisbjerg and Gissel-Nielsen, 1969, Bitterli et al, 2010] and both forms may be present within the same soil. The presence of organic matter also affects the availability of selenium for uptake into plants; selenium binds to organic matter rendering it less available for uptake by plants [Keskinen et al, 2009]. By assessing the uptake of selenium from soils to plants by use of a single CR value, important relationships between bioavailability and selenium speciation cannot be represented leading to a large degree of uncertainty associated with the model output.

Aside from the issue of the bioavailability of selenium species, there are a number of additional factors that complicate the use of a single CR in modelling studies. For example, data from multiple analyses are often combined in the derivation of an 'appropriate' CR to apply within a model. By combining multiple data, potentially derived for a multitude of different environmental conditions, important factors governing selenium behaviour may be lost. For example, variation in soil pH and Eh may not be accurately recorded such that relationships between these important parameters and CR values are not considered and are represented only by the uncertainty associated with any derived CR (e.g. scatter around a mean or median point).

A further factor that should be considered in relation to the uptake of selenium by plants from soils is the potential for plants to discriminate against uptake and/or preferentially take up selenium from what would be considered biologically unavailable pools [Limer and Thorne, 2010]).

3.3 EXAMPLE MODEL APPROACHES

A number of models are available for evaluating the solid-liquid partitioning of radionuclides, including Se-79, in soils and subsequent uptake to plants. Three models, which have been applied to the hypothetical scenario outlined in Section 5, are described below.

3.3.1 NDA RWMD SPREADSHEET MODEL

The NDA RWMD Spreadsheet model represents a conventional approach to modelling the behaviour of radionuclides in soils with results then being used to compute plant concentrations. The model consists of two soil compartments (surface soil and subsoil). The model considers two geosphere-biosphere interfaces: natural groundwater discharge and abstraction of groundwater for use in irrigation practices.

The hydrology of the two soil compartments are represented by the following parameters that are balanced within the model:

- Precipitation plus irrigation;
- Percolation from surface soil to subsoil;
- Evapotranspiration;
- Capillary rise from subsoil to surface soil to replenish soil moisture deficit;
- Throughflow gains from surface soil;
- Baseflow gains from subsoil;
- Throughflow losses from surface soil;
- Baseflow losses from subsoil; and
- Groundwater discharge to subsoil.

Typically in application, the interface with the underlying geosphere model will be at rockhead, i.e. the total depth of the surface soil plus subsoil will be taken equal to the depth of unconsolidated material. However, this is not a requirement of the model and the total depth of surface soil plus subsoil may be less than the depth of unconsolidated material.

Transport of radioactivity within the soil is represented by soil-water partition coefficients applied to the surface soil and subsoil. Total activity in soil is computed, both on a volumetric basis and a dry mass basis (Bq kg^{-1}). Activity concentrations on soil solids and in soil solution can also be computed.

For the groundwater pathway, contamination of plants occurs only by root uptake, through application of a soil to plant concentration ratio.

For well abstraction, contamination of plants can be via root uptake or direct foliar contamination by spray irrigation.

Foliar contamination allows for the effects of wash-off and is effectively treated as a single event occurring at some specified time before harvest. The interception and retention of activity on or in the plant does not reduce the amount going to soil. This is consistent with the approach that is adopted for root uptake, as a cautious approach is adopted that does not take cropping losses into account.

3.3.2 ANDRA AQUABIOS MODEL

The Andra Aquabios model [Albrecht and Miguel, 2010] uses a conventional approach to modelling the behaviour of radionuclides in soils and uptake into plants. A schematic of the model is presented in Figure 2.

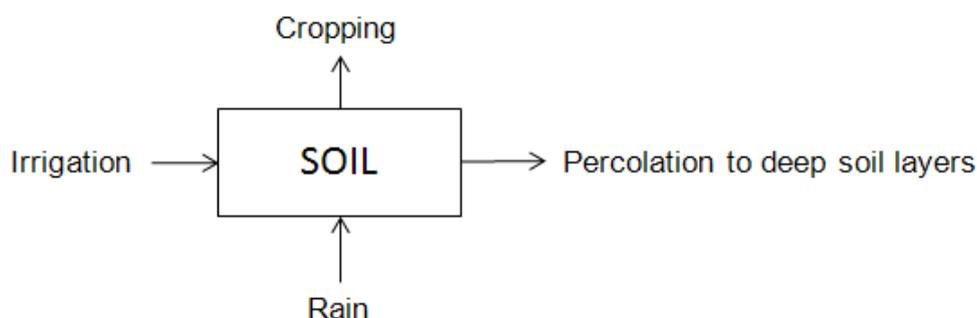


Figure 2 Schematic of Aquabios model

The behaviour of radionuclides within the soil column is evaluated on the basis of the water budget within a soil column with a solid/liquid distribution coefficient (K_d) being applied to calculate retention. The model 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 dynamics of radionuclide accumulation in soil depend on the system losses (the higher the losses the more quickly equilibrium is achieved).

Irrigation by contaminated well water is one of the modelled sources of contamination and is dependent upon the water budget of the soils. Local information on monthly precipitation and evapotranspiration are used to assess the yearly need for irrigation. Release of contamination into river water and the effects of river flooding can also be modelled.

Fluxes of radionuclides are modelled using yearly averaged irrigation and the concentration in the irrigation water.

Under humid conditions, particularly during the colder season, when precipitation exceeds evapotranspiration, excess water in the system exits via percolation. The amount of excess water is set equal to that derived from the hydraulic budget and has the ability to transport dissolved radionuclides into deeper sections of the soil where they become unavailable for root uptake.

An additional loss term is related to the export of radionuclides at crop harvest, which can be modelled based on the yield of cultivated plants, an empirically determined soil to plant transfer coefficient for those plants and soil physical parameters (depth of homogenisation and soil density).

Interception of radionuclides in irrigation water and transfer from leaves to the edible portion can be considered in Aquabios, in addition to root uptake.

3.3.3 CONVENTIONAL MODEL USED BY IRSN

When using the conventional model IRSN considers a single soil compartment with the behaviour of Se-79 in relation to that compartment being governed by a Kd parameter. All Se-79 is considered to be labile and thus bioavailable.

The model does not explicitly consider irrigation, precipitation and evapotranspiration, but rather the water flux in the system is considered as a net flux of water (percolation rate) which is calculated by subtracting the evapotranspiration flux from the combined flux of irrigation and precipitation. No loss of Se by evapotranspiration was considered. The amount of Se-79 in percolate water is calculated according to the equation:

$$\begin{aligned} & \text{Se79 in percolate water (Bq L}^{-1}\text{)} \\ &= \frac{\text{Irrigation rate (m y}^{-1}\text{)} * \text{Se79 concentration in irrigation water (Bq L}^{-1}\text{)}}{\text{Percolation rate (m y}^{-1}\text{)}} \end{aligned}$$

The concentration of Se-79 in the bioavailable pool is calculated from:

$$\begin{aligned} & \text{Se79 available pool (Bq kg}^{-1}\text{)} \\ &= \frac{\text{total Se79 sorbed on soil (Bq kg}^{-1}\text{)} * \text{dry soil weight of soil layer (kg)} \\ &+ \text{Se79 in solution (Bq L}^{-1}\text{)} * \text{volume of solution in soil layer (L)}}{\text{dry soil weight of soil layer (kg)}} \end{aligned}$$

Plants obtain Se-79 via root uptake, which is represented by an equilibrium concentration ratio applied to the bioavailable pool of Se-79.

Volatilisation and the groundwater below the soil compartment are not considered.

3.4 ADDRESSING UNCERTAINTIES

The applicability of a single Kd to long-term assessment models is debatable and was one of the key parameters identified in Smith et al [2009] as requiring further consideration. In particular, it was recommended that an alternative approach to Kd estimation for long-term modelling studies that takes due account of the different pools of selenium, including fractions that are reversibly and non-reversibly bound and in solution. The approach could make use of Kd, but would have to apply appropriate kinetic parameters to take account of the behaviour of the different selenium pools. The influence of soil redox conditions on Kd was also identified as an important process requiring consideration.

In order to address some of the uncertainties relating to the uptake of selenium from soils by plants, Smith et al [2009] also noted that it may be appropriate to consider multiple soil layers within the rooting zone of soils to allow the bio-geochemical factors affecting the vertical profile of selenium to

be taken into account. This would also allow for more detailed consideration of the different selenium species present in soils and their relative bioavailability under different hydrological conditions. Such an approach may lead to a greater understanding of selenium behaviour and how it affects root uptake. Site-specific information on soil type and hydrology may be important in this respect.

By taking account the behavioural complexities relating to soil behaviour it may be possible to determine whether:

- Kd is an appropriate approach for long-term modelling studies – for example, it may be that the use of Kd represents an average of selenium behaviour (including the different selenium species) over the long-term;
- The large range in Kd can be reduced through consideration of the behaviour of selenium under differing environmental conditions; and,
- CR ranges can be recommended that apply to different soils under a range of conditions by accounting for the different selenium species that are likely to be present and their respective bioavailability.

These different approaches have been developed independently of BIOPROTA. The resultant models are discussed in Section 4.

4. ALTERNATIVE MODELLING APPROACHES

Since publication of Smith et al [2009], three separate models for the behaviour of selenium in soils and plants have been developed, which aim to address some of the uncertainties identified with regard to the complex behaviour of selenium in soils. These are the CIEMAT model [Thorne, 2010; Perez-Sanchez et al, 2012], the ANDRA SAMP model for all radionuclides, including Se-79 [Miquel and Basso, 2010] and the IRSN Se-79 model [Coppin et al, 2011]. The CIEMAT and ANDRA SAMP models are comprised of multiple soil layers that enable differences in selenium behaviour with soil depth to be addressed thus enabling soil hydrological and redox conditions to be taken into account in a time-dependent way. The IRSN model uses equilibrium and kinetic parameters to take account of labile (reversibly bound) and recalcitrant (non-reversibly bound) pools of selenium in soils.

Each model is described below.

4.1 THE CIEMAT SE-79 SOIL-PLANT MODEL

Thorne [2010] and Perez-Sanchez et al [2012] describe a two-component model, implemented in MS Excel and the AMBER modelling tool that was specifically designed for addressing the complex behaviour of Se-79 in the soil-plant system. The overall model, which includes kinetic parameterisation, enables the effects of changing soil hydrological conditions (used as a surrogate for redox) on sorption, root uptake and volatilisation to be assessed and incorporates cropping losses and the recycling of Se-79 to soil organic matter.

The information presented below is derived from Thorne [2010] and Perez-Sanchez et al [2012].

4.1.1 CALCULATION OF SOIL PROFILE DISTRIBUTION COEFFICIENTS AND WATER FLUXES

The first component to the model is implemented in an MS Excel spreadsheet that computes the spatially and temporally varying hydrological characteristics of ten soil layers, representative of the soil rooting zone, and provides spatially and temporally distributed distribution coefficients. This is achieved through the simulation of the time-dependent height of the water table relative to each soil layer. Water table height relative to the soil profile is calculated through consideration of average monthly precipitation and evapotranspiration. The position of the water table is then used to assign vertically varying distribution coefficients taking account of redox conditions (anoxic conditions are assumed to prevail below the water table whereas oxic conditions prevail above the capillary fringe) for what can be considered equilibrium conditions (a 10 year simulation period)^a.

The spreadsheet model also enables water flux between soil layers to be calculated which allows for the subsequent evaluation of radionuclide flux. The water flux into the top soil layer is defined by the rate of precipitation resulting in a downward flow. Loss occurs via drainage from the lowest soil layer such that drainage and precipitation control the downward flux of water (upward flux into the lowest soil layer can also be represented, so that discharge areas can be represented as well as recharge areas). Upward flux is driven by movements of the water table.

^a As noted in Perez-Sanchez et al [2012], the 10 year simulation period is applied to ensure that the water table height, which is set to an arbitrary value at the beginning of year 1, stabilises into a repeating annual cycle.

4.1.2 RADIONUCLIDE TRANSPORT

The transport of Se-79 between soil layers and into plants is calculated in a model encoded within the AMBER modelling package.

Selenium transport between soil layers is calculated in relation to water flux, water contents and distribution coefficients from the MS Excel spreadsheet model with data defined as time-series elements repeating annually.

Volatilisation

Volatilisation is incorporated into the model, both directly from soil and as a result of evapotranspiration in plants.

In soils, production of volatile forms of selenium is assumed to be microbially mediated with methylation being the predominant pathway. As such, it is assumed that activity will be concentrated at or below the water table. Potentially volatile selenium produced below the water table is likely to be solubilised and, potentially, reutilised rather than being released from soil to air. The following generalised loss-rate model is therefore assumed:

- Highly unsaturated soils are subject to a constant low rate of loss from volatilisation;
- Maximum volatilisation rates are assumed for soils close to saturation; and,
- A lower constant rate of volatilisation is assumed for soil layers below the water table.

