



Strålsäkerhets
myndigheten

Swedish Radiation Safety Authority

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Research

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C-14 Long-Term Dose Assessment:
Data Review, Scenario Development,
and Model Comparison

SSM perspective

Background

BIOPROTA (www.bioprota.org) is an international collaboration forum which seeks 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. Particular emphasis is placed on 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.

This study was supported financially by the following sponsoring organisations: Agence Nationale pour la Gestion des Déchets Radioactifs (Andra, France), Electricité de France (EDF, France), LLW Repository Ltd (LLWR, UK), the National Cooperative for the Disposal of Radioactive Waste (Nagra, Switzerland), the Nuclear Decommissioning Authority Radioactive Waste Management Directorate (NDA RWMD, UK), Nuclear Waste Management Organisation of Japan (NUMO, Japan), Svensk Kärnbränslehantering AB (SKB, Sweden) and Strålsäkerhetsmyndigheten (SSM, Sweden).

This report provides a summary of a programme of work, commissioned within the BIOPROTA collaborative forum, to compare and contrast different assessment models for the behaviour of C-14 in soil and plants.

Objectives

C-14 is present in solid radioactive wastes arising from the nuclear power industry, in reactor operating wastes and in graphite and activated metals that will arise from reactor decommissioning. Its half-life of 5730 years is one of the factors that may enable releases of C-14 to the biosphere from deep and near-surface radioactive waste repositories. These releases may occur as C-14 bearing gases, especially methane, or as aqueous species, and enter the biosphere from below via natural processes or via groundwater pumped from wells. Assessment of radiation doses to humans due to such releases must take account of the major role of carbon in biological processes, requiring specific C-14 assessment models to be developed. The overall objective of this study is to perform an inter-comparison between five C-14 assessment models to understand the processes involved and identify areas where further research is required to address some of the uncertainties.

Results

The inter-comparison identified significantly different results for the activity concentrations in the soil, atmosphere and plant compartments, based upon the different modelling approaches. The major source of uncertainty was related to the identification of conditions under which mixing occurs and isotopic equilibrium are established. Furthermore, whilst the assumed release area plays a role in determining the calculated atmospheric C-14 concentrations, the openness of the plant canopy and the wind profile in and above that canopy are the key drivers.

Need for further research

Future considerations may include forms of carbon other than methane (i.e. dissolved organic substances) entering soil, and uptake via aquatic pathways.

Project information

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Strålsäkerhetsmyndigheten

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This report concerns a study which has been conducted for the Swedish Radiation Safety Authority, SSM. The conclusions and viewpoints presented in the report are those of the author/authors and do not necessarily coincide with those of the SSM.

BIOPROTA

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

C-14 Long-Term Dose Assessment in a Terrestrial Agricultural Ecosystem: FEP Analysis, Scenario Development, and Model Comparison

FINAL REPORT

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G M Smith, M C Thorne and S Xu**

**Version 3.0, Final
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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 work undertaken between January 2010 and March 2011 to compare and contrast different assessment models for the behaviour of C-14 in soil and plants. Some initial discussions of the topic were reported in the 2008 Annual BIOPROTA workshop report, leading to some preliminary work being carried out between October 2008 and October 2009. This preliminary work was documented and reported at a workshop, hosted by Electricité de France (EDF) in Paris, in February 2010.

The subsequent study reported here was carried out by staff from the Agence Nationale pour la Gestion des Déchets Radioactifs (Andra, France), EDF, the Swedish Radiation Safety Authority (SSM) and the technical support team, made up from GMS Abingdon, Limer Scientific Consulting, Quintessa, Eden Nuclear and Environment and Mike Thorne and Associates.

The study was supported financially by the following sponsoring organisations: Andra, EDF, LLW Repository Ltd (UK), the National Cooperative for the Disposal of Radioactive Waste (Nagra, Switzerland), the Nuclear Decommissioning Authority (NDA) Radioactive Waste Management Directorate (RWMD), Nuclear Waste Management Organisation of Japan (NUMO), Svensk Kärnbränslehantering AB (SKB, Sweden) and SSM.

Additional technical input and comments are gratefully acknowledged from staff at the Institut de Radioprotection et de Sûreté Nucléaire (IRSN, France), the University of Nottingham, UK, the University of East Finland, the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT, Spain), EcoMatters (Canada), the National Institute of Radiological Sciences (NIRS, Japan), Facilia AB (Sweden) and the Helmholtz-Zentrum, München.

The report is presented as working material for information. The content may not be taken to represent the official position of the organisations involved, and the models cited in relation to any particular organisation are not necessarily that organisation's current position. All material is made available entirely at the users' risk.

Version History

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Version 3.0: Final report, incorporating review comments from project sponsors, prepared by Limer Scientific Consulting Ltd with review by Mike Thorne & Associates, GMS Abingdon and Eden Nuclear and Environment and distributed 14 November 2011.

EXECUTIVE SUMMARY

This report describes a study undertaken within the BIOPROTA international collaboration forum, to compare and contrast models for C-14 dynamics in the soil-plant system and consider the implications for dose assessment for long-term C-14 release to the biosphere. Some initial discussions of the topic were reported in the 2008 Annual BIOPROTA workshop report leading to some preliminary work being carried out between October 2008 and October 2009. This work was documented and reported at a workshop hosted by EDF in Paris in February 2010. The current project was developed at that workshop and is described below.

The overall objective of the project is to improve confidence in dose assessments for long-term releases into the biosphere of C-14 disposed in radioactive waste repositories. This project has compared quantitative estimates of the C-14 concentrations in specific components of the dose assessment models (soil, plant-canopy atmosphere, and plants), for various release scenarios linked to abstraction of contaminated water used for irrigation of agricultural crops and gaseous release from the geosphere.

The models included in the comparison exercise were those developed by or on behalf of Andra, EDF, LLW Repository Ltd (LLWR), NDA RWMD and the model of Avila and Pröhl, which was developed with support from SKB and Posiva. Note that these models are not necessarily those used by any of these organisations.

The FEP analysis, discussion of the models and examination of results highlights important differences in the conceptual models employed, which feed through to large differences in estimates of C-14 concentrations in different parts of the system. The differences and their significance are considered in relation to the major model subcomponents addressing C-14 behaviour in: the soil, the plant canopy atmosphere and the plant itself.

Within the soil subsystem, it is possible to store a fraction of C-14 in recalcitrant organic pools that are not readily bioavailable. Such an approach is supported by a substantial body of empirical evidence which demonstrates the existence of a wide range of carbon compounds in soil, some as readily degradable materials, such as cellulose, with others in less biologically available forms (e.g. humic and fulvic substances). It is also possible to include more elaborate soil irrigation sub-models, but a comparison with a simpler approach, in which irrigation depends only on yearly averaged precipitation and evaporation with no distinction between plants, shows the small impact of this additional detail.

The conceptualisation of the canopy atmosphere varies between the models used in this study, and this is the cause of the majority of the differences in calculated plant C-14 concentrations. When the atmospheric C-14 concentration is fixed the difference in calculated plant C-14 concentration for a given field size dropped from three or more orders of magnitude to less than a factor of five.

The final link in the sequence of uptake of carbon by plants in a soil-plant-atmosphere system involves uptake into the plants and uncertainty in the canopy atmosphere results is carried through into the plant concentration results. All models use the same isotope ratio approach with comparable stable carbon concentrations in both air and plant. Possible additional uncertainty linked to C-14 root uptake or translocation of leaf-deposited bicarbonates does not show, because these processes do not contribute more than 2% of plant carbon in any of the models.

There is not an agreed "right way forward" on C-14 biosphere considerations, which is a reasonable position given the research-level status of current knowledge. Thus, this study is not a traditional benchmarking exercise, but an intercomparison of research models that take different approaches as their bases.

Overall, the results presented in this study show clearly how important the conceptualisation of the dynamics of C-14 (and stable C) within the plant canopy atmosphere is upon the calculated plant

C-14 concentrations. The approach of some models, in which the air the plant uses for photosynthesis is assumed to be subject to a relatively small degree of mixing, naturally leads to higher calculated plant C-14 concentrations than in the approach adopted in other models, in which the air the plant uses is subject to a greater degree of mixing with uncontaminated air. Whilst the assumed contaminated field size can, and does, play a role in determining the calculated atmospheric C-14 concentrations, it is the assumed degree of openness of the canopy and the wind profile both in and above the plant canopy that are more likely to be the key drivers in determining the concentration of C-14 in the atmosphere used by the plant for photosynthesis. Furthermore, there is an interaction between these factors, with field size being of greater importance for a well-ventilated, open plant canopy.

This study, whilst providing information with respect to the dynamics of the models currently used by various waste management organisations, is not able to address all the uncertainties in the dose assessment. These may be addressed taking into account site-specific information but may also be addressed by consideration of the outcomes of additional on-going studies, examples of which are discussed at the end of this report.

This study does not imply that the approach taken by any contributory organisation is “right” or “wrong”. Rather, the study output should be used, going forward, to develop a consensus on processes that should/should not be considered in research models for C-14 biosphere studies, and the circumstances when their inclusion is/is not justified on a site basis or on the basis of the current status of a national programme.

CONTENTS

PREFACE II

EXECUTIVE SUMMARY	IV
1. INTRODUCTION	1
1.1 Background	1
1.2 Report structure	1
2. FEP ANALYSIS	3
2.1 Methodology	3
2.2 Results of the FEP analysis	4
2.3 COMPARISON with pre-existing interaction matrices and FEP analysis for the same system (terrestrial)	16
2.4 Comparison of interaction matrices	20
3. MODEL DESCRIPTIONS	22
3.1 AquaC_14	22
3.2 SA_Carbon14	29
3.3 Simplified enhanced RIMERS	33
3.4 Avila and Pröhl model	39
3.5 Thorne-Limer model	41
3.6 Comparison of mathematical representation of C-14 dynamics in the canopy atmosphere	46
3.7 Implementation of the models for this study	48
4. FEP AUDIT OF MODELS USED IN THIS PROJECT	49
4.1 Features	49
4.2 Events and processes	49
5. SCENARIO DESCRIPTION	51
5.1 Source Term	51
5.2 Media in which C-14 concentrations were calculated	52
5.3 Additional scenario information	52
6. DATA AND PARAMETER VALUES	54
6.1 Climate, soil and irrigation data	54
6.2 AquaC_14 (Andra model) parameters	54
6.3 SA_Carbon14 (EDF) model	56
6.4 Simplified enhanced RIMERS	58
6.5 Avila and Pröhl model	58

6.6	Thorne-Limer model	59
7.	RESULTS	61
7.1	AquaC_14	61
7.2	SA_Carbon14	64
7.3	Simplified enhanced RIMERS	67
7.4	Avila and Pröhl Model	69
7.5	Thorne-Limer model	72
7.6	Comparison of results	74
8.	DISCUSSION	80
8.1	Overview	80
8.2	Specific considerations of the environmental media	80
8.3	Outlook and ongoing work	81
9.	REFERENCES	82

1. INTRODUCTION

1.1 BACKGROUND

The global carbon cycle and the long-term implications of continued C-14 discharges from the nuclear fuel cycle have been studied for several decades [Ekdahl et al, 1972; Killough, 1980] and the need to address radiological impacts from disposal of radioactive waste containing C-14 has also been recognised for some time [Bush et al, 1983]. Particular interest remains in improving the assessment of possible annual individual doses to members of potential exposure groups arising from releases to the biosphere of C-14 from deep and shallow radioactive waste disposal facilities, e.g. the Swedish SFR facility [Thomson et al, 2008] and for a variety of waste types, especially reactor operating wastes [Magnusson et al, 2008] and graphite [Limer et al, 2010].

Models for C-14 behaviour in the biosphere were reviewed under the BIOPROTA programme in BIOPROTA [2005]. That review considered the modelling of releases of C-14 both to aquatic (freshwater and marine) and to terrestrial ecosystems. It is acknowledged that, even with very pessimistic assumptions in a terrestrial model, the highest assessed possible doses associated with a C-14 release come from modelling releases to aquatic ecosystems [e.g. Bergström et al., 2008]. As noted by Limer and Thorne [2011], the high doses associated with aquatic releases can, to some extent, be attributed to the assumptions made with respect to the uptake of C-14 by fish. Although there was some interest expressed in further investigating the modelling of C-14 releases to aquatic ecosystems, the overall consensus of the organisations within BIOPROTA was that the greater interest lay in the modelling of releases to terrestrial ecosystems. Thus, a more detailed, quantitative C-14 model comparison exercise, focussing on terrestrial agricultural ecosystems, was completed under BIOPROTA in 2009, and documented in an internal project progress report [Limer et al, 2009a]. This identified significantly different results in terms of calculated C-14 concentrations in plants arising from different modelling approaches in long-term dose assessment models for application to release of C-14 to the terrestrial environment from solid radioactive waste disposal. A follow-up study within BIOPROTA has therefore been undertaken, commencing with a workshop to discuss those attributes of C-14 of particular relevance to radiological assessments for geological waste repositories. The workshop was held on 16-17 February 2010 and was hosted by EDF, Paris, France. The workshop involved presentation and discussion of both data and model developments needed to improve the representation of C-14 behaviour in the soil-plant-atmosphere system, with additional discussions relating to freshwater ecosystems. Presentations and discussions are reported in Limer and Thorne [2010].

This report describes activities subsequent to the February 2010 report, including:

- a FEP (Features, Events and Processes) analysis and development of an interaction matrix that details linkages between FEPs;
- An audit of available models against the FEP list;
- Development of a model scenario;
- Application of models, by different users, to the model scenario; and
- Analyses of model calculation results to evaluate similarities and differences in output resulting from the different model approaches employed.

1.2 REPORT STRUCTURE

Section 2 presents an audit of a recognised list of FEPs relating to the assessment of radionuclides in the biosphere, the FEP list developed in the BIOMASS project [IAEA, 2003], with respect to the behaviour of C-14 in the biosphere. For completeness this audit was compared with other similar

FEP analyses carried out by members of the IUR Waste Working Group [IUR, 2006] and CIEMAT [Aguero et al, 2006]. From this, a suggested list of FEPs for C-14 was developed, such that organisations with C-14 models might be better able to demonstrate and justify which FEPs are considered in their model, and why, if applicable, certain FEPs have been disregarded. The FEP analysis is used to underpin the development of an interaction matrix (IM), which forms the overall conceptual model for the environmental transfer of C-14 in the soil-plant system.

Section 3 presents descriptions of the models that have been applied in this project and Section 4 provides the results of an audit of each model against those FEPs outlined in Section 2.

In this project, consideration has been given to C-14 entering the biosphere either in contaminated irrigation water, or as a result of the upwelling of gas from below the soil zone. The descriptions relating to both of these scenarios are given in Section 5. The data used in each model when applied to the scenarios are presented in Section 6. Results and conclusions of the comparison of model outputs are presented in Sections 7 and 8, respectively.

2. FEP ANALYSIS

2.1 METHODOLOGY

Before commencing a FEP analysis, it is important to clearly identify the question that the model being developed is supposed to address. For a radioactive waste disposal facility, a typical question might be of the form:

What are the annual individual doses to members of potentially exposed groups (PEGs) arising from the release of C-14 advected with, or diffusing in, gas or groundwater into (rooting zone) agricultural soil from below or via irrigation?

The preceding BIOPROTA C-14 project [Limer et al, 2009a] focussed upon the soil-plant-atmosphere system, and found that differences in conceptual models lead to striking differences in the estimated C-14 concentrations in both the plant canopy atmosphere and the plants. It therefore seemed reasonable to maintain the focus of the modelling activities in this project on the same system, meaning that our assessment question would be of the form:

What are the concentrations of C-14 in foodstuffs consumed by PEGs from release of C-14 advected with, or diffusing in, gas or groundwater into (rooting zone) agricultural soil from below or via irrigation?

It is recognised that the question as posed disregards releases to aquatic systems. Aquatic systems were outside the scope of the current project. Nonetheless, the behaviour of C-14 in freshwater systems may warrant further consideration.

Once the assessment question has been posed, the following steps are used to carry out the FEP analysis.

1. Take a generic FEP list for the biosphere and screen that for relevance to the question, so as to refine the FEP list.
2. Choose a set of key conceptual model objects (CMO's), which make up the leading diagonal elements (LDE's) of the IM.
3. Go through all the off-diagonal elements (ODE's) to identify processes that affect transfer of C-14 among those CMOs. This is done in two steps:
 - a. Consider each LDE in turn and how C-14 might be transferred to other leading diagonal elements.
 - b. Check that all the FEPs in the refined list are somewhere in the IM, or document why the FEP is not included.
4. This process may identify redundant LDE's or the need to create new leading diagonal elements, such that step 3 may be repeated.

The objective of this process is that, after some iteration, the generic FEP list should be annotated with all the FEPs either in the IM (with a note of where) or excluded with a documented reason why. In doing so, the conceptual model is also defined; a non-quantitative description of all compartments (or mesh points) in the environment and the processes of radionuclide transfer, or affecting radionuclide transfer, between them.

The mathematical model development and the search for data to support parameter value choice then follows on from this FEP analysis and conceptual model development. Where data gaps are highlighted, this may signal the need to go back and simplify the processes being modelled, or the need to instigate a research programme.

The generic FEP list chosen for this analysis was the one developed during the IAEA BIOMASS programme [IAEA, 2003]. The BIOMASS FEP list contains some 135 FEPs. These FEPs are divided into four broad categories:

- Assessment context;
- Biosphere system features;
- Biosphere system events and processes; and
- Human exposure features, events and processes.

This list was initially screened to determine which of those FEPs were regarded as being of relevance to our assessment question; this screening was subsequently updated as a result of iterations of stages 3 and 4 of the process given above. A summary of this screening, together with the reasoning behind the inclusion or exclusion of a given FEP, is given in Section 2.2.1. The conceptual model objects were defined (Section 2.2.2) and IM's developed (Section 2.2.3) independent of the FEP audit. The iterative process of comparing the FEP list with the IM's was then carried out until a satisfactory conclusion as to the final FEP list and IM's for C-14 was reached. Descriptions of those FEPs included in the final IM's are given in Section 2.2.4.

2.2 RESULTS OF THE FEP ANALYSIS

2.2.1 Screening of BIOMASS FEP list

The tables given below present the summary of the FEP audit for each of the three broad categories of FEPs given in the BIOMASS FEP list relevant to the assessment question^a:

- Assessment context (Table 1);
- Biosphere system features (Table 2); and
- Biosphere system events and processes (Table 3).

These tables contain both the initial screening (the Y/N column for their inclusion in the model) and a summary of either where the FEP is represented in the model, be it in the assessment context description or a location in an IM, or else a reason for why that FEP has been disregarded.

^a A fourth category of FEPs relate to human exposure, but these were not considered in this BIOPROTA study as they are outside the scope of the assessment question.

Table 1 Assessment Context

FEP number	FEP name	Y/N?	Where/why?
1.1	Assessment Purpose	Y	Not in IM as that is the question defined in Section 2.1
1.2	Assessment Endpoints	Y	Not in IM as that is the question defined in Section 2.1
1.2.1	Annual Individual Dose	N	Out of context
1.2.2	Lifetime Individual Dose	N	Out of context
1.2.3	Annual Individual Risk	N	Out of context
1.2.4	Lifetime Individual Risk	N	Out of context
1.2.5	Collective Dose/Risk	N	Out of context
1.2.6	Dose to Non-human Biota	N	Out of context
1.2.7	Modification of the Radiation Environment	Y	Encompassed in whole IM
1.2.8	Fluxes	N	Out of context
1.2.9	Non-radiological Endpoints	N	Out of context
1.2.10	Uncertainties and/or Confidence	Y	Part of the model assessment rather than the IM ^a
1.3	Assessment Philosophy	Y	Underpins the selection of the question being addressed.
1.4	Repository System	N	Needs to be accounted for in site-specific assessment as may affect dominant form of release, but out of scope of current context.
1.5	Site Context	Y	Assessment context. The IMs in Section 2.2.3 are for a temperate agricultural ecosystem. Differences for other ecosystems are noted.
1.6	Source Term	Y	A1 ^b
1.6.1	Geosphere/Biosphere Interface	Y	Ground water - B1 (upwelling & irrigation), K1 (interception of irrigation water), A2 (percolation). Gas - B1 (dissolution), E1 (advection/diffusion)
1.6.2	Release Mechanism	Y	Example release mechanisms include: groundwater release to land and surface water bodies via natural aquifer discharge; groundwater release via extraction of well water; and gaseous release. ^c
1.6.3	Source Term Characteristics	Y	Exclusively C-14 for this context, which may occur in the form dissolved CO ₂ , bicarbonate, DOC, dissolved carbonate, CH ₄ gas, CO ₂ gas, and/or CO.
1.7	Time Frames	Y	Current context assumes continuous release until equilibrium is reached
1.8	Societal Assumptions	Y	Modern cultivation practice (small farm rather than subsistence smallholder)

^a Participants generally have an interest in addressing doses to average members of critical groups or reference persons, and in a best estimate of doses, but need to consider the uncertainties around that, or the confidence in the best estimate.

^b It is recognised that the source could be in groundwater or in gaseous form, and the release could be diffusive or advective.

^c Release of solid materials as a result of human intrusion or natural erosion are also potential mechanisms, but are outside the current scope.

Table 2 Biosphere System Features

FEP number	FEP name	Y/N?	Where/why?
2.1	Climate	Y	Assessment context (focus on temperate but highlight differences for other climatic zones.)
2.1.1	Description of Climate Change	N	Out of context. May need to be considered for site specific assessments.
2.1.2	Identification and Characterisation of Climate Categories	Y	Assessment context
2.2	Human Society	Y	Assume similar societal behaviour as for today (see FEP 1.8).
2.3	Systems of Exchange	N	Out of context
2.3.1	Environment Types Natural and Semi-natural, Agricultural, Urban and Industrial	Y	Assessment context (agricultural)
2.3.2	Ecosystems	Y	Assessment context
	Living Components of Ecosystems	Y	F6, G7, J10, K11
	Non-living Components of Ecosystems	Y	B2, C3, D4, E5, H8, I9

Table 3 Biosphere Events and Processes

FEP number	FEP name	Y/N?	Where/why?
3.1	Natural Events and Processes	Y	See ODEs
3.1.1	Environmental Change Physical, Chemical and Ecological Changes	N	Equilibrium conditions considered for current context
3.1.2	Environmental Dynamics	Y	e.g. climate-dependent farming practices.
	Diurnal Variability	Y	Important for C-14 uptake dynamics
	Seasonal Variability	Y	Important for C-14 uptake dynamics
	Inter-annual and Longer Timescale Variability	N	Timescale is beyond scope of assessment
3.1.3	Cycling and Distribution of Materials in Living Components	Y	See sub-FEPs
	Transport Mediated by Flora and Fauna	Y	See sub-FEPs
	Root Uptake	Y	J2, J5
	Respiration	Y	I11, H11, E10, E7, E6
	Transpiration	Y	H11, I11
	Intake by Fauna	Y	F2, F3, F4
	Interception	Y	K1
	Weathering	Y	L3, L4, L11
	Bioturbation	Y	E8 general. Soil layers: H3, I4, K6, C8, D9, F11
	Metabolism by Flora and Fauna	Y	See sub-FEPs
	Translocation	Y	K10, J11
Animal Metabolism	Y	F5	
3.1.4	Cycling and Distribution of Materials in Non-living Components	Y	See sub-FEPs
	Atmospheric Transport	Y	I8, H9, L9
	Evaporation	Y	H2
	Gas Transport	Y	General: H5 (degassing) Soil layers: J5, E10
	Aerosol Formation and Transport	N	Trivial for carbon
	Precipitation	N	May be important as dilution mechanism but is not in the IM
	Washout and Wet Deposition	N	Trivial for carbon
	Dry Deposition	N	Trivial for carbon
	Water-borne Transport	Y	See below
	Infiltration	N	Not important for carbon
	Percolation	Y	General: A2, L2. Soil layers: A2, B7.

Table 3 Biosphere Events and Processes

FEP number	FEP name	Y/N?	Where/why?
	Capillary Rise	Y	General: B1 (also mean upwelling). Soil layers: G2
	Groundwater Transport	Y	L2
	Multiphase Flow	N	May be important for C (not considered in simplified models), CO ₂ , CH ₄ and dissolved.
	Surface Run-off	N	May be important for C (not considered in simplified models).
	Discharge	Y	L2
	Recharge	Y	A2 (groundwater sources only)
	Transport in Surface Water Bodies	N	Out of context
	Erosion	N	Irrelevant for carbon in current models (loss of organic matter)
	Solid-phase Transport	N	Irrelevant for carbon in current models (loss of organic matter)
	Landslides and Rock Falls	N	Out of context
	Sedimentation	N	Out of context
	Sediment Suspension	N	Out of context
	Rain Splash	N	Irrelevant for carbon
	Physicochemical Changes	Y	See below
	Dissolution/Precipitation	Y	B5
	Adsorption/Desorption	Y	C2, D2, B3, B4
	Colloid Formation	N	May be important for C (not considered in simplified models).
3.2	Events and Processes Related to Human Activity	Y	See sub-FEPs
3.2.1	Chemical Changes	Y	See sub-FEPs
	Artificial Soil Fertilisation	Y	May be important but does not fit in IM. Any that occurs will be implicitly taken into account through the definition of soil properties.
	Chemical Pollution	N	Out of context
	Acid Rain	N	Out of context
3.2.2	Physical Changes Construction, Water Extraction by Pumping, Water Recharge by Pumping, Dam Building, Land Reclamation	N	Out of context
3.2.3	Recycling and Mixing of Bulk Materials	Y	See sub-FEPs
	Ploughing	Y	General: C10, D10 Soil layers: H3, I4, C8, D9, F11, K6
	Well Supply	Y	Assessment context (water scenario)

Table 3 Biosphere Events and Processes

FEP number	FEP name	Y/N?	Where/why?
	Other Water Supply	Y	B1 (upwelling) (water scenario)
	Irrigation	Y	B1, K1 (water scenario)
	Recycling of Bulk Solid Materials	Y	C10, C11, D10, D11, C7, D7, D3, C4
	Artificial Mixing of Water Bodies	N	Out of context
	Dredging	N	Out of context
	Controlled Ventilation	N	Out of context. This FEP could be important if the plants were grown in a greenhouse rather than in open air; the best way to get a minimally mixed atmosphere is in a greenhouse.
3.2.4	Redistribution of Trace Materials Water Treatment, Air Filtration, Food Processing	N	Carbon is bulk not trace. Other aspects are outside context.

2.2.2 Conceptual model objects (CMO's)

Conceptual model objects are a means of compartmentalising a system into features; when using an IM to describe a model such features are often referred to as Leading Diagonal Elements (LDE's), as that is where they are located in the IM. Twelve conceptual model objects were identified; these are described in Table 4. The atmosphere has been split into two compartments; the height that separates the two compartments is the height at which the mixing of air becomes significantly affected by the wind. This height is variously referred to as the roughness height, or zero displacement height. In this BIOPROTA study it is represented with the symbol z_d .

It was considered that the soil could be further broken down into two layers: an upper layer (UL) which is subject to ploughing, and a lower layer (LL) which is not disturbed by human activity. Soil CMO's are similar for both layers.

Soil macrobiota were considered for inclusion as a CMO, since they are responsible for the process of bioturbation in the soil. As the soil macrobiota would otherwise behave identically to soil microbiota in terms of the carbon cycling in the soil, rather than including macrobiota as an explicit CMO their presence is implicit in the inclusion of the FEP "bioturbation" in the soil layer IM presented in the following section (Figure 3).

Table 4 Conceptual Model Objects

Conceptual model object	Leading diagonal element position in Gas and Water IM's	Leading diagonal element position in the Soil layer IM	Description
Source	A1	A1	Water: Groundwater contaminated with 1 Bq/l, used for irrigation and upwelling into soil of interest. Gas: $^{14}\text{CH}_4$ or $^{14}\text{CO}_2$ (1 Bq m^{-3}). Scenario specific flux rates are defined.
Soil water	B2	B2, G7	Liquid water in the soil pores. Agricultural soil (depth, texture, pH, Eh)
Soil solids – recalcitrant	C3	C3, H8	Slow turnover of non-living carbon (residence time of greater than 10 years)
Soil solids – labile	D4	D4, I9	Fast turnover of non-living carbon (residence time of less than 10 years)
Soil gas	E5	E5, J10	CO_2 and CH_4 in the soil pores, as gas or dissolved
Soil microbes	F6	F6, K11	Microbes
Mycorrhizae	G7	L12	Mycorrhizae
Plant canopy atmosphere below z_d	H8	-	Within the canopy (without lateral air flow)
Plant canopy atmosphere above z_d	I9	-	Within the canopy (with lateral air flow)
Below-ground plant material	J10	-	Roots
Above-ground plant material	K11	-	Stems and leaves and fruits and grains
Sink	L12	-	Anything outside volume of interest

2.2.3 C-14 interaction matrices

The water and gas source IM's that have been developed are given in Figure 1 and Figure 2, respectively. The two soil layers are considered further, in particular how they interact with each other, in Figure 3; the yellow boxes indicate the lower soil layer (LL) and the grey boxes indicate the upper soil layer (UL).