The model allows for volatilisation from soils to be suppressed if required. By increasing assumptions relating to volatilisation from a rate of 0.03 y^{-1} to 0.3 y^{-1} , penetration of Se-79 from surface soil layers to layers at depth is greatly reduced due to the loss of Se-79 from the system.

Volatilisation from plants is governed by the rate of uptake of soluble Se-79 from soils. Volatilisation is therefore represented in the model as a loss process from the plants.

Uptake into Plants

The AMBER model has a single plant compartment with root uptake arising from all soil layers. However, the density of roots varies throughout the soil profile and also varies seasonally. Uptake by roots will also be variable with respect to the degree of soil saturation. These factors are encoded within the model by soil-layer specific considerations.

Once within plants, losses from volatilisation and cropping can occur. Both cropping and volatilisation losses are represented by time-dependent rate coefficients. In the case of volatilisation, the rate coefficient is defined for each month of the year. For cropping losses, rates and timings throughout the year are varied depending upon whether soils are used for pasture or arable crops. Account is taken of the return of Se-79 in organic matter to soils through an additional model compartment. This compartment transfers Se-79 to the upper soil layer representing release following the mineralisation of organic matter.

4.1.3 MODEL PARAMETERISATION

The key parameters included within the Se-79 model are as follows:

- Distribution coefficients in oxic and anoxic conditions (Kd_{min} and Kd_{max});
- Soil volatilisation rates in oxic and anoxic conditions and within the capillary fringe;
- Root uptake rate per unit depth of soil and parameters for modification of uptake in relation to soil water content;
- Volatilisation rate from plants; and,
- Rate of loss of Se-79 from organic matter in soil.

4.1.4 INITIAL ANALYSIS OF MODEL SENSITIVITY TO CHANGING PARAMETER VALUES

A number of analyses have been undertaken to determine the sensitivity of the model to changing parameter values, specifically distribution coefficients, volatilisation rates and root uptake.

Distribution coefficients

By assuming a low Kd for oxic soil layers and moderately low Kd for anoxic layers, Se-79 was shown to migrate rapidly down the soil column such that plant concentrations reduce after the initial few years. If a high Kd is applied to the oxic zone and a very high Kd to the anoxic zone, Se-79 concentrations are maximal in the top soil layer with little downward migration throughout the simulation period. The main losses from soil are through volatilisation and plant uptake. If differences in Kd are maximised between the oxic and anoxic layers, Se-79 accumulates in the lower soil layers. However this retention acts to support concentrations in the upper layers such that plant concentrations do not differ significantly from the initial case described above. Overall, results indicate that plant uptake is maximised by assuming a relatively high Kd for the oxic zone.

Volatilisation rate from soil

Changes in volatilisation rate were found to have only limited effects with plant and soil concentrations decreasing as volatilisation rates increase. Effects were limited because of the dominance of plant uptake as a means of removing Se-79 from soil as compared with direct volatilisation.

Root uptake

Increasing root uptake was found, as expected, to increase plant concentrations. However the effect was not directly linear due to increased losses due to plant volatilisation and cropping and a greater return of Se-79 bound to organic matter to the soil. Resultant mineralisation leads to increased soil Se-79 concentrations in the uppermost soil layers.

4.2 ANDRA SAMM MODEL

The ANDRA SAMM (SCM – ANDRA Multi-compartment Model) approach [Miquel and Basso, 2010] has some similar concepts to those described above for the CIEMAT model, but rather than representing the dynamic hydrology of multiple soil layers, SAMM integrates advection and diffusion

processes between soil layers; the number of soil layers being user-defined. The model approach is illustrated in Figure 3.

Contamination can take place via the application of irrigation water to the surface of the soil and subsequent advective movement downward to an aquifer or through upward movement from a contaminated aquifer to the surface depending on the surface and soil hydrological balance. Downward advection occurs if the hydraulic balance of the soil (precipitation plus irrigation minus evapotranspiration) is positive. Upward advection (i.e. via capillary rise) occurs if evapotranspiration is greater than precipitation plus irrigation input.

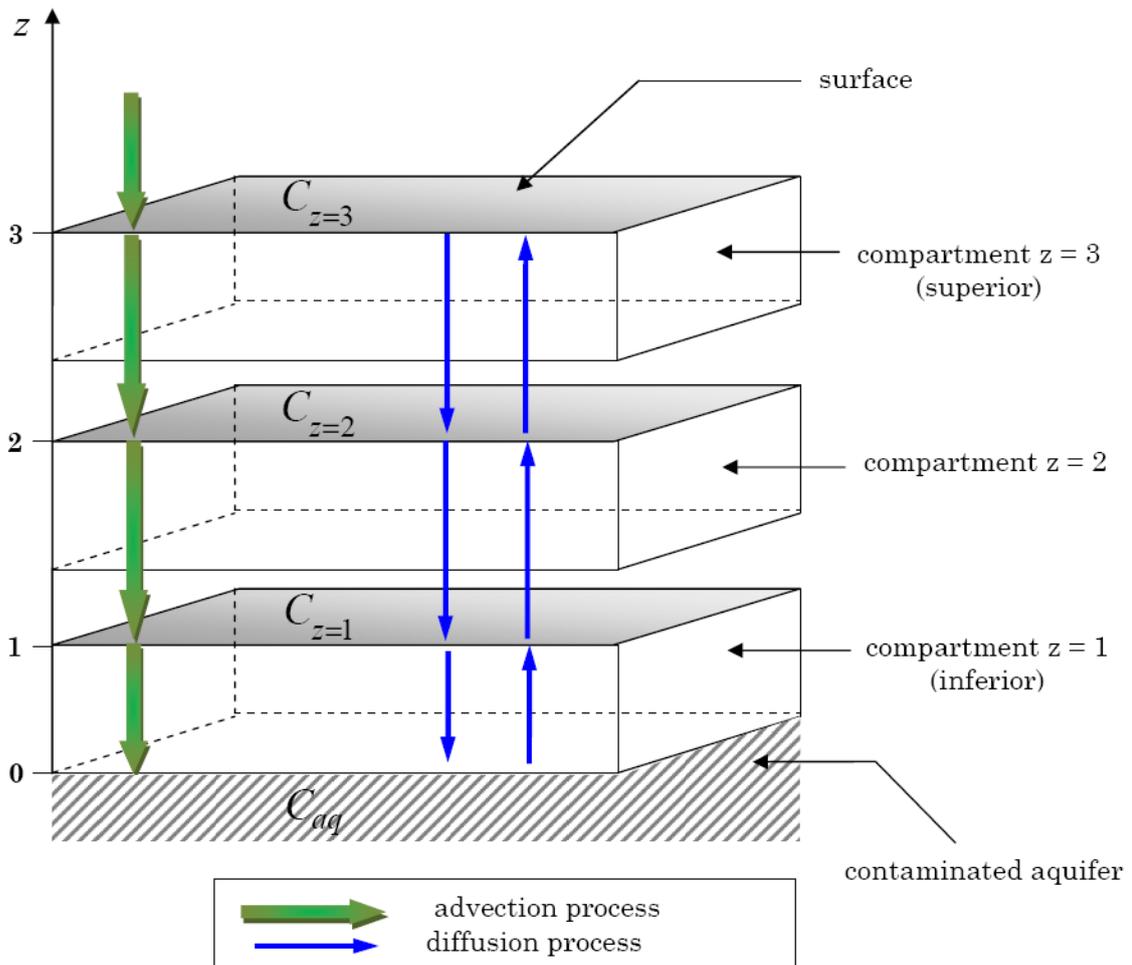


Figure 3 SMM: Discretised vertical soil column with three compartments

The model uses a one-dimensional representation of soil, with multiple soil layers, each user defined but of equal thickness, with an aquifer present below the lowest soil layer. Porosity and the degree of saturation of each soil layer are defined and the Kd value for each soil layer can be varied (Figure 4).

soil parameters			
Depth (m)	Degree of saturation (-)	Soil porosity (-)	Root distribution (%)
0.2	0.5	0.5	48.8
0.4	0.5	0.5	29.6
0.6	0.5	0.5	15.2
0.8	0.5	0.5	5.6
1	0.5	0.5	0.8
1.2	0.5	0.5	
1.4	0.5	0.5	
1.6	0.5	0.5	
1.8	0.5	0.5	
2	0.5	0.5	
Aquifer	1	0.5	

Figure 4 Example of a 2 m soil column with a defined root distribution and user defined constant soil physics

Diffusion of Se-79 in soils is controlled by the local concentration gradients defined on the basis of Kd in each soil layer (Figure 5).

λ (y)	Depth (m)	Kd (m ³ /kg)
1.07000E-05	0.2	0.02
	0.4	0.02
Diffusion coefficient in pure water (m ² /y)	0.6	0.02
	0.8	0.02
0.038	1	0.02
	1.2	0.02
	1.4	0.02
	1.6	0.02
Soil-to-plant transfer factor (kg _{dry} /kg _{wet})	1.8	0.02
	2	0.02
2.226	Aquifer	0.02

Figure 5 Definition of transfer parameters, of which Kd may vary as a function of depth

Uptake of Se-79 from soils to plants occurs via root uptake, with transfer to plants being represented by a soil-plant transfer factor (depending on the kind of agricultural plant) that takes into account root distribution and total Se-79 concentration within soil layers. Transfer from soils to plants is limited in

this example to the upper five soil compartments^a with root distribution decreasing as a function of depth from ca. 50% in the top soil layer to ca. 1% in soil layer 5.

The model allows the concentration in each soil layer to be calculated over multiple time steps such that migration between soil layers can be determined. Volatilisation is conservatively excluded from the model.

4.3 E-K MODEL DEVELOPED AT IRSN

The E-K model developed at IRSN as a Kd alternative model [Coppin et al, 2011] is implemented in the SYMBIOSE platform. The model has recently been developed and is in the process of being validated. The model can be used to represent a single or multiple soil layers: the depth of each soil layer being user-defined. Furthermore, for the multiple soil layers it is possible to define the root distribution in each soil layer.

The selenium soil/solution distribution within a soil layer is defined by using an equilibrium/kinetic model (E-K) as illustrated in Figure 6. Briefly the E-K model considers that the sorbed selenium is divided in two fractions: one labile fraction (S1) for which selenium is in equilibrium with the solution (i.e. Kd' is applied) and another fixed (recalcitrant) fraction (S2) for which the sorption and desorption are kinetically limited (k+ and k- parameters). The nature of the fixed component is not explicit in this model.

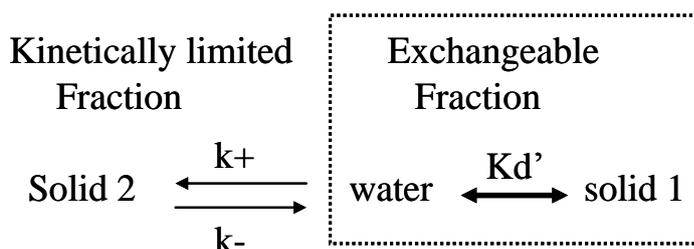


Figure 6 Schematic representation of the E-K model

The E-K model does not explicitly consider irrigation, precipitation or evapotranspiration. Water flux in the system is a net flux of water (percolation rate), calculated by subtracting evapotranspiration flux from the combined flux from irrigation and precipitation. Losses of Se-79 from volatilisation and by cropping are not considered. Only losses by percolation to below the lowest soil layer are considered.

As with the conventional Kd model, Se-79 in percolate water is calculated according to the following equation:

^a Depth of root penetration and root distribution within each layer can be varied. Values presented are those applied within the model application to the hypothetical scenario outlined in Section 5.

$$\begin{aligned} & \text{Se79 in percolate water (Bq.L}^{-1}\text{)} \\ &= \frac{\text{Irrigation rate (m y}^{-1}\text{)} * \text{Se79 concentration in irrigation rate (Bq.L}^{-1}\text{)}}{\text{Percolation Rate (m y}^{-1}\text{)}} \end{aligned}$$

The available Se-79 activity includes that sorbed onto site S1 of the soil, rather than the total sorbed to soil, and is calculated according to the equation:

Se79 available pool (Bq kg⁻¹) =

$$\begin{aligned} & \text{Se79 sorbed on site S1 of soil (Bq kg}^{-1}\text{)} \times \text{dry soil weight of soil layer (kg)} \\ & + \frac{\text{Se79 in solution (Bq L}^{-1}\text{)} \times \text{volume of solution in soil layer (L)}}{\text{dry soil weight of soil layer (kg)}} \end{aligned}$$

Transfer of Se-79 from soil to plant is calculated by using a constant transfer factor for selenium but considering that the selenium plant uptake is restricted to a bioavailable pool (i.e. the exchangeable fraction of Figure 6) constituted by the selenium present in soil solution and the selenium sorbed on site S1. Plant uptake does not affect the concentration of selenium in the bioavailable pool because the model considers that the selenium content of this pool is infinite compared with that taken up by plants.

5. APPLICATION OF MODELS TO A HYPOTHETICAL SCENARIO

In order to assess the relative capabilities of the models described in Section 4 a hypothetical scenario has been developed. The objectives of applying models to the scenario were to provide:

- a mechanism for exploring the significance of different model and data assumptions as a means of investigating the level of complexity required to provide confidence in the long-term modelling of Se-79, and,
- practical experience in applying the scientific information available to an assessment question.

Each of the models described in Sections 3 and 4 has been applied. Details of the application of models to the scenario and results are presented.

5.1 SCENARIO DESCRIPTION

5.1.1 SOURCE TERM

Two source terms were suggested, which were to be considered separately.

1. Unit activity concentration of Se-79 (1 Bq/L) in irrigation water is applied to soil to meet agricultural requirements. Interception by growing crops was excluded from consideration such that 100% direct deposition to the soil is assumed. Irrigation water is applied year on year.
2. Unit activity concentration of Se-79 (1 Bq/L) in upwelling groundwater. Upwelling occurs due to seasonal variation in water table level or due to capillary rise during relevant parts of the year. Account to be taken of the 5 years of weather data supplied, as an illustration of variation within each year and from year to year.

The chemical form of Se-79 was to be advised by those carrying out the calculation of release of Se-79 in groundwater from the geosphere. (This will clearly have been a requirement for the geosphere part of the assessment, but may vary from site to site, and the variations may be important).

5.1.2 SOIL PROPERTIES

Proposed soil properties are given in Table 5; these being the same as those used in the recently completed CI-36 studies [Bytwerk et al, 2011; Limer et al, 2009].

Table 5 Proposed soil properties

Parameter	Value
Topsoil thickness (m)	3.0E-1
Soil Porosity (-)	4.5E-1
Soil water content ($m^3_{\text{water}} m^{-3}_{\text{soil}}$)	2.5E-1
Soil grain density ($kg m^{-3}$)	2.6E+3

Assumptions for soil mineralogy, organic content and microbial activity as are relevant to the soil types of most interest to sites were to be considered.

5.1.3 CLIMATE AND WATER DATA

Relevant data, based on the CI-36 study assumptions, are provided in Table 6.

Table 6 Climate data

Parameter	Units	Value*	References / Notes
Precipitation	$m y^{-1}$	5.94E-1	Météo France [2008], detailed 5 y data were supplied separately for those who could use them.
Irrigation rate	$m y^{-1}$	Cereal: 1.44E-1	The focus on a single crop type is to avoid complications of different crop type water requirements.
Evapotranspiration	$m y^{-1}$	7.12E-1	Or varied according to Meteo Data.
Percolation Rate	$m y^{-1}$	Cereal: 2.6E-2	Or varied according to Meteo Data.

*The data imply no net annual upwelling, but there can be upwelling within seasonal variation, resulting in radionuclide transfer.

5.1.4 CALCULATION ENDPOINTS

For each source term:

- Concentration of Se-79 in bulk soil in top 30 cm as a function of time from onset of release, Bq/kg, until quasi-equilibrium occurs.

- Concentration of Se-79 in the grain part of the harvested crop at the time of harvest for successive years (ignoring any contribution from interception of irrigation water), Bq/kg fresh weight.

5.2 MODEL APPLICATION AND RESULTS

The output from each model for the scenario(s) to which they have been applied are detailed in this section. Where relevant, descriptions of the approach taken to applying the models to the scenario are provided.

5.2.1 CIEMAT MODEL

The CIEMAT model was applied to the irrigation scenario only. This was achieved by adapting the reference scenario from the sensitivity study described in Pérez-Sánchez et al [2012]. The modifications required in order to represent the scenario are presented below.

Hydrology

Five year detailed meteorological data (from Meteo France [2008]) were employed as input to the MS Excel hydrology spreadsheet model. Specifically, the mean monthly temperature, rainfall and potential evapotranspiration (PE) were averaged over the five years of the data and the rainfall and PE were then multiplied by twelve to give the rainfall rate and PE rate on an annual basis for each calendar month of the year. Results are listed in Table 7, where month 1 is taken to be January.

Table 7 Averaged monthly climate data

Month	Temperature (°C)	Rainfall Rate (m/y)	PE Rate (m/y)
1	5.38E+00	1.07E+00	3.40E-01
2	2.97E+00	1.03E+00	1.87E-01
3	7.09E+00	4.83E-01	5.64E-01
4	1.05E+01	7.32E-01	8.52E-01
5	1.30E+01	7.65E-01	7.07E-01
6	1.80E+01	4.45E-01	1.27E+00
7	1.82E+01	5.19E-01	1.34E+00
8	1.70E+01	5.50E-01	9.03E-01
9	1.65E+01	8.42E-01	1.04E+00
10	1.44E+01	5.75E-01	7.25E-01
11	6.53E+00	9.89E-01	5.00E-01
12	3.72E+00	9.00E-01	2.28E-01

For months 5-8 (May, June, July, August), the rainfall rate R was augmented by the specified irrigation rate (0.144 m for cereals). Assuming a uniform rate of irrigation over months 5-8, the effective irrigation rate is 0.432 m y⁻¹ applied over four months of the year. Thus, the rainfall rates for months 5-8 were increased by 0.432 m y⁻¹ to allow for the contribution from irrigation. Thus, the total

rates were 1.197, 0.877, 0.951 and 0.982 m y⁻¹ in months 5, 6, 7 and 8, respectively, as detailed in Table 8.

The actual evapotranspiration (AE)/PE ratio was taken as 0.4, as discussed in Thorne [2010]. However, with a = 0.8 and b = 1.5, this led to substantial drainage of the upper layers of the soil and an average value of the soil water content (θ) of 0.25 m³ m⁻³ could not be achieved in the top two soil layers (0-0.2 m depth and 0.2-0.4 m depth, respectively). To deal with this, the value of a was reduced to 0.3, i.e. the soil was treated as less freely draining than the sandy soil assumed in the reference calculations.

As is standard practice with the CIEMAT hydrology model, a ten year period was simulated, duplicating the meteorological data each year. The first nine years are a spin-up period and results from the tenth year are transferred to the radionuclide transport model; the annual cycle in soil hydrology had stabilised by year 10. The year 10 hydrological characteristics, showing the height of the water table above the base of the 2 m soil column (h) are set out in Table 8 (note that the times are the beginning of the period for which the data are given).

Table 8 Water table height and input hydrological data in year 10

Time (y)	9.00	9.08	9.17	9.25	9.33	9.42	9.50	9.58	9.67	9.75	9.83	9.92
h (m)	1.57	1.66	1.73	1.63	1.57	1.65	1.59	1.54	1.55	1.52	1.45	1.51
R (m/y)	1.07	1.03	0.48	0.73	1.20	0.88	0.95	0.98	0.84	0.58	0.99	0.90
PE (m/y)	0.34	0.19	0.56	0.85	0.71	1.27	1.34	0.90	1.04	0.73	0.50	0.23
AE	0.14	0.075	0.23	0.34	0.28	0.51	0.54	0.36	0.42	0.29	0.2	0.09

By this adjustment, the water table remains at a roughly constant level of 1.45 to 1.73 m above the base of the soil column (0.55 to 0.27 m below the ground surface) throughout the year; it does not fall in the summer months because the augmentation of rainfall by irrigation compensates for enhanced actual evapotranspiration over the summer months.

The water contents of the ten 0.2 m thick soil layers are shown in Table 9.

Table 9 Soil Water Contents in Year 10 (Note Theta 1 is the θ value for the first 0.2 m soil layer closest to the surface and all soil layers are 0.2 m in thickness)

Time (y)	9.00	9.08	9.17	9.25	9.33	9.42	9.50	9.58	9.67	9.75	9.83	9.92
Theta 1	0.14	0.21	0.28	0.18	0.14	0.20	0.15	0.12	0.13	0.11	0.10	0.11
Theta 2	0.32	0.40	0.44	0.38	0.32	0.39	0.34	0.29	0.30	0.27	0.20	0.26
Theta 3	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.44	0.44	0.43	0.39	0.43
Theta 4	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45
Theta 5	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45
Theta 6	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45
Theta 7	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45
Theta 8	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45
Theta 9	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45

Theta 10	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45	0.45
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The water table is typically located close to the top of layer 3, hence θ values are typically 0.45 for layers 3-10. Note that Theta1 was modified in the spreadsheet from its original value of 0.5 to 0.45. This was because the scenario specified that the total soil porosity is 0.45. Thus, by definition, $\theta = 0.45$ at saturation. Theta-v was corresponding reduced from 0.4 to 0.35.

In soil layer 1, θ varies from 0.10 to 0.28 over the year. In soil layer 2, θ varies from 0.20 to 0.44. There is no obvious seasonal variation and the fluctuations are most appropriately attributed to the variability in hydrological inputs. The mean annual values of θ for soil layers 1 and 2 are 0.15 and 0.33, respectively. The average over these two layers is 0.24, which is in reasonable agreement with the value of 0.25 specified in the scenario.

Water fluxes between the soil layers are shown in Figure 7 (note that positive fluxes are directed downward). From Figure 7 it is evident that all fluxes are positive, i.e. the inclusion of irrigation during the summer months more than compensates for the enhanced evapotranspiration over that period and maintains a net downward water flux. No strong seasonal trend is evident in the derived fluxes.

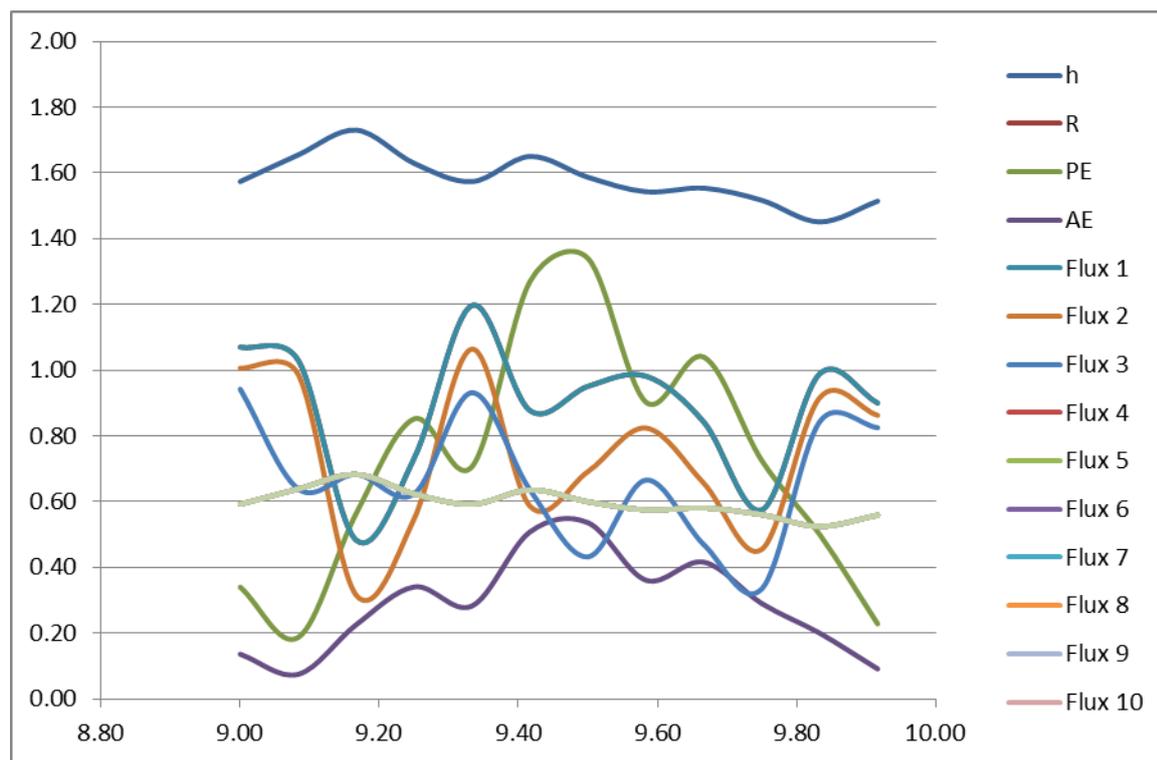


Figure 7 Soil Water Fluxes in Year 10 (Units m/y; positive fluxes are directed downward)

The water table in winter lies relatively close to the soil surface and, in the summer, the soil moisture deficit without irrigation would be relatively limited; as such the need for irrigation of cereal crops would be minimal (cereals are capable of developing root systems to a depth of 1 m or more, so could

readily extract water from the capillary fringe of a deeper water table). However, because the water table is maintained close to the surface by irrigation, cereal roots would be mainly confined to the upper part of the soil column. In the transport model, it was therefore assumed that roots are present only in soil layer 1, as the maintained moisture content of this layer during the summer is above the wilting point.