	A	B	C	D	E	F	G	H	I	J	K	L
1	SOURCE (Water)	irrigation upwelling capillary rise	X	X	X	X	X	X	X	X	interception of irrigation water	X
2	percolation	SOIL WATER	adsorption	adsorption	exsolution?	ingestion	uptake	evaporation	X	root uptake	X	percolation to groundwater (& groundwater flow from area)
3	X	desorption	SOIL SOLIDS - Recalcitrant	decomposition?	decomposition	ingestion	uptake	X	X	X	X	weathering
4	X	desorption	decomposition	SOIL SOLIDS - Labile	decomposition	ingestion	uptake	X	X	X	X	weathering
5	X	gas sorption	X	X	SOIL GAS	inhalation / assimilation / metabolism	X	degassing	X	root uptake	aerenchyma photosynthesis (assuming CO2)	X
6	X	excretion	X	death excretion?	respiration decomposition methane oxidation	SOIL MICROBES	X	X	X	X	X	X
7	X	excretion?	death & decomposition	death & decomposition	respiration decomposition	X	MYCORRHIZAE	X	X	X	X	cropping (fruiting body) / dispersal
8	X	X	X	X	diffusion and barometric pumping bioturbation	X	X	CANOPY ATMOSPHERE - slow air flow (below zd)	diffusion / advective transport	X	photosynthesis (CO2)	X
9	X	X	X	X	X	X	X	diffusion / advective transport	CANOPY ATMOSPHERE faster air flow (above zd)	X	photosynthesis (CO2)	free air
10	X	root exudation	death & decomposition (UL & LL) ploughing	death & decomposition (UL & LL) ploughing	root respiration	X	root exudation and uptake	X	X	BELOWGROUND PLANT MATERIAL	translocation (assuming root uptake)	cropping loss
11	X	X	death & decomposition (UL)	death & decomposition (UL)	X	X	X	respiration transpiration	respiration transpiration	translocation	ABOVEGROUND PLANT MATERIAL	cropping loss weathering
12	X	X	X	X	X	X	X	X	X	X	X	SINK

Figure 1 Water Source Interaction Matrix

	A	B	C	D	E	F	G	H	I	J	K	L
1	SOURCE (Gas)	dissolution	X	X	advection / diffusion	X	X	X	X	X	X	X
2	X	SOIL WATER	adsorption	adsorption	exsolution / evaporation	ingestion	ingestion / uptake	evaporation	X	root uptake	X	percolation to groundwater (& groundwater flow from area)
3	X	desorption	SOIL SOLIDS - Recalcitrant	decomposition?	decomposition	ingestion	ingestion / uptake	X	X	X	X	weathering
4	X	desorption	decomposition	SOIL SOLIDS - Labile	decomposition	ingestion	ingestion / uptake	X	X	X	X	weathering
5	X	gas sorption	X	X	SOIL GAS	inhalation / assimilation / metabolism (CH4)	X	degassing	X	root uptake	aerenchyma photosynthesis (assuming CO2)	X
6	X	excretion	X	death excretion?	respiration (CO2) decomposition methane oxidation (CO2)	SOIL MICROBES	X	X	X	X	X	X
7	X	excretion?	death & decomposition	death & decomposition	respiration (CO2) decomposition	X	MYCORRHIZAE	X	X	X	X	cropping (fruiting body) / dispersal
8	X	X	X	X	diffusion and barometric pumping bioturbation	X	X	CANOPY ATMOSPHERE - slow air flow (below zd)	diffusion / advective transport	X	photosynthesis (CO2)	X
9	X	X	X	X	X	X	X	diffusion / advective transport	CANOPY ATMOSPHERE faster air flow (above zd)	X	photosynthesis (CO2)	free air
10	X	root exudation	death & decomposition (UL & LL) ploughing	death & decomposition (UL & LL) ploughing	root respiration	X	transfer	X	X	BELOWGROUND PLANT MATERIAL	translocation (assuming root uptake)	cropping loss
11	X	X	death & decomposition	death & decomposition	X	X	X	respiration transpiration	respiration transpiration	translocation	ABOVEGROUND PLANT MATERIAL	cropping loss weathering
12	X	X	X	X	X	X	X	X	X	X	X	SINK

Figure 2 Gas Source Interaction Matrix

	A	B	C	D	E	F	G	H	I	J	K	L
1	SOURCE (Water)	upwelling	X	X	X	X	irrigation	X	X	X	X	X
2	percolation	SOIL WATER	adsorption	adsorption	exsolution / evaporation?	ingestion	capillary rise	X	X	X	X	X
3	X	desorption	SOIL SOLIDS - Recalcitrant	decomposition?	decomposition	ingestion	X	bioturbation ploughing	X	X	X	X
4	X	desorption	decomposition	SOIL SOLIDS - Labile	decomposition	ingestion	X	X	bioturbation ploughing	X	X	X
5	X	gas sorption	X	X	SOIL GAS	inhalation / assimilation / metabolism (CH4)	X	X	X	diffusion / advection	X	X
6	X	excretion	X	death excretion?	respiration (CO2) decomposition methane oxidation (CO2)	SOIL MICROBES	X	X	X	X	bioturbation ploughing	X
7	X	percolation	X	X	X	X	SOIL WATER	adsorption	adsorption	exsolution / evaporation?	ingestion	ingestion / uptake
8	X	X	bioturbation ploughing	X	X	X	desorption	SOIL SOLIDS - Recalcitrant	decomposition?	decomposition	ingestion	ingestion / uptake
9	X	X	X	bioturbation ploughing	X	X	desorption	decomposition	SOIL SOLIDS - Labile	decomposition	ingestion	ingestion / uptake
10	X	X	X	X	diffusion	X	dissolution	X	X	SOIL GAS	inhalation / assimilation / metabolism (CH4)	X
11	X	X	X	X	X	bioturbation ploughing	excretion	X	death excretion?	respiration (CO2) decomposition methane oxidation (CO2)	SOIL MICROBES	X
12	X	X	X	X	X	X	excretion?	death & decomposition	death & decomposition	respiration (CO2) decomposition	X	MYCORRHIZAE

Figure 3 Soil Layer Interaction Matrix (water source). The yellow boxes indicate the lower soil layer (LL) and the grey boxes indicate the upper soil layer (UL).

2.2.4 FEP descriptions

In this section, descriptions of the FEPs included within the interaction matrices are given, specifying how they relate to C-14 behaviour in the biosphere. The descriptions are derived primarily from IUR [2006], with additional text relating to C-14 where appropriate.

Aerenchyma

Aerenchyma are inter-connected gas-filled pathways found in some plants, e.g. rice, which grow on water-logged soils. Aerenchyma are a potential route of transport for C-14 from soil atmosphere to plant tissues.

Bioturbation

Bioturbation is the redistribution and mixing of soil by the activities of plants and burrowing animals. Bioturbation is not a C-14 specific issue, but is potentially relevant. It will be linked to soil type, climatic conditions and soil depth of interest. Bioturbation may involve recycling of sub-soil materials and incorporation of surface soil materials (e.g. detritus).

Capillary rise

Capillary rise is the upward movement of water through soil layers above the water table. The process arises as a result of capillary forces relating to evaporation and transpiration. Capillary rise is important in the overall water and nutrient dynamics in soil-crop systems and is a potential transport route of C-14 in groundwater to the soil rooting zone.

Cropping loss (plants & animals)

Potentially, cropping provides an important removal process, at least for the higher values of root uptake. Some models have conservatively ignored this process on the assumption that radionuclides taken up into crops would eventually be returned to the same soil through a variety of processes (including plant senescence and degradation or animal excretion).

Death and Decomposition

The death of animals or plants (e.g. plant roots) leads to the release of radionuclides to the immediate environment during decomposition. During plant senescence and decomposition, changes in the location and chemical form of carbon may be identified (e.g. transfer from above-ground to below-ground storage organs during senescence or incorporation within detritivorous animals or decomposing micro-organisms).

Degassing / Volatilisation

Water to air degassing can be a significant environmental transport pathway and may be significant for carbon dynamics, notably in the soil-solution or at the soil-atmosphere interface. Carbon may be lost from soils as CO₂ or CH₄. That part released as CO₂ will be available for uptake and incorporation into plants via photosynthesis. C-14 released from soils as CH₄ is likely to be lost from the system as it will not enter the stomata and become involved in the photosynthetic reactions. Rates of volatilisation will be dependent upon soil conditions, including moisture content, microbial activity, the form in which carbon is present and climatic factors such as temperature and humidity.

Diffusion

Diffusion is a physical process whereby material moves under the influence of a concentration gradient, resulting in a net flux from high to low concentration regions.

Discharge from below (upwelling)

In assessing the discharge of C-14 in groundwater from below, consideration would be required as to chemical processes associated with changes in redox conditions as groundwater migrates from sub-soil to surface soil. Additional geosphere-biosphere interface zone (GBIZ) processes may be important in this context.

Environmental change

In the long-term, climatic and associated environmental changes may alter plant uptake dynamics, through both physical changes (e.g. water regimes) and biological changes (e.g. vegetation species present). This may be important for long-term modelling of soil-plant C-14 dynamics.

Erosion

Not a C-14 specific issue. Should be included in models if the site context suggests wind or water erosion is a process for removing soil from the area of interest.

Evaporation

Transfer of water from the ground directly to the atmosphere which may include the transfer of C-14 in the gas phase.

Soil additives

One example of this is the addition of lime (CaCO_3) to agricultural soils, which could affect the behaviour of C-14.

Foliar uptake and photosynthesis

Foliar uptake is likely to be the major pathway for carbon uptake in the soil-plant system. Carbon volatilised from soils in the form of CO_2 will be incorporated into plant material following transport across stomata through the process of diffusion and then incorporated into biomass via photosynthesis. Absorption of dissolved CO_2 in irrigation water across stomata and subsequent incorporation into plant material by photosynthesis cannot be precluded.

Gas sorption

C-14 in soil gas may dissolve in soil water, or sorb to solid matter.

Infiltration

The process by which C-14 in irrigation water enters soil. The balance of infiltration, run-off and evaporation will be determined by a number of factors including soil type, topography, climate and rate of input. For most controlled systems, run-off is likely to be negligible.

Ingestion

Incorporation of C-14 in dissolved or solid form into micro-organisms or soil macrofauna.

Inhalation

Incorporation of C-14 in the gaseous phase into soil macrofauna as a result of breathing.

Interception

C-14 in groundwater applied to plants via spray irrigation may be intercepted thus preventing direct transport of water (and C-14) to the soil. Dissolved C-14 in intercepted water may bind to plant material leading to surface contamination or be taken up through stomata and be incorporated into plant material. Alternatively, intercepted water and C-14 may subsequently be transferred to soil as a result of plant run-off. Interception is a process that is largely considered in current models.

Irrigation

The use and application of abstracted water (containing C-14) for (agricultural) crops, to supplement natural water supplies (e.g. precipitation). Irrigation may involve spraying of water directly onto plants or application to soils (surface soil or flood irrigation).

Micro-organism metabolism and assimilation

Micro-organisms play important roles in the environmental fate of many elements, with a multiplicity of physico-chemical and biological mechanisms effecting changes in mobility and speciation. Physico-chemical mechanisms of removal include adsorption, ion exchange and entrapment. Microbial activity may be particularly important for the C-14 gas scenario whereby C-14 enters the biosphere in the form of methane (CH_4). Methanotrophs present in the soil may convert CH_4 to CO_2 which, when volatilised from soil, can be incorporated into plant material through the photosynthetic process. Some degree of assimilation of C-14 into microbes may occur as a result of the CH_4 metabolic process.

Microbial activity is dependent on a number of factors, including nutrient availability, temperature, water content, degree of aeration. Thus, for example, nutrient-deficient soils may have a slower rate of microbially induced speciation than nutrient-rich soils.

Percolation

Percolation is the process by which C-14 in soil water moves downwards into deeper horizons. This will depend on the proportion of carbon in the dissolved form (i.e. not bound to soil solids or incorporated in soil organic matter).

Precipitation

The rate of precipitation drives water flow from the soil surface to depth and acts to dilute C-14 in groundwater.

Respiration

Respiration is a consequence of metabolic processes that result in the oxidation of organic compounds, resulting in the release of C-14 as CO₂. In the case of plants, this includes root respiration. There is no distinction between the process of respiration from the roots and from the above-ground parts, but the receptor of the resultant CO₂ differs.

Root exudation

Roots of vegetation can release organic compounds containing C-14 directly to soil or soil water which may then be available for uptake by soil fauna and flora.

Root uptake

Although some data suggests that up to 10% of plant carbon requirements may be met by root uptake (see IUR [2006] and references therein), it is more generally accepted that the contribution to plant carbon uptake from roots is more like 1-2% [IUR, 2006; Sheppard et al, 1991; Vuorinen et al, 1989]. This uptake mechanism may therefore be important where the source of C-14 is from below when combined with an open canopy that permits rapid dilution of C-14 released from the soil surface.

Sorption (adsorption and desorption)

Currently, sorption (accounting for both adsorption and desorption) is considered as an instantaneous and reversible process through the application of K_d values. However, non-reversible sorption may also occur, or it may be necessary to consider the dynamic exchange between bound (i.e. organic carbon) and unbound carbon, such as CO₂. This should not be confused with carbon uptake into organic matter. Given the long-term nature of the release, it may be necessary to consider relatively slow processes which re-release bound/unavailable C-14 back into the unbound/available form.

Translocation

Translocation involves the transfer of C-14 from one (non-edible) part of a plant to another (edible) part.

Weathering

Weathering involves the loss of C-14 from the system. It can involve loss of surficial C-14 from leaf surfaces or physical loss of C-14 associated with surface soils as a result of atmospheric processes.

2.3 COMPARISON WITH PRE-EXISTING INTERACTION MATRICES AND FEP ANALYSIS FOR THE SAME SYSTEM (TERRESTRIAL)

The IM's presented in the previous section were developed following a two-day project workshop that involved a limited number of participants (three in total). In order to have confidence that the IM's presented are complete, they have been compared to published IM's for C-14 in a terrestrial ecosystem. Results of the comparison are given in this section.

2.3.1 IUR "Radioecology and Waste" Task Force

The International Union of Radioecology (IUR) created a Task Force "Radioecology and Waste" with the overall objective to promote the cooperation between radioecologists for research in the field of radioactive waste management. This Task Force produced a report [IUR, 2006] which provided an overview of the available knowledge, as of 2006, related to the behaviour of C-14, Cl-36, Tc-99,

Np-237 and U-238 in both terrestrial and aquatic ecosystems. An overview of the behaviour of the studied radionuclides was presented with the help of IM's, developed for both terrestrial and aquatic environments. Potentially relevant processes were identified for each radionuclide. The matrix that this Task Force developed for C-14 is shown in Figure 4.

The fourth workshop of this Task Force was hosted by CIEMAT in Madrid, Spain, between the 9th and 10th of October 2007, and centred on the terrestrial environment. Following a review of the IUR Report 6 findings [IUR, 2006], existing IM's for specific radionuclides were revised and some new IM's for additional radionuclides developed. The revised C-14 IM developed during that IUR workshop is reported in IUR [2007] and is shown in Figure 5. In this matrix, only those processes identified as being important are shown; in IUR [2006] all possible processes from the general matrix were shown. Three processes were added: sorption (J11), recharge by surface waters (K2), and desorption (K10).

2.3.2 CIEMAT analysis

Agüero et al [2006] present a C-14 IM and FEP analysis relevant to Spanish terrestrial conditions. The methodology used was based on the BIOMASS "Reference Biospheres Methodology", which provides a logical and systematic approach with supplementary documentation that helps to support the decisions necessary for model development. The methodology was also applied to the radionuclides Cl-36, Pu-239 and Tc-99. For each radionuclide, the physical and chemical characteristics were reviewed, and consideration given as to how those characteristics affected the behaviour of each radionuclide in various environmental media (the LDE's of the IM).

The IM developed for C-14 is almost identical to the one which resulted from the IUR Workshop in 2007. For uptake of C-14 into plants, root uptake is recognised as a potential pathway. Nonetheless, photosynthesis is considered to dominate. The potential for loss of C-14 to atmosphere during the use of contaminated groundwater in the spray irrigation process is acknowledged.

Atmosphere	1)Deposition	1)Deposition 2)Photosynthesis	1)Inhalation	1)Dry deposition 2)Precipitation 3)Gas sorption	1)Diffusive exchange 2)Pressure pumping				1)Diffusive exchange 2)Pressure pumping (both at outcrop)
1)Evaporation 2) Gas evolution 3)Droplet production	Water Bodies	1)Root uptake 2)Irrigation	1)Ingestion	1)Irrigation 2)Recharge by surface waters	1)Release from solution				1)Recharge by surface waters
1)Transpiration 2)Respiration 3)Pollen and seed release 4)Leaf fall 5)Release of other organic	1)Root exudation 2)Senescence and death	Vegetation	1)Ingestion	1)Root exudation	1)Root respiration	1)Litter fall 2)Senescence and death	1)Symbiotic association		1)Root exudation 2)Litter fall (at outcrop) 3)Senescence and death 4) Biological weathering
1)Exhalation 2) Eructation	1)Excretion 2)Death	1)Excretion 2)Death	Animals	1)Excretion		1)Excretion 2)Death and decomposition	1)Excretion of gut microbiota	1)Excretion	1)Excretion 2)Death and decomposition (both at outcrop)
1)Evaporation	1)Seepage 2)Throughflow 3)Groundwater recharge	1)Root uptake	1)Ingestion	Soil Solution	1)Ion exchange 2)Degassing	1)Sorpton 2)Fixation	1)Uptake	1)Sorpton 2)Fixation 3)Diffusion 4)Mineral precipitation	1)Advection 2)Diffusion
1)Diffusive exchange 2)Pressure pumping	1)Solution at boundaries	1)Root uptake and transport in aerenchyma)	1)Inhalation (burrowing animals)	1)Isotopic exchange 2)Solution	Soil Atmosphere	1)Adsorption	1)Uptake	1)Adsorption 2)Carbonate production	1)Diffusive exchange 2)Pressure pumping (both for unsaturated parent material)
1)Resuspension		1)External contamination	1)Ingestion	1)Desorption 2) Release during degradation	1)Degassing	Soil Organic Matter	1)Ingestion 2)Utilization	1)Complex formation	1)Particle transport 2)Colloid transport
1)Resuspension		1)Symbiotic association	1)Ingestion	1)Leaching 2)Mineralization 3)Excretion	1)Respiration 2)Fermentation	1)Fertilization 2)Death and decomposition 3)Biofilms	Soil Microbiota		1)Transport 2)Biological weathering
1)Resuspension		1)External contamination	1)Ingestion	1)Desorption 2)Mineral dissolution	1)Degassing	1)Microbial metabolism	1)Ingestion 2)Utilization	Soil Inorganic Matter	1)Particle transport 2)Colloid transport 3)isotopic exchange
1)Resuspension (at outcrop)	1)Desorption 2)Mineral dissolution	1)External contamination 2)Irrigation	1)Ingestion 2)Bioturbation	1)Diffusion 2)Advection 3)Colloid transport	1)Degassing	1)Microbial metabolism	1)Ingestion 2)Utilization	1)Chemical and mechanical weathering 2)isotopic exchange	Interface with geosphere

Figure 4 The general interaction matrix for the terrestrial environment [IUR, 2006], with the processes of potential importance for C-14 highlighted in bold

	A	B	C	D	E	F	G	H	I	J	K
1	Atmosphere		Photosynthesis	Inhalation		1)Diffusive exchange 2)Pressure pumping					
2	Gas evolution	Water Bodies	Irrigation	Ingestion	1)Irrigation 2)Recharge by surface waters						1)Recharge by surface waters
3	Respiration		Vegetation	Ingestion		Root respiration	1)Litter fall 2)Senescence and death	Symbiotic association			
4	1)Exhalation 2) Eructation			Animals	Excretion		1)Excretion 2)Death and decomposition				
5			Root uptake		Soil Solution	1)Ion exchange 2)Degassing		Uptake			
6	1)Diffusive exchange 2)Pressure pumping		1)Root uptake 2)Transport in aerenchyma		1)Isotopic exchange 2)Solution	Soil Atmosphere		Uptake			
7					1)Desorption 2)Release during degradation	Degassing	Soil Organic Matter	1)Ingestion 2)Utilisation			
8			Symbiotic association		Excretion	1)Respiration 2)Fermentation	1)Fertilisation 2)Death and decomposition 3)Biofilms	Soil Microbiota			
9									Soil Inorganic Matter		
10										Interface with geosphere -parent material	Desorption
11										Sorption	Groundwater of the bedrock

Figure 5 IUR [2007] Workshop C-14 Terrestrial Ecosystem Interaction Matrix

2.4 COMPARISON OF INTERACTION MATRICES

The IM's presented in the current report, developed during the two-day project workshop, are largely comparable with those previously developed [IUR, 2006, 2007; Agüero et al, 2006] for the terrestrial ecosystem. Nonetheless, some notable differences are evident. These differences largely arise as a result of the focus of the FEP analysis and IM development. The focus of both the IUR and CIEMAT analyses was on all key pathways relevant to human dose assessments; thus transport to animals was also considered. The current BIOPROTA study focuses only on processes up to, and including, transport of C-14 into plants. The different focus enables more detailed breakdown of soil and plant relevant compartments in the current study; thus a greater number of leading diagonal elements (LDE) are defined leading to greater transparency in the processes considered important for the transport of C-14 in soils and plants.

The comparison focuses on the current groundwater IM (Figure 1).

2.4.1 Leading diagonal elements

Both Agüero et al [2006] and IUR [2006, 2007] consider only one atmospheric LDE, whereas the BIOPROTA IM divides the canopy atmosphere into two compartments, one above and one below the z_d . It is considered important at this stage to clearly delineate the two zones to recognise the differences in air-plant interactions and C-14 transport dynamics.

The CIEMAT [Agüero et al, 2006] and IUR matrices apply a single vegetation LDE, whereas the current IM considers both below-ground and above-ground plant material as two separate compartments. Again, important differences are noted between the C-14 processes occurring within these two compartments. The importance of root uptake of C-14 has not yet been clarified and, until further consideration is given to this process, it is considered necessary for plant material to remain divided, thus allowing clear differentiation between the process of C-14 uptake by roots and uptake by leaf stomata.

The current IM also divides soil organic matter into labile and recalcitrant compartments; both CIEMAT and IUR consider a single LDE for organic matter. Again, the current differentiation is aimed at increasing clarity in relation to the different processes and timescales relevant to C-14 turnover in soils.

In the present IM, mycorrhizae are specifically considered as a separate LDE. Mycorrhizae are acknowledged to play an important role in the cycling of nutrients between soils and plants, being associated with plant roots. C-14, as glucose or other carbohydrates is passed from the plant to mycorrhizae, via roots, in return for enhanced provision of water and soil nutrients as part of a mutually beneficial symbiotic relationship. No individual LDE for mycorrhizae is included within the CIEMAT or IUR IM's, but rather they are incorporated within the soil microbiota LDE. Due to the important role of soil microbiota in their own right in relation to C-14 (the microbial oxidation of methane to carbon dioxide for subsequent uptake by plants and incorporation into plant material by photosynthesis), the decision was made to keep these two components separate in the current IM.

Both the CIEMAT and IUR IM's consider soil microbiota and farm animals. As noted previously, animals are excluded from the current IM due to the focus of the current project on transport of C-14 in soils and uptake into plants. However, it is acknowledged that animals may represent an important loss mechanism for C-14 through the processes of exhalation and eructation.

IUR [2006] considers the interface with the geosphere as an LDE, which is further sub-divided into two separate LDE's – interface with geosphere-parent material and groundwater of the bedrock^a. Groundwater is considered within the current groundwater IM, but the interface with geosphere-parent material is not incorporated. However, the focus of the current IM and FEP analysis is on soil and plant processes; thus interactions between the geosphere and groundwater may be argued to be

^a Agüero et al [2006] also considers two LDE's, but simplifies the terminology to 'parent material' and 'groundwater'

outside the scope of the current study. Nonetheless, interactions between the geosphere and groundwater may impact upon C-14 concentrations in groundwater; thus the availability of C-14 for transport into the biosphere, and may therefore warrant further consideration. The current IM considers a 'sink' LDE, to which loss processes from the system can be directed. Such an LDE is not incorporated in the Agüero et al [2006] or IUR matrices.

2.4.2 Processes linking LDE's

In IUR [2006, 2007] and Agüero et al [2006] the key process linking groundwater directly to vegetation is irrigation, with the potential for direct uptake by leaves. Remaining irrigation water is transported to soils. Indirectly, groundwater can enter plants from root uptake of soil solution. For transport between atmosphere and vegetation, the key processes are photosynthesis for uptake into plants and respiration for the transfer of C-14 from plants to atmosphere. Root exudation and respiration are mechanisms for plants to transfer C-14 to soil solution and soil atmosphere, respectively. These processes are all considered in the current IM.

The main differences noted in processes between LDE's relate to differences in terminology. Differences are detailed in the following table for the current IM (groundwater) and that of IUR [2007], unless otherwise stated. Where new processes, not considered to date in the current IM, are identified, these are emboldened. These may warrant further consideration.

Table 5 Differences between the BIOPROTA IM and previous IM's

Process position ^a	Current IM	IUR [2007] or other (stated)
B3/B4	Desorption	Desorption (IUR [2006] & Agüero et al [2006] only) Release during degradation
D6	Death & decomposition Excretion?	Fertilisation Death & decomposition Biofilms
H5	Degassing	Diffusive exchange Pressure pumping
F3/F4	Ingestion	Ingestion Utilisation
H2	Evaporation	No direct process – soil water to soil gas then release to atmosphere Evaporation considered in IUR [2006] and Agüero et al [2006]
C11/D11	Death & decomposition	Litterfall Senescence & death
B10	Root exudation	No pathway Root exudation included in IUR [2006] and Agüero et al [2006] only

^a Relates to position within the current groundwater IM (Figure 1)

There are no direct processes linking the geosphere-parent material LDE with biosphere LDE's within the IUR or CIEMAT IM's. Thus, by excluding this LDE from the current IM, no important processes are considered to have been missed within the biosphere-specific IM.

As noted in section 2.3.2, Agüero et al [2006] recognises the potential for loss of C-14 in groundwater to atmosphere during spray irrigation. This process has not been considered in the current IM and may warrant further consideration. Since spray irrigation would occur in the above-canopy atmospheric compartment, loss to the sink LDE would be considered appropriate.

3. MODEL DESCRIPTIONS

This section contains descriptions of the models used in the quantitative model inter-comparisons. There are five models in total:

- The model developed for and by Andra, AquaC_14;
- The model developed by EDF, SA_Carbon14;
- The C-14 specific model developed for NDA RWMD, enhanced RIMERS;
- The model developed for SKB and Posiva, called the Avila and Pröhl model; and
- The model developed for the LLW Repository Ltd (LLWR), referred to herein as the Thorne-Limer model.

Although all of these models can be, and have been, used to calculate potential impacts to humans, the focus of the descriptions presented in this study is the soil-plant system. A further four models were identified, but have not been used for this study. Those models are the UK Food Standard's Agency model PRISM [Walke et al, 2010], IRSN's model TOCATTA^a, the Nagra C-14 model, and the CIEMAT C-14 model.

3.1 AQUAC_14

The Andra C-14 model was originally developed by Penfold and Watkins [1998] and transcribed into the general radionuclide biosphere transfer model, Aquabios^b, jointly with equations describing transfer of "classical" radionuclides. During transcription, minor changes were carried out to allow parallel treatment of C-14 and classical radionuclides [van Hecke, 2001]. However, due to difficulties associated with parallel transcription, the C-14 model was later extracted from Aquabios, without changing the model equations given by van Hecke [2001]. The current version of the model is stand-alone and known as AquaC_14.

The mathematical model is mostly based on the original formulations by Penfold and Watkins [1998]. A detailed analysis of the Andra AquaC_14 model was carried out in 2010, primarily based upon the outcome of the initial C-14 intercomparison. The equations of this recent, slightly updated, model have been summarised in an Andra technical note [Albrecht, 2010].

3.1.1 Model equations

The sequence used in this description is identical to the one which appears within the model structure of AquaC_14. This sequence of equations is controlled by the rank of the equation in the model management software (MoM). The AquaC_14 model is characterised by a large number of parameters that are specific to carbon, such as stable carbon concentrations in the various compartments and components.

In the current model, only one type of equation (type A) is used. The equations are all for the same form of carbon (carbon dioxide, ¹⁴CO₂). It is possible to enhance the model by considering methane (¹⁴CH₄), which is very different in biogeochemical behaviour from its oxidised form. In this case, two other types of equations will be needed, one for CO₂ and one for CH₄.

^a A paper about the TOCATTA model parameterisation for C-14 and testing against measurements performed in the vicinity of the La Hague fuel recycling plant in France is currently in preparation.

^b For details on the Aquabios model see Albrecht and Miquel [2010].

Water contamination

As for the model Aquabios, two sources of water may be contaminated; water from a well and/or river water. The first two equations read the concentrations of the river (C_{riv}) and ground water (C_{nap}) in the associated input files ($rejet_{rivie}$, $rejet_{nappe}$):

$$C_{riv} = rejet_{rivie} \quad (1)$$

$$C_{nap} = rejet_{nappe} \quad (2)$$

In the current model three additional input variables are available: (i) via upwelling water (or gas flow); (ii) deposition related to gaseous discharges; and (iii) concentration in a passing plume.

Drinking water may have its source in a well (pts) or in a river (riv). Where drinking water is a mixture of well and river water, the concentration ($C_{boisson}$, Bq L⁻¹) is estimated with the following equation:

$$C_{boisson} = \frac{(1 - FH_{eaupts}) \cdot C_{riv} + (FH_{eaupts}) \cdot C_{nap}}{1 + Kd_{MES} \cdot MES_{Riv} \cdot 1E-6 + 1 + Kd_{MES} \cdot MES_{Nap} \cdot 1E-6} + \left[\frac{(-) \times \frac{Bq}{L}}{(-) + \frac{L}{kg} \times \frac{mg}{L} \times 1E-6 \frac{kg}{mg}} + \frac{(-) \times \frac{Bq}{L}}{(-) + \frac{L}{kg} \times \frac{mg}{L} \times 1E-6 \frac{kg}{mg}} \right] = \frac{Bq}{L} \quad (3)$$

FH_{eaupts} represents the fraction of drinking water from the wells, C_{riv} , C_{nap} (Bq L⁻¹) the concentrations of C-14 in the river and well waters, MES_{Riv} , MES_{Nap} the mass concentrations of suspended matter in the river and the well water (mg L⁻¹, the factor 1E-6 converts mg to kg) and Kd_{MES} the distribution factor between water and suspended matter (Bq kg_{dry}⁻¹ per Bq L⁻¹). This factor considers not only the overall sorption of carbon, but also other processes that transfer carbon from the aqueous phase to the solid phase (e.g. precipitation of carbonate, organic matter formation etc.). No distinction is made between the sorption to suspended solids in the river compared with those in the well (cf. use of a single Kd_{MES}).