The hydrology model also calculates Kd values on a normalised range of [0, 1]. The actual Kd values are then estimated in the transport model by scaling these normalised values to the specified Kd range.

The normalised Kd values are listed by soil layer in Table 10.

Table 10 Normalised Kd Values for Year 10

Time (y)	9.00	9.08	9.17	9.25	9.33	9.42	9.50	9.58	9.67	9.75	9.83	9.92
Kd1	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Kd2	0.14	0.53	0.88	0.40	0.14	0.50	0.19	0.05	0.07	0.01	0.00	0.01
Kd3	0.98	1.00	1.00	1.00	0.98	1.00	1.00	0.92	0.95	0.83	0.51	0.82
Kd4	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Kd5	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Kd6	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Kd7	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Kd8	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Kd9	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Kd10	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00

Note that normalised Kd values are 1.0 below the water table and are close to zero in soil layer 1. That layer is substantially under-saturated throughout the year, permitting ready oxygen penetration from the surface. Soil layer 2 is generally substantially under-saturated, but can sometimes lie within the capillary fringe, resulting in reduced oxygen availability.

Radionuclide Transport

All the above hydrological data were transferred into the radionuclide transport model. In addition, the following hydrological adjustments were made to the transport model.

- Th_{max} (water content of the soil at saturation) was reset from 0.5 to 0.45 to conform to the specified total porosity of the soil, but Th_{min} (minimum water content of the soil well above the capillary fringe) was maintained at 0.1;
- Th_a , Th_b , Th_c and Th_d (relating to volatilisation from soil) were scaled by a factor of 0.45/0.5 to reflect the altered total soil porosity – the resultant values were 0.27, 0.36, 0.43 and 0.45.

Cereals were taken to have a growth period starting at the beginning of April (month 4) and finishing in September (month 9). The standing biomass, biomass production rates and removal rates adopted (based on a reference model area of 1 m²) are listed in Table 11.

Harvesting is taken as occurring at the beginning of October (month 10, see also below) and is set to match annual production. The standing biomass after harvest (0.05 kg d.w.) represents roots and stubble. These degrade to soil organic matter over the winter months.

As the scenario specifies that the soil particle density is 2600 kg m⁻³ and the porosity is 0.45, the soil bulk density is set to 2600*0.55 = 1430 kg m⁻³.

Root uptake is parameterised using the parameter G_x (x = 1, 10 for the various soil layers). Due to the high water table, roots are taken only to develop in soil layer 1. Thus, G₂ to G₁₀ are set to zero for all months. G₁ is also zero during the winter. From the analysis in Thorne [2010], G is typically 0.05 m⁻¹ y⁻¹ for a rapidly growing crop. Taking account of reduced growth during final grain maturation, G₁ was taken as the following vector over months 1-12 {0, 0, 0, 0.05, 0.05, 0.05, 0.05, 0.05, 0.02, 0, 0, 0}.

Table 11 Standing Biomass, Biomass Production Rate and Biomass Removal Rate

Month	Standing Biomass (kg d.w.)	Production Rate (kg d.w. per year)	Removal Rate (kg d.w. per year)
1	0.0500	0.0000	0.0000
2	0.0500	0.0000	0.0000
3	0.0500	0.0000	0.0000
4	0.0708	0.5000	0.0000
5	0.1250	0.8000	0.0000
6	0.2208	1.5000	0.0000
7	0.3667	2.0000	0.0000
8	0.5750	3.0000	0.0000
9	0.7833	2.0000	0.0000
10	0.4583	0.0000	9.8000
11	0.0500	0.0000	0.0000
12	0.0500	0.0000	0.0000
Total		9.8000	9.8000

The scenario specified a source term of unit activity concentration of Se-79 (1 Bq L⁻¹) in irrigation water with direct deposition to soil. As the irrigation rate is 0.144 m y⁻¹, so the time-averaged input rate to soils is 144 Bq y⁻¹. However, as discussed above, irrigation is taken to occur only in the summer months (May to August), so the activity input is taken as 432 Bq y⁻¹ over these four months and 0 Bq y⁻¹ over the other eight months of the year.

The CIEMAT model was designed for a well-drained agricultural soil rather than one with significant upwelling groundwater and, as such, a correction was needed to the hydrological data. As there is 0.1 m y⁻¹ of water entering from below that is unaccounted for, there was a need to increase F_{10_S} in the AMBER model by 0.1 m y⁻¹ in each month to allow for loss of this additional water to the drain, as it effectively decreases the soil water residence time in soil layer 10. This is a relatively small (~20%) perturbation to F_{10_S}.

Results

Results from the model using the above parameterisation are set out below. The model was run for a period of 50 years with an output time step set at 0.02 y. Figure 8 shows plant concentrations over this period, Figure 9 shows soil concentrations (excluding the contribution from soil organic matter) in soil layer 1 and Figure 10 shows the amount of Se-79 in soil organic matter.