For well water, no filtration is considered such that the concentration of suspended solids (MES_{Nap}) is set to zero. For river water, the impact of sorption and retention by filtration is modelled for human drinking water, an approach that is not followed for the drinking water of livestock (C_{abreuv} , Bq L⁻¹):

$$C_{abreuv} = (1 - FH_{eaupts}) \cdot C_{riv} + (FH_{eaupts}) \cdot C_{nap} \quad (4)$$

The water brought onto cultivated soils ($C_{eaucult}$, Bq L⁻¹) and grassland soils ($C_{eauprai}$, Bq L⁻¹) in the form of irrigation water (or flooding) may have its source in the well or the river. The factors $F_{ceaupuits}$ and $F_{peaupuits}$ indicate fractions of well water used for the cultivated (c) or grassland soils (p). The fraction for river water is calculated knowing that the sum of fractions from the wells and the river equals unity:

$$C_{eaucult} = (1 - F_{ceaupuits}) \cdot C_{riv} + (F_{ceaupuits} \cdot C_{nap}) \quad (5)$$

$$C_{eauprai} = (1 - F_{peaupuits}) \cdot C_{riv} + (F_{peaupuits} \cdot C_{nap}) \quad (6)$$

Soil contamination

In the current AquaC_14 model, soil contamination is a function of irrigation quantified by a rate ($teaucult$, $L\ m^{-2}\ y^{-1}$). Mass concentration in soil ($Bq\ kg^{-1}$) is modelled as for traditional radionuclides [Albrecht and Miquel, 2010] using a box model approach and the quantification of flows:

$$\frac{dC}{dt} = Q - \lambda C \quad (7)$$

whereby Q is the input ($Bq\ kg^{-1}\ y^{-1}$) and λC the output. In the case of C-14, which is significantly retained by vegetation (the fraction reaching the ground, $FEIS$ (-) is equal to 0.5 for the current setting), two inflows must be distinguished; the fraction directly reaching the soil and the fraction leached from vegetation:

$$\underbrace{input \cdot FEIS}_{direct} + \underbrace{input \cdot \frac{LPSS}{LPSS + LPSP + LPSA}}_{indirect} (1 - FEIS) \quad (8)$$

Of the fraction intercepted by the plant, some is transported by translocation to the leaves ($LPSP$, y^{-1}), some evaporated ($LPSA$, y^{-1}) and some leached ($LPSS$, y^{-1}). The sum of these losses (LPS) is quantified using the following equation:

$$LPS = LPSS + LPSP + LPSA \quad (9)$$

It is used to calculate the input function in the equation for quantification of soil activity (Q_{cult} , $Bq\ kg^{-1}\ y^{-1}$), with the example of cultivated soil given here:

$$Q_{cult} = \frac{teaucult \cdot Ceaucult}{\underbrace{(1 - poro_cult) \cdot rhopart \cdot PROFCULT}_{=rhocult}} FEIS + \frac{teaucult \cdot Ceaucult}{(1 - poro_cult) \cdot rhopart \cdot PROFCULT} \cdot \frac{LPSS}{LPS} (1 - FEIS) \quad (10)$$

The parameters used in this equation, their units and descriptions are listed in Table 6. Soil density ($rhocult$, $kg.m^{-3}$) is not directly specified, but is calculated on the basis of porosity and particle density.

Table 6 Parameters used to quantify the soil contamination

Parameter	Units	Description
teaucult	$L\ m^{-2}\ y^{-1}$	Irrigation rate
FEIS	(-)	Fraction of irrigation reaching the soil
LPSS	y^{-1}	Loss via percolation
LPSP	y^{-1}	Loss via translocation
LPSA	y^{-1}	Loss via evaporation
poro_cult	(-)	Porosity of cultivated soil
rhopart	$kg\ m^{-3}$	Density of particles
profcult	m	Depth of cultivated soil
tensoileau	(-)	Degree of saturation

To calculate system losses (λ), several additional parameters must be determined starting with the water balance, which is the sum of water input via precipitation ($rain$, $L\ m^{-2}\ y^{-1}$) and irrigation ($teaucult$, $L\ m^{-2}\ y^{-1}$), minus evapotranspiration (ETP , $L\ m^{-2}\ y^{-1}$), which may be adapted, depending on the type of crop, using a crop coefficient ($coef_{cult}$, -):

$$Bihy_{cult} = pluie + teau_{cult} + Taux_{cult} - coef_{cult} \cdot etp \quad (11)$$

The retention factor of C-14 in soil ($retent_{soilc}$, -) is a parameter required to calculate accumulation of C-14 in the soil:

$$retent_{solc} = 1 + \frac{Kdsol \cdot (1 - poro_{cult}) \cdot rhopart}{tensolceau \cdot poro_{cult} \cdot 1000} \cdot \left[\frac{\frac{L}{kg} (-) \frac{kg}{m^3}}{(-) \cdot \frac{L}{m^3}} \right] \quad (12)$$

where $poro_{cult}$ is the porosity (volume/volume) of the cultivated soil, $rhopart$, the density of soil particles ($\sim 2600 \text{ kg m}^{-3}$), and $tensolceau$, the degree of saturation (with $tensolceau = 1$ for saturated soil). A similar equation exists for grassland soil with p ($prai$) replacing c ($cult$) in the given equations.

Leaching losses are calculated based on the retention factor ($retent_{solc}$), the water balance ($Bihycult$) and the thickness of the soil compartment in question ($profcult$):

$$\lambda_{lix} = \frac{Bihycult}{tensolceau \cdot poro_{cult} \cdot profcult \cdot retent_{solc}} \quad (13)$$

Quantification of total soil losses needs to consider, in addition, soil degassing described by a term for soil-air degassing (LSA):

$$L_{culture} = \frac{Bihycult}{tensolceau \cdot poro_{cult} \cdot profcult \cdot retent_{solc}} + LSA \quad (14)$$

Equations identical to those described for the cultivated soil, but applicable for grassland, need to be mentioned. They consider the total input to the soil ($Qprairie$), the water balance ($Bihyprai$) and leaching and out-gassing losses ($Lprairie$).

To calculate the dynamic evolution of C-14 in soil, the integrated form of the differential equation given at equation 7 is used:

$$C_{sol}(t_j) = \frac{Q}{\lambda_{tot}} + \left[\left(C_{sol}(t_i) - \frac{Q}{\lambda_{tot}} \right) e^{-(\lambda_{tot})(t_j - t_i)} \right] \quad (15)$$

Integration is possible, when λ_{tot} and Q are constant; this is the case for step-wise calculations. The analytical solutions as transcribed in the model for cultivated and grassland soils are:

$$CSolparai = Qculture / (Lculture + lambda) + (conc_{cult_1} - Qculture / (Lculture + lambda)) \cdot EXP(-(Lculture + lambda) \cdot (tj - ti)) \quad (16)$$

$$CSolprai = Qprairie / (Lprairie + lambda) + (conc_{prai_1} - Qprairie / (Lprairie + lambda)) \cdot EXP(-(Lprairie + lambda) \cdot (tj - ti)) \quad (17)$$

*MoM performs the calculations step by step; soil concentrations of the previous step ($conc_{cult_1}$ for example) are managed by specific functions called UDF, which store these values for use during the next step.

3.1.2 Uptake into plants

As previously indicated, uptake of C-14 into plants occurs through interception and translocation as well as by root transfer and photosynthesis. To estimate the portion intercepted, it is necessary to quantify the surface activity related to interception ($Inter_{cult}$, Bq m⁻²), which depends on the contamination flux, the intercepted fraction (1-FEIS) and losses (LPS). The example given here is for the cultivated soil:

$$Inter_{cult} = \frac{Ceau_{cult} \cdot teau_{cult} \cdot (1 - FEIS)}{LPS} \cdot \left[\frac{\frac{Bq}{L} \cdot \frac{L}{m^2 y} \cdot (-)}{\frac{1}{y}} \right] \quad (18)$$

To estimate the contamination via incorporation of ¹⁴CO₂ during photosynthesis requires information on the volume concentration in the canopy of plants (Bq m⁻³), which depends on the concentration of C-14 in the soil (C_{soil} in Bq kg⁻¹_{dry}), the soil density [(1-porosity) × density of particles; kg_{dry} m⁻³] and the rate of outgassing (y⁻¹). A geometric factor related to the wind direction ($Fgeo$, -), the wind speed ($Vvent$, m s⁻¹), seconds per year (3.16E7 s y⁻¹) and the fetch must also be taken into account. The latter allows the size of the fields to be taken into account:

$$fetch = \frac{length \cdot height}{volume}; [m^{-1}] \quad (19)$$

By making the field wider, the fetch is reduced and the activity of the canopy increases:

$$Ccano_{cult} = \frac{Csol_{cult} \cdot \overbrace{(1 - poro_{cult}) \cdot Rhocult}^{Rhopart} \cdot LSA}{Fgeo \cdot fetch \cdot Vvent \cdot 3.16e7} \cdot \left[\frac{\frac{Bq}{kg_{dry}} \cdot \frac{kg_{dry}}{m^3} \cdot \frac{1}{y}}{(-) \cdot \frac{1}{m} \cdot \frac{m}{s} \cdot \frac{s}{y}} \right] \quad (20)$$

A similar procedure is used for the canopy of a grassland area.

With these parameters it is possible to quantify the three types of contamination of the plant:

- (1) via root transport;
- (2) via contamination by photosynthesis; and
- (3) via interception.

The example given here is that for cereals. Root transfer from soil to plant is quantified on the basis of an isotopic ratio approach. As a reminder:

$$\left(\frac{^{14}C}{C_{tot}} \right)_{plant} = \left(\frac{^{14}C}{C_{tot}} \right)_{sol} \cdot \left[\frac{Bq/kg_{fresh}}{Bq/kg_{fresh}} \right] = \left[\frac{Bq/kg_{dry}}{Bq/kg_{dry}} \right] \quad (21)$$

As the concentration of total carbon (= stable carbon) in plants is normally given in kg kg_{dry}⁻¹ there is a need to have a conversion factor from dry to fresh plant kg_{dry}/kg_{fresh} which is calculated based on the water content of a fresh plant ($Eau_{plante} = 0.8$ kg.kg_{fresh}⁻¹) and the content of dry matter (1 - Eau_{plante}):

$$Conv_pl_sec_frais = \frac{1 - eau_plante}{(1 - eau_plante) + eau_plante} = \frac{1 - eau_plante}{1} ; \left[\frac{kg_{dry}}{kg_{fresh}} \right] \quad (22)$$

A similar equation exists for considering the mass of water in grass ($eauher = 0.9 \text{ kg kg}_{fresh}^{-1}$):

$$Conv_her_sec_frais = \frac{1 - eauher}{1} ; \left[\frac{kg_{dry}}{kg_{fresh}} \right] \quad (23)$$

The use of the isotopic approach to estimate the C-14 root transfer to plants is shown here in detail for cereals:

$$\left(\frac{{}^{14}C}{C_{stable} \cdot Conv_pl_sec_frais} \right)_{cer} = \left(\frac{{}^{14}C}{C_{stable}} \right)_{soil_dry} ; \left[\frac{\frac{Bq}{kg_{fresh}}}{\frac{kg}{kg_{dry}} \cdot \frac{kg_{dry}}{kg_{fresh}}} \right] = \left[\frac{Bq/kg_{dry}}{Bq/kg_{dry}} \right] \quad (24)$$

Rearrangement and application of parameter names used in AquaC_14 gives the following with 1 indicating the contribution of the root pathway:

$$Ccer1 = \frac{CSolcult \cdot CcarbCer \cdot Conv_pl_sec_frais}{CcarbSol} ; \left[\frac{\frac{Bq}{kg_{dry}} \cdot \frac{kg}{kg_{dry}} \cdot \frac{kg_{dry}}{kg_{fresh}}}{\frac{kg}{kg_{dry}}} \right] = \left[\frac{Bq}{kg_{fresh}} \right] \quad (25)$$

The fraction of carbon that cereals acquire through roots (fC_{dusol} , -) and the concentration of carbon in the soil ($C_{carbSoil}$, kg kg_{dry}^{-1}) are required parameters. Consideration of these parameters yields the equation integrated in MoM for cereals (1.10.1.3):

$$Ccer1 = \frac{CSolcult \cdot CcarbCer \cdot Conv_pl_sec_frais \cdot fC_{dusol}}{CcarbSol} \quad (26)$$

For the part of the contamination coming from plant photosynthesis ($Ccer2 \text{ Bq kg}_{fresh}^{-1}$) the isotopic approach is again applied:

$$\left(\frac{{}^{14}C}{C_{tot}} \right)_{plant} = \left(\frac{{}^{14}C}{C_{tot}} \right)_{canopy} ; \left[\frac{\frac{Bq}{kg_{fresh}}}{\frac{kg}{kg_{dry}} \cdot \frac{kg_{fresh}}{kg}} \right] = \left[\frac{Bq/m^3}{kg/m^3} \right] \quad (27)$$

Using the carbon concentration in air ($C_{carbair}$ in kg m^{-3}) and the plant carbon fraction coming from the air (fC_{deAir} , -):

$$Ccer2 = \frac{CcarbCer \cdot Conv_pl_sec_frais \cdot fC_{deAir} \cdot Ccanocult}{CcarbAir} \quad (28)$$

It is again necessary to convert the total carbon concentration of the plant (e.g. cereals) measured in kg kg_{dry}^{-1} , to $\text{kg kg}_{fresh}^{-1}$, hence the need to introduce in each equation the conversion factor $Conv_pl_sec_frais$.

The part of contamination coming from interception (C_{cer3} , Bq kg_{fresh}⁻¹) depends on the interception factor previously quantified ($Intercult$, Bq m⁻²), the translocation factor ($LPSP$, y⁻¹) the number of crop rotations per year (T_{crop} , y⁻¹), the leaf area index (LAI_{plant} , -) and the annual yield of the plant ($Rendcult$, kg m⁻²).

$$C_{cer3} = Intercult \cdot \frac{LPSP}{T_{crop}} \cdot \frac{LAI_{plant}}{Rendcult} \cdot \left[\frac{Bq}{m^2} \cdot \frac{1}{y} \cdot \frac{m^2}{m^2} \cdot \frac{kg_{fresh}}{m^2} \right] \quad (29)$$

The total concentration of C-14 in cereals ($CCer$ in Bq kg_{fresh}⁻¹) is equal to the sum of these three components:

$$CCer = CCer1 + CCer2 + CCer3 \quad (30)$$

The same approach is used for all agricultural crops and grass. A list of the parameters, abbreviations and dimensions is given in Table 7.

Table 7 Parameters, Abbreviations and Units Used for the Contamination of Crops

Parameter / abbreviation	Definition	Units
C_{cer}	Contamination of cereals	Bq kg _{fresh} ⁻¹
C_{fruit}	Contamination of fruits	Bq kg _{fresh} ⁻¹
$C_{feuille}$	Contamination of leafy vegetables	Bq kg _{fresh} ⁻¹
C_{rac}	Contamination of root vegetables	Bq kg _{fresh} ⁻¹
C_{pdt}	Contamination of potatoes	Bq kg _{fresh} ⁻¹
C_{her}	Contamination of grass	Bq kg _{fresh} ⁻¹
C_{foin}	Contamination of hay $C_{her} / (1 - EAU_{her})$	Bq kg _{dry} ⁻¹

For the quantification of the contamination of grass and hay the specific parameters for grassland soil and grass must be applied as follows:

$$C_{her1} = C_{solprai} \cdot C_{carbprai} \cdot Conv_{her_sec_frais} \cdot f_{cdusol} / C_{carbSol} \quad (31)$$

$$C_{her2} = (C_{carbPrai} \cdot Conv_{her_sec_frais} \cdot f_{CdeAir} \cdot C_{canoprai}) / C_{carbAir}$$

$$C_{her3} = (Intercult \cdot LPSP / T_{crop}) \cdot LAI_{prai} / Rendprai$$

$$C_{her} = C_{her1} + C_{her2} + C_{her3}$$

Table 8 lists model-relevant parameters, with abbreviations used in equations, the dimensions and the meaning.

Table 8 C-14 Model Parameters, Units and Definitions

Parameter / abbreviation	Units	Definition
$C_{carbair}$	kg m ⁻³	Carbon mass fraction in air
$C_{carbcer}$	kg kg _{dry} ⁻¹	Carbon mass fraction in cereals
$C_{carbfeuille}$	kg kg _{dry} ⁻¹	Carbon mass fraction in leafy vegetables
$C_{carbfruit}$	kg kg _{dry} ⁻¹	Carbon mass fraction in fruits
$C_{carbpdt}$	kg kg _{dry} ⁻¹	Carbon mass fraction in potatoes
$C_{carbprai}$	kg kg _{dry} ⁻¹	Carbon mass fraction in prairie grass
$C_{carbrac}$	kg kg _{dry} ⁻¹	Carbon mass fraction in root vegetables
$C_{carbsoil}$	kg kg _{dry} ⁻¹	Carbon mass fraction in soil

Parameter / abbreviation	Units	Definition
<i>Coefcult</i>	(-)	Water uptake coefficient of cultured plants
<i>Eauher</i>	L kg ⁻¹ _{fresh}	Water content of grass
<i>Fcdeair</i>	(-)	Fraction of stable C in plants coming from air via photosynthesis
<i>Fcdusoil</i>	(-)	Fraction of stable C in plant coming from the soil
<i>Feaucarb</i>	(-)	Mass fraction of carbon in water
<i>Fetch</i>	m ⁻¹	Fetch (non-standard definition)
<i>Fgeo</i>	(-)	Geometric factor
<i>Laicer</i>	(-)	Leaf area index cereals
<i>Laifeuille</i>	(-)	Leaf area index leafy vegetables
<i>Laifruit</i>	(-)	Leaf area index fruit
<i>Laipdt</i>	(-)	Leaf area index potatoes
<i>Laiprai</i>	(-)	Leaf area index prairie grass
<i>Lairac</i>	(-)	Leaf area index of root vegetables
<i>Poro_cult</i>	(-)	Porosity of cultivated soils
<i>Poro_prai</i>	(-)	Porosity of prairie soil
<i>Profcult</i>	m	Rooting depth of cultivated soils
<i>Profprai</i>	m	Rooting depth of prairie soils
<i>Rendcult</i>	kg _{fresh} m ⁻² y ⁻¹	Yield of cultivated soils
<i>Rendprai</i>	kg _{fresh} m ⁻² y ⁻¹	Yield of prairie soils
<i>Rho_plante</i>	kg _{fresh} m ⁻³	Fresh density of plant material
<i>Rhopart</i>	kg m ⁻³	Density of soil particles
<i>Tcrop</i>	y ⁻¹	Number of crop rotations per year
<i>Vvent</i>	m s ⁻¹	Wind speed

3.2 SA_CARBN14

A river model was proposed by EDF to compute the potential dose from C-14 to members of the public at any location along the Loire River in France [Ciffroy et al, 2001; Damois et al, 2002].

Dilution of C-14 along the river is estimated in a separate model and this work has indicated that C-14 concentrations vary on an annual cycle [Sheppard et al, 2006a]. In this model, lateral and vertical mixing is assumed to be complete. Thus, the input of relevance to the present model is the concentration of C-14 in the water at the time and place where the assessment is to be done. It is, therefore, possible to apply the model described below to the scenario of contaminated well water that is used to irrigate crops.

Due to the effects of seasonality, it was considered important to use time-dependent models representative of C-14 transfers in the food-chain. Wirth [1982] noted that, if equilibrium specific-activity models are used instead of time-dependent specific-activity models, “*the radiation exposure is always overestimated for local short-time inputs*”^a. Here, specific activity is defined as the activity of C-14 per unit mass of total C.

^a However, in the solid waste disposal context, it is not clear that this is a significant argument against their use.

The conceptual model used by EDF is an adaptation of the equations described in Sheppard et al [2006a] and is summarised here for convenience. Some parameters for the model are given in Sheppard et al [2006b]. The model is represented in Figure 6.

Calculation of the water content in soil, the percolation rate and the irrigation rate are based on the same equations used in the model for CI-36, SA_36Cl [Limer et al, 2008a].

3.2.1 C-14 in soil

Input of C-14 to soil is from irrigation using contaminated surface water. There is evidence [Sheppard et al, 1991; Sheppard and Evenden, 1996a, b] that C-14 in soil can be partitioned between labile and relatively recalcitrant forms, where the recalcitrant forms may be inorganic (carbonate minerals) or organic (insoluble humic substances). The parameter f_f can be used to reflect the recalcitrant long-term soil organic C when C-14 is added in an inorganic form. Soil organic matter is not simple; it ranges from easily decomposed freshly added material through to forms that are very recalcitrant. Typically, only a small fraction of the plant material added each year contributes to the recalcitrant fraction; most is decomposed rapidly and lost as a result of respiration by soil decomposer organisms. The second term of the following equation corresponds to the increase of C-14 in organic form in soil due to the residue of crops after harvest, which contributes to the soil organic pool. The third term corresponds to the proportion of the pool of organic matter that is mineralised each year. The present model simply partitions a portion of the input to recalcitrant or fixed forms:

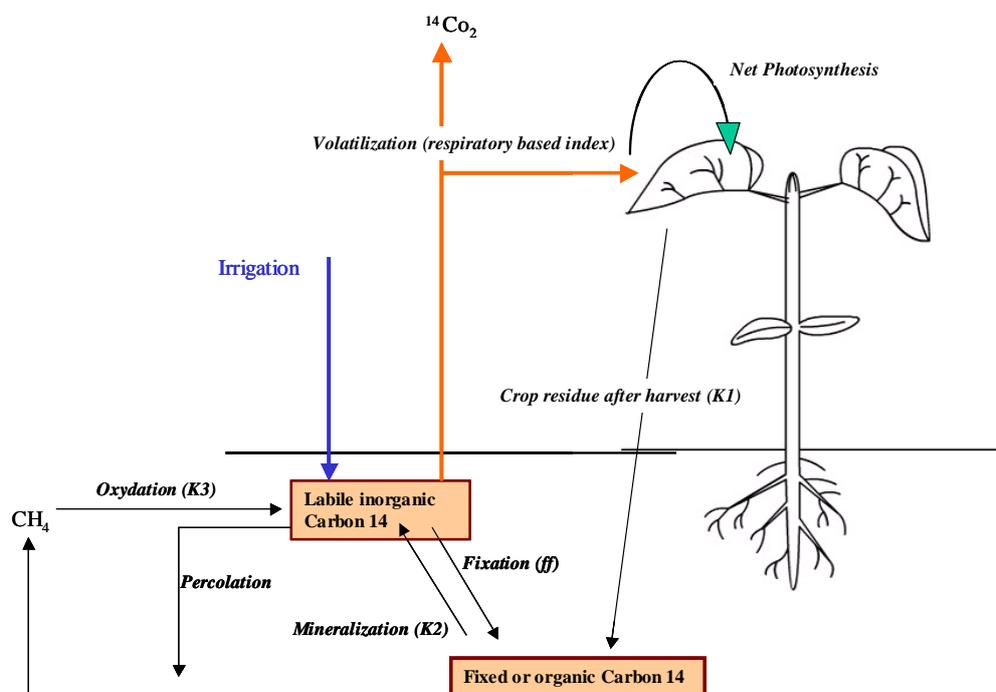


Figure 6 EDF conceptual model for the uptake of C-14 into crops (SA_Carbon14)

$$C_{soil}^{C14-fixed}(t_n) = C_{soil}^{C14-fixed}(t_{n-1}) + (f_f \times Q_{irr_n} \times C_{Water}^{14} + \sum_{crop} S_{crop} \times A_{crop}^{C14,spc} \times C_{crop}^C \times n_{crop} \times \frac{B_{crop} \times Per_{not_harvested}}{\Delta t} \times K_1 - K_2 \times C_{soil}^{C14-fixed}(t_{n-1})) \Delta t \quad (32)$$

where:

- $C_{soil}^{C14-fixed}$ is the activity concentration of fixed or recalcitrant C-14 in soil (Bq m⁻²);
- f_f is the fraction of C-14 that becomes fixed when applied to soil (-);

- Q_{irr} is the irrigation rate ($m\ d^{-1}$);
- C_{water}^{C14} is the activity concentration in the irrigation water ($Bq\ m^{-3}$);
- $A_{crop}^{C14, spe}$ is the specific activity in crop ($Bq\ kg\ C^{-1}$);
- B_{crop} is the yield of the crop per time unit ($kg\ dry\ matter\ m^{-2}\ d^{-1}$);
- $Per_{not_harvested}$ is the percentage of crop not removed (e.g., straw and roots for cereals);
- K_1 is the isohumic coefficient (fraction of fresh organic matter that becomes stable);
- C_{crop}^C is the concentration of C in plant dry matter ($kg\ C\ kg^{-1}\ dry\ matter$);
- n_{crop} is the number of germination events per year for each crop;
- K_2 is the mineralisation rate of organic matter (d^{-1}); and,
- Δt is defined by $t_n - t_{n-1}$.

The other fraction of C-14 in soil is labile, subject to loss due to radioactive decay, volatilisation and leaching:

$$C_{soil}^{C14_labile}(t_n) = C_{soil}^{C14_labile}(t_{n-1})e^{-(\lambda_{leach} + \lambda_{vol})\Delta t} + \frac{((1 - f_f)Q_{irr}(\Delta t)C_{water}^{C14} + K_2C_{soil}^{C14-fixed} + F_{CO2})}{h_{soil}(\lambda_{leach} + \lambda_{vol})} (1 - e^{-(\lambda_{leach} + \lambda_{vol})\Delta t}) \quad (33)$$

where:

- $C_{soil}^{C14-labile}$ is the activity concentration of labile C-14 in soil at time n ($Bq\ m^{-3}$);
- λ_{vol} is the volatilisation from soil rate constant (d^{-1});
- h_{soil} is the depth of the ploughing zone;
- F_{CO2} is the flux of CO_2 entering the root zone; and,
- λ_{leach} is the leaching rate from soil rate (d^{-1}).

The leaching (percolation) rate is calculated with the model presented in Limer et al [2008b].

The flux of CO_2 (F_{CO2}) can be a scenario value or it can be calculated from the flux of CH_4 entering the soil zone (F_{CH4}):

$$F_{CO2} = K_3 \times F_{CH4} \quad (34)$$

where K_3 (-) is the fraction of CH_4 that is oxidised in the soil.

The volatilisation rate used in this model is an empirical quantity, assumed to include release of C-14 from soil liquids (e.g., dissolved $^{14}CO_2$), or indirectly from soil solids (e.g., dissolution of carbonate C-14) and soil organic matter (decomposition of organic C-14). Once the activity concentration of C-14 in soil is known, the flux density of C-14 ($Bq\ m^{-2}\ soil\ d^{-1}$), assumed to be $^{14}CO_2$, leaving the soil, $Q_{volatilized}^{C14}$, can be computed as:

$$Q_{\text{volatilized}}^{\text{C14}} = \lambda_{\text{vol}} C_{\text{soil}}^{\text{C14_labile}} \quad (35)$$

3.2.2 C-14 in the plant-canopy atmosphere

Carbon dioxide from respiration can be dissipated in two ways: photosynthesis and turbulent mixing with the atmosphere. There are not many, if any, generalised models of the mixing of soil and free atmospheric air in the plant canopy, and a rigorous treatment may not be possible given the geometric complexity of the many different types of plant canopies. The EDF model therefore utilises the simplifying assumption that only a fraction of CO₂ in the plant canopy originates as CO₂ released from the soil. Two approaches have been used by EDF in this BIOPROTA study, and are described below.

First approach: Sheppard approach [Sheppard et al, 2006a]

The specific activity (Bq C-14 g⁻¹ C) of CO₂ volatilised from the soil into the plant-canopy atmosphere $A_{\text{volatilized}}^{\text{C14}}(t)$ is a ratio of the flux densities:

$$A_{\text{volatilized}}^{\text{C14}}(t) = CD \left[Q_{\text{volatilized}}^{\text{C14}}(t) / Q_{\text{volatilized}}^{\text{C}}(t) \right] \quad (36)$$

where CD is a canopy dilution factor (-) to indicate the degree to which the canopy air is diluted by free-atmosphere air that is assumed to have no contaminant C-14. The canopy dilution factors proposed in the publication of Sheppard et al [2006b] are based on the publication of Amiro et al [1991] considering that the height of the crop is 0.3 m and that the z_d is one sixth of the crop height. In this approach, it is assumed that the size of the garden is 10 *10 m and that the cereal field can have two sizes (100 m* 100 m or 1000m*1000m).

Second approach: Respiratory approach

The second approach utilises the simplifying assumption that only a fraction of CO₂ in the plant canopy originates from CO₂ released from the soil. This fraction, called the respiratory recycling index ($I_{\text{respiratory_recycling}}$, -) refers to the flux of respired CO₂ re-fixed by photosynthesis relative to the total respiratory flux [Sternberg 1989; Greaver et al. 2005]. Research by Greaver et al [2005] indicates that values of gross photosynthesis and respiration are often estimated as being unrealistically low if no recycling of CO₂ within the plant canopy is assumed. The respiratory recycling index can be determined from measurements of the isotopic ratio C-13/C-12 in soil/air/plant.

For this study, two different values have been taken from the literature in the parameterisation of $I_{\text{respiratory_recycling}}$. One value is associated with cereals and the other with the three other crop types considered in this study. The reason for this is the anticipated similarity in the crop canopies: dense for the cereal and more open for the other crops. The specific activity (Bq C-14 kg⁻¹ C) of CO₂ in the air canopy $A_{\text{volatilized}}^{\text{C14}}$ is then equal to the ratio of C-14 volatilised from the soil and the net photosynthesis flux, P (kg C m⁻² d⁻¹):

$$A_{\text{volatilized}}^{\text{C14}} = I_{\text{respiratory_recycling}} \left[Q_{\text{volatilized}}^{\text{C14}} / P \right] \quad (37)$$

P can be calculated from the dry matter production rate and the carbon content in the considered plant.

3.2.3 C-14 plant uptake

In SA_Carbon14, the absorption of C as CO₂ during photosynthesis is considered as the key route of C uptake by the plant, with all other sources being negligible in comparison. The decomposition of organic matter in the soil, combined with root and soil organism respiration mean that the gas-filled soil pore space is often enriched in CO₂ relative to the free above-ground atmosphere. The air in the

plant canopy is a mixture of air from the soil pore space and the free atmosphere, and so the plant C-14 specific activity is also a mixture of the two sources. However, SA_Carbon14 deals exclusively with contaminated water, so that the free atmosphere is not considered a source. Any cosmogenic C-14 in the atmosphere would contribute to background dose from C-14, rather than to the dose attributed to any technical facilities.