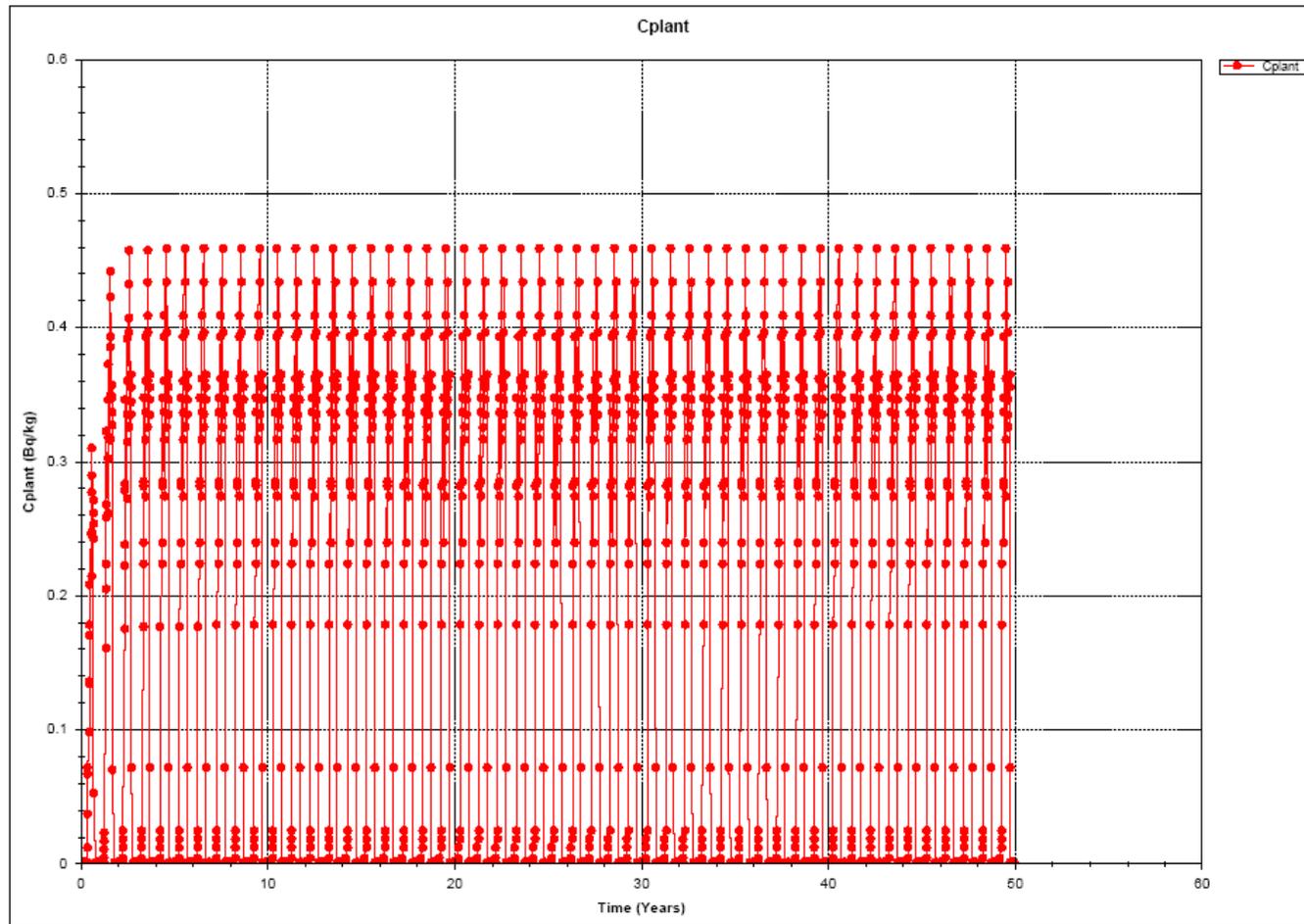


Figure 8 Plant Concentrations (Bq/kg d.w.) for Irrigation Input

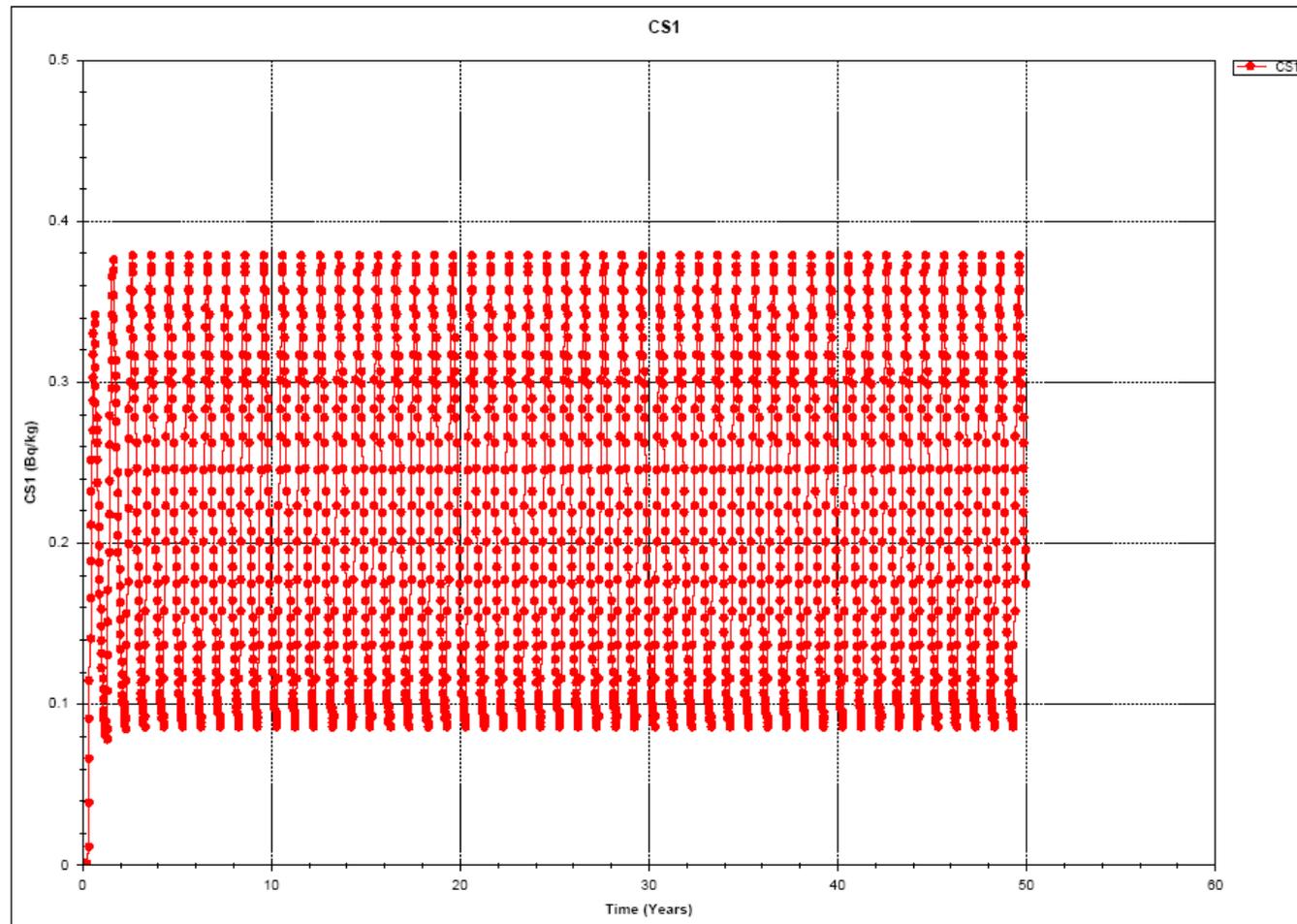


Figure 9 Soil Layer 1 Concentrations (Bq/kg d.w.) for Irrigation Input

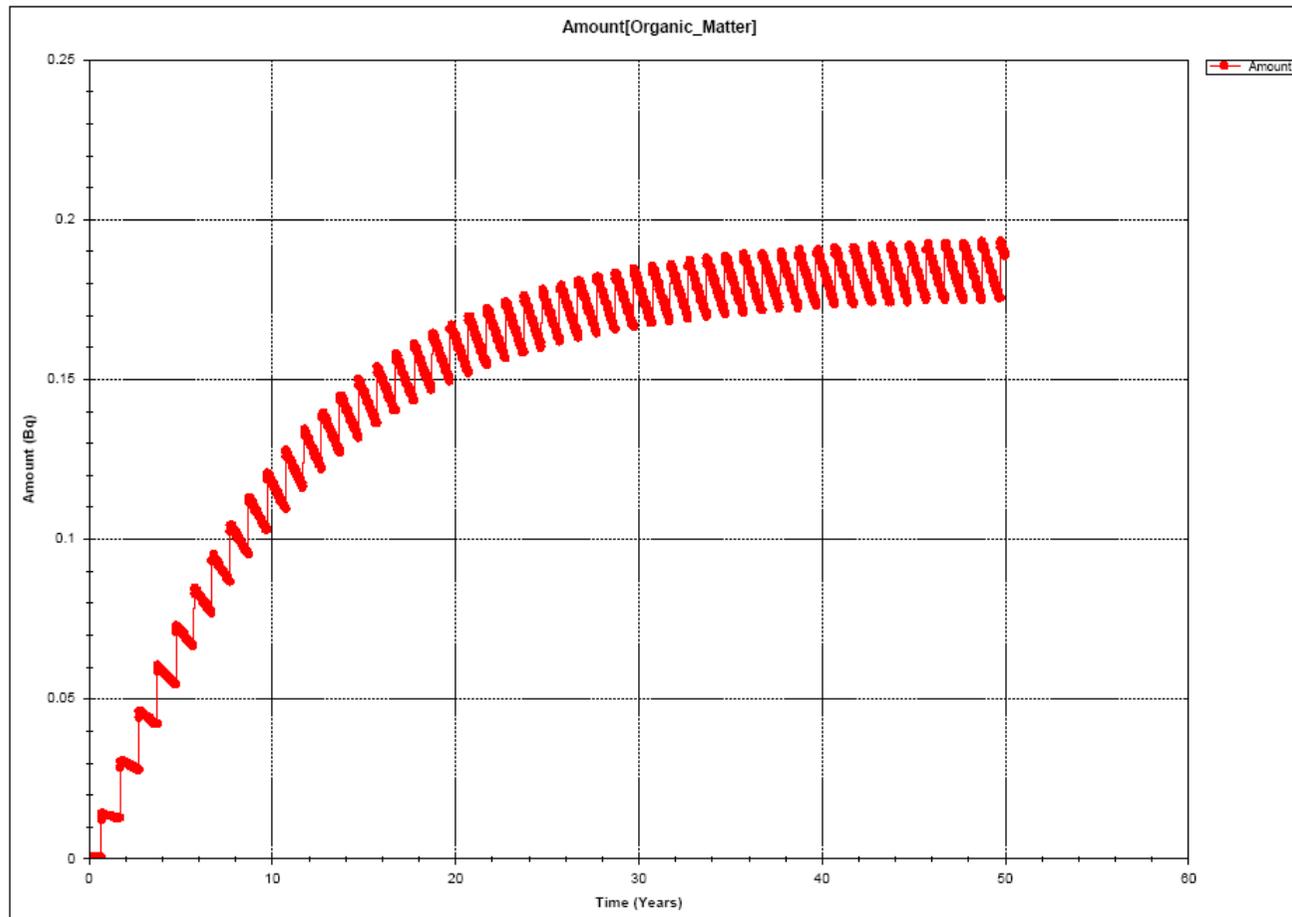


Figure 10 Soil Organic Matter Content (Bq/kg d.w.) for Irrigation Input

Figure 8 to Figure 10 indicate saturation of the system on a timescale of a few years to decades. Thus, a 50 year simulation period is considered adequate. Figure 11 displays results on an expanded timescale in order to illustrate the intra-annual variability in plant concentrations.

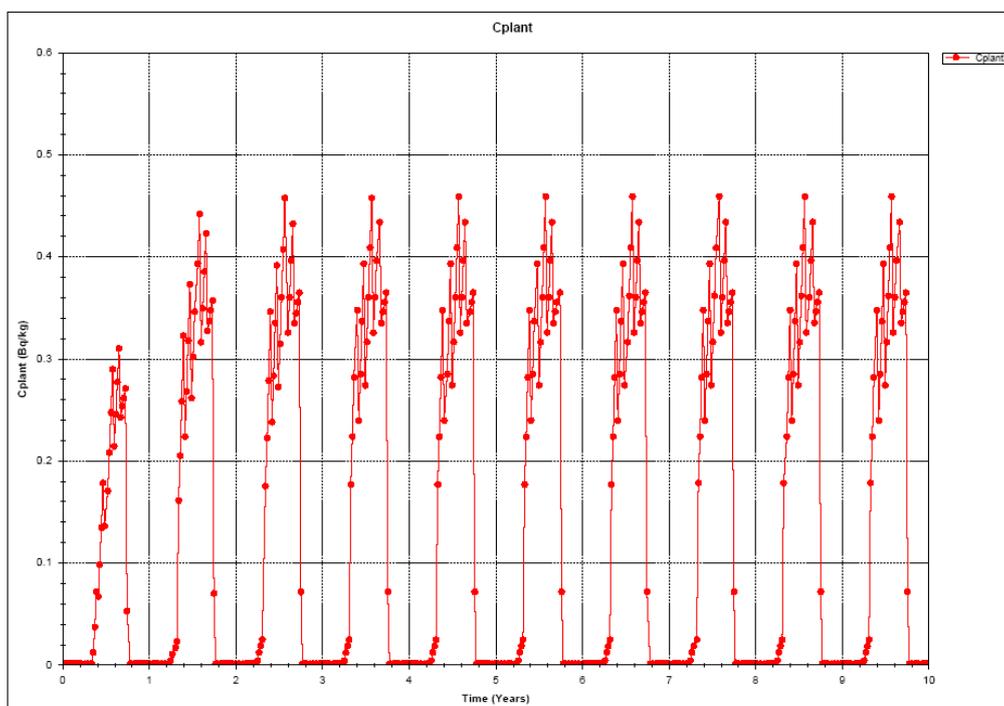


Figure 11 Plant Concentrations (Bq/kg d.w.) over the First 10 Years for Irrigation Input

Plant concentrations begin to increase from the start of each growing season and peak at harvest. The detailed structure before harvest is due to the competing effects of on-going root uptake and growth dilution, but overall root uptake dominates over growth dilution. At harvest, the plant activity is completely removed either through cropping or return to soil organic matter. The detailed picture for soil organic matter activity (Figure 12) shows increases in the organic matter activity at each harvest followed by a decrease due to mineralisation.

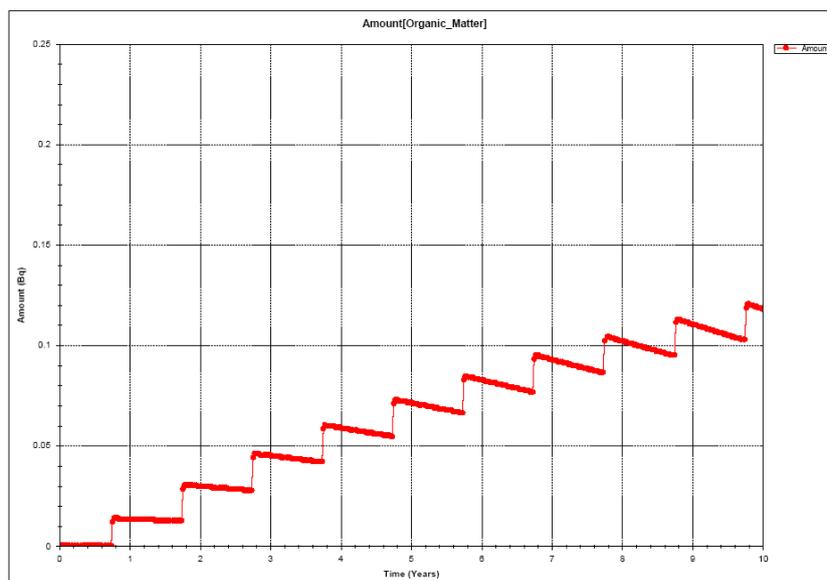


Figure 12 Activity (Bq/kg d.w.) in Organic Matter for the Irrigation Case

Results from the time of harvest in Year 50 of the calculations in all the model compartments are detailed in 0.

Because of the low K_d values adopted for Se-79 and the dominant downward vertical flow in the system, the activity is distributed to a substantial depth in the soil column and significant losses to the soil sink have occurred. Substantial volatilisation from soils has also occurred, with 0.435 of the added Se-79 having been lost by this route, which is consistent with volatilisation rates observed in the literature. However, because of the limited activity inventory of Se-79 in plants, volatilisation from plants is of limited importance.

If the activity content of plants at harvest 50 is taken as indicative of the whole of the simulated period, total cropping loss plus transfer to organic matter to 49.74 years can be estimated as $49 \times 0.286 = 14$ Bq. The total found in cropping loss plus organic matter is 13.4 Bq, in good agreement with this crude calculation. The fraction in organic matter is $0.175 / 13.4 = 0.013$. This is rather less than the amount of $14 \times 0.064 = 0.896$ Bq that would be expected. This arises because Se-79 in organic matter is subject to mineralisation and returned to the inorganic pool in soils. The difference between 0.896 Bq and 0.175 Bq is 0.721 Bq. If this is added to the total found in cropping loss plus organic matter of 13.4 Bq, the result is 14.1 Bq, bringing the estimate into better agreement with the total assessed as being lost from plants over 49 harvests. These mass balance calculations serve to demonstrate the internal consistency of the model.

Table 12 Activity Contents at Harvest in Year 50 (49.74 years of simulation)

Compartment	Activity Content (Bq)	Fraction
Plants	2.86E-01	3.96E-05
Soil 1	9.36E+01	1.30E-02
Soil 2	1.49E+02	2.07E-02
Soil 3	5.57E+02	7.72E-02
Soil 4	5.37E+02	7.44E-02
Soil 5	4.70E+02	6.52E-02
Soil 6	4.08E+02	5.66E-02
Soil 7	3.52E+02	4.89E-02
Soil 8	3.01E+02	4.17E-02
Soil 9	2.53E+02	3.50E-02
Soil 10	1.82E+02	2.52E-02
Organic Matter	1.75E-01	2.43E-05
Cropping Loss	1.32E+01	1.83E-03
Soil Sink	7.60E+02	1.05E-01
Volatile Sink from Soil	3.14E+03	4.35E-01
Volatile Sink from Plants	3.12E-02	4.32E-06
Total	7.21E+03	1.00E+00

The soil and plant concentrations at harvest 50 (49.74 y) are:

- 0.33 Bq/kg for soil layer 1;
- 0.52 Bq/kg for soil layer 2; and
- 0.36 Bq kg⁻¹ for plants on a dry weight basis.

The contribution from Se-79 in organic matter in soil is small in comparison with the inorganic contribution and is neglected.

Taking the soil layer 1 concentration and that in plants at harvest 50 gives a soil to plant concentration ratio of 1.12 (on a dry weight plant, dry weight soil basis). Values of this order of magnitude have been reported in the literature and a soil to plant concentration ratio ~1 is assumed in the derivation of G values reported in Thorne [2010].

5.2.2 NDA RWMD SPREADSHEET MODEL

The NDA RWMD model has been applied to the irrigation scenario.

Hydrological properties applied within the NDA RWMD Spreadsheet model and their values, based on 5-year meteorological data, are listed in Table 13. The parameters are considered representative of a well-drained agricultural soil that would be suitable for growing cereals.