The specific activity of C-14 in plants (Bq kg C⁻¹) depends on existing C-14 in the plant and volatilised C-14:

$$A_{crop}^{C14,spe}(t_n) = A_{crop}^{C14,spe}(t_{n-1})e^{-(g/B)(\Delta t)} + A_{volatilised}^{C14}(t_n)(1 - e^{-(g/B)(\Delta t)}) \quad (38)$$

Here g is the plant dry matter production rate (kg m⁻² d⁻¹) and B is the total plant dry matter (kg m⁻²) at time t assuming that the growth function is linear. The ratio g/B is the same as a relative growth rate (RGR) and is the fraction of total plant dry matter that is produced per unit time. This growth rate is used to take account of the C-14 dilution in the plant that would follow as a result of plant growth. If the gross photosynthetic rate is used to estimate the flux of carbon used by the crop, then the respiration of roots and shoots should also be taken into account to describe correctly the turnover of carbon in the crop. Here only the net photosynthesis via the use of the plant dry matter production rate is used; therefore respiration is implicitly taken into account.

Over longer time periods (larger values of Δt) and for faster growth rates (large values of P/B) the contribution of previous plant specific activity lessens. The other fraction, defined by $(1 - e^{-(P/B)(\Delta t)})$, has a specific activity that reflects the specific activity of labile C volatilised from the soil.

The plant dry matter production rate (kg m⁻² d⁻¹) is equal to:

$$g_{crop} = \frac{B_{harv,crop}}{t_{harvest,crop} - t_{germ,i}} \quad (39)$$

Here $B_{harv,crop}$ is the biomass of the crop at harvest and t_{germ} and $t_{harvest}$ are the dates of germination and harvest in the year. The value of $A_{crop}^{C14,spe}$ corresponds only to crops from land irrigated with contaminated water. In both the human and animal diets, there will be plant-based foods that are not contaminated. The activity in crop (A_{crop}^{14C} , Bq. kg⁻¹ dry weight) is then calculated according to Equation 40.

$$A_{crop}^{14C} = A_{crop}^{14C,spe} \cdot C_{crop}^C \quad (40)$$

3.3 SIMPLIFIED ENHANCED RIMERS

The NDA RWMD funded the development of the enhanced RIMERS model to represent the transport of gaseous forms of C-14 in the accessible environment [Thorne, 2005, 2006]. The enhanced RIMERS model has been implemented in GoldSim^a.

The model comprises 10 compartments, as shown in Figure 7. In this model, standing biomass (1) degrades to either decomposable plant material (DPM, 2) or resistant plant material (RPM, 3). Both DPM and RPM degrade by the same pathways, though at different rates, to generate microbial biomass in soil (BIO, 4), physically stabilised organic matter (POM, 5) and chemically stabilised organic matter (COM, 6), as well as CO₂ in soil solution (7). The microbial biomass also respire CO₂, and both physically and chemically stabilised organic matter can eventually degrade to yield CO₂.

^a Details of GoldSim are available at GoldSim Technology Group LLC, 22516 SE 64th Place, Suite 110, Issaquah, Washington 98027-5379, USA, see also <http://www.goldsim.com/>.

When CH₄ enters the soil system, it is available to soil microbes and is metabolised by them to CO₂. Carbon dioxide in soil solution (7) exchanges with the soil atmosphere (8), which in turn exchanges with the below-canopy atmosphere (9). Carbon dioxide in the below-canopy atmosphere is available for uptake by plants and incorporation in tissues through photosynthesis. Additionally, it may exchange with the above-canopy atmosphere (10) and be rapidly advected away by the wind; there is also some (return) exchange with the below-canopy atmosphere.

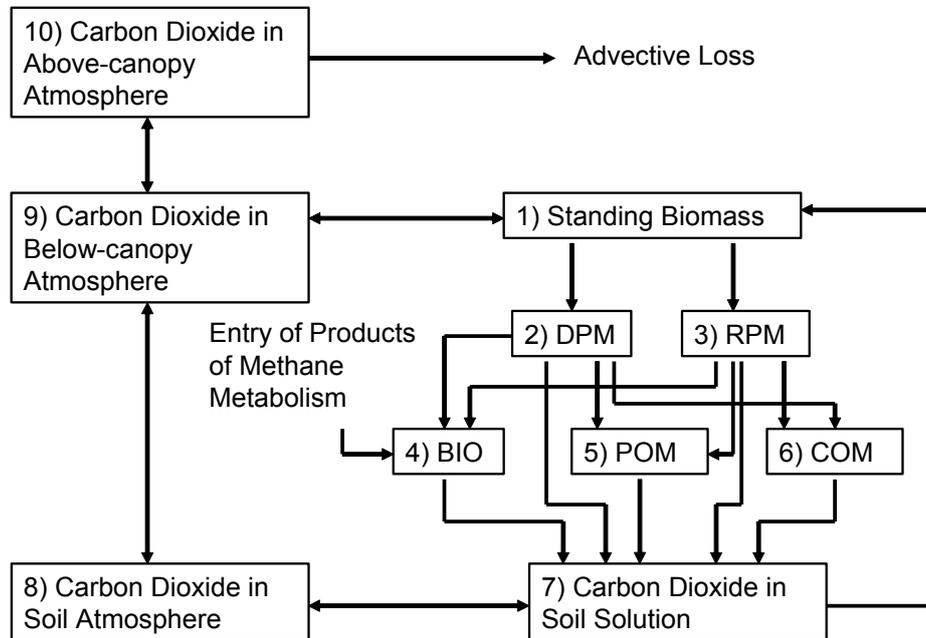


Figure 7 Compartmental Structure of the RIMERS Model (double-headed arrows represent bi-directional exchanges)

Full details of the enhanced RIMERS model can be found in Thorne [2005, 2006]. In this study, a simplified version of the model has been used; the simplified model is shown in Figure 8. This model treats the rapidly exchanging soil solution and soil atmosphere as being in equilibrium and considers only atmospheric transfers, i.e. it neglects the effects of plant uptake in depleting the soil atmosphere and below-canopy atmosphere. The volumes of the compartments are denoted by V (m³) and the flow rates between compartments by k_{ij} (m³ d⁻¹), where the transfer is from compartment i to compartment j . The rate coefficients for transfers between compartments, denoted λ_{ij} are calculated as k_{ij}/V_i .

Defining the activity contents of the compartments as q_i , the relevant equations are:

$$\frac{dq_1}{dt} = I + \lambda_{21}q_2 - \lambda_{12}q_1 \quad (41)$$

$$\frac{dq_2}{dt} = \lambda_{12}q_1 - \lambda_{21}q_2 - \lambda_{23}q_2 + \lambda_{32}q_3 \quad (42)$$

$$\frac{dq_3}{dt} = \lambda_{23}q_2 - \lambda_{32}q_3 - \lambda_{3L}q_3 \quad (43)$$

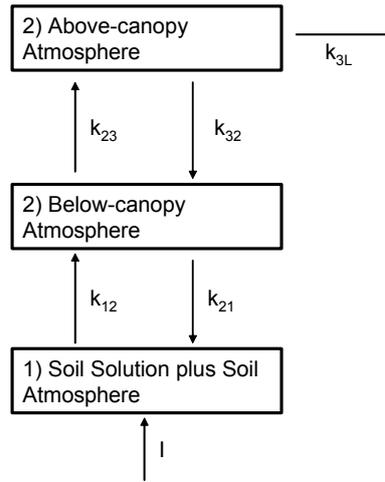


Figure 8 Structure of the Simplified Version of the Enhanced RIMERS Model

This model is readily solved analytically at equilibrium (which occurs rapidly) to give:

$$q_3 = I/\lambda_{3L} \quad (44)$$

$$q_2 = (\lambda_{3L} + \lambda_{32})q_3/\lambda_{23} \quad (45)$$

$$q_1 = (I + \lambda_{21}q_2)/\lambda_{23} \quad (46)$$

Here q_i (Bq) is the content of compartment i , I (Bq d^{-1}) is the rate of input into soil and λ_{ij} (d^{-1}) is the rate coefficient from compartment i to compartment j or as loss ($j = L$). Values of λ_{ij} are calculated as k_{ij}/V_i , where k_{ij} ($\text{m}^3 \text{d}^{-1}$) is the flow of CO_2 from compartment i to compartment j or to loss (L), and V_i (m^3) is the volume of CO_2 in the compartment. Note that volumes and flows are defined at standard temperature and pressure, and the model is defined for a reference area of 1 m^2 .

The concentration of C-14 in each compartment, C_i (Bq m^{-3}) is given by $C_i = q_i/V_i$.

It also follows from balance considerations that $k_{21} = k_{12}$ and that $k_{32} = k_{23} + \delta$, where δ is an allowance for the net uptake of CO_2 by plants (photosynthesis less respiration).

The concentration of C-14 in plants, C_p (Bq m^{-3}), is a weighted average of C_1 and C_2 . This can be written $C_p = \alpha C_1 + (1 - \alpha)C_2$, where α is the fraction of plant carbon obtained by root uptake. C_p can be converted to a mass basis by noting that, for plants with a dry weight to wet weight ratio of 0.1 and taking the dry weight component to be mainly cellulose, 1 kg (fresh weight) plants corresponds to 0.04 kg C. At STP, this corresponds to a volume of CO_2 of $(40/12) \times 0.0224 = 7.467\text{E-}2 \text{ m}^3$. Thus, defining $C_{p\text{-mass}}$ (Bq per kg (f.w.)):

$$C_{p\text{-mass}} = 7.46710^{-2} \times C_p \quad (47)$$

Sensitivity studies using the model were run with $I = 1 \text{ Bq m}^{-2} \text{ d}^{-1}$ and with compartment volumes V_1 , V_2 and V_3 of $5.6\text{E-}4$, $1.65\text{E-}4$ and $1.65\text{E-}3 \text{ m}^3$, respectively. The value of δ was taken as $2\text{E-}4 \text{ m}^3 \text{ d}^{-1}$, this being 50% of the net photosynthetic uptake in the reference model to allow for losses in respiration. The value of α was taken as 0.0215. Results from these sensitivity studies are shown in Figure 9, where $C_{p\text{-mass}}$ is denoted C_p' .

When both k_{12} and k_{23} are small, the value of k_{3L} does not substantially affect $C_{p\text{-mass}}$, provided that it is sufficiently large. This is because the important processes are exchanges within the soil

atmosphere and canopy and removal from the above-canopy atmosphere at any reasonable rate effectively ensures that negligible reflux occurs from the above-canopy atmosphere to the below-canopy atmosphere. However, when k_{23} is large, there is an effect of k_{3L} and this effect is greatest when k_{12} is also large.

In the context of the sensitivity study described above, it is relevant to examine plausible transfer rates between the below-canopy and above-canopy atmosphere, as these are a primary determinant of the results obtained. This was done using an approach described in Section 5 of Limer et al [2008b]. There it was argued that transport across the canopy boundary can be treated as a diffusion-like process, where the diffusion coefficient D ($\text{m}^2 \text{s}^{-1}$) can be represented by:

$$\begin{aligned} D &= D_{air} & z \leq z_d \\ &= D_{air} + k u_* (z - z_d) & z > z_d \end{aligned} \quad (48)$$

where k is von Karman's constant, u_* is the friction velocity that depends on the wind speed away from the surface and the surface roughness length, which is in turn related to the height of the roughness elements (here, the plant canopy), z is the height above the surface (determined by the compartment geometry), z_d is the height above the surface where the wind speed is taken to fall to zero (the zero displacement plane), and D_{air} is the diffusion coefficient in air. The zero displacement plane is assumed to lie within the canopy of the 'dominant' plant species that determines the wind profile close to the surface.

Diffusion-like processes can be considered at the relevant interfaces of compartments containing gas. The relevant transport resistance between compartments i and j and the associated diffusion-like transfer rate between compartments can be represented in a number of different ways; there is no unique representation. In the analysis reported here an expression for the transport resistance that takes account of variations in the diffusion coefficient in the donor and receptor compartments in the direction of transport is used.

$$\Omega^{ij} = \frac{1}{2A} \int_{\zeta-h^i}^{\zeta+h^j} \frac{dz}{D(z)} \quad (49)$$

$$\lambda^{ij} = \frac{1}{V^i \Omega^{ij}} \quad (50)$$

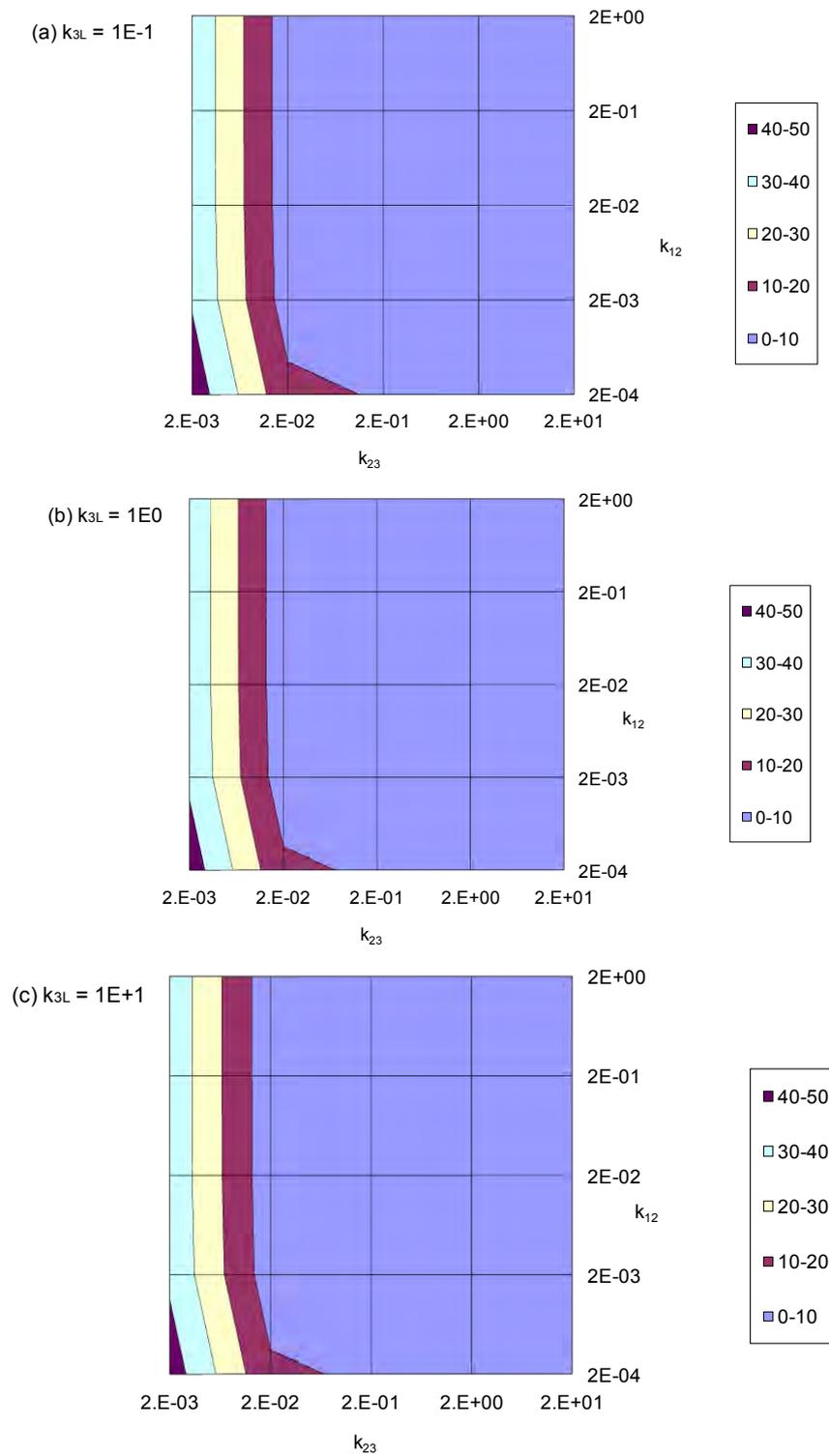


Figure 9 Sensitivity studies with the simplified enhanced RIMERS model

Here Ω^j ($s\ m^{-1}$) is diffusive transport resistance, A (m^2) is the interface area between the compartments in the direction of transport (here the z-axis), ζ represents the position of the interface, h (m) is the length of the compartment in the direction of transport, λ_{ij} (s^{-1}) is the rate coefficient and

V_i (m^3) is the donor compartment volume. If constant diffusion coefficients are employed in each compartment one has:

$$\begin{aligned}\Omega^{ij} &= \frac{1}{2A} \left(\frac{h^i}{D^i} + \frac{h^j}{D^j} \right) \\ &= \frac{(h^i + h^j)}{2AD^{ij}} \quad \text{if } D^{ij} = D^i = D^j\end{aligned}\tag{51}$$

Using this form for the effective diffusion coefficient one obtains the following expression for the diffusive-like transport resistance when the upper limit of integration is above the zero displacement height:

$$\begin{aligned}\Omega^{ij} &= \frac{1}{2A} \left[\frac{z_d - z_L}{D_{air}} + \frac{\ln\left[1 + \frac{k u_* (z_U - z_d)}{D_{air}}\right]}{k u_*} \right] \quad z_d > z_L \\ &= \frac{1}{2A} \left[\frac{\ln\left[1 + \frac{k u_* (z_U - z_d)}{D_{air}}\right]}{k u_*} - \frac{\ln\left[1 + \frac{k u_* (z_L - z_d)}{D_{air}}\right]}{k u_*} \right] \quad z_d \leq z_L\end{aligned}\tag{52}$$

In the case that is of interest here, z_L is zero and $z_d > z_L$ by definition. Thus, treating the below-canopy atmosphere as compartment 2 and the above-canopy atmosphere as compartment 3:

$$\Omega^{23} = \frac{1}{2A} \left[\frac{z_d - z_L}{D_{air}} + \frac{\ln\left[1 + \frac{k u_* (z_U - z_d)}{D_{air}}\right]}{k u_*} \right]\tag{53}$$

Also, $V_2 = Az_c$ where z_c is the height of the canopy. Thus:

$$\lambda_{23} = \frac{1}{A \cdot z_c \cdot \Omega^{23}}\tag{54}$$

and the area, A , cancels out.

Following Limer et al [2008b], $k = 0.4$, $D_{air} = 1.4 \cdot 10^{-5} \text{ m}^2 \text{ s}^{-1}$ and $u^* = 0.2 \text{ m s}^{-1}$, results for various z_U , z_d and z_c are as shown in Table 9.

Table 9 Values of λ_{23} obtained by physically based modelling of diffusion

z_u	z_d	z_c	$\lambda_{23} (d^{-1})$
10	0.4	0.5	12.0
10	0.3	0.5	15.9
10	0.2	0.5	23.7
10	0.2	0.3	39.1
10	0.1	0.3	75.8
5	0.4	0.5	12.0
5	0.3	0.5	15.9
5	0.2	0.5	23.8
5	0.2	0.3	39.2
5	0.1	0.3	76.1

As expected, the height of free air above the canopy makes very little difference to the results obtained. With values of λ_{23} in the range 12.0 to 76.1 d^{-1} and a value of V_2 of $1.65E-4 m^3$, the corresponding range of values of k_{23} is $1.98E-3$ to $1.26E-2 m^3 d^{-1}$. These rates are toward the bottom end of the range examined in the sensitivity study, as would be expected for a closed canopy, as is represented in this model.

Overall, based on the results reported above, for a release rate of $1 Bq m^{-2} d^{-1}$ of C-14 labelled CO_2 , a reasonable range of estimates of the C-14 concentration in plants is 1 to 50 $Bq kg^{-1}$ (f.w.) with a best estimate of $5 Bq kg^{-1}$ (f.w.). For comparison, Thorne [2005], using the more complex extended-RIMERS model estimated that for an input of $1 Bq m^{-2} d^{-1}$, the C-14 activity in standing biomass is $127 Bq kg^{-1} C$ (range 18 to $1530 Bq kg^{-1} C$). As biomass is taken to comprise 4% carbon [Thorne, 2005], this corresponds to $5.08 Bq kg^{-1}$ (f.w.) (range 0.72 to $61.2 Bq kg^{-1}$ f.w.). Thus, these results are very similar to those from the subsequent sensitivity study reported above.

3.4 AVILA AND PRÖHL MODEL

In 2008, SKB and Posiva jointly commissioned a study to develop a set of simplified models for assessment of human exposures resulting from potential underground releases of C-14 [Avila and Pröhl, 2008]. This study considered models for both terrestrial and aquatic ecosystems. To be consistent with the approach adopted for the release of other radionuclides within the SKB and Posiva safety assessments, the models are based on the specific activity approach, although the specific activity consideration here is very different from that in those models that examine global circulation and time frames of hundreds of years and averages over huge carbon pools [Killough, 1980]. The description of the model presented here is adapted from Avila and Pröhl [2008].

The primary assumption of this model is that all C-14 that is input to the system with irrigation water will be immediately released to the mixing layer where it can be assimilated by the irrigated plants via photosynthesis (Figure 10).

3.4.1 Application to irrigation scenario

The input of C-14 with contaminated irrigation water, $ReleaseRate$ ($Bq y^{-1}$), is calculated as:

$$ReleaseRate = C_{water} V_{irr} Area \quad (55)$$

Here C_{water} is the C-14 concentration in irrigation water ($Bq m^{-3}$), V_{irr} is the volume of irrigation water per unit area used in a year ($m^3 y^{-1}$) and $Area$ is the irrigated area (m^2).

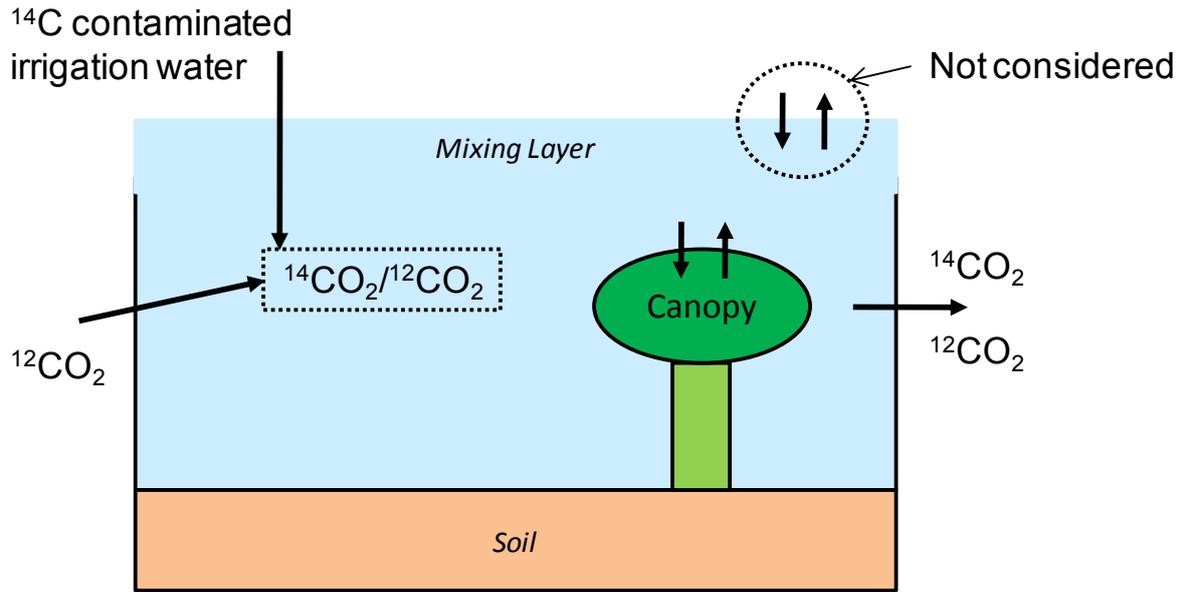


Figure 10 Schematic of the Avila and Pröhl Model (adapted from Avila and Pröhl [2008])

The incorporation of C-14 into the plant via photosynthesis can then be estimated by calculating the excess ¹⁴C/¹²C ratio (specific activity) at equilibrium ($R_{^{14}\text{C}/^{12}\text{C}}$, Bq kg⁻¹ C):

$$R_{^{14}\text{C}/^{12}\text{C}} = \frac{\text{ReleaseRate}}{\text{Area}} = \frac{C_{\text{water}} \cdot V_{\text{irr}}}{h \cdot \lambda_{\text{ex}} \cdot C_{\text{CA}} + \text{NPP}} \quad (56)$$

Here

- h is the mixing height (m);
- λ_{ex} is the air exchange rate in the mixing layer (y⁻¹);
- C_{CA} is the stable C content in the air (kg C m⁻³); and
- NPP is the net primary production in the ecosystem (kg C m⁻² y⁻¹).

The air exchange rate in the mixing layer can be obtained by dividing the wind speed at the vegetation height by the fetch of the affected area:

$$\lambda_{\text{ex}} = \frac{v}{r} \quad (57)$$

Here v is the wind speed at the vegetation height (m y⁻¹) and r is the fetch of the release area (m). Under the assumption that the release area is circular, the fetch r can be determined using the following equation:

$$r = \sqrt{\text{Area}/\pi} \quad (58)$$

The vegetation height, h_{veg} (m), is defined as the height at which photosynthetic assimilation of carbon is most efficient. Empirical values of the wind speed at this height are not usually available, but can be estimated from commonly measured values at the height of 10 m (v_{10} , m y⁻¹), assuming an exponential wind profile and a vegetation-specific roughness length (z_d , m) [Seinfeld, 1986]. The

roughness length is defined as the height at which the wind speed becomes zero when the wind profile above the canopy is extrapolated:

$$v = v_{10} \frac{\ln(h_{veg}/z_d)}{\ln(10/z_d)} \quad (59)$$

The C-14 concentration in the plant, C_p (Bq kg⁻¹ f.w.), is then given by:

$$C_p = C_{CP} R_{14C/12C} \quad (60)$$

Here C_{CP} is the stable C content of the plant (kg C kg⁻¹ f.w. plant).

3.4.2 Application to gaseous release scenario

For the gaseous release scenario *ReleaseRate* is simply a pre-defined value (Bq y⁻¹). The specific activity in the plant is then calculated using the following equation:

$$R_{14C/12C} = \frac{E \cdot \frac{ReleaseRate}{Area}}{h \cdot \lambda_{ex} \cdot C_{CA} + NPP} \quad (61)$$

Here E is the effective release fraction – fraction of C-14 released to the mixing layer that occurs in a period when photosynthesis can take place (-). In the application of this model to the scenarios considered in this study, the values of *ReleaseRate* and *Area* set equal to each other to ensure a gaseous release rate of 1 Bq m⁻² y⁻¹.

3.5 THORNE-LIMER MODEL

Previous discussions within BIOPROTA have led to the recognition of limitations with respect to all of the models considered in the earlier part of this study [Limer et al, 2009a]. LLWR has previously used existing models developed for the NDA RWMD to perform calculations relating to the potential impacts of C-14 released in gaseous form from the LLW facility. These models were RIMERS and enhanced RIMERS [Thorne, 2007], and were used in the 2002 Environmental Safety Case (ESC) [McGarry, 2003] and the 2008 submission to the Environment Agency in accordance to Schedule 9 Requirement 2 [Ball et al, 2008]. For the 2011 ESC, the LLWR decided to fund the development of a new C-14 model, which took into account the work within BIOPROTA, so that it would increase the capabilities of the model used in their assessment to addresses the exchange of gas in a soil-plant-atmosphere system using an approach that includes an enhanced and more physically based representation of the processes involved.

In particular, this model (referred to simply as the “Thorne-Limer model” in this study) considers two regions in the above-ground atmosphere and utilises concepts from the field of micrometeorology to describe the exchange of air between these regions and losses from the area of interest. The lower layer only experiences molecular diffusion processes in relation to the movement of molecules of CO₂, whereas the upper layer experiences some degree of turbulent mixing as a result of winds which flow over the area of interest. The thickness of these layers, and the degree of plant uptake of carbon from them, is dependent upon the canopy density, which affects the light intensity and thus the rate of photosynthetic uptake of carbon in the canopy profile. Below, a description of the plant uptake model is summarised from Limer et al [2011].

3.5.1 C-14 in soil

C-14 bearing gas is assumed to enter the soil in the forms of CH₄ and CO₂. Any fluxes of CH₄ are not available directly to plants, so they have to be converted to CO₂ fluxes by microbial metabolism in the

soil zone. The degree of metabolism may be affected by the mass flux of CH₄. Thus, the total plant available fluxes of C-14 labelled and bulk CO₂, Q_p (Bq s⁻¹) and G_p (kg [C] s⁻¹) are given by:

$$Q_p = Q_c + \mu \cdot Q_m \quad (62)$$

$$G_p = G_c + \mu \cdot G_m \quad (63)$$

The conversion efficiency μ lies in the range [0, 1] and is determined by the flow of CH₄ per unit area. This is represented as $\mu(\varphi)$, where φ (kg[C] m⁻² s⁻¹) is defined as:

$$\varphi = \frac{G_m}{w \cdot X} \quad (64)$$

The functional form of $\mu(\varphi)$ is not well defined, but it should be ~ 1 at small values of φ and decline to zero when the microbial capacity of the soil to metabolise CH₄ is exceeded. A simple parameterisation is:

$$\mu(\varphi) = \min(1, \exp\{-k \cdot (\varphi - a)\}) \quad (65)$$

where k and a are calibration coefficients. Thus, for $\varphi \leq a$, $\mu(\varphi) = 1$ and for $\varphi > a$, $\mu(\varphi)$ declines exponentially to zero.

3.5.2 C-14 in the plant-canopy atmosphere

From the studies undertaken in support of BIOPROTA [Limer et al, 2009a], it was shown that a single pass model is appropriate to representing the flow of C-14 labelled CO₂ through the plant canopy and its uptake in photosynthesis. However, in the model used, the canopy was treated as only a single layer structure and horizontal transport was neglected. In the model described here, a multi-layer system is adopted. This allows more complex canopy structures (both open and closed) to be represented. Furthermore, the layer heights are defined identically in each area of the model, so that horizontal transport can be represented as transfers between corresponding layers of adjacent compartments. The geometry adopted for a single model area is shown in Figure 11.

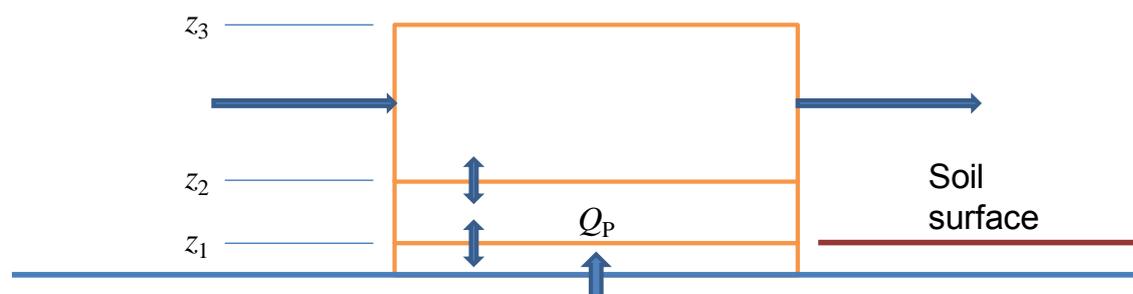


Figure 11 Vertical Structure of the Biosphere Model

The vertical structure shown in Figure 11 distinguishes the base of the model from the soil surface. Thus, the region [0, z₁] corresponds to soil solution plus soil atmosphere. The height of this compartment corresponds to the thickness of the soil plus subsoil of the cap. The height of the second layer is chosen to be equal to the height where turbulent mixing in the plant canopy commences, and thus is plant specific. Layer 3 will contain the top of the plant canopy; its height is fixed for all plant types to ensure that there is a minimum of a 10 m thickness of free air that overlies the canopy.

Air exchanges can occur between the layers and also a horizontal flow of air can occur both within and above the canopy. Note that a horizontal flow of air is not expected in the soil zone. However, to

make the formulation of the model identical for each layer, such a flow is included in the model formulation, though it would be usual to set the value to zero.

It is noted that the movement of C-14 labelled CO₂ through the canopy will be fairly rapid. Therefore, variations in wind direction do not need to be considered. Furthermore, it is only the component of the wind direction along the line of the 2D section that is relevant. In a 2D model the transverse component does not move the activity out of the region of interest. As the component of wind along the section can be in either sense, it is recommended that separate calculations be undertaken for the situations in which the wind blows from left to right along the line of the section and when it blows from right to left and that the arithmetic mean of the two results should be used for assessment purposes. It may also be appropriate to perform calculations for different wind speeds.

Computationally, the model is fully defined by the following ordinary differential equations. Note that these equations are specified only for C-14. The stable carbon contents of the various layers are considered to be input parameters (see below).

For the lowest layer, the governing equation is:

$$w \cdot X \cdot \theta \cdot \frac{dC_1}{dt} = Q_p - U_1 \cdot z_1 \cdot w \cdot C_1 + w \cdot X \cdot V_{2,1} \cdot C_2 - w \cdot X \cdot V_{1,2} \cdot C_2 \quad (66)$$

The left hand side corresponds to the rate of change of the activity content of the layer, i.e. C₁ (Bq m⁻³) is the activity concentration in the soil atmosphere and θ (-) is the porosity of the soil (the volumetric concentrations in soil atmosphere and soil solution are taken as identical, but different concentrations can be accommodated by defining an effective porosity).

The first term on the right hand side of the equation, Q_p, is the vertical flux of plant available C-14 labelled gas from the base of the model. The second term corresponds to advective horizontal transport out of the layer, with U (m s⁻¹) representing the wind velocity. The final two terms on the right correspond to vertical transport into and out of the overlying layer. This is represented by the concept of an effective velocity V (m s⁻¹), though, in practice, the process is treated diffusively (see below).

For layer 2, though vertical exchanges from both the layer below and the layer above have to be represented, porosity does not apply and there is no explicit source term. Thus, the relevant equation for layer 2 is:

$$w \cdot X \cdot (z_2 - z_1) \cdot \frac{dC_2}{dt} = -U_2 \cdot (z_2 - z_1) \cdot w \cdot C_2 + w \cdot X \cdot V_{1,2} \cdot C_1 + w \cdot X \cdot V_{3,2} \cdot C_3 - w \cdot X \cdot V_{2,1} \cdot C_2 - w \cdot X \cdot V_{2,3} \cdot C_2 \quad (67)$$

For the thick topmost layer, the relevant equation is:

$$w \cdot X \cdot (z_3 - z_2) \cdot \frac{dC_3}{dt} = -U_3 \cdot (z_3 - z_2) \cdot w \cdot C_3 + w \cdot X \cdot V_{2,3} \cdot C_2 - w \cdot X \cdot V_{3,2} \cdot C_3 \quad (68)$$

All the terms in these equations are well-defined, except for V and U. These are discussed below.

Representation of vertical air flow

Transport vertically in and through the canopy is treated as a diffusion-like process, where the diffusion coefficient D (m² s⁻¹) is represented by:

$$\begin{aligned}
D &= D_{air} & z \leq z_d \\
&= D_{air} + k u_* (z - z_d) & z > z_d
\end{aligned} \tag{69}$$

where k is von Karman's constant, u_* is the friction velocity that depends on the wind speed away from the surface and the surface roughness length, which is in turn related to the height of the roughness elements (here the plant canopy), z is the height above the surface (determined by the compartment geometry), z_d is the height above the surface where the wind speed is taken to fall to zero (the zero displacement plane), and D_{air} is the diffusion coefficient in air.

The zero displacement plane is assumed to lie within the canopy of the 'dominant' plant species that determines the wind profile close to the surface, i.e. $z_d < z_c$, where z_c (m) is the canopy height.

The relevant transport resistance between layers j and $j+1$ and the associated diffusion-like transfer rate between compartments can be represented in a number of different ways; there is no unique representation. Here, an expression is used for the transport resistance that takes account of variations in the diffusion coefficient in the donor and receptor compartments in the direction of transport. Note that the subscript i indicating the specific region is not relevant to this analysis in the vertical direction and has been omitted.

$$\Omega^{j,j+1} = \frac{1}{2} \int_{\zeta+h^j}^{\zeta+h^{j+1}} \frac{dz}{D(z)} \tag{70}$$

Here $\Omega^{j,j+1}$ ($s \text{ m}^{-1}$) is diffusive transport resistance, ζ represents the position of the interface, and h (m) is the length of the compartment in the direction of transport.

If constant diffusion coefficients are employed in each compartment one has:

$$\Omega^{j,j+1} = \frac{1}{2} \left(\frac{h^j}{D^j} + \frac{h^{j+1}}{D^{j+1}} \right) \tag{71}$$

Finally, it remains to relate $\Omega^{j,j+1}$ to the required velocity of transfer. As wX_j is the area between the compartments, the rate constant for flow between them is $1/h^j \Omega^{j,j+1} s^{-1}$. This rate constant can also be written as $wX_j V_{j,j+1} / wX_j h^j = V_{j,j+1} / h^j$. Thus:

$$V_{j,j+1} = \frac{1}{\Omega^{j,j+1}} \tag{72}$$

Representation of horizontal velocity

Within the plant canopy, it is appropriate to take $U = u_*$ above the zero displacement plane and to zero below the zero displacement plane. However, above the canopy the air flow regime is different. Here, it is assumed that the wind speed at 10 m, U_{10} (m s^{-1}) is provided as input. The wind speed is then calculated at the mid-point of the layers using:

$$\begin{aligned}
U_j &= U_{10} \left(\frac{u_*}{U_{10}} \right)^{(10-z)/(10-z_c)} & \text{with } z = z_j & \text{ for } z_j > z_c \\
&= u_* & & \text{for } z_d < z_j \leq z_c \\
&= 0 & & \text{for } z_j \leq z_d
\end{aligned} \tag{73}$$

3.5.3 C-14 plant uptake

The equations given in Section 3.5.2 allow the time-dependent concentrations of C-14 in air to be calculated. These values are expressed for the soil atmosphere (C_1) or for the above-soil atmosphere (C_2 and C_3) in units of Bq m^{-3} . The CO_2 concentration in these compartments is taken as $f_j C_s \text{ m}^3 \text{ m}^{-3}$ where C_s is the volumetric fraction of CO_2 in free air (0.00038) and f_j (-) is an enrichment factor (30 to 60 for the soil atmosphere and 1.0 for the above-ground atmosphere). The density of dry air at STP is 1275.4 g m^{-3} and the average molecular weight of air is approximately 29. Therefore, the mass concentration of carbon in each compartment is $0.00038 \times 1275.4 \times 12/29 \times f_j = 0.0002 f_j \text{ kg[C] m}^{-3}$. From this it follows that the specific activity of C-14 in each compartment, S_{j_i} , is $C_j/0.0002 f_j \text{ Bq kg}^{-1}[\text{C}]$. Assuming that plants obtain fractions g_j of their carbon from the various layers j , the specific activity of plant carbon from that area, S_P ($\text{Bq kg}^{-1}[\text{C}]$), is given by:

$$S_P = \sum_j g_j S_j \quad (74)$$

Note that the possibility of some plant carbon being obtained by root uptake is included in this model.

Calculating the g_j

When defining how much carbon the plant takes up from the various compartments, one possible assumption is that the plant takes up carbon uniformly, e.g. if the plant was 1 m high then the plant would take up $1/100^{\text{th}}$ of its carbon per cm height.

However, it is accepted that the dominant means of carbon uptake by the plant is via photosynthesis, i.e. that root uptake can be neglected, and that photosynthesis does not occur at a uniform rate through the plant height. Anten [1997] noted that experimental means alone are not sufficient to be able to characterise the many factors which influence canopy photosynthesis, and that modelling can facilitate a more detailed investigation of the processes involved by using the models to evaluate the empirical data.

One particular factor which will influence C uptake in the canopy is the light intensity. Monsi and Saeki [1953; an English translation is available as Monsi and Saeki, 2005] used an extinction curve to represent the light intensity in the canopy, specifically Beer's Law.

$$I = I_0 \cdot \exp(-K \cdot LAI) \quad (75)$$

Here I is the shaded light intensity under the leaf area index LAI , I_0 is the original incoming light intensity (i.e. the light intensity at the top of the canopy), and K is the extinction coefficient. Using experimental data, Monsi and Saeki [1953] observed that the extinction coefficients K largely fall between 0.3–1.5, and that the K -values of the majority of communities fall into two groups. In particular, they noted that the smaller values of approximately 0.3–0.5 occur almost always in grass formations, and that the larger values of approximately 0.7–1.0 occur in herb or shrub formations. In other words, the light extinction by a given leaf layer is somewhat faster with broad-leaf types than with grass-types.

Using the substitution $v = z_c - z$, it is possible to implement this in the Thorne-Limer model as the following:

$$I(v) = I_0 \cdot \exp\left(-K \cdot LAI \cdot \frac{v}{v+z}\right) \quad (76)$$

$$I(0) = I_0 \quad \text{[top of canopy]}$$

$$I(z_c) = I_0 \cdot \exp(-K \cdot LAI) \quad \text{[bottom of canopy]}$$

Anten [1997] proposed that the light saturated rate of leaf photosynthesis through a plant canopy, P , would follow a similar curve:

$$P = P_0 \cdot \exp\left(-\frac{K_N}{K_{df}} \cdot K_{df} \cdot LAI\right) \quad (77)$$

Here P_0 is the photosynthetic uptake at the top of the canopy, K_N is the coefficient of nitrogen allocation, and K_{df} is the extinction coefficient for diffuse light. It is appropriate to assume that K_{df} is equal to K in Equation 76. Using empirical data, Anten [1997] observed that the ratio between K_N and K_{df} is approximately 0.4, so that Equation 77 simplifies to the following.

$$P = P_0 \cdot \exp(-0.4 \cdot K \cdot LAI) \quad (78)$$

Using the same substitution as previously, one arrives at the following equations. This model means that the degree of carbon uptake will decrease as one moves from the top to the bottom of the canopy; the rapidity of the decrease will depend upon the canopy density.

$$P(v) = P_0 \cdot \exp\left(-0.4 \cdot K \cdot LAI \cdot \frac{v}{v+z}\right) \quad (79)$$

$$P(0) = P_0 \quad \text{[top of canopy]}$$

$$P(z_c) = P_0 \cdot \exp(-0.4 \cdot K \cdot LAI) \quad \text{[bottom of canopy]}$$

K and LAI can be obtained from experimental literature. In the Thorne-Limer model interest lies not in the absolute rate of photosynthetic uptake, but rather the fractional C-14 uptake per unit height through the canopy. To determine this it is necessary to integrate P over the canopy profile, i.e. over the range $[z_1, z_c]$. The value for P_0 can then be determined by normalising $P(v)$ to integrate to 1 over the range $[z_1, z_c]$.

$$G(P) = \int_{z_1}^{z_c} P(z) = 1$$

$$= \int_{z_1}^{z_c} P_0 \cdot \exp\left[-K_2(z_c - z) \frac{LAI}{z_c}\right]$$

$$= \frac{P_0 \cdot z_c}{0.4 \cdot K \cdot LAI} \cdot [1 - \exp(-0.4 \cdot K \cdot LAI)] \quad (80)$$

Solving this equation allows P_0 to be derived. It is then possible to determine the fraction of carbon taken up by the plants from each compartment by solving Equation 65 for each compartment. Note that the limits of the integral need to be constrained to ensure that any fraction of the compartment that is above the plant canopy height is not considered for plant uptake of carbon.

3.6 COMPARISON OF MATHEMATICAL REPRESENTATION OF C-14 DYNAMICS IN THE CANOPY ATMOSPHERE

In the earlier part of the BIOPROTA C-14 study it was agreed that, given the emphasis the models described in the preceding sections typically place upon the plant uptake of C-14 from the canopy atmosphere, it was necessary to understand the conceptual differences in the various approaches adopted [Limer et al, 2009a]. The purpose of this section is to summarise those differences.

As noted in Section 3.1, in the Andra model AquaC_14, in order to estimate the contamination via incorporation of $^{14}\text{CO}_2$ during photosynthesis, information on the volume concentration in the canopy of plants (Bq m^{-3}) is required. In the Andra model this depends on the concentration of C-14 in the soil, the soil density and the rate of degassing. A geometric factor related to the wind direction, the wind speed and a parameter relating the size and volume of the canopy ("fetch") must also be taken into account.

As noted in Section 3.2, in the EDF model SA_Carbon14, two approaches are adopted to calculate the atmospheric C-14 concentration. Both methods are based upon empirical data in which consideration has been given to the crop height, the wind speed and also the lateral extent of the field.

In the Enhanced RIMERS model (Section 3.3), the calculated plant canopy C-14 concentration depends on air exchange rates between the soil atmosphere, the plant canopy atmosphere and the above-canopy atmosphere. These exchange rates are calculated based on the wind velocity within the canopy. Clearance of the above-canopy atmosphere is by downwind transport, as in the Avila and Pröhl model, but this does not much affect C-14 concentrations within the canopy, so these are essentially independent of the linear dimensions of the release area.

As noted in Section 3.4, in the Avila and Pröhl model the calculated atmospheric C-14 concentration depends upon the crop height and the height of the mixing layer, the wind speed at the top of the mixing layer and also at the vegetation height, the size of the area of consideration, the stable carbon content of the air and also the net primary productivity of the ecosystem.

In the Thorne-Limer model (Section 3.5), the calculated atmospheric C-14 concentration depends upon the crop height, the height of the zero velocity layer (z_d) and the wind speed. The calculated atmospheric C-14 concentrations are independent of the field size.

The dependency of the calculated plant canopy atmosphere concentration on the wind speed, crop height, and the definition of fetch used in each model, is summarised in Table 10. From this it is clear that the term “fetch” is not really applicable for the Andra model, AquaC_14, as it has a very different mathematical representation than the classical fetch term.

Table 10 Dependency of the Calculated Plant Canopy Atmosphere Concentration on the Wind Speed, Crop Height, and the Definition of Fetch in Each Model

Model	Factors taken into account for calculating plant canopy atmosphere C-14 concentration				
	Wind speed	Crop height	“Fetch”	Fetch definition	Fetch units
AquaC_14	Yes	No	Yes	$\frac{length \times height}{volume}$	m ⁻¹
SA_Carbon14 (Sheppard approach)	Yes	Yes	Yes	Length parallel to the wind direction [Amiro et al, 1991]	m
SA_Carbon14 (Respiratory approach)	The canopy concentration depends upon a recycling index, which itself was determined from experiments in which the crop height and field size varied. Thus there is some implicit dependence upon the crop height and a parameter relating to the field size.				
Simplified Enhanced RIMERS	Parameter values set using similar arguments to those adopted in the LLWR model. As the model is one dimensional, the effects of fetch are not represented.				
Avila and Pröhl	Yes	Yes	Yes	$\sqrt{Area/\pi}$	m
Thorne-Limer	Yes	Yes	No	N/A	N/A

3.7 IMPLEMENTATION OF THE MODELS FOR THIS STUDY

In the quantitative comparison aspect of this study the models were either implemented by participants belonging to one of the sponsoring organisations, or were implemented on behalf of a sponsoring organisation by the project technical support team (TST). This is summarised in Table 11.

Table 11 Responsibility for the Implementation of the Models in this Study

	“Owner” organisation	Responsibility for calculations	Software used
Aqua_C14	Andra	Andra	MoM
SA_Carbon14	EDF	EDF	Ecolego
Simplified Enhanced RIMERS	NDA RWMD	TST (author of the model)	Analytical calculations
Avila and Pröhl	SKB and Posiva	TST	Excel
Thorne-Limer	LLW Repository Ltd.	TST (authors of the model)	Excel and Matlab

4. FEP AUDIT OF MODELS USED IN THIS PROJECT

In this section the individual models are audited against the features (Section 4.1) and events and processes (Section 4.2) described in Section 2. Such an audit might be considered as an initial stage in assessing the structure of these models, which is appropriate given the nature of the open question being addressed in this study. For more in-depth safety assessments, this initial audit would form one stage of the iterative process referred to in Section 2. Examples of more detailed analyses of safety assessment models and the associated interaction matrices include SKB SR-Site assessment [SKB, 2010] and NWMO's deep geological repository assessment [Quintessa et al, 2011a,b].

4.1 FEATURES

Twelve features (or CMO's) were identified in section 2.2.2. These are listed in Table 12. It was considered that the soil could be further broken down into two layers: an upper layer (UL) which is subject to ploughing, and a lower layer (LL) which is not disturbed by human activity. Soil CMO's are similar for both layers. The features used in the models described in Section 2 are audited in Table 12. Where a feature is considered explicitly in the model this is denoted by "EXP", and where a FEP is implicitly considered "imp" is used. None of the models considered in this study explicitly represent mycorrhizae.

Table 12 Conceptual model objects (compartment) audit

Feature	Model				
	AquaC_14	SA_Carbon14	Enhanced RIMERS	Avila and Pröhl	Thorne-Limer
Source	EXP	EXP	EXP	EXP	EXP
Soil water	imp	EXP	EXP		
Soil solids – recalcitrant		EXP	EXP		
Soil solids – labile		EXP	EXP		
Soil gas	imp	imp	EXP		EXP
Soil microbes			imp		
Mycorrhizae					
Plant canopy atmosphere below z_d	EXP	imp	EXP	imp	EXP
Plant canopy atmosphere above z_d		imp	EXP	imp	EXP
Below-ground plant material	imp	imp	imp		imp
Above-ground plant material	EXP	imp	EXP	imp	imp
Sink		EXP	EXP	imp	EXP

4.2 EVENTS AND PROCESSES

The events and processes included in the models are audited against those identified in Section 2.2.3. Results of the audit are presented in Table 13. Where an event or process is considered explicitly in the model this is denoted by "EXP", and where a FEP is implicitly considered "imp" is used.

The following events and processes considered to be of potential relevance to C-14 in the FEP analysis part of this project, which are not included in any of the models in this study are: aerenchyma, bioturbation, capillary rise, environmental change, the use of soil additives and weathering.

Table 13 Events and Processes Audit

Events / Processes	Model				
	AquaC_14	SA_Carbon14	Enhanced RIMERS	Avila and Pröhl	Thorne-Limer
Aerenchyma					
Bioturbation					
Capillary rise					
Cropping loss (plants & animals)	imp	EXP			
Death and decomposition			EXP		
Degassing / volatilization	EXP	EXP	EXP		EXP
Diffusion			imp		EXP
Discharge from below (upwelling)	EXP	EXP	EXP		
Environmental change					
Evaporation	EXP				
Soil additives					
Foliar uptake and Photosynthesis	EXP	EXP	EXP		EXP
Gas sorption					
Infiltration	EXP	EXP			
Ingestion	EXP	EXP		EXP	EXP
Inhalation	EXP	EXP		EXP	EXP
Interception	EXP				
Irrigation	EXP	EXP		EXP	
Micro-organism metabolism and assimilation	imp	EXP	EXP		
Percolation	EXP	EXP			
Precipitation	EXP	EXP			
Respiration	EXP	imp	imp		
Root exudation					
Root uptake	EXP		EXP		EXP
Root respiration			imp		
Sorption (adsorption and desorption)	EXP	EXP			
Translocation	EXP				
Weathering					

5. SCENARIO DESCRIPTION

The scenario description presented to participants of the project is reproduced below.

5.1 SOURCE TERM

The source terms given in this section have been selected to facilitate an understanding of the different behaviours of the models in this study. It is acknowledged that in “real” performance assessment calculations the source term is unlikely to be constant in time, nor is the source term likely to be spatially homogenous (if the assessment model considers multi-dimensional landscapes).

5.1.1 Groundwater (irrigation) source

For the groundwater contamination scenario, it was assumed that C-14 in groundwater reaches the rooting zone of agricultural soil via abstraction and irrigation. It was assumed that the concentration of activity in the irrigation water is 1 Bq L^{-1} and plant interception of the irrigation water occurs; the stable carbon concentration of the irrigation water is also fixed. The amounts of irrigation water and how and when in the season irrigation occurs depend upon the climate, and also how each individual model considers irrigation (e.g. monthly irrigation events, or an annual average). Climate data from an inland site in France were used [Météo France, 2008]; this site was the same as used for the BIOPROTA CI-36 study [Bytwerk et al, 2011; Limer et al, 2008a]. From these climate data, crop-specific irrigation rates were then determined according to the methodology detailed in Limer et al [2008a].

The definition of the source water concentration (Bq L^{-1}) and the irrigation rates (m y^{-1}) mean that the calculated soil C-14 concentrations for the irrigation scenario should be expected to be independent of the field size assumed (Section 5.3.2), since the source term has effective units of $\text{Bq m}^{-2} \text{y}^{-1}$.

5.1.2 Gaseous source

For a gas scenario, it was assumed that C-14 bearing gas (CH_4) enters the soil zone at a rate of $1 \text{ Bq m}^2 \text{y}^{-1}$. Reference calculations assumed that the entirety of this flux is converted to CO_2 in the rooting zone, thus making it available for plant uptake after degassing. Variant calculations were also performed using some of the models, which considered only part of the C-14 bearing CH_4 to be oxidised in the soil zone to CO_2 .

Methane oxidation can occur in soils over a range of soil CH_4 concentrations. In a review of the production, oxidation, emission and consumption of CH_4 in soils, Le Mer and Roger [2001] made a distinction between the “high affinity oxidation” which occurs at CH_4 concentrations close to atmospheric levels ($< 12 \text{ ppm}$), and “low affinity oxidation” which occurs in soils with CH_4 concentrations higher than 40 ppm .

High affinity oxidation is reportedly ubiquitous in soils that have not been exposed to high NH_4^+ concentrations [Topp and Hanson, 1991], and results in around 10% of the total soil CH_4 being consumed [Topp and Pattey, 1997]. Methane oxidation in methanogenic environments (rice fields, peat soils, landfills, etc.) is generally considered as low affinity oxidation. It has been observed that greater than 90% of the CH_4 produced in the anaerobic part of rice fields can later be re-oxidised by methanotrophs in the aerobic soil layers [e.g. Frenzel et al, 1992; Oremland and Culbertson, 1992]. On the basis of the above information, the variant calculations which have been performed have assumed that only 9-11% of the CH_4 released into the soil is oxidised. Such a value is applicable only if the release of CH_4 from the waste is small compared to the atmospheric concentration. Indeed, it has been shown that an incubation under CH_4 leads to an enhancement of methanotrophic population in soils (rice fields, forests and grassland).

5.1.3 Fixed concentration assumptions

In addition to the above calculations, a series of calculations which assumed a fixed concentration of C-14 in various compartments were made (soil, plant canopy atmosphere). These fixed concentration assumptions were either $1 \text{ Bq C-14 kg}^{-1} \text{ C}$ in all soil pools, or $1 \text{ Bq kg}^{-1} \text{ C}$ in the canopy atmosphere.

5.2 MEDIA IN WHICH C-14 CONCENTRATIONS WERE CALCULATED

Calculations of the concentration of C-14 in the following crops were required: root vegetables, leafy green vegetables, cereals and fruit. These crops were chosen because the edible parts are very different, i.e. for root vegetables, the edible part is below ground; for lettuce and fruit, the edible part is above ground; whereas for grain it is only the seed body that is eaten. These are the same crops that were used in the BIOPROTA CI-36 study [Bytwerk et al, 2011; Limer et al, 2008a].

The best estimates of concentration of C-14 in the crops at time of harvest in Bq kg⁻¹ carbon were to be calculated, using the first year of the climate data [Météo France, 2008], together with estimates of the potential range of concentrations given uncertainties associated within an average year. If concentrations were thought to be potentially higher in subsequent years of chronic input, a continuous sequence of five years of climate data was provided [Météo France, 2008], repeated if more than 5 years was needed.

In addition to the C-14 concentration in the crops, the C-14 concentration in the top soil (Bq kg⁻¹ C) and plant-canopy atmosphere was also to be calculated. The focus is on the endpoint of C-14 levels in crops because ingestion of food derived from such crops is the dominant exposure pathway for C-14 releases [BIOPROTA, 2005].

5.3 ADDITIONAL SCENARIO INFORMATION

5.3.1 Crop uptake of carbon

In reference calculations it was assumed that the plant obtains 2% of its carbon from soil and 98% from the atmosphere, via photosynthesis or foliar uptake [Amiro et al, 1991]. It was noted that participants needed to be clear in reporting their assumed stable carbon content of the plants; e.g. Sheppard et al [2006b] assume 500 g C kg⁻¹ dry weight for all crops, whereas Penfold and Watkins [1998] have crop-specific values.

A reference crop height of 1 m was assumed. Additional calculations using alternative crop heights were also reported for some of the models.

5.3.2 Site geometry and wind profile

Calculations were carried out assuming a range of field sizes, all of which are square:

- 1 m² (1 m * 1 m)
- 100 m² (10 m * 10 m)
- 10,000 m² (100 m * 100 m)
- 1,000,000 m² (1000 m * 1000 m)

Although it may be argued that the field sizes considered in this study do not relate directly to those that might be used in some safety assessments, they provide a useful means to assess the sensitivity of the models in this study to the site geometry. Participants were asked to report clearly how the geometry is managed within their conceptual model, including use of the term “fetch”. As noted in section 3.6, this is because although the term is widely used, it is not always for the same entity, nor with the same units.

With respect to wind speed, in the November Madrid project workshop it became clear that there are varying assumptions as to the shape of the wind profile and also the values assumed within the canopy. Some consider an average wind speed throughout the entire canopy, others assume the wind speed to be equal to that at the top of the canopy throughout, and some participants assume a wind speed profile within the canopy which depends upon the location within the canopy. It was noted that often the wind speed is given for a certain height (e.g. 10 m) and then scaled to the vegetation height; in this instance a zero plane, z_d (m), is also given, below which there is zero wind speed. The wind at the height of the vegetation is then given by the following equation:

$$v = v_{10} \frac{\ln(h_{veg}/z_d)}{\ln(10/z_d)} \quad (81)$$

The height of z_d is often defined with respect to the height of the vegetation. In some literature sources, z_d is given as a sixth or tenth of the vegetation height [e.g. Amiro et al, 1991]. Allen et al [1998] observed that for a wide range of crops the zero plane displacement height, z_d (m) can be estimated from the canopy height, z_c (m), by assuming that it is two-thirds of the canopy height. For a study of barley in Estonia it was concluded that the calculated z_d could be taken as three-quarters and two-thirds of z_c for a dense and a moderate canopy respectively [Mölder, 1997]. Avila and Pröhl [2008] report z_d for farmlands and forests, which are $\frac{1}{4}$ and $\frac{1}{2}$ of the vegetation height respectively.

Applying this equation to a vegetation of height 1 m, using a range of assumptions about the position of z_d with respect to the canopy height gives a range of wind speeds which might be assumed for a 1 m vegetation height; these values are displayed in Table 14 and Figure 12. It was therefore suggested that participants apply the wind speeds and z_d given in Table 14 for their calculations. Participants were asked to report what assumption(s) they made with respect to the position of z_d with respect to the vegetation height. If alternative crop heights were used, participants were also asked to report the wind speeds they derived.