Table 13 Hydrological Quantities and Reference Values

Quantity	Units	Description	Value
h_1	$m\ y^{-1}$	Precipitation plus irrigation	0.884
l_2	$m\ y^{-1}$	Percolation to subsoil	0.884
E_1	$m\ y^{-1}$	Evapotranspiration	0.25
E_2	$m\ y^{-1}$	Capillary rise to replenish soil moisture deficit	0.25
S_{in}	$m\ y^{-1}$	Throughflow gains from surface soil	0
B_{in}	$m\ y^{-1}$	Baseflow gains from subsoil	0
S_{out}	$m\ y^{-1}$	Throughflow losses from surface soil	0
B_{out}	$m\ y^{-1}$	Baseflow losses from subsoil	0.634
G	$m\ y^{-1}$	Groundwater discharge to subsoil	0

The relevant soil characteristics are as set out in Table 14. Note that the water-filled soil porosity is not required since the spreadsheet assumes that the soil is maintained at field capacity.

Table 14 Soil Characteristics

Parameter	Symbol	Reference Value	Units
Depth of topsoil	d_1	0.3	m
Porosity of topsoil	ϕ_1	0.45	N/A
Density of topsoil	ρ_1	1430	kg/m^3
Depth of subsoil	d_2	5	m
Porosity of subsoil	ϕ_2	0.5	N/A
Density of subsoil	ρ_2	1325	kg/m^3

A range of Kd values have been applied within the model (0.001, 0.003, 0.01, 0.03, 0.1, 0.3 and 0.49 $m^3\ kg^{-1}$) to give an appreciation of the sensitivity to this parameter value.

The plant to soil concentration ratio for Se-79 is 1.0. However, it should be noted that this is defined on a fresh weight basis and corresponds to a value ~ 10 for most crops and ~ 2 for cereal grain on a dry weight basis. As cropping losses are not included in the model, the reference value can be left unchanged at 1.0, but the results can be interpreted as applying on a dry weight basis rather than a fresh weight basis.

Results

Values of CT_1 (total Se-79 in surface soil) and CT_2 (total Se-79 in subsoil) for different values of K_d are listed in Table 15.

Table 15 Total Soil Concentrations

K_d Value ($m^3 kg^{-1}$)	CT_1 ($Bq kg^{-1}$)	CT_2 ($Bq kg^{-1}$)
0.001	0.299	0.299
0.003	0.753	0.753
0.01	2.34	2.34
0.03	6.88	6.86
0.1	22.7	22.5
0.3	67.4	65.7
0.49	109	105

A compatible range of soil K_d values to those within the CIEMAT model of 0.001 to 0.01 $m^3 kg^{-1}$ have been applied in the NDA RWMD spreadsheet model, resulting in total soil concentrations in topsoil of 0.299 to 2.34 $Bq kg^{-1}$. For comparison, at harvest 50, the CIEMAT model gave 0.327 $Bq kg^{-1}$. This value is toward the lower end of the calculated range because soil layer 1 in the CIEMAT model is generally unsaturated and the lower end of the range of K_d values is taken to apply in unsaturated conditions. Results from the NDA RWMD spreadsheet model therefore correspond very closely to those obtained with the detailed CIEMAT model.

5.2.3 ANDRA SAMM MODEL

The ANDRA SAMM model was applied to both the groundwater and irrigation scenarios.

Groundwater scenario

The model was applied with respect to two irrigation rates; the given irrigation rate of 0.144 $m y^{-1}$ and the estimated irrigation rate of 0.491 $m y^{-1}$. Both irrigation rates have been applied either with or without the export of crops from the modelled area (Figure 13). The model has been applied taking account both of seasonal variability in climate data and through the application of annual climate data.

Results from the application of seasonal climate data

A similar trend is evident between soil and plant concentrations over time, with cereal concentrations ($Bq kg^{-1}$ fresh weight) being approximately 3 times greater than soil concentrations ($Bq kg^{-1}$ dry weight) when an irrigation rate of 0.144 $m y^{-1}$ is assumed.

Increasing the irrigation rate from 0.144 $m y^{-1}$ to 0.491 $m y^{-1}$ effectively reduced the ability of contaminated groundwater to reach the upper soil compartments that equate to the soil rooting zone; hence cereal Se-79 concentrations remain at zero throughout the simulation period irrespective of cropping processes. Deficit in the hydrological balance at an assumed irrigation rate of 0.144 $m y^{-1}$

however results in the advection of contaminated groundwater to the surface soils and Se-79 is then available for plant uptake. Where cropping is assumed to occur, such that 100% of the crop is exported, equilibrium in soil and plant concentrations is achieved within 500 years. If it is assumed that cropping does not occur such that recycling of organic matter occurs, both soil and plant concentrations continue to rise throughout the simulation period.

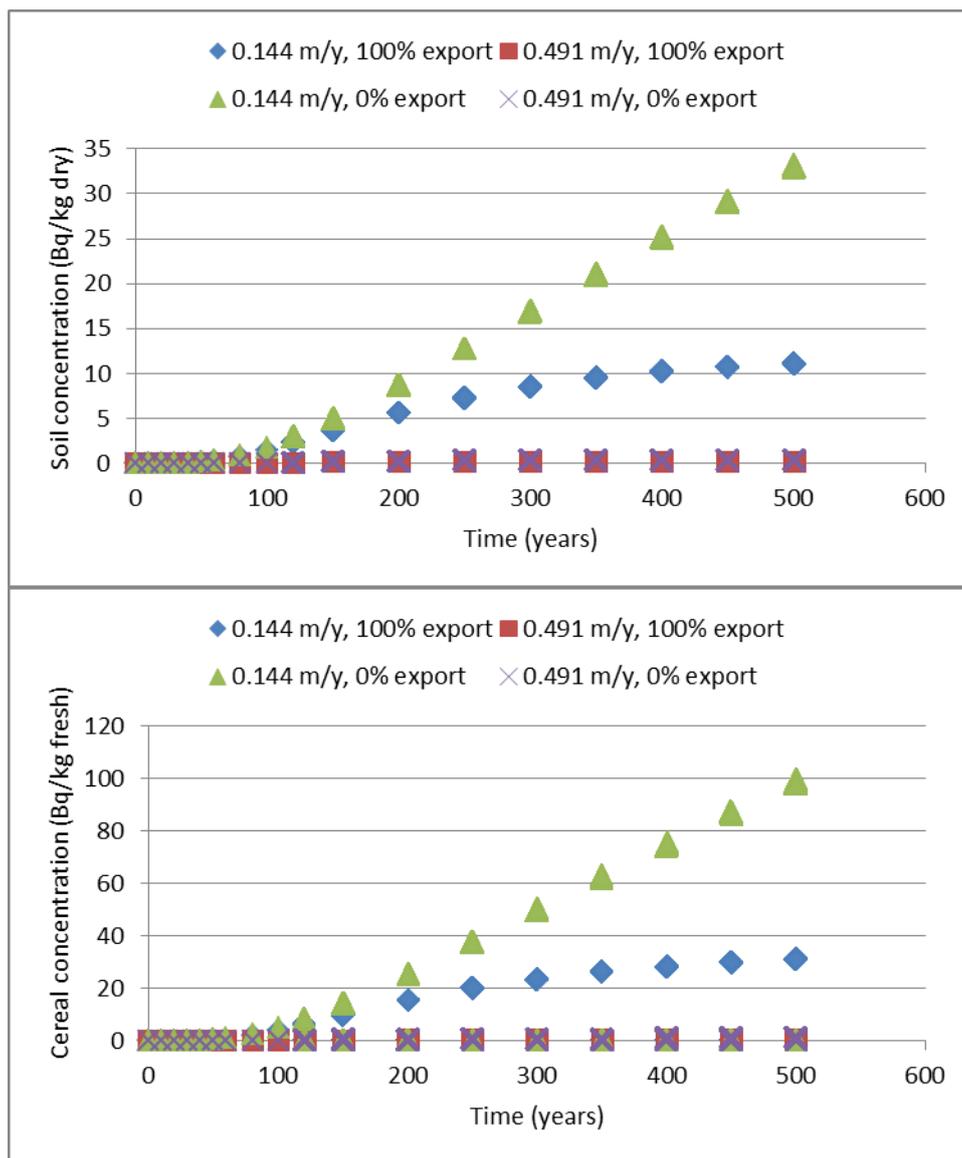


Figure 13 Results of the ANDRA SAMM model for the groundwater scenario for soil (upper) and cereals (lower).

Results from the application of annual climate data

When seasonal variability is excluded from the model, i.e. irrigation rates and soil hydrological conditions throughout the year are assumed constant, results indicate that the time required for any Se-79 in groundwater to reach the soil rooting zone is extended by approximately 200 years as compared against the results of the seasonal climate variation scenario (Figure 14). Activity concentrations reaching the upper soil layers are substantially suppressed by the exclusion of seasonal variation from the model. Equilibrium soil and plant concentrations are not achieved within the 500 year simulation period.

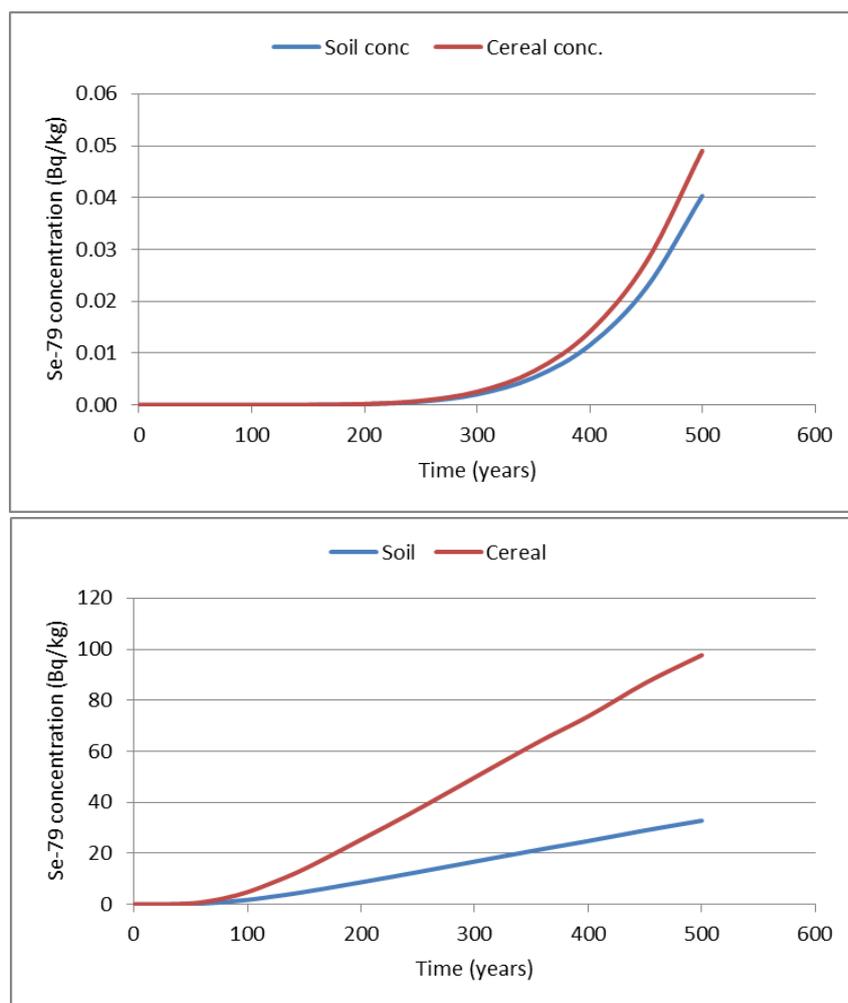


Figure 14 Se-79 concentrations in soil (Bq kg^{-1} dry weight) and cereals (Bq kg^{-1} fresh weight) when seasonal variation is excluded (above) or included (below) from the groundwater scenario (irrigation rate 0.144 m y^{-1}), no cropping losses.

Irrigation scenario

The irrigation scenario again considered differences in model output in relation to whether or not cropping losses are assumed (Figure 15). The model was applied taking account of seasonal variation in climate data.

The removal of 100% of the crop at harvest time resulted in a large reduction in overall soil and plant concentrations compared with a 0% cropping assumption whereby Se-79 cycling between plant and soil organic matter could occur. When cropping is assumed, equilibrium conditions are achieved within the first few years of simulation. In the absence of cropping losses, equilibrium is not achieved within the 500 year simulation period.