Table 14 Effect of z_d on Calculated Effective Wind Speed at 1 m

Parameter	Units	Assumptions about z_d				
		Amiro et al [1991]	Avila and Pröhl [2008]	Mölder [1997]	0.67	0.75
z_d^a	m	0.17	0.25	0.5	0.67	0.75
v (1 m)	$m\ s^{-1}$	2.82706	1.87902	1.15689	0.74863	0.55531

^a Associated with a crop height of 1 m

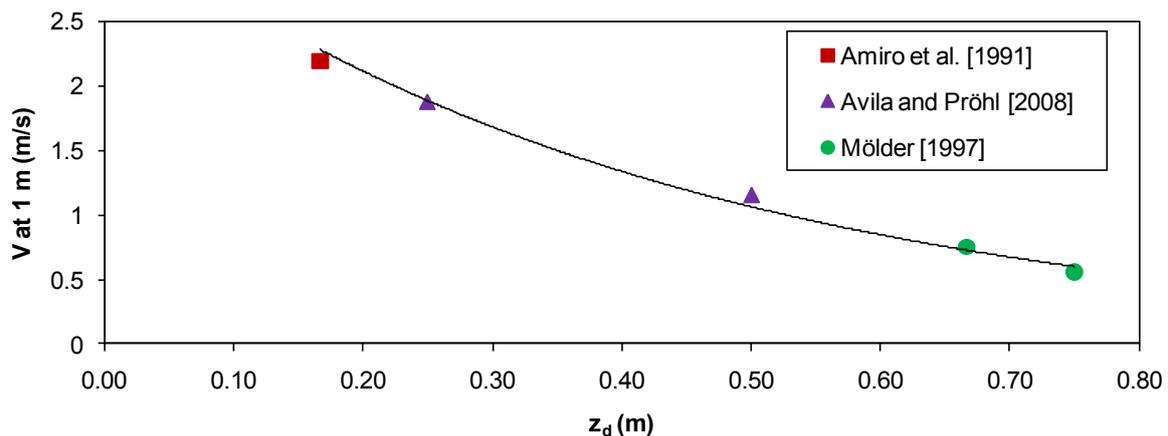


Figure 12 Effect of z_d on Calculated Effective Wind Speed at 1 m

6. DATA AND PARAMETER VALUES

The purpose of this section is to summarise the data used as input to the calculations presented in this report. The first section considers the climate, soil and irrigation data that were used by all models. The subsequent section details the values of the parameters used by each model.

6.1 CLIMATE, SOIL AND IRRIGATION DATA

In this study, climate and soil data from the Bure site in France were used [Andra, 2005; Météo France, 2008]. The soil data are provided in Table 15. Since the EDF model has the most involved method for calculating the irrigation rates for the crops, crop-specific irrigation rates have been provided by EDF (Table 16); Andra used a single irrigation rate for all crops. Crop-specific percolation rates have also been derived using the crop specific irrigation rates from EDF and a methodology devised by Andra (the derivation is explained further in the previous C-14 report, Limer et al [2009a]). The stable carbon content of the air was assumed to be $1.7\text{E-}4 \text{ kg C m}^{-3}$ and the mass fraction of stable carbon in the irrigation water was assumed to be $2\text{E-}5$ (i.e. 0.002 %). The reference assumption for the distribution coefficient of carbon in soil, K_d , is $3.0\text{E-}3 \text{ m}^3 \text{ kg}^{-1}$ [Sheppard et al, 2006b].

Table 15 Properties of Rendosol Soil (Profile “1B”) at Bure [Andra, 2005]

Parameter	Description	Units	Value
d	Soil depth	m	2.5E-1
pH	pH (with water)	-	8.0E0
C_stable	Stable C content	%	2.9E0
Calcaire	Calcium carbonate content	%	3.0E0

Table 16 Climate and Hydrology Data

Input / output	Parameter	Crop	Value (m y^{-1})
Inputs	Precipitation	-	5.94E-1
	Irrigation rate	Leafy vegetables	1.2E-1
		Root vegetables	1.8E-1
		Fruit	8.4E-2
		Cereal	1.44E-1
Outputs	Evapotranspiration	-	7.12E-1
	Percolation Rate	Leafy Vegetables	2.0E-3
		Root vegetables	6.2E-2
		Fruit	0E0
		Cereal	2.6E-2

6.2 AQUAC_14 (ANDRA MODEL) PARAMETERS

Andra carried out four default calculations (with four “fetch” parameter values based on four widths of the fields) with all parameter values as specified in the scenario description and summarised in Table 16.

Table 17 Deterministic Parameter Values Used by Andra

Parameter name	Description	Units	Value
Soil characterisation			
profcult	thickness of soil layer	m	0.25
poro_cult	soil porosity	(-)	0.5
tensolceau	soil water content	(-)	0.5
Climate data			
coefcult	coefficient to scale irrigation needs	(-)	1
teaucult	average irrigation need for all four plant types	L m ⁻² y ⁻¹	132
ETP	evapotranspiration	L m ⁻² y ⁻¹	712
pluie	average precipitation	L m ⁻² y ⁻¹	594
Geometry data			
“fetch”	inverse of the width of the field (100 m), with three supplemental calculations with fetch (width) = 0.1 (1m), 0.001 (100m), and 0.0001 (1000m)	m ⁻¹	0.01
vvent	wind velocity in canopy	m s ⁻¹	2
Contamination			
Cnap	activity concentration in contaminated aquifer irrigation water	Bq L ⁻¹	1
FEIS	fraction of irrigation water reaching the soil	(-)	0.8
fluxgaz	upward flux of ¹⁴ CH ₄ into the soil column	Bq m ⁻² y ⁻¹	1
trans_CH4_CO2_c	degree of transformation of methane to carbon dioxide	(-)	0.11
fCdeAir	fraction of plant carbon coming from the air	(-)	0.98
fCdusol	fraction of plant carbon coming from the soil via root uptake	(-)	0.02
LSA	soil degassing	y ⁻¹	14.6
LPSS	wash-off from leaves	y ⁻¹	3000
LPSP	translocation factor	y ⁻¹	1.8
LPSA	evaporation losses of leaves	y ⁻¹	6000
Stable C			
Ccarbfeuil	stable C content of leafy vegetables	kg kg ⁻¹ _{dry}	0.325
Ccarbfruit	stable C content of fruits	kg kg ⁻¹ _{dry}	0.415
Ccarbcer	stable C content of cereals	kg kg ⁻¹ _{dry}	0.7
Ccarbrc	stable C content of root vegetables	kg kg ⁻¹ _{dry}	0.5
Ccarbair	stable C content of the air	kg m ⁻³	1.70E-04
C_carbSol	stable C content of soil	kg kg ⁻¹ _{dry}	0.029

Furthermore, Andra carried out probabilistic calculations, using the calculations both as an uncertainty and a sensitivity tool. Table 18 lists the parameters, and their distributions, used in the probabilistic approach.

Table 18 Parameter Ranges Used in the Stochastic Calculations by Andra

Parameter name	Units	Deterministic value	Min.	Max.	Description
Climate data					
coefcult	(-)	1	0.47	1	coefficient to scale irrigation needs
teaucult	L m ⁻² y ⁻¹	132	130	850	average irrigation need based on 5 year EDF data sheet
Geometry data					
fetch1	m ⁻¹	0.01	0.1	0.0001	field width varying between 1 and 1000 m
Contamination					
Kdsol	m ³ kg ⁻¹	3E-3	3E-3	4E-2	particle - solution distribution coefficient in soil
Ccarbfeuil	kg kg ⁻¹ _{dry}	0.325	0.1	0.7	stable C content of leafy vegetables
Ccarbfruit	kg kg ⁻¹ _{dry}	0.415	0.1	0.7	stable C content of fruits
Ccarbcer	kg kg ⁻¹ _{dry}	0.7	0.1	0.7	stable C content of cereals
Ccarbrc	kg kg ⁻¹ _{dry}	0.5	0.1	0.7	stable C content of root vegetables
Vvent	m s ⁻¹	2	0	10	wind velocity in canopy
LSA	y ⁻¹	14.6	10	20	soil degassing
LPSS	y ⁻¹	3000	15	3000	wash-off from leaves
LPSP	y ⁻¹	1.8	1.8	36.5	translocation factor
LPSA	y ⁻¹	6000	5000	6000	evaporation losses of leaves

6.3 SA_CARBO14 (EDF) MODEL

For this study, two areas are considered: one where cereals are cultivated and a second representative of a vegetable garden with leafy vegetables, root vegetables and fruits. The crop-independent and crop-dependent parameters used in the deterministic calculations with the EDF model, SA_Carbon14, are given in Table 19 and Table 20. The distributions used for the stochastic calculations performed with this model are given in Table 21.

Table 19 Crop-independent Parameters in the EDF Model, SA_Carbon14

Parameter name	Description	Unit	Value	References
h _{soil}	Thickness of the soil layer	m	0.22	-
ρ _p	Soil (particle) density	kg m ⁻³	2600.0	-
f _f	Fraction of C-14 that becomes fixed when applied to soil	-	0.02	Sheppard et al [2006b]
K ₂	Mineralisation rate of organic matter	month ⁻¹	1.1 E-3	Angers et al [2010]
K ₃	Rate constant of CH ₄ uptake in soil	(-)	0.90	Le Mer and Roger [2001]
K _d	Sorption coefficient	m ³ kg ⁻¹	0.003	Sheppard et al [2006b]
λ _{vol}	Volatilisation from soil rate constant	month ⁻¹	1.2	Sheppard et al [2006b]

Table 20 Crop specific parameters in the EDF model, SA_Carbon14 (Deterministic values)

Parameter name	Description	Units	Crop	Value	References
$B_{\text{harv, crop}}$	Harvest biomass of the crop	kg m^{-2}	Cereals	1.2	Ciffroy et al [2005]
			Leafy Green Vegetables	0.9	
			Fruit	0.775	
			Root Vegetables	0.9	
C_{plant}^C	Stable C content in plant	$\text{g C kg}^{-1}_{\text{dry}}$	All crops	500	Sheppard et al [2006b]
CD	Canopy dilution factor	-	Cereal	0.3 / 0.2	Sheppard et al [2006b]
			Vegetable garden	0.1	
$I_{\text{respiratory_recycling}}$	respiratory recycling index	-	Cereals	0.45	Sternberg [1989]; Striegl and Wickland [2001]; Greaver et al [2005]
			Leafy Green Vegetables / Fruits / Root vegetables	0.12	
K_1	Isohumic coefficient	-	Cereals	0.15 (grain or straw)	Angers et al [2010]
			Leafy Green Vegetables	0.16	
			Fruit	0.16	
			Root Vegetables	0.10	
$\text{Per}_{\text{not_harvested}}$	Percentage of crop not removed	-	Cereals	0.62 (grain) 0.27 (straw)	Angers et al [2010]
			Leafy Green Vegetables	0.60	
			Fruit	0.60	
			Root Vegetables	0.35	
t_{germ}	Date of germination of the plant	month	Cereals	3	Ciffroy et al [2005]
			Leafy Green Vegetables	3, 5.5, 7.5	
			Fruit	4	
			Root Vegetables	3	
t_{rec}	Date of harvest of the plant	month	Cereals	9	Ciffroy et al [2005]
			Leafy Green Vegetables	5, 7, 9	
			Fruit	8	
			Root Vegetables	9	
n_{crop}	Number of harvest cycles in one year	-	Cereals, Fruit, Root vegetables	1	Ciffroy et al [2005]
			Leafy Green Vegetables	3	
$\text{Surf}_{\text{crop}}$	Percentage of cultivated surface which is occupied by a specific crop	-	All Crops	1.0	-
			Leafy Vegetables	0.95	
			Fruit	0.015	
			Root Vegetables	0.035	

Table 21 Parameter Distributions Used for Stochastic Calculations

Parameter	Units	Crop	Best estimate value	Probability density function
h_{soil}	m	-	0.22	Uniform (0.15;0.3)
K_d	$m^3 kg^{-1}$	-	0.003	Log-Normal (0.003;2.3)
f_f	-	-	0.02	Triangular (0;0.02;0.04)
K_3	(-)	-	0.9	Triangular (0;0.90;0.97)
λ_{vol}	month ⁻¹	-	1.2	Triangular (0.12;1.2;12)
$I_{respiratory_recycling}$	-	Cereals	0.45	Triangular (0.33;0.45;0.71)
		Leafy Green Vegetables / Fruits / Root Vegetables	0.12	Triangular (0.03;0.12;0.29)
CD	-	Cereal	0.3	Uniform (0.2-1)
		Vegetable garden	0.1	Uniform (0.1-1)
$B_{harv, crop}$	kg m ⁻²	Cereals	1.2	Uniform (1 ;1.4)
		Leafy Green Vegetables	0.9	Uniform (0.8;1)
		Fruit	0.775	Uniform (0.75;0.8)
		Root Vegetables	0.9	Uniform (0.8;1)

6.4 SIMPLIFIED ENHANCED RIMERS

Compartment volumes and transfer rates between compartments are expressed in terms of the equivalent volume of CO₂ at standard temperature and pressure (m³ and m³ d⁻¹, respectively), assuming a density of gaseous CO₂ of 1.9647 kg m⁻³. The resulting compartment sizes are given in Table 22, whilst the transfer fluxes and resulting rates are given in Table 23.

Table 22 Compartment Size for the Simplified Enhanced RIMERS Model

Name	Volume (m ³)
(1) Soil solution plus soil atmosphere	5.6E-4
(2) Below-canopy atmosphere	1.65E-4
(3) Above-canopy atmosphere	1.65E-3

Table 23 Transfer Fluxes and Rates for the Simplified Enhanced RIMERS Model

Transfer		Rate (m ³ d ⁻¹)	Rate (d ⁻¹)
From	To		
Soil solution and soil atmosphere	Below-canopy atmosphere	1.87E-3	3.34
Below-canopy atmosphere	Soil solution and soil atmosphere	1.87E-3	11.33
Below-canopy atmosphere	Above-canopy atmosphere	1.85E-2	112.10
Above-canopy atmosphere	Below-canopy atmosphere	1.87E-2	11.33
Above-canopy atmosphere	Sink	4.52E-1	273.90

6.5 AVILA AND PRÖHL MODEL

Table 24 and Table 25 contain the crop-independent and crop-dependent parameters used in the calculations using the Avila and Pröhl model. The crop irrigation rates used are as given in Table 16.

Table 24 Crop-independent Parameters Used in Calculations with the Avila and Pröhl Model

Parameter name	Description	Units	Value
<i>A</i>	Area of field	m ²	1, 100, 10000, 1000000
<i>E</i>	Effective release fraction	-	0.5
<i>v</i> ₁₀	Wind speed at 10 m	m s ⁻¹	5
<i>h</i> _{veg}	Vegetation height	m	1.0 (0.3, 0.4 and 2.0 used in variant calculations)
<i>h</i>	Mixing height	m	10
<i>z</i> _d	Roughness length	m	Calculated as a fraction of the vegetation height (one-sixth or two-thirds)

Table 25 Crop-dependent Parameters Used in Calculations with the Avila and Pröhl Model

Parameter name	Description	Units	Crop Type	Value
<i>NPP</i>	Net primary productivity (these are the same as used in SA_Carbon14)	kg C m ⁻² y ⁻¹	Cereals	1.2
			Green Leafy Vegetables	0.9
			Fruit	0.775
			Root Vegetables	0.9

The wind speed at the top of the vegetation was derived using Equation 32; Table 26 shows the results.

Table 26 Derived Wind Speed at the Top of the Vegetation Used with the Avila and Pröhl Model

Assumption about <i>z</i> _d	Wind Speed (m s ⁻¹) at the top of vegetation of height			
	0.3 m	0.4 m	1 m	2 m
<i>z</i> _d = 1/6 vegetation height	1.69E+00	1.79E+00	2.19E+00	2.63E+00
<i>z</i> _d = 2/3 vegetation height	5.18E-01	5.59E-01	7.49E-01	1.01E+00

6.6 THORNE-LIMER MODEL

Table 27 and Table 28 contain the crop-independent and crop-dependent parameters used in the calculations using the Thorne-Limer model. As with the Avila and Pröhl model, the wind speeds were derived using Equation 32, yielding identical results.

Table 27 Crop Independent Parameters Used in Calculations with the Thorne-Limer Model

Parameter name	Description	Units	Value
<i>A</i>	Area of field	m ²	1, 100, 10000, 1000000
<i>v</i> ₁₀	Wind speed at 10 m	m s ⁻¹	5
<i>h</i> _{veg}	Vegetation height	m	1.0 (0.3, 0.4 and 2.0 used in variant calculations)
<i>h</i>	Mixing height	m	10
<i>z</i> _d	Zero displacement height	m	Calculated as a fraction of the vegetation height (one-sixth or two-thirds)
<i>K</i> _r / <i>K</i>	Ratio of <i>K</i> _r to <i>K</i>	-	0.4
<i>d</i>	Depth of soil	m	0.6
<i>θ</i>	Porosity	-	0.5
<i>ρ</i>	Bulk grain density	kg m ⁻³	2600

Table 28 Crop Dependent Parameters Used in Calculations with the Thorne-Limer Model

Parameter name	Description	Units	Value	
			Broad-leafed crops	Narrow-leafed crops
<i>LAI</i>	Crop leaf area index	-	3.62	1.71
<i>K</i>	Extinction coefficient	-	0.85	0.4

7. RESULTS

7.1 AQUAC_14

The model used by Andra, AquaC_14, was applied to both the irrigation and gas upwelling scenarios, with calculations for fields of four different sizes. The system reached equilibrium within one year.

7.1.1 Irrigation scenario

For this scenario, the manner in which the C-14 labelled water enters the system means that the calculated concentration of C-14 in the soil is independent of the field size. For irrigation water contaminated with 1 Bq L^{-1} , and applied at a rate of 132 L y^{-1} , the soil C-14 concentration is $4.32\text{E-}1 \text{ Bq kg}^{-1} \text{ C}$. The activity concentration in the canopy atmosphere increases linearly with length of the field parallel to the wind direction (Figure 13).

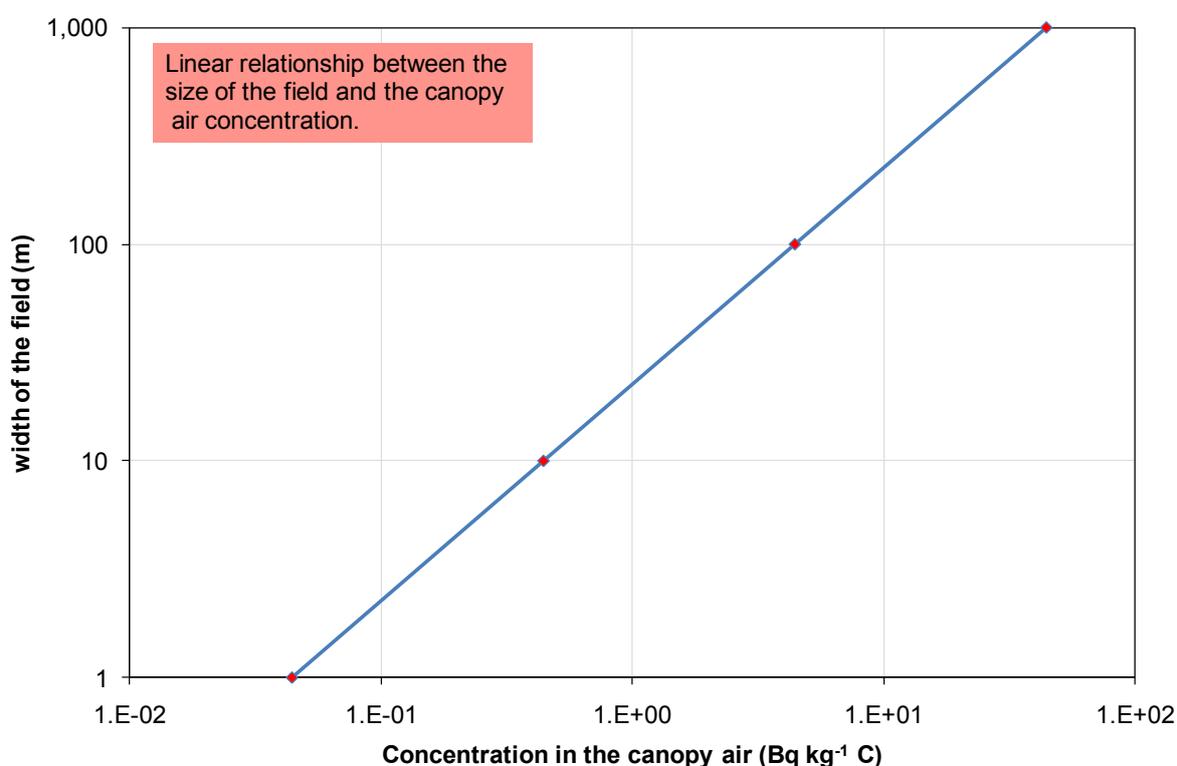


Figure 13 Effect of Field Width on Calculated Canopy Atmosphere C-14 Concentration ($\text{Bq kg}^{-1} \text{ C}$)

However, this perfect linearity is somewhat altered, when depicting the concentration of C-14 in cereals as a function of the width of the field (Figure 14). If the width of the field is small, the diffusive losses are more significant; this is reflected directly by the canopy concentration (Figure 13). It is only for a field width of 1 m, where the impact of the photosynthetic pathway is much reduced due to strong diffusive losses, that the additional pathways of contamination, root uptake and direct interception, known generally to be of much lower significance than the photosynthetic pathways, have an observable input (Figure 14). This can be used as a visual confirmation for the dominance of the photosynthetic contamination pathway and the insignificance of the other pathways in situations of larger field sizes.

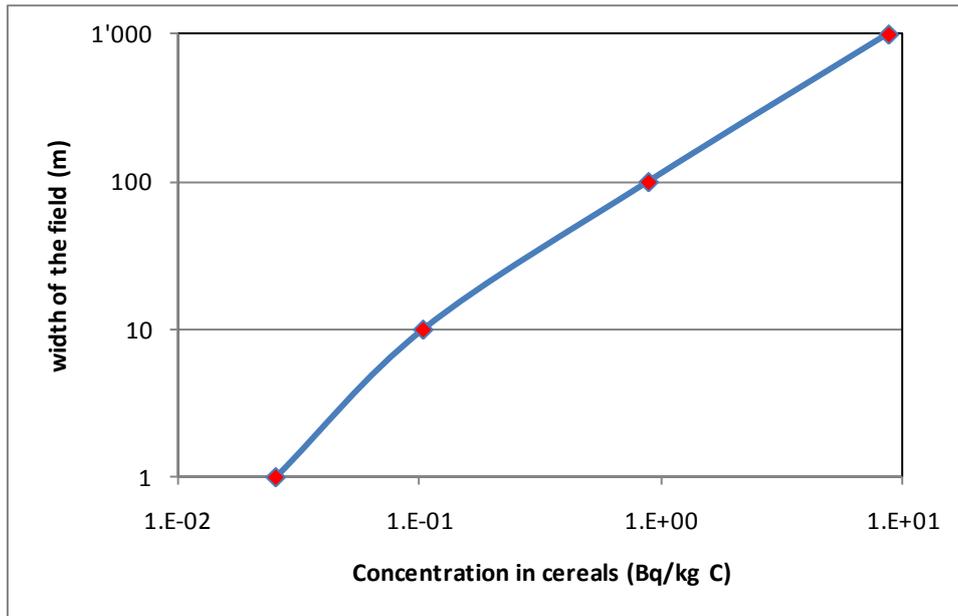


Figure 14 Effect of Field Width on Calculated Cereal C-14 Concentration (Bq kg⁻¹ C)

The calculated concentrations of C-14 (Bq kg⁻¹ C) for all crop types are shown in Table 29, again accounting for the effect of differing field dimensions.

Table 29 Calculated C-14 Concentration in Crop (Bq kg⁻¹ C) for Fields of Varying Sizes, Assuming Contaminated Irrigation Water Applied to the Site

Crop type	Calculated C-14 concentration in crop (Bq kg ⁻¹ C) for fields of varying sizes			
	Length 1 m Fetch = 1	Length 10 m Fetch = 0.1	Length 100 m Fetch = 0.01	Length 1000 m Fetch = 0.001
Cereals	2.55E-02	1.04E-01	8.86E-01	8.71E+00
Green leafy vegetables	4.29E-02	1.21E-01	9.04E-01	8.73E+00
Fruit	3.59E-02	1.14E-01	8.97E-01	8.72E+00
Root vegetables	3.15E-02	1.10E-01	8.93E-01	8.72E+00

For large fields, where the C-14 contamination is mostly via photosynthesis, differences between plant types become much reduced (Table 29, last column). This is a consequence of equal irrigation and thus equal soil concentration and degassing and equal contamination of the canopy atmosphere in the case of the four plant types. The remaining difference is a consequence of minor differences in the stable plant carbon content defined by the user. The more significant differences for the small field case (Table 29, first column) are due to the relative impact of the root uptake pathway, which is again dependent on the plant stable carbon content. The interception and translocation pathway on the other hand does not generate differences between individual plant types, because the key parameters, such as interception, translocation, leaf area index and yield have been given identical parameter values in the current exercise.

When the soil C-14 concentration is fixed at 1 Bq kg⁻¹ C, the calculated plant C-14 concentrations increase with increasing field size as before (Table 30). However, for the same reasons as given above, the increase in the crop C-14 concentrations is not perfectly linear with the increase in C-14 content of the canopy atmosphere (Table 30).

Table 30 Calculated C-14 Concentration in Crop (Bq kg⁻¹ C) for Fields of Varying Sizes, Assuming Soil C-14 Concentration is 1 Bq kg⁻¹ C

Crop type	Calculated C-14 concentration in crop (Bq kg ⁻¹ C) for fields of varying sizes			
	Length 1 m Fetch = 1	Length 10 m Fetch = 0.1	Length 100 m Fetch = 0.01	Length 1000 m Fetch = 0.001
Canopy atmosphere	1.03E-01	1.03E+00	1.03E+01	1.03E+02
Cereals	3.92E-02	2.20E-01	2.03E+00	2.02E+01
Green leafy vegetables	5.66E-02	2.38E-01	2.05E+00	2.02E+01
Fruit	4.96E-02	2.31E-01	2.04E+00	2.02E+01
Root vegetables	4.53E-02	2.26E-01	2.04E+00	2.02E+01

When the soil concentration is considered to be affected by irrigation (giving a calculated C-14 concentration of 4.32E-1 Bq kg⁻¹ C, as before), but the atmospheric C-14 concentration is fixed (to 1.70E-4 Bq m⁻³, i.e. 1 Bq C-14 kg⁻¹ C per kg m⁻³ stable C in the atmosphere), the calculated plant C-14 concentrations are independent of the field size. In these circumstances, the calculated C-14 concentrations in the crops are 2.13E-1, 2.30E-1, 2.23E-1 and 2.19E-1 Bq kg⁻¹ C for cereals, leafy vegetables, fruit and root vegetables respectively.

7.1.2 Gaseous release scenario

For this scenario, Andra considered releases of C-14 labelled CH₄ to the soil zone. In one variant 100% was assumed to be converted to CO₂ and hence available to plant uptake after degassing, whilst in another variant 11% of the CH₄ was presumed to be converted. The results from this scenario are reported in Table 31. As with the irrigation scenario, the calculated soil concentration of C-14 was independent of the field size. In contrast to the irrigation scenario, the calculated plant concentrations of C-14 are the same for each of the crops. This is because contamination via the gas flux does not have a root uptake or interception component.

Table 31 Calculated C-14 Concentration in Model Components (Bq kg⁻¹ C) for Fields of Varying Sizes, Assuming Gaseous Release of C-14 Labelled CH₄

Gaseous release assumption	Model component	Calculated C-14 concentration in crop (Bq kg ⁻¹ C) for fields of varying sizes			
		Length 1 m Fetch = 1	Length 10 m Fetch = 0.1	Length 100 m Fetch = 0.01	Length 1000 m Fetch = 0.001
100% CH ₄ oxidised to CO ₂	Soil	3.78E-03	3.78E-03	3.78E-03	3.78E-03
	Canopy Atmosphere	3.88E-04	3.88E-03	3.88E-02	3.88E-01
	Crop	3.88E-04	3.88E-03	3.88E-02	3.88E-01
11% CH ₄ oxidised to CO ₂	Soil	4.15E-04	4.15E-04	4.15E-04	4.15E-04
	Canopy Atmosphere	4.27E-05	4.27E-04	4.27E-03	4.27E-02
	Crop	1.00E-05	8.53E-05	8.38E-04	8.36E-03

7.1.3 Stochastic calculations

Andra performed a set of stochastic calculations to consider the impact of assumptions with respect to the stable carbon content of the crops, and the magnitude of the transfer rates upon the calculated C-14 concentrations in the six modelled output variables; the summary statistics from 10,000 runs are given in Table 32. Table 33 shows the correlations of the modelled variables to these parameters. As was observed in the Cl-36 study [Limer et al, 2008a; Limer et al, 2009b; Bytwerk et al, 2011], the

calculated plant concentrations of the radio-isotope are inversely correlated with the assumed stable element (carbon or chlorine) content. It is also as expected that the soil and plant C-14 concentrations are positively correlated to the rate of application of contaminated irrigation water to the soil (*teacult*).

Table 32 Summary statistics from Andra stochastic calculations

Model Compartment	C-14 Concentration (Bq kg ⁻¹ C)							
	Mean	Geo. mean	Median	Min.	Max.	Lower quartile	Upper quartile	Std. dev.
Canopy atmosphere	5.07E-1	5.78E-2	4.52E-2	1.08E-3	3.09E+2	2.03E-2	1.28E-1	5.61E+0
Cereal	1.88E+0	1.18E+0	1.25E+0	3.29E-2	6.21E+1	6.13E-1	2.36E+0	2.17E+0
Leafy vegetables	1.87E+0	1.18E+0	1.26E+0	3.16E-2	6.17E+1	6.16E-1	2.35E+0	2.13E+0
Fruit	1.86E+0	1.18E+0	1.27E+0	2.93E-2	6.51E+1	6.25E-1	2.30E+0	2.14E+0
Root vegetables	1.86E+0	1.18E+0	1.26E+0	2.83E-2	6.24E+1	6.14E-1	2.31E+0	2.12E+0
Soil	4.44E-1	3.80E-1	4.28E-1	3.15E-2	1.12E+0	2.65E-1	6.05E-1	2.20E-1

Table 33 Sensitivity of Modelled C-14 Concentrations to Input Parameters. Correlations Highlighted in red are significant at p < 0.05 for N = 10,000.