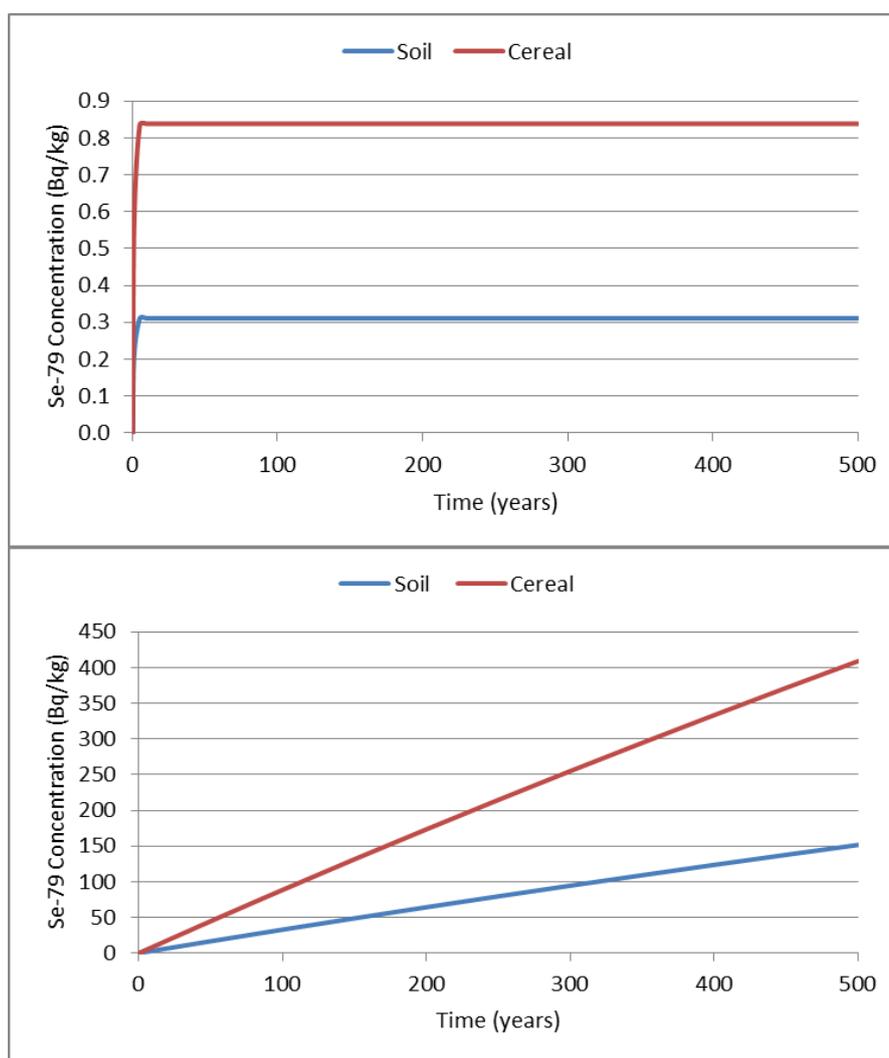


Figure 15 Results of the ANDRA SAMM model for the irrigation scenario with (upper) and without (lower) cropping losses. Results are presented as Bq kg⁻¹ dry weight for soils and Bq kg⁻¹ fresh weight for cereals.

5.2.4 ANDRA AQUABIOS MODEL

The ANDRA Aquabios model was applied to the irrigation scenario. Results from the application of the Aquabios model to the Se-79 irrigation scenario are presented in Figure 16.

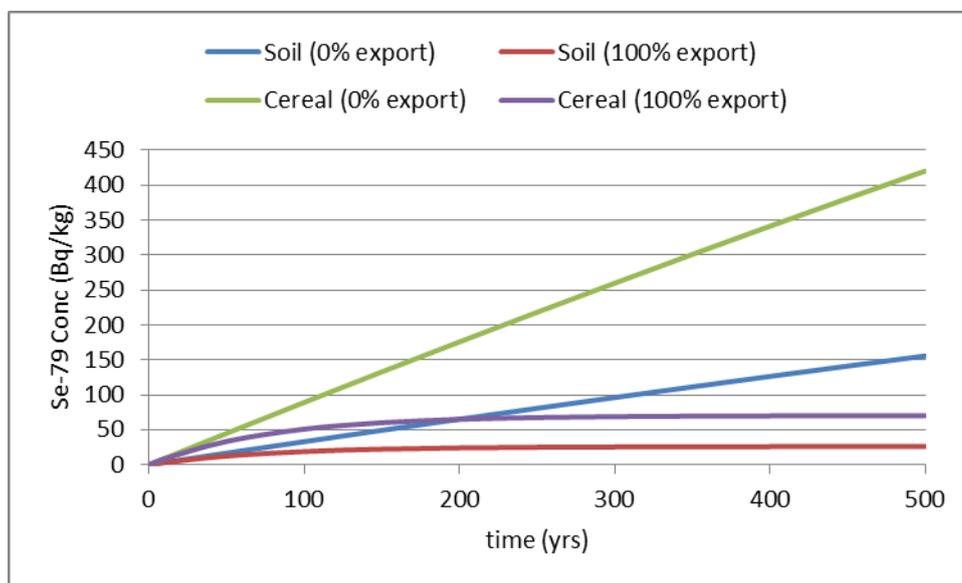


Figure 16 Se-79 concentrations in soils (Bq kg^{-1} dry weight) and cereals (Bq kg^{-1} fresh weight) as calculated by the Aquabios model.

The overall trend in results from Aquabios is similar to that achieved with the SAMM model in that, with the assumption of cropping losses, equilibrium conditions are achieved, but in the absence of cropping losses, soil and plant concentrations continue to rise throughout the simulation period. The time required for equilibrium to be achieved when cropping losses are assumed is greater for the Aquabios model (approximately 150 years) compared with SAMM (approximately 5 years). Activity concentrations in soils and plants calculated by the Aquabios model are a factor of around 100 greater than those calculated by SAMM when cropping losses are assumed; yet activity concentrations in soils and plants are consistent between the models in the absence of cropping losses.

5.2.5 E-K MODEL

The E-K model [Coppin et al, 2010] was applied to the irrigation scenario. The chemical form of selenium for the simulation was selenite, Se(IV).

The E-K model was applied in single soil compartment mode since the multiple soil layer version considers annual tillage resulting in the homogenisation of the upper 30 cm of soil, which was not considered necessary on the basis of the scenario description.

Specific model parameters for the E-K model are provided in Table 16.

Table 16 Specific parameters for the E-K model

Parameter	Units	Value	References / Notes
Se-79 concentration in percolate water	Bq L ⁻¹	5.54	Based on the 1 Bq/L concentration of irrigation water
Kd'	L kg ⁻¹	6	Coppin et al [2011]
k+	day ⁻¹	26.16	
k-	day ⁻¹	7.2E-3	
Ft (soil-plant transfer factor)	Bq.kg ⁻¹ fresh weight/ Bq kg ⁻¹	0.022	Value used in SYMBIOSE platform for Se-79
Dry soil in the considered soil layer	kg	429	
Volume of solution in the considered soil layer Topsoil layer : thickness 0.3 m and area 1 m ²	L	75	

Results

Results of the simulation are presented in Figure 17.

Results indicate that equilibrium conditions will be achieved within around 40,000 years, at which point the Se-79 activity concentrations are calculated to be:

- 3.5E+03 Bq kg⁻¹ in the upper soil layer; and
- 0.75 Bq kg⁻¹ fresh weight in cereal grain.

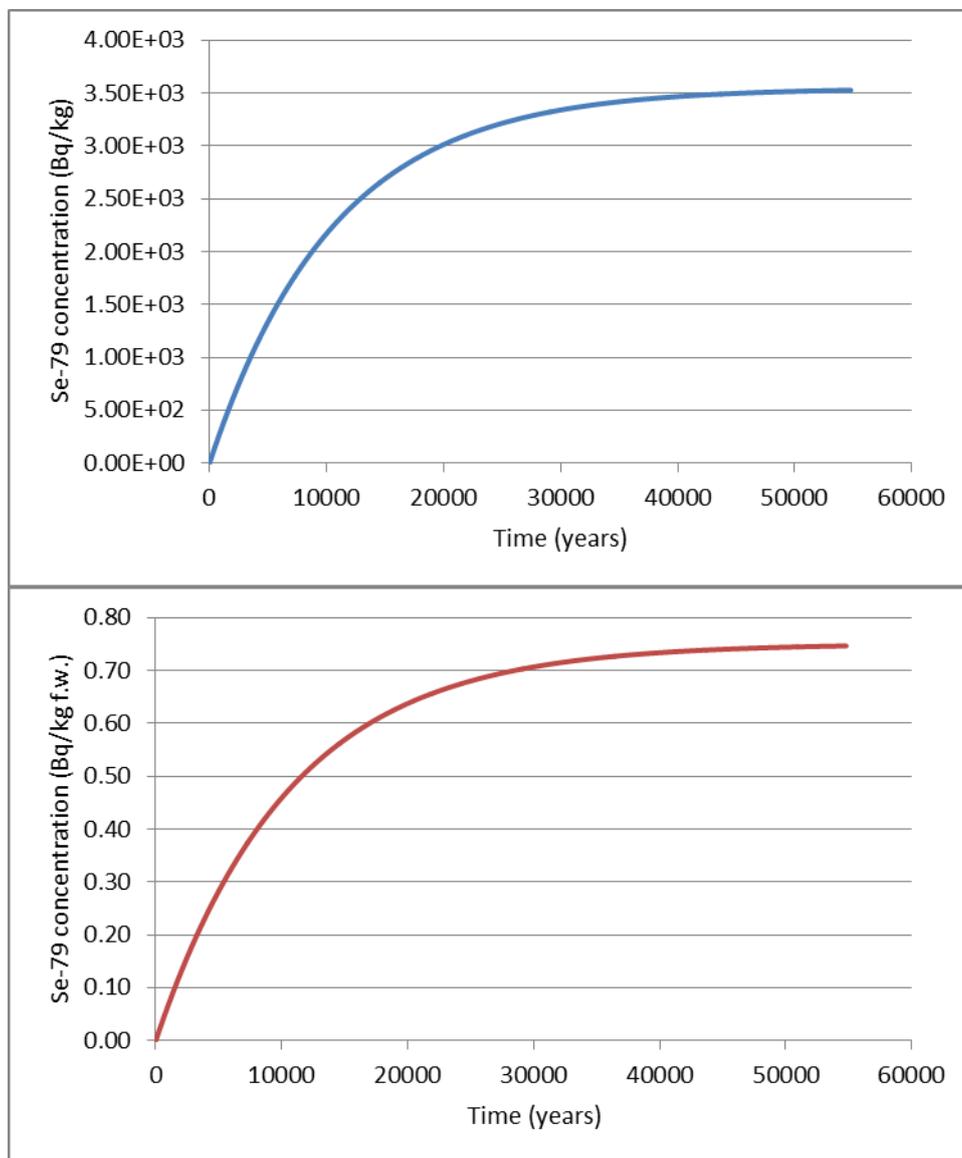


Figure 17 Se-79 concentrations in surface soil (upper) and cereal grain (lower) calculated by the E-K model

5.2.6 CONVENTIONAL Kd MODEL (IRSN)

Parameters applied to represent the irrigation scenario through a conventional Kd approach model are detailed in Table 17.

Table 17 Specific parameters for the Kd model

Parameter	Units	Value	References / Notes
Se-79 concentration in percolate water	Bq L ⁻¹	5.54	Based on the 1 Bq/L concentration of irrigation water
Kd	L kg ⁻¹	20	Coppin et al. 2011
Ft (soil-plant transfer factor)	Bq kg ⁻¹ fresh weight / Bq kg ⁻¹	0.022	Value used in Symbiose platform for Se-79
Dry soil in the considered soil layer	kg	429	
Volume of solution in the considered soil layer	L	75	
Topsoil layer : thickness 0.3 m and area 1 m ²			

Results

Results of the Kd model simulation are presented in Figure 18.

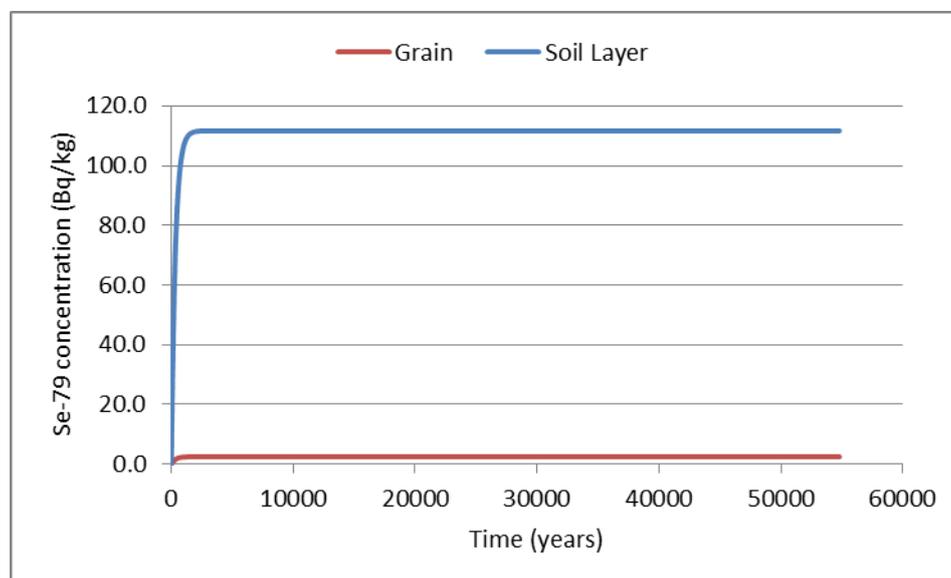


Figure 18 Se-79 activity concentrations in soil (Bq kg⁻¹) and cereal grain (Bq kg⁻¹ fresh weight) calculated using the conventional Kd model

Equilibrium conditions are achieved within around 1,500 years, at which point Se-79 concentrations are calculated as:

- 111 Bq kg⁻¹ in the surface soil layer; and,
- 2.4 Bq kg⁻¹_{fresh weight} in the grain of cereal.

A large difference in Se-79 concentrations in soil is evident between the E-K and Kd models (Figure 19). Soil Se-79 concentrations are more than an order of magnitude greater in the E-K model compared with the Kd model. The opposite trend is evident for cereal grain concentrations (Figure 20) where concentrations are calculated to be around 3 times greater with the Kd model as compared with the E-K model.

However, similar results between E-K and Kd models can be obtained for Se-79 concentration in soil by considering a Kd value of 642 L.kg⁻¹. This value can be calculated from the E-K parameters ($K_d = K_d' + k_+/k_- \cdot V/M$). Despite the same Se-79 concentration in soil being obtained for the two models, the concentration in grain is different: 0.75 Bq kg⁻¹_{fresh weight} with the E-K model and 77.5 Bq kg⁻¹_{fresh weight} for the Kd (642) model.

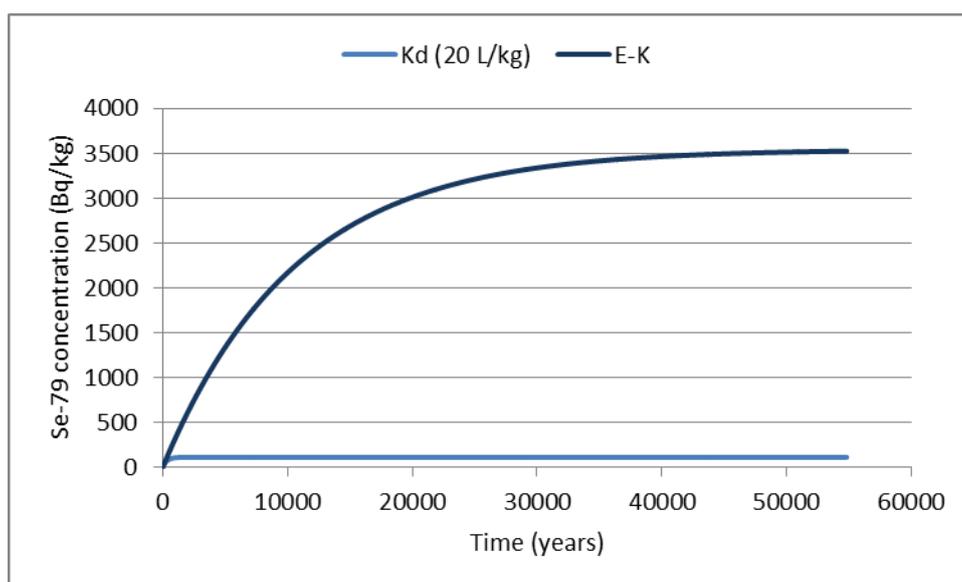


Figure 19 Comparison of Se-79 concentrations in soils as calculated by the E-K and Kd approaches used by IRSN

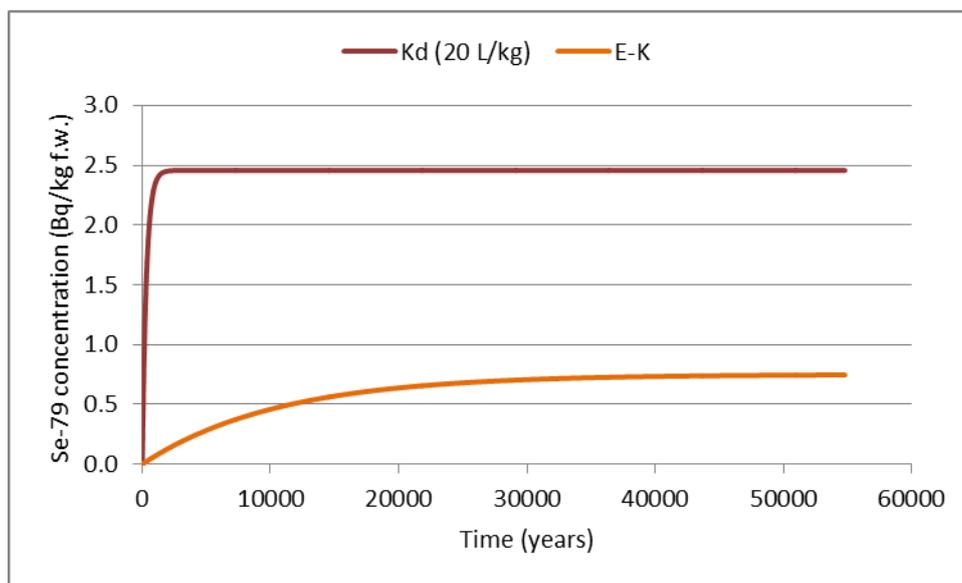


Figure 20 Comparison of Se-79 concentrations in cereal grain as calculated by the E-K and Kd approaches used by IRSN

The differences observed in the two model compartments arise from the inclusion of a kinetically-limited (recalcitrant) Se-79 compartment within the E-K model which effectively retains Se-79 within the soil yet renders this unavailable to plants. As such, for the same Se-79 concentration in soil, the bioavailable pool is reduced due to the slow release of bound Se-79 to the bioavailable pool.

5.3 SUMMARY OF MODEL RESULTS

Only one model (Andra SAMM) was applied to the upwelling groundwater scenario. As such, comparison of results has focussed on the irrigation scenario, results from which are summarised in Table 18.

Table 18 Summary of model output for the irrigation scenario

Model	time to equilibrium (years)	soil concentration (Bq kg ⁻¹)	plant concentration (Bq kg ⁻¹ f.w.)
<i>Conventional Approaches</i>			
NDA Spreadsheet ^a	5not applicable	0.299 (Kd 0.001)	0.598
		0.753 (Kd 0.003)	1.506
		2.34 (Kd 0.01)	4.68
		6.88 (Kd 0.03)	13.76
		22.7 (Kd 0.1)	45.4
		67.4 (Kd 0.3)	134.8
		109 (Kd 0.49)	218
Conventional Kd (20 L/kg)	1500	111	2.4
Aquabios (plant export)	200	70	26
Aquabios (no plant export)	500 ^b	420	155
<i>New Approaches</i>			
CIEMAT hydrological	<50	0.33-0.52	0.52 ^c
Andra SAMM (plant export)	5	0.3	0.84
Andra SAMM (no plant export)	500 ^b	151	409
E-K ([Coppin et al, 2010], IRSN)	40000	3500	0.75

^a plant concentrations calculated from soil surface activity concentrations using a concentration ratio to cereals of 2 (fresh weight basis) as detailed in Section 5.2.2.

^b equilibrium not achieved in simulation period

^c reported on dry weight basis

As indicated by Table 18, results vary considerably between the different models and a number of different factors contribute to the variation observed. Factors resulting in large variation in model output include Kd (as particularly evident from the results of the NDA Spreadsheet model) and whether or not cropping losses (plant export) is considered, as evidenced by the results from the Aquabios and ANDRA SAMM models.

Considerable variation was also evident in the time to reach equilibrium with the inclusion or exclusion of cropping loss being an important factor governing whether equilibrium was reached. When cropping was included as a loss process, the time taken for equilibrium conditions to be reached varied from 5 to 200 years. When excluded as a process, equilibrium conditions were not observed within a 500 year simulation period. However, volatilisation losses were excluded from these calculations. The Kd value, the percolation rate, the value of Ft and inclusion (or not) of loss by cropping explain the differences between the model results, rather than the mathematical construction of each model.

When considering specifically the new approaches to modelling (i.e. the CIEMAT hydrological model, the ANDRA SAMM model, and the E-K model (IRSN)), plant concentrations (taking into account plant export in the case of the ANDRA SAMM model) at equilibrium were similar (although the presentation of results from the CIEMAT model in terms of plant dry weight must be taken into consideration). This is particularly interesting considering the variation in soil concentrations between the CIEMAT and Andra SAMM models (with plant export) (range 0.3 to 0.52 Bq/kg) and that resulting from the E-K model [Coppin et al, 2010] (3500 Bq/kg at equilibrium). It is interesting to note that for the same considered timescale of observation similar Se-79 concentrations in soil can be obtained with E-K [Coppin et al, 2010] and Andra SAMM (without plant export) models (respectively 165 and 150 Bq/kg at 500 years). The fact that the E-K model [Coppin et al, 2010] does not consider export by plants explains the difference in the calculated long term equilibrium soil activity concentrations.

On the basis of the results presented, it is evident that the more complex models serve to limit the amount of Se-79 available to plants such that uptake is greatly reduced (although inclusion of plant export is also relevant for the ANDRA SAMM model). Further evaluation of these models may therefore be warranted; in particular it would be valuable to validate their predictive capabilities against field data were such a data set to be identified. Nonetheless, it is likely that use of more complex modelling approaches, which take account of the redox behaviour of selenium in soils and resultant bioavailability will reduce the radiological significance of Se-79 in repository safety assessments. It may also be interesting to compare the models while assuming more precise constraints on the different parameters and other assumptions, notably, cropping or not, biomass, different percolation fluxes, different timescale of observation, K_d and F_t values. Only IRSN stated the form in which selenium was added to the system (selenite) and the influence of chemical form on model output cannot therefore be considered on the basis of these results.

6. SUMMARY AND CONCLUSIONS

This report has outlined a number of factors considered to be of importance in modelling Se-79 entry into the food chain (i.e. behaviour in soils and uptake into plants) and a range of modelling approaches, both conventional and bespoke, has been presented. In order to evaluate the different approaches to modelling, each model has been applied to a hypothetical release scenario and results compared.

It is considered that the key to accurately representing selenium behaviour in soils is to understand and account for soil redox conditions and their influence on the partitioning of selenium between solid and liquid phases. Selenium binding to soil (including organic matter and minerals) effectively renders selenium unavailable to plants.

Conventional models largely consider a single soil compartment such that differences in selenium behaviour throughout the soil column, as driven by changing hydrological (and hence redox) conditions, are unaccounted for. As such, more selenium tends to be considered as available to plants resulting in high Se-79 activity concentrations in plants that may then enter the food chain. The behaviour of selenium in soils is largely represented in such models through the application of a single K_d with plant uptake then being represented by a soil to plant CR.

The alternative, bespoke, model approaches presented take different approaches to addressing selenium behaviour in soils. Two models (ANDRA SAMM and the CIEMAT hydrological model) consider multiple soil layers which enable different selenium retention properties to be assigned that are consistent with the soil layer hydrological conditions. Whilst a single soil to plant concentration ratio is applied, the application takes account of the bioavailable fraction of selenium in each soil layer and the density of roots associated with that soil compartment. The alternative approach presented (the E-K model [Coppin et al, 2010]) splits the soil into two components with a kinetic parameter applied to link between labile and recalcitrant selenium. Plant uptake is then restricted to the labile fraction.

Whilst results indicate wide variation in soil Se-79 activity concentrations between these models, plant activity concentrations are reassuringly similar (within a factor of 2 when plant cropping is taken into account) suggesting that the use of these more process-orientated models, whether or not they include volatilisation, may serve to (justifiably) reduce the importance of Se-79 in repository safety assessments. Nonetheless, there may be merit in applying the different approaches to a real scenario where both soil and plant activity concentrations are known to evaluate further the predictive capabilities of the different approaches. Such data may arise from sites to which agronomic biofortification practices are applied, such as in Finland, for which soil monitoring may be routine. However, the requirement to continually add selenium to soils suggests continued loss from the system. This may arise from methylation and subsequent volatilisation of the added selenium. Alternatively, labile selenium may be lost via surface water run-off (as indicated by Winkel et al [2012]). Evaluation of the potential consequences of loss to surface waters and subsequent accumulation in aquatic plants and animals may be warranted, although this process may be associated with further dilution.

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