Input Parameters	Modelled variables: C-14 concentration in model compartment (Bq kg ⁻¹ C)					
	Canopy atmosphere	Cereal	Leafy vegetables	Fruit	Root vegetables	Cultivated soil
Stable C in cereal	-0.01	-0.45	-0.02	-0.01	-0.01	-0.02
Stable C in leafy vegetables	0.02	-0.01	-0.44	0.01	0.00	-0.02
Stable C in fruit	0.00	0.01	0.01	-0.43	0.01	-0.01
Stable C in root vegetables	0.00	0.01	0.01	0.01	-0.43	0.01
Cultural coefficient	0.02	0.02	0.02	0.02	0.02	0.35
Fetch	-0.10	-0.07	-0.07	-0.06	-0.06	-0.01
K _d	0.04	0.03	0.03	0.03	0.01	0.85
LPSA	0.00	-0.03	-0.03	-0.03	-0.03	0.00
LPSP	-0.02	0.42	0.42	0.41	0.42	-0.01
LPSS	0.00	-0.13	-0.12	-0.12	-0.11	0.04
LSA	0.01	0.00	-0.01	0.00	0.01	-0.09
teacult	0.01	0.36	0.37	0.37	0.37	0.26
V _{vent}	-0.12	-0.06	-0.05	-0.06	-0.06	0.00

7.2 SA_CARBN14

The EDF model, SA_Carbon14, was applied to the irrigation and gaseous release scenarios. Consideration was given to a range of field sizes for the cereal, whilst calculations for the other crops focussed upon a 10 m by 10 m field.

7.2.1 Irrigation scenario

SA_Carbon14 has two soil components: labile and fixed carbon. As with AquaC_14, the calculated C-14 concentrations in the soil were independent of the field size. The calculated C-14 concentration of the fixed soil compartment was 1.51E-1 Bq kg⁻¹ C for all crops; the calculated C-14 concentration of the labile soil compartment was 1.27E0 and 1.39E0 Bq kg⁻¹ C for the cereal and vegetable soils respectively.

The calculated atmosphere and plant C-14 concentrations for this scenario are given in Table 34. The respiratory approach leads to calculated atmosphere and plant C-14 concentrations up to a factor of 1.73 lower than those calculated using the Sheppard approach (see Section 3.2.2 for an explanation of these approaches).

Table 34 Calculated Atmosphere and Plant C-14 Concentrations by SA_Carbon14 for the Irrigation Scenario

Model variable	Crop type	Calculated C-14 concentration (Bq kg ⁻¹ C)			
		Respiratory approach ⁺	Sheppard approach		
			10 m * 10 m	100 m * 100 m	1000 m * 1000 m
Atmosphere C-14 concentration	Cereals	4.93E+01	-	5.65E+01	8.48E+01
	Other crops	1.31E+01	2.26E+01	-	-
Crop C-14 concentration	Cereals	4.01E+01	-	4.60E+01	6.90E+01
	Green leafy vegetables	6.82E+00	1.18E+01	-	-
	Fruit	9.46E+00	1.64E+01	-	-
	Root vegetables	8.94E+00	1.55E+01	-	-

⁺ The following field sizes have been assumed: 30*30 m for cereal and 10*10 m for garden crop

7.2.2 Gaseous scenario

For the gaseous release scenario consideration was given to either 100% or 9% of any C-14 labelled gas released into the soil as being in the form of CO₂ and hence available for plant uptake. The calculated C-14 concentrations in the three soil compartments for these two scenarios are given in Table 35. The calculated atmosphere and plant C-14 concentrations for this scenario are given in Table 36.

Table 35 Calculated Soil C-14 Concentrations by SA_Carbon14 for the Gaseous Release Scenario

Release assumption	Calculated soil C-14 concentration (Bq kg ⁻¹ C)		
	Labile C in cereal soil	Labile C in vegetable garden soil	Fixed C in soil
100% CO ₂	3.93E-03	3.95E-03	5.66E-04
91% CH ₄ and 9% CO ₂	3.54E-04	3.56E-04	5.09E-05

Table 36 Calculated Atmosphere and Plant C-14 Concentrations from the EDF Model for the Gaseous Release Scenarios

Release assumption	Model variable	Crop type	Calculated C-14 concentration (Bq kg ⁻¹ C)			
			Respiratory approach ⁺	Sheppard (2006) approach		
				10 m * 10 m	100 m * 100 m	1000 m * 1000 m
100% CO ₂	Atmosphere C-14 concentration	Cereals	3.72E-01	-	4.27E-01	6.40E-01
		Other crops	4.41E-02	7.64E-02	-	-
	Crop C-14 concentration	Cereals	2.35E-01	-	2.70E-01	4.05E-01
		Green leafy vegetables	2.80E-02	4.85E-02	-	-
		Fruit	2.80E-02	4.84E-02	-	-
		Root vegetables	2.79E-02	4.83E-02	-	-
91% CH ₄ and 9% CO ₂	Atmosphere C-14 concentration	Cereals	3.35E-02	-	3.84E-02	5.74E-02
		Other crops	3.97E-03	6.88E-03	-	-
	Crop C-14 concentration	Cereals	2.12E-02	-	2.43E-02	3.63E-02
		Green leafy vegetables	2.52E-03	4.37E-03	-	-
		Fruit	2.51E-03	4.36E-03	-	-
		Root vegetables	2.51E-03	4.35E-03	-	-

⁺ The following field sizes have been assumed: 30*30 m for cereal and 10*10 m for garden crop

7.2.3 Fixed activity calculations

The calculated atmosphere and plant C-14 concentrations for a fixed soil concentration (1 Bq kg⁻¹ C) are given in Table 37. Consideration was given to fields with length 10 and 1000 m.

Table 37 Calculated Atmosphere and Plant C-14 Concentrations by the EDF Model for the Fixed Soil C-14 Concentration Scenario

Model variable	Crop type	Calculated C-14 concentration (Bq kg ⁻¹ C)		
		Respiratory approach	Sheppard (2006) approach	
			10 m * 10 m	1000 m * 1000 m
Atmosphere C-14 concentration	Cereals	9.47E+01	-	1.63E+02
	Other crops	1.12E+01	1.94E+01	-
Crop C-14 concentration	Cereals	5.98E+01	-	1.03E+02
	Green leafy vegetables	7.15E+00	1.24E+01	-
	Fruit	7.10E+00	1.23E+01	-
	Root vegetables	7.08E+00	1.23E+01	-

When the concentration in the atmosphere was fixed to 1 Bq kg⁻¹ C, the calculated plant C-14 concentrations were 6.3E-01 Bq kg⁻¹ C for cereals, fruit and root vegetables, and 6.4E-01 Bq kg⁻¹ C for leafy green vegetables. The plant concentrations calculated for this case are independent of the method used to determine the atmospheric C-14 concentration.

7.2.4 Stochastic calculations

A series of stochastic calculations were carried out using SA Carbon14 for the irrigation scenario. Table 38 below presents the mean (with the 5th and 95th percentiles in brackets) calculated

concentrations of C-14 in the various model compartments; the parameter distributions used are given in Table 21. As with the deterministic calculations, the mean calculated C-14 concentrations for the numerous model variables are greater when the Sheppard (2006b) approach to determining atmospheric C-14 concentrations is used.

Table 38 Results of Stochastic Calculations Using SA_Carbon14 for the Irrigation Scenario

Region	Model variable	Approach used to determine atmospheric C-14 concentration	
		Respiratory approach	Sheppard (2006b) approach
Soil	Labile C in cereal soil	5.22E-01 (1.69E-01, 1.32E+00)	5.42E-01 (1.74E-01, 1.29E+00)
	Labile C in vegetable garden soil	6.33E-01 (1.70E-01, 1.84E+00)	6.63E-01 (1.76E-01, 1.85E+00)
	Fixed C in soil	1.70E-01 (9.93E-02, 2.43E-01)	3.21E-01 (1.59E-01, 4.99E-01)
Atmosphere	Cereal	5.75E+01 (2.81E+01, 9.67E+01)	1.76E+02 (6.03E+01, 3.46E+02)
	Vegetable garden	1.61E+01 (6.83E+00, 2.78E+01)	1.27E+02 (3.29E+01, 2.33E+02)
Crop	Cereal	4.58E+01 (3.08E+01, 6.40E+01)	1.41E+02 (5.72E+01, 2.31E+02)
	Green leafy vegetables	1.03E+01 (4.26E+00, 1.80E+01)	8.12E+01 (2.06E+01, 1.52E+02)
	Fruit	1.10E+01 (4.75E+00, 1.88E+01)	8.70E+01 (2.25E+01, 1.57E+02)
	Root vegetables	1.08E+01 (4.70E+00, 1.84E+01)	8.57E+01 (2.23E+01, 1.55E+02)

7.3 SIMPLIFIED ENHANCED RIMERS

7.3.1 Gas scenario

As described in Section 3.3, for a source term of $1 \text{ Bq m}^{-2} \text{ d}^{-1}$ of C-14 as CO_2 to the soil zone, the concentration in plants is estimated to be in the range 1 to 50 Bq kg^{-1} (f.w.) with a best estimate of 5 Bq kg^{-1} (f.w.). This is based on plants being $0.04 \text{ kg C kg}^{-1}$ (f.w.). Thus, the corresponding concentrations expressed on a carbon mass basis are 25 to $1250 \text{ Bq kg}^{-1} \text{ C}$, with a best estimate of $125 \text{ Bq kg}^{-1} \text{ C}$.

For the gas scenario, the source term is $1 \text{ Bq m}^{-2} \text{ y}^{-1}$, so the concentration in plants is estimated to be 0.068 to $3.42 \text{ Bq kg}^{-1} \text{ C}$, with a best estimate of $0.34 \text{ Bq kg}^{-1} \text{ C}$.

Previous studies have not reported C-14 concentrations in soil atmosphere and below-canopy atmosphere, but these are readily derived. The compartment volumes are as described in Section 6.4, and the transfers assumed are also not varied ($k_{12} = 1.87\text{E-}3 \text{ m}^3 \text{ d}^{-1}$, $k_{23} = 1.85\text{E-}2 \text{ m}^3 \text{ d}^{-1}$ and $k_{3L} = 4.52\text{E-}1 \text{ m}^3 \text{ d}^{-1}$). The value of δ is taken as $2\text{E-}4 \text{ m}^3 \text{ d}^{-1}$, this being 50% of the net photosynthetic uptake in the reference model to allow for losses in respiration. The value of α is taken as $2.15\text{E-}2$. The value of l is taken as $1/365.25 = 2.74\text{E-}3 \text{ Bq m}^{-2} \text{ d}^{-1}$ ($1 \text{ Bq m}^{-2} \text{ y}^{-1}$).

The transfer rates between compartments are listed in Section 6, Table 23. The activity, q_j (Bq), of each compartment is then:

- $q_3 = 0.00274/273.9 = 1\text{E-}5 \text{ Bq}$
- $q_2 = (273.9 + 11.33) \times 10^{-5}/112.1 = 2.544\text{E-}5 \text{ Bq}$
- $q_1 = (0.00274 + 11.33 \times 2.544 \times 10^{-5}) / 3.34 = 9.067\text{E-}4 \text{ Bq}$

Now recall that V (m^3) is the volume of CO_2 in each compartment at STP. Thus, the mass of carbon in each compartment is $(V/0.0224) \times 0.012 = 0.5357V$ kg C, since a mole of gas occupies $0.0224 m^3$ at STP and a mole of CO_2 contains 0.012 kg C. Thus, the activity concentrations in the three compartments are:

- Compartment 1: $3.02 \text{ Bq kg}^{-1} \text{ C}$
- Compartment 2: $2.88E-1 \text{ Bq kg}^{-1} \text{ C}$
- Compartment 3: $1.13E-2 \text{ Bq kg}^{-1} \text{ C}$

The high degree of dilution in the above-canopy atmosphere is evident from these results. The concentration in plants is given by:

- $C_p = \alpha C_1 + (1 - \alpha)C_2 = 0.0215 \times 3.02 + 0.9785 \times 0.288 = 0.0649 + 0.2818 = 3.47E-1 \text{ Bq kg}^{-1} \text{ C}$

Note that about 19% of the C-14 in the plant derives from root uptake in this reference case. Thus, finally for the gas scenario, the following results are applicable in the reference case:

- Concentration in soil gas (and also in soil solution, which is considered to be in equilibrium with soil gas): $3.02 \text{ Bq kg}^{-1} \text{ C}$
- Concentration in the sub-canopy atmosphere: $2.88E-1 \text{ Bq kg}^{-1} \text{ C}$
- Concentration in the above-canopy atmosphere: $1.13E-2 \text{ Bq kg}^{-1} \text{ C}$
- Concentration in plants: $3.47E-1 \text{ Bq kg}^{-1} \text{ C}$

Note that the below-canopy atmosphere corresponds approximately to the atmosphere below the zero velocity plane in the Thorne-Limer model and the above-canopy atmosphere corresponds approximately to the atmosphere above the zero velocity plane.

7.3.2 Irrigation scenario

The results for the gas scenario are for $1 \text{ Bq m}^{-2} \text{ y}^{-1}$. They can be applied to the groundwater scenario by scaling by the relevant input fluxes. These are: leafy vegetables $120 \text{ Bq m}^{-2} \text{ y}^{-1}$; root vegetables $180 \text{ Bq m}^{-2} \text{ y}^{-1}$; fruit $84 \text{ Bq m}^{-2} \text{ y}^{-1}$; and cereal $144 \text{ Bq m}^{-2} \text{ y}^{-1}$. Results are shown in Table 39.

Table 39 Simplified Enhanced RIMERS Results for the Irrigation Pathway

Component	Concentration ($\text{Bq kg}^{-1} \text{ C}$)			
	Green leafy vegetables	Root vegetables	Fruit	Cereal
Soil atmosphere and solution	362.4	543.6	253.7	434.9
Canopy atmosphere	34.6	51.8	24.2	41.5
Above-canopy atmosphere	1.36	2.03	0.95	1.63
Plants	41.6	62.4	29.1	49.9

7.3.3 Fixed activity calculations

In the case of a fixed soil concentration of $1 \text{ Bq kg}^{-1} \text{ C}$, the input rate was set to give the following concentrations by scaling:

- Concentration in soil gas (and also in soil solution, which is considered to be in equilibrium with soil gas): $1.0 \text{ Bq kg}^{-1} \text{ C}$
- Concentration in the sub-canopy atmosphere: $9.54E-2 \text{ Bq kg}^{-1} \text{ C}$
- Concentration in the above-canopy atmosphere: $3.74E-3 \text{ Bq kg}^{-1} \text{ C}$

- Concentration in plants: $1.15\text{E-}1 \text{ Bq kg}^{-1} \text{ C}$

In the case of a fixed canopy atmosphere concentration, the input rate is set to give the following concentrations by scaling:

- Concentration in soil gas (and also in soil solution, which is considered to be in equilibrium with soil gas): $1.05\text{E+}1 \text{ Bq kg}^{-1} \text{ C}$
- Concentration in the sub-canopy atmosphere: $1.0 \text{ Bq kg}^{-1} \text{ C}$
- Concentration in the above-canopy atmosphere: $3.92\text{E-}2 \text{ Bq kg}^{-1} \text{ C}$
- Concentration in plants: $1.204 \text{ Bq kg}^{-1} \text{ C}$

Note that the concentration in plants is slightly higher than the concentration in the sub-canopy atmosphere because of the contribution from root uptake. This would not occur if the C-14 were released at the soil surface rather than into the underlying soil. In that case, the concentration in plants would be $9.785\text{E-}1 \text{ Bq kg}^{-1} \text{ C}$.

7.4 AVILA AND PRÖHL MODEL

The Avila and Pröhl model has been applied to both the irrigation and gas scenarios. Consideration has been given to the effect of different assumptions relating to field size and wind speed at the top of the vegetation and also to the relationship between canopy height and z_d for which two relationships have been assumed ($\frac{1}{6}$ and $\frac{2}{3}$), which is consistent with the assumptions for the Thorne-Limer model (Section 7.5).

The Avila and Pröhl model is based on a specific activity approach. As such, plant concentrations ($\text{Bq kg}^{-1} \text{ C}$) and canopy air concentrations ($\text{Bq kg}^{-1} \text{ C}$) are identical. For presentation purposes, results are given only for plants.

7.4.1 Irrigation scenario

In the irrigation scenario crop specific irrigation rates, consistent with the values in the scenario description, are applied. Two wind velocity assumptions are made – a wind velocity scaled from 5 m s^{-1} at 1 m and a wind velocity set at 2 m s^{-1} .

Table 40 gives the results for crops of different heights being grown in fields of varying size for a wind velocity scaled from 5 m s^{-1} at 10 m when the roughness layer (z_d) was set at $\frac{1}{6}$ of the crop height. Similar results are presented in Table 41 for a roughness layer set at $\frac{2}{3}$ of the crop height. Assumptions relating to the height of the z_d have limited effect with results from $z_d = \frac{1}{6}$ and $z_d = \frac{2}{3}$ being within a factor of around three. Increasing field size had a greater influence on the result, with plant concentrations increasing by an order of magnitude for each increase in field size. Increasing crop height reduces plant C-14 concentrations with the effect being most pronounced when z_d is set to $\frac{2}{3}$ of the crop height.

If a constant wind velocity of 2 m s^{-1} is assumed, no difference is evident in plant concentrations when z_d is set to $\frac{1}{6}$ crop height or $\frac{2}{3}$ crop height (Table 42). Equally, crop height does not affect the calculated plant C-14 concentrations.

Table 40 Results of the Avila and Pröhl Model for Crops of Differing Height in Fields of Varying Size (wind velocity scaled from 5 m s⁻¹ at 10m; z_d = 1/6).

Field size	Crop height (m)	Plant C-14 concentration (Bq kg ⁻¹ C)			
		Cereal	Green leafy vegetables	Root vegetables	Fruit
1 m * 1 m	0.3	8.96E-04	7.46E-04	1.12E-03	5.22E-04
	0.4	8.47E-04	7.06E-04	1.06E-03	4.94E-04
	1	6.92E-04	5.77E-04	8.65E-04	4.04E-04
	2	5.75E-04	4.79E-04	7.19E-04	3.35E-04
10 m * 10 m	0.3	8.96E-03	7.46E-03	1.12E-02	5.22E-03
	0.4	8.47E-03	7.06E-03	1.06E-02	4.94E-03
	1	6.92E-03	5.77E-03	8.65E-03	4.04E-03
	2	5.75E-03	4.79E-03	7.19E-03	3.35E-03
100 m * 100 m	0.3	8.95E-02	7.46E-02	1.12E-01	5.22E-02
	0.4	8.46E-02	7.05E-02	1.06E-01	4.94E-02
	1	6.92E-02	5.77E-02	8.65E-02	4.04E-02
	2	5.75E-02	4.79E-02	7.18E-02	3.35E-02
1000 m * 1000 m	0.3	8.89E-01	7.42E-01	1.11E+00	5.20E-01
	0.4	8.41E-01	7.02E-01	1.05E+00	4.92E-01
	1	6.88E-01	5.74E-01	8.61E-01	4.02E-01
	2	5.72E-01	4.77E-01	7.16E-01	3.34E-01

Table 41 Results of the Avila and Pröhl Model for Crops of Differing Height in Fields of Varying Size (wind velocity scaled from 5 m s⁻¹ at 10m; z_d = 2/3).

Field size	Crop height (m)	Plant C-14 concentration (Bq kg ⁻¹ C)			
		Cereal	Green leafy vegetables	Root vegetables	Fruit
1 m * 1 m	0.3	2.92E-03	2.44E-03	3.65E-03	1.70E-03
	0.4	2.71E-03	2.26E-03	3.38E-03	1.58E-03
	1	2.02E-03	1.69E-03	2.53E-03	1.18E-03
	2	1.51E-03	1.25E-03	1.88E-03	8.78E-04
10 m * 10 m	0.3	2.92E-02	2.43E-02	3.65E-02	1.70E-02
	0.4	2.71E-02	2.26E-02	3.38E-02	1.58E-02
	1	2.02E-02	1.69E-02	2.53E-02	1.18E-02
	2	1.50E-02	1.25E-02	1.88E-02	8.78E-03
100 m * 100 m	0.3	2.92E-01	2.43E-01	3.65E-01	1.70E-01
	0.4	2.70E-01	2.25E-01	3.38E-01	1.58E-01
	1	2.02E-01	1.68E-01	2.53E-01	1.18E-01
	2	1.50E-01	1.25E-01	1.88E-01	8.77E-02
1000 m * 1000 m	0.3	2.85E+00	2.39E+00	3.59E+00	1.68E+00
	0.4	2.65E+00	2.22E+00	3.33E+00	1.56E+00
	1	1.99E+00	1.66E+00	2.50E+00	1.17E+00
	2	1.49E+00	1.24E+00	1.86E+00	8.71E-01

Table 42 Results of the Avila and Pröhl Model for Crops of Differing Heights in fFields of Varying Size (wind velocity 2 m s^{-1}).

Field size	Plant concentration ($\text{Bq kg}^{-1} \text{ C}$)			
	Cereals	Green leafy vegetables	Root vegetables	Fruit
1 m * 1 m	7.57E-04	6.31E-04	9.46E-04	4.42E-04
10 m * 10 m	7.57E-03	6.31E-03	9.46E-03	4.42E-03
100 m * 100 m	7.57E-02	6.31E-02	9.46E-02	4.42E-02
1000 m * 1000 m	7.52E-01	6.28E-01	9.42E-01	4.40E-01

Plant C-14 concentrations are directly related to field size when constant air velocity is assumed (Figure 15) although differences are noted between crops such that the rate of increase in C-14 uptake by plants with increasing field size is greatest for root vegetables and lowest for fruit. Differences arise as a result of the irrigation rates assumed for each crop type.

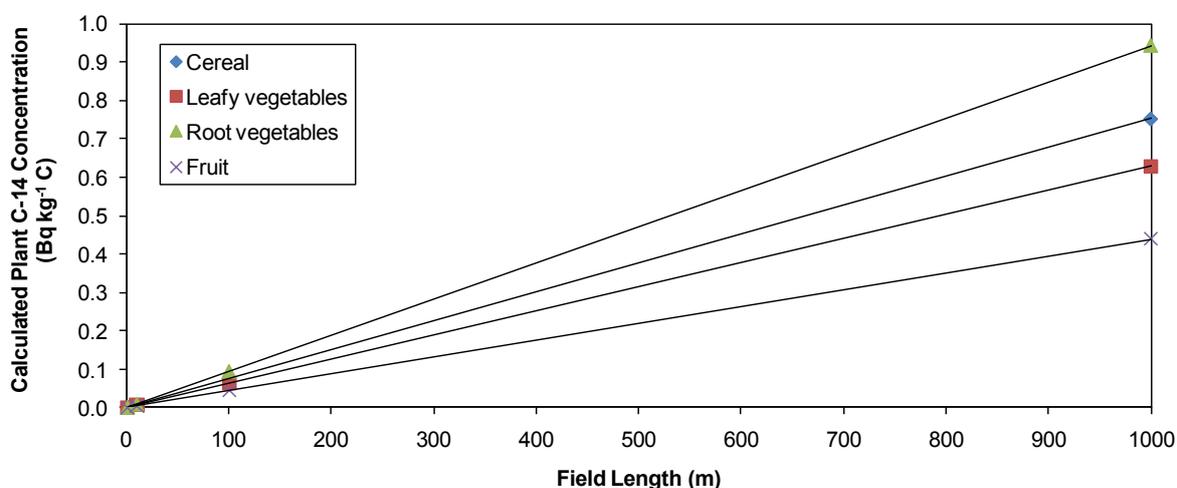


Figure 15 Variation in C-14 Concentration in Crops with Field Length under Constant Wind Velocity (2 m s^{-1}).

7.4.2 Gaseous release scenario

Similar assumptions relating to wind velocity and roughness height (z_d) to those of the irrigation scenario were applied to the gaseous release scenario. Results are shown in Table 43.

Consistent with the irrigation scenario, results are within a factor of 3.25 for the two roughness heights assumed. However, within each roughness height category, no significant variation in plant C-14 concentrations by crop is evident (up to a factor of two). The greatest variation in crop C-14 concentrations again relates to the field size assumed, with an increase in field size corresponding to an increase in plant C-14 concentration. Increasing crop height again served to reduce plant C-14 concentrations, with a greater reduction being observed when z_d is set to $\frac{2}{3}$ rather than $\frac{1}{6}$ of the crop height.

7.4.3 Fixed activity calculations

A calculation with the fixed activity concentration in air ($1 \text{ Bq kg}^{-1} \text{ C}$) can also be made with the Avila and Pröhl model by considering that $R, {}^{14}\text{C}/{}^{12}\text{C}$, is equal to 1, so that the calculated activity in the crop is $1 \text{ Bq kg}^{-1} \text{ C}$.

Table 43 Results of the Avila and Pröhl Model for Crops of Differing Height in Fields of Varying Size for a Gaseous Release Scenario.

Field size	Crop height (m)	Plant concentration – all crops (Bq kg ⁻¹ C)			
		Scaled wind velocity		Fixed wind velocity	
		$z_d = 1/6$	$z_d = 2/3$	$z_d = 1/6$	$z_d = 2/3$
1 m * 1 m	0.3	3.11E-06	1.01E-05	2.63E-06	2.63E-06
	0.4	2.94E-06	9.40E-06	2.63E-06	2.63E-06
	1	2.40E-06	7.02E-06	2.63E-06	2.63E-06
	2	2.00E-06	5.23E-06	2.63E-06	2.63E-06
10 m * 10 m	0.3	3.11E-05	1.01E-04	2.63E-05	2.63E-05
	0.4	2.94E-05	9.40E-05	2.63E-05	2.63E-05
	1	2.40E-05	7.02E-05	2.63E-05	2.63E-05
	2	2.00E-05	5.23E-05	2.63E-05	2.63E-05
100 m * 100 m	0.3	3.11E-04	1.01E-03	2.63E-04	2.63E-04
	0.4	2.94E-04	9.38E-04	2.63E-04	2.63E-04
	1	2.40E-04	7.01E-04	2.63E-04	2.63E-04
	2	2.00E-04	5.22E-04	2.63E-04	2.63E-04
1000 m * 1000 m	0.3	3.09E-03	9.91E-03	2.61E-03	2.61E-03
	0.4	2.92E-03	9.19E-03	2.61E-03	2.61E-03
	1	2.39E-03	6.91E-03	2.61E-03	2.61E-03
	2	1.99E-03	5.16E-03	2.61E-03	2.61E-03

7.5 THORNE-LIMER MODEL

The Thorne-Limer model was applied to the gas scenario only.

Three assumptions were employed with respect to the uptake of carbon by plants. Two relate to the rate at which light is extinguished in moving from the top of the plant downwards and the effect of this on the rate of photosynthesis, distinguished according to whether broad leaf or narrow leaf crops are assumed. Uptake is greatest per unit plant height from the upper part of the canopy. The third uptake assumption is that carbon uptake is uniform throughout the canopy. Carbon uptake is thus independent of crop type.

Two roughness height, z_d , assumptions have been considered whereby z_d is $1/6$ or $2/3$ of the crop height: results are presented in Table 44 and Table 45, respectively.

Table 44 Results of the Thorne-Limer Model Assuming a Roughness Height Equivalent to $1/6$ Crop Height

Crop height (m)	z_d (m)	C-14 concentration (Bq kg ⁻¹ C)					
		Soil	Within canopy air (below z_d)	Within and above canopy (above z_d)	Plant (uniform uptake)	Plant (broad leaf uptake)	Plant (narrow leaf uptake)
0.3	0.05	2.10E-06	2.87E-01	3.66E-03	5.08E-02	3.03E-02	4.56E-02
0.4	0.07	2.20E-06	3.81E-01	3.68E-03	6.65E-02	3.91E-02	5.96E-02
1	0.17	2.80E-06	9.45E-01	3.86E-03	1.61E-01	9.23E-02	1.43E-01
2	0.33	3.81E-06	1.89E+00	4.19E-03	3.18E-01	1.81E-01	2.83E-01

Table 45 Results of the Thorne-Limer Model Assuming a Roughness Height Equivalent to $\frac{2}{3}$ Crop Height

Crop height (m)	z_d (m)	C-14 concentration (Bq kg ⁻¹ C)					
		Soil	Within canopy air (below z_d)	Within and above canopy (above z_d)	Plant (uniform uptake)	Plant (broad leaf uptake)	Plant (narrow leaf uptake)
0.3	0.20	3.00E-06	1.13E+00	3.67E-03	7.57E-01	5.96E-01	7.22E-01
0.4	0.27	3.40E-06	1.51E+00	3.70E-03	1.01E+00	7.94E-01	9.61E-01
1	0.67	5.81E-06	3.77E+00	3.90E-03	2.51E+00	1.98E+00	2.40E+00
2	1.33	9.81E-06	7.53E+00	4.28E-03	5.02E+00	3.95E+00	4.79E+00

Results indicate that the assumptions relating to the uptake of carbon in relation to light attenuation through the canopy has minimal effect on plant C-14 concentrations; results are all within a factor of two (Figure 16). Nonetheless, a very slight reduction is observed in C-14 concentrations in broad leaf plants compared with narrow leaf plants due to broad leaf LAI increasing the amount of light attenuation when compared with narrow leaf plants.

For a fixed atmospheric C-14 concentration of 1 Bq kg⁻¹ C in both layers of the atmosphere, the calculated plant C-14 concentration is 1 Bq kg⁻¹ C also.

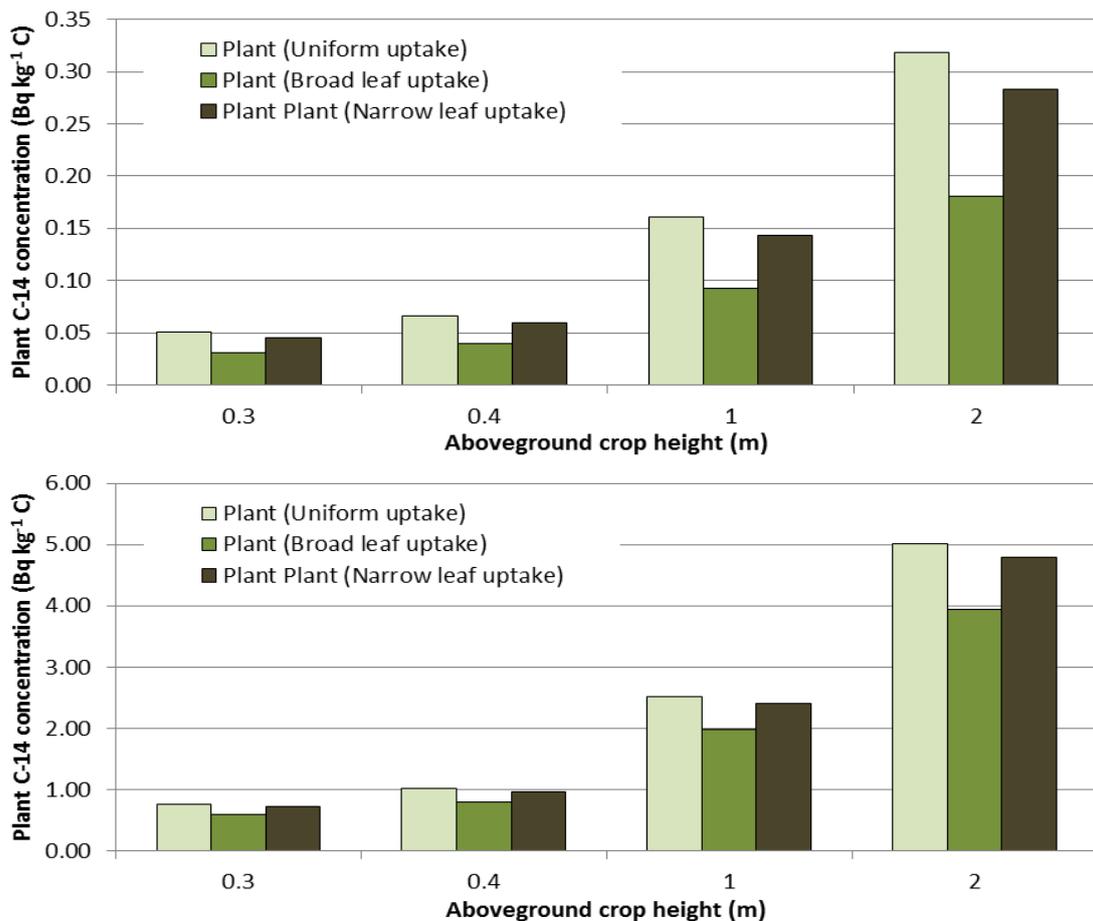


Figure 16 Calculated Plant C-14 Concentrations Using the Thorne-Limer Model. (a) Zero plane height 1/6; and (b) Zero plane height 2/3.

7.6 COMPARISON OF RESULTS

In this section a comparison is made between the calculated C-14 concentrations in the various environmental media for the scenarios considered. There are many consistencies between the models, such as increasing atmospheric and plant C-14 concentrations for larger fields in those that have a dependency on the field size. Such an observation results from the shared assumption of these models that increasing the field size will increase the time it takes for the contaminated air above the field to exchange with uncontaminated air, and thus the dilution of C-14 in the atmosphere is reduced. Independent of any assumed dependence of the atmospheric C-14 concentration on the field size, it is also clear that those models which assume a lesser degree of exchange of the contaminated air which the plant “sees” and ‘free’ uncontaminated air will naturally calculate higher plant C-14 concentrations for a given C-14 flux entering the system.

7.6.1 Irrigation scenario

Results for the soil compartment

Four of the five models were applied to the irrigation scenario, of which three reported the calculated C-14 concentration in carbon in the soil; these are shown in Table 46. As would be anticipated given the definition of the source term, the calculated soil C-14 concentrations are independent of the field size assumed. Note that the soil C-14 concentrations reported from AquaC_14 and SA_Carbon14 relate to soil solids, whereas the soil C-14 concentrations reported from the simplified enhanced RIMERS model relate to the soil gas (which is assumed to be in equilibrium with the soil solution). As such it is only the AquaC_14 and SA_Carbon14 results that can be compared; these agree within a factor of 3.3.

Table 46 Summary of Calculated C-14 Concentrations in Soil Carbon for the Irrigation Scenario

Soil below crop type	Calculated C-14 concentration in soil (Bq kg ⁻¹ C)		
	AquaC_14	SA_Carbon14	Simplified enhanced RIMERS
Cereals	4.32E-01	Labile: 1.27E0 Fixed: 1.51E-01	4.39E+2
Green leafy vegetables		Labile: 1.39E0 Fixed: 1.51E-01	3.62E+2
Fruit			2.54E+2
Root vegetables			5.44E+2

Results for the atmosphere and plant compartments

For the smallest field size, the calculated atmospheric C-14 concentrations vary over six orders of magnitude; the Avila and Pröhl model reporting the lowest values and the EDF model reporting the highest (Table 47). However, as the field size considered increases, so the difference in the calculated canopy atmosphere C-14 concentrations decreases, such that for a field of length 1000 m the difference in calculated values is two orders of magnitude. These differences are reflected in the variation in calculated plant C-14 concentrations. As an example the calculated C-14 concentrations in cereal for field sizes of length 10 m and 1000 m are shown in Figure 17.

It is those models that have the most cautious assumptions about the plant canopy, e.g. a dense canopy, which generate the highest reported calculated canopy and plant C-14 concentrations. However, some of those models appear to be less sensitive to the field size assumed than the models which have a less cautious plant canopy atmosphere approach. This is to be expected. With a dense canopy, the above-canopy wind is not seen within the canopy where movement is largely by vertical diffusion.

Table 47 Summary of Calculated C-14 Concentrations in the Canopy Atmosphere Carbon for the Irrigation Scenario

Field Length (m)	Calculated C-14 concentration in the atmosphere (Bq kg ⁻¹ C) for a given field length (m)			
	1	10	100	1000
AquaC_14	4.44E-02	4.44E-01	4.44E+00	4.44E+01
Avila and Pröhl (z _d = 1/6) - cereal	3.46E-04	3.46E-03	3.46E-02	3.44E-01
Avila and Pröhl (z _d = 2/3) - cereal	2.02E-03	2.02E-02	2.02E-01	1.99E+00
Avila and Pröhl (2 m/s wind speed) - cereal	7.57E-04	7.57E-03	7.57E-02	7.52E-01
SA_Carbon14 (Sheppard approach)	-	2.26E+01	5.65E+01	8.48E+01
SA_Carbon14 (Respiratory approach)	-	Cereal: 4.01E+01		
Simplified enhanced RIMERS	Results are given for each crop as a couplet: <Canopy>, <Above canopy> Cereal: 4.15E+1, 1.63E0 Green leafy vegetables: 3.46E+1, 1.36E0 Fruit: 2.42E+1, 9.50E-1 Root vegetables: 5.18E+1, 2.03E0			

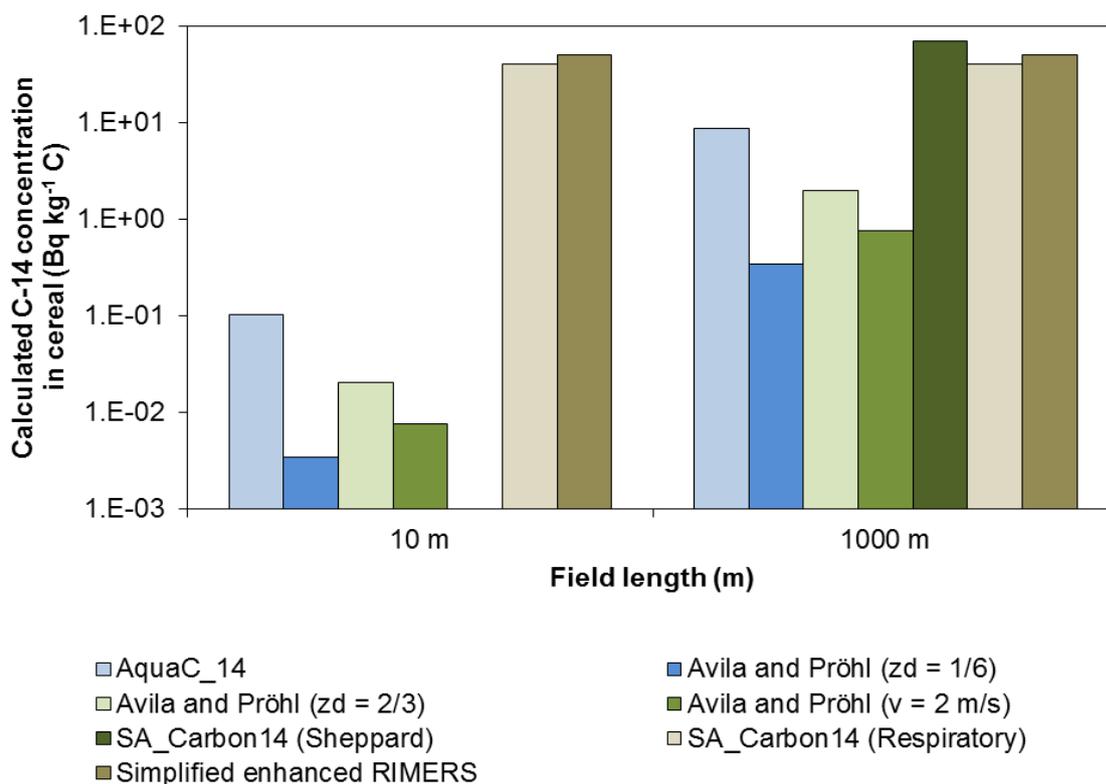


Figure 17 Calculated Cereal C-14 Concentrations for the Irrigation Scenario – Effect of Field Size

7.6.2 Gaseous release scenario

Results for the soil compartment

The calculated soil C-14 concentrations, reported by the AquaC_14 and SA_Carbon14 models, are very similar. When the C-14 labelled gas entering the soil is assumed to be 100% CO₂, the calculated soil C-14 concentration is of the order 3.9E-03 Bq kg⁻¹ C in an available form; SA_Carbon14 also reports 5.7E-04 Bq kg⁻¹ C in a more recalcitrant form. The calculated soil C-14 concentrations in the Thorne-Limer model are much lower than for the other models; the calculated soil C-14 concentration associated with a plant of height 1 m is 2.80E-6 Bq kg⁻¹ C. As with the irrigation scenario, the soil C-14 concentrations reported thus far relate to soil solids. The simplified enhanced RIMERS model reports soil gas C-14 concentrations of 3.02E0 Bq kg⁻¹ C.

Results for the atmosphere and plant compartments

As with the irrigation scenario, differences between the calculated atmospheric C-14 concentrations for a field of any given size decrease as the field size increases (Table 48). Thus for a field of length 1 m the difference is five orders of magnitude, whilst for a field of length 1000 m the difference drops to two orders of magnitude.

Table 48 Summary of Calculated C-14 Concentrations in the Canopy Atmosphere Carbon for the Gaseous Release Scenario

Field length (m)	Calculated C-14 Concentration in the atmosphere (Bq kg ⁻¹ C)			
	1	10	100	1000
AquaC_14 (100% CO ₂)	3.88E-04	3.88E-03	3.88E-02	3.88E-01
AquaC_14 (11% CO ₂)	4.27E-05	4.27E-04	4.27E-03	4.27E-02
Avila and Pröhl (z _d = 1/6)	2.40E-06	2.40E-05	2.40E-04	2.39E-03
Avila and Pröhl (z _d = 2/3)	7.02E-06	7.02E-05	7.01E-04	6.91E-03
Avila and Pröhl (2 m s ⁻¹ wind speed)	2.63E-06	2.63E-05	2.63E-04	2.61E-03
SA_Carbon14 (Sheppard approach) - 100% CO ₂	-	7.64E-02	4.27E-01	6.40E-01
SA_Carbon14 (Sheppard approach) - 9% CO ₂	-	6.88E-03	3.84E-02	5.74E-02
SA_Carbon14 (Respiratory approach) - 100% CO ₂	-	Cereal (30 m by 30 m field): 3.72E-01 Other crops (10 m by 10 m field): 4.41E-02		
SA_Carbon14 (Respiratory approach) - 9% CO ₂	-	Cereal: 3.35E-02 Other crops 3.97E-03		
Simplified enhanced RIMERS	Below z _d : 2.88E-01 Above z _d : 1.13E-02			
Thorne-Limer (z _d = 1/6)	Below z _d : 9.45E-01 Above z _d : 3.86E-03.			
Thorne-Limer (z _d = 2/3)	Below z _d : 3.77E+00 Above z _d : 3.90E-03			

As with the irrigation scenario, this is then reflected in the calculated plant C-14 concentrations, as shown in Figure 18 (a field size of 10 m is used as an example).

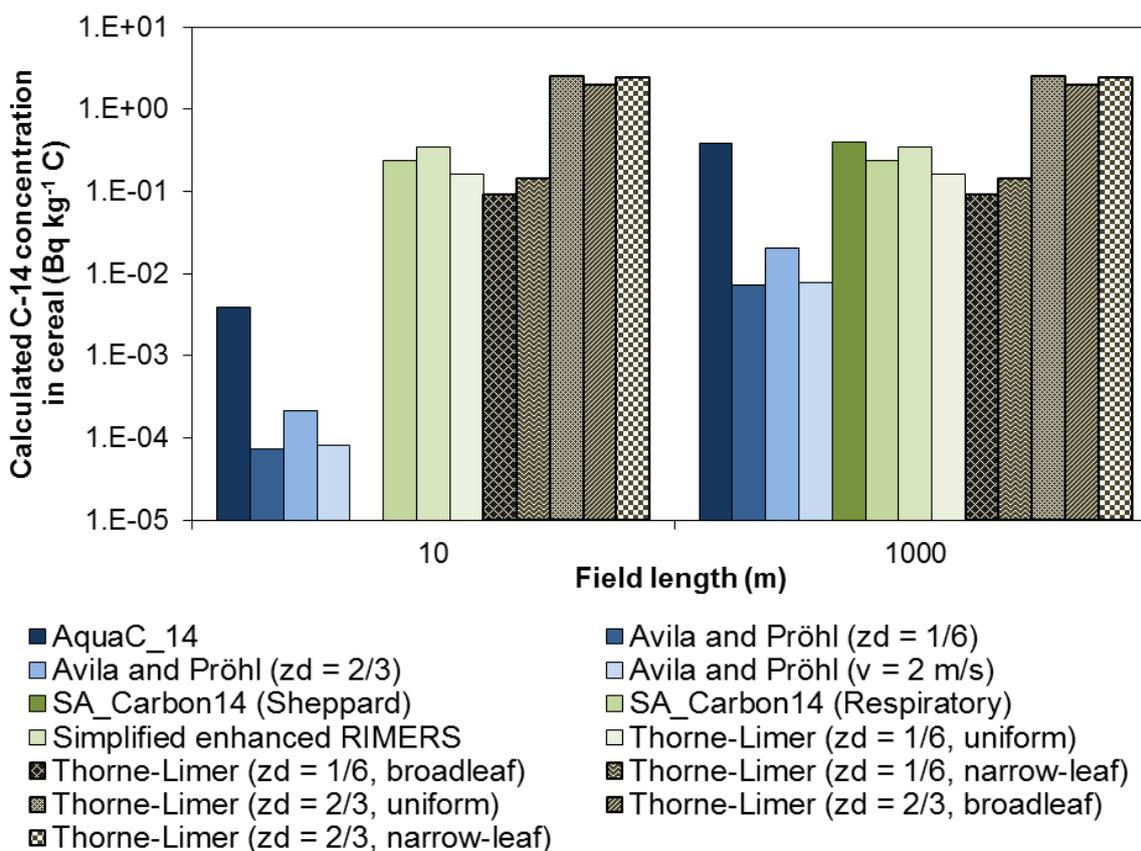


Figure 18 Calculated Plant C-14 Concentrations for the Gaseous Scenario – Fields of 10 m and 1000 m in Length

7.6.3 Fixed concentration calculations

Fixing the soil C-14 concentration in carbon in soil does not reduce the variability in the calculated plant C-14 concentrations for fields of a given size (e.g. the results for a field of length 10 m are shown in Table 49).

Table 49 Calculated Plant C-14 Concentrations when the Soil C-14 Concentration is Fixed to 1 Bq kg⁻¹ C (10 m field length)

Crop Type	Calculated plant C-14 concentration (Bq kg ⁻¹ C)				
	AquaC_14	SA_Carbon14 – resp. approach	SA_Carbon14 – Sheppard approach	Simplified enhanced RIMERS	Avila and Pröhl
Cereals	2.20E-01	5.98E+01	-	1.15E-1	1.00E+00
Green leafy vegetables	2.38E-01	7.15E+00	1.24E+01		
Fruit	2.31E-01	7.10E+00	1.23E+01		
Root vegetables	2.26E-01	7.08E+00	1.23E+01		

When the C-14 concentration in carbon in the air is fixed, the variability in the calculated plant C-14 concentrations drops significantly (as shown in Figure 19). This demonstrates that in these models, the key processes responsible for model variability in calculated plant C-14 concentrations are volatilisation and the exchange of gas in the atmosphere and the way they are represented in the models.

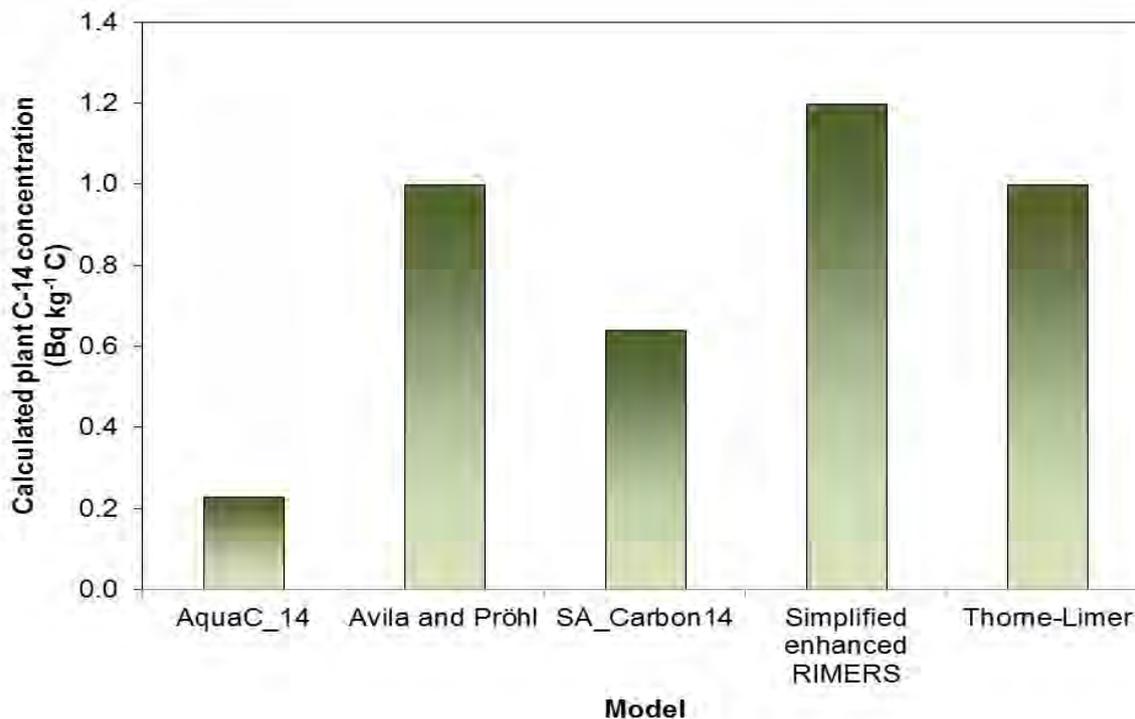


Figure 19 Calculated Plant C-14 Concentrations if the Atmosphere is Assumed to have a Concentration of 1 Bq kg⁻¹ C

7.6.4 Influence of other factors

In the calculations presented in this report, it is not only the source term which was varied. Participants also gave consideration as to the effects of the size of the field, height of crops, values of z_d , depth of soil layer, etc., on calculated C-14 concentrations in the soil, atmosphere and plants. Three of the five models are formulated such that the results associated with them will have a dependence upon the field size (see Section 3.6). Specifically, in AquaC_14, the Avila and Pröhl and SA_Carbon14 models there is a positive correlation between the field size assumed and the calculated C-14 concentrations in each of the model compartments (calculated C-14 concentrations in cereal are shown in Figure 20 as an example).

The calculated plant C-14 concentrations in the Avila and Pröhl and Thorne-Limer models are dependent upon the plant height and the position of z_d with respect to the plant height. In the Avila and Pröhl model, the correlation between plant height and calculated plant C-14 concentration is negative (e.g. Table 40), whilst for the Thorne-Limer model it is positive (e.g. Table 44). In the Avila and Pröhl model, increasing the plant height lowers the calculated ratio of C-14/C-12 in the atmosphere (see equations 56 and 61). In the Thorne-Limer model, increasing the plant height automatically both increases the thickness of the compartment of air which sees only diffusive air mixing and decreases the thickness of the compartment which is subject to turbulent mixing. Examination of Table 44 shows that such a change leads to an almost linear increase in the calculated C-14 concentration in the lower atmosphere compartment, with a less marked change in the calculated C-14 concentration in the upper atmosphere compartment.

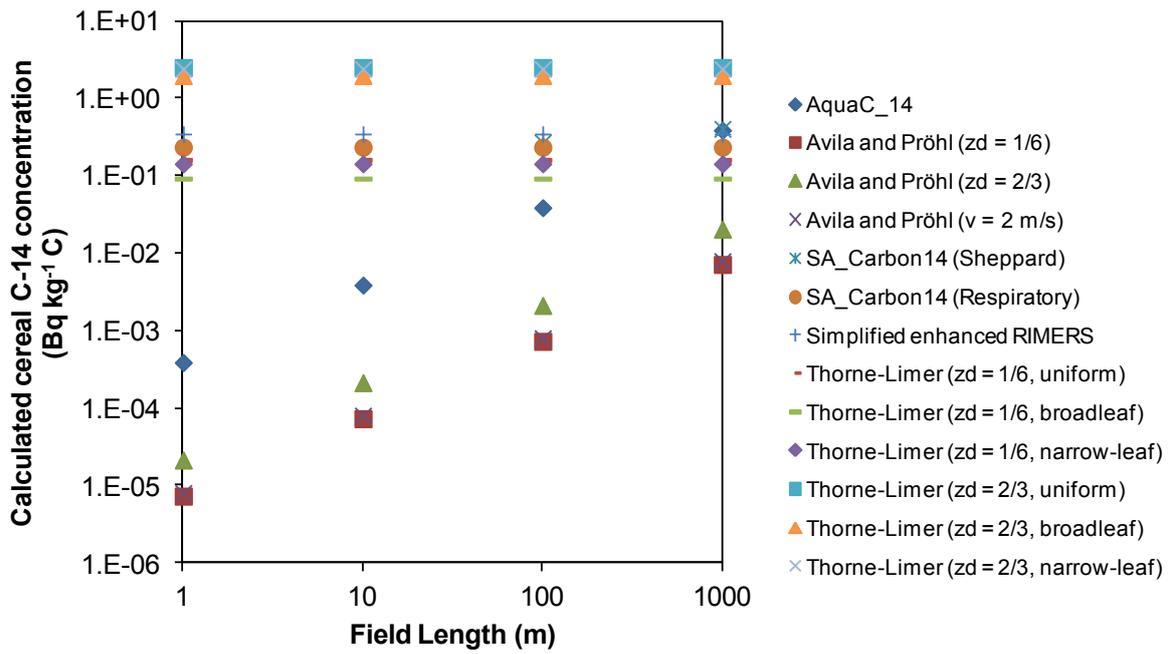


Figure 20 Effect of Field Size on Calculated Cereal C-14 Concentration for the Gas Release Scenario (Bq kg⁻¹ C)

8. DISCUSSION

8.1 OVERVIEW

As stated in the introduction, the overall objective of the project has been to investigate what are the key uncertainties in dose assessments for long-term releases into the biosphere of C-14 present in radioactive waste repositories. This project has compared quantitative estimates of the C-14 concentration in specific components of the dose assessment models (soil, plant-canopy atmosphere, plants), for agreed geosphere to biosphere release scenarios, the irrigation well tapping contaminated groundwater and gaseous release from below.

The FEP analysis, discussion of the models and examination of results highlights important differences in the conceptual models employed, which feed through to large differences in estimates of C-14 concentrations in different parts of the system. The differences and their significance are considered in relation to the major model subsections addressing C-14 behaviour in: the soil, the plant canopy atmosphere and the plant itself.

The results presented here show a considerable range in calculated C-14 concentrations in all the environmental media considered (soil, atmosphere, and plants). The largest variability is in the canopy atmosphere C-14 concentrations, which impact directly upon the calculated plant C-14 concentrations.

8.2 SPECIFIC CONSIDERATIONS OF THE ENVIRONMENTAL MEDIA

Within the soil subsystem it is possible to store a fraction of C-14 in recalcitrant organic pools that are not readily bioavailable. Such an approach is justified as phenomenological information indicates, for example, the formation of biologically unavailable humic substances. It is also possible to include more elaborate soil irrigation sub-models as SA_Carbon14 does, but the comparison with the simpler approach adopted in AquaC_14, where irrigation depends only on yearly averaged precipitation and evaporation with no distinction between plants shows only a small impact.

The conceptualisation of the canopy atmosphere varies between the models used in this study, and this is the cause of the majority of the variability in calculated plant C-14 concentrations. When the atmospheric C-14 concentration was fixed, the variability in calculated plant C-14 concentration for a given field size dropped from three or more orders of magnitude to less than a factor of five.

The uncertainty analysis carried out using AquaC_14 indicates 5.5 orders of magnitude between the minimum and maximum values for the canopy C-14 concentration, 1.5 for the soil and 3.3 for the plant concentrations; thus the uncertainty of one model based on input parameter uncertainty is not much higher than inter-model uncertainty. For SA_Carbon14, the stochastic calculations show that there are less than two orders of magnitude between the minimum and maximum. It should be noted that in the AquaC_14 uncertainty analysis the stable carbon content of the plants was allowed to vary, whereas it was not varied in the SA_Carbon14 stochastic calculations. However, this is a relatively limited source of uncertainty and is not sufficient to explain the difference in uncertainty ranges between the two models.

The final link in the sequence involves uptake into the plants; the uncertainty in the canopy atmosphere results are carried through into the plant concentration results. All modellers use the same isotope ratio approach with comparable stable carbon concentrations in both air and plant. Possible additional uncertainty linked to C-14 root uptake or translocation of leaf deposited bicarbonates does not generally show because these processes do not contribute more than 2% of plant carbon in any of the models. However, the effects of root uptake can be seen in the simplified enhanced RIMERS model (see Section 7.3).

8.3 OUTLOOK AND ONGOING WORK

Overall, the results presented in this study show clearly how important the conceptualisation of the dynamics of C-14 (and stable C) within the plant canopy atmosphere is upon the calculated plant C-14 concentrations. The approach of some models, in which the air the plant uses for photosynthesis is assumed to be subject to a relatively small degree of mixing, naturally leads to higher calculated plant C-14 concentrations than in the approach adopted by other models in which the air the plant uses is subject to a greater degree of mixing with uncontaminated air. Whilst the assumed field size (and thus fetch, irrespective of the chosen definition an organisation chooses to adopt) can, and does, play a role in determining the calculated atmospheric C-14 concentrations, in reality it is the assumed degree of openness of the canopy and the wind profile both within and above the plant canopy which are more likely to be the key drivers in determining the C-14 concentration in the CO₂ that the plants absorb for photosynthesis.

This study has provided information with respect to the workings of the models used by various waste management organisations and, thus, identified where key uncertainties lie and given some confidence for future model developments and application. Additional work is needed, however, to determine appropriate values of key parameters and test the conditions under which individual models, and model settings, may be most applicable. Such increased confidence will be built by consideration of the outcomes of this study with those of additional studies that are being funded by individual waste management organisations. Two examples are given below.

Independently of the results obtained in this inter-comparison, though the results presented here support the opinion, NDA RWMD has considered it to be important to develop an understanding of the transport of C-14 behaviour in soil-plant systems in field conditions [NDA RWMD, 2009]. However, use of significant amounts of C-14 in field plots is precluded by radiological protection considerations. Therefore, NDA RWMD has commissioned Serco and the University of Nottingham to undertake a programme using C-13 as a surrogate for C-14. Experimentally, this requires the use of isotope ratio mass spectroscopy (IRMS) to determine C-13/C-12 ratios. Currently, small-scale laboratory investigations are being carried out in the first year of the project. The laboratory experiments will provide guidance and confidence in the design of the field experiments, which will be carried out during the second and third years. These latter experiments will focus on the behaviour and fate of labelled CH₄ introduced into subsurface soil and its subsequent incorporation into vegetation. The field experiments will make use of methods developed as part of the Artificial Soil Gassing and Response Detection (ASGARD) facility situated on the University of Nottingham's Sutton Bonington campus. The equipment associated with this facility allows gases such as CO₂ and CH₄ (from cylinders or mains sources) to be supplied to experimental plots or containers to which gas flow is individually regulated by mass flow controllers before being injected from diffusive outlets situated below the ground surface. However, a virgin site at the University Farm will be used for this project to avoid issues of cross-contamination with other experimental work. The results from these experiments will provide information that will help to further bound the uncertainties regarded as having the most influence in the behaviour of C-14 in agricultural ecosystems, and as such support future safety assessment calculations relating to C-14.

SSM also recognise the importance of being able to model C-14 appropriately in terrestrial ecosystems, both from the context of geological disposal of solid radioactive wastes and also releases from operating nuclear power plants. To this end, building upon both internal interests and the BIOPROTA C-14 work, SSM is undertaking a detailed review of C-14 models that are used in both these areas to understand the requirements of models to address these assessment contexts, to investigate whether it is possible to develop a model which might be applicable to both.

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2012:47

The Swedish Radiation Safety Authority has a comprehensive responsibility to ensure that society is safe from the effects of radiation. The Authority works to achieve radiation safety in a number of areas: nuclear power, medical care as well as commercial products and services. The Authority also works to achieve protection from natural radiation and to increase the level of radiation safety internationally.

The Swedish Radiation Safety Authority works proactively and preventively to protect people and the environment from the harmful effects of radiation, now and in the future. The Authority issues regulations and supervises compliance, while also supporting research, providing training and information, and issuing advice. Often, activities involving radiation require licences issued by the Authority. The Swedish Radiation Safety Authority maintains emergency preparedness around the clock with the aim of limiting the aftermath of radiation accidents and the unintentional spreading of radioactive substances. The Authority participates in international co-operation in order to promote radiation safety and finances projects aiming to raise the level of radiation safety in certain Eastern European countries.

The Authority reports to the Ministry of the Environment and has around 270 employees with competencies in the fields of engineering, natural and behavioural sciences, law, economics and communications. We have received quality, environmental and working environment certification.

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