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# Transport of C-14 in terrestrial and aquatic environments

BIOPROTA project report

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## **BIOPROTA project report**

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This report concerns a study which was conducted by BIOPROTA. The conclusions and viewpoints presented in the report are those of the authors. SKB may draw modified conclusions, based on additional literature sources and/or expert opinions.

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# Preface

BIOPROTA is an international collaboration forum aiming to support resolution of key issues in biosphere aspects of assessments of the long-term impact of contaminant releases associated with radioactive waste and contaminated land management. It is understood that there are radioecological and other data and information issues that are common to assessments required in many countries. Collaboration through projects focused on mutual research needs 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](http://www.bioprota.org).

The general objectives of BIOPROTA are to make available the best sources of information to justify modelling assumptions made within long-term safety assessments. Particular emphasis is placed on key data for the assessment of long-lived radionuclide migration and accumulation in the biosphere, and the associated radiological impact, following discharge or release to the surface environment. 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 collaborative effort can efficiently be applied to finding solutions.

This report is the final deliverable in the 2021–2022 BIOPROTA work programme on ‘Transport of C-14 in Terrestrial and Aquatic Environments’. It details the findings from a literature review of the biogeochemical carbon cycle and its relevance in the development of models for assessing the radiological impacts of releases of C-14. It then goes on to describe the development of conceptual and mathematical models based on that review.

Financial support for the project was provided by the following BIOPROTA members: the French National Radioactive Waste Management Agency (ANDRA), the Swiss Federal Nuclear Safety Inspectorate (ENSI), Nuclear Waste Management Organization (NWMO, Canada), Nuclear Waste Services (NWS, UK), Posiva Oy (Finland), the Swedish Nuclear Fuel and Waste Management Company (SKB), and the Swedish Radiation Safety Authority (SSM).

The technical work has been supported by four project meetings including wider input from BIOPROTA members, and has been discussed at the 2022 annual BIOPROTA meeting. These wider contributions are acknowledged, as well as review of contributing draft material by BIOPROTA members. The report is presented as working material for information. The content may not be taken to represent the official position of the organisations involved. All material is made available entirely at the user’s risk.

The report has been published by SKB on behalf of the BIOPROTA forum, providing further quality assurance checks and a publication platform to facilitate its proper library registration and enhance its availability. SKB bears no responsibility for the content and the conclusions do not represent the official position of SKB.



## Executive Summary

The importance of using the best available scientific information in the development of assessment models of key radionuclides, such as C-14, has obvious advantages for the optimised design of radioactive waste management programmes. The avoidance of inappropriate caution in model assumptions has been an important driver in this study.

Understanding of physical and chemical forms of C-14 in relevant source terms was a key starting point for this study. Significant among the waste streams are those waste streams for which it is not clear which disposal option is most appropriate, such as graphite waste.

The importance of physical and chemical form then runs through the entire process of conceptual and mathematical model development, right up to the point of exposure. Noting this, an Appendix has been provided on the selection of appropriate dose coefficients that takes into account the latest scientific information relevant to the biokinetic models applied by the International Commission on Radiological Protection.

In the next part of the study, a review of the secondary literature on the distribution and transport of isotopes of carbon in the environment has provided the basis for development of a set of site-generic conceptual models of carbon distribution and transport in terrestrial and aquatic ecosystems. In turn, it is shown how the conceptual models for agricultural and freshwater ecosystems can be combined and used as the basis for a mathematical model that can be used to develop and parameterise a computational model of the environment that may be used for assessment purposes. The approach to model development has followed the approach suggested in output from programs of the International Atomic Energy Agency.

The amount of information on behaviour of carbon in the environment is enormous. In order to achieve a practical model solution, it is necessary to make simplifying assumptions. Simplifications are appropriate when the level of temporal or spatial discrimination is sufficient to meet the purposes of the assessment, as set out in the assessment context. However, where simplifications have been made and are not self-evident, comments have been included in the tables describing the conceptual models. Provision of such comments supports transparency of decisions on model assumptions. It also supports the independent auditing of the model and revision of the model as may be necessary if and when the assessment context changes.

In many assessment models used for C-14, the specific activities of various environmental media of relevance are assumed to be the same (or related through fixed ratios) and independent of time. These so-called specific activity models have advantages in simplicity of application. However, they commonly rely on assumptions about equilibrium over temporal and spatial scales and should not be applied without consideration of whether they are appropriate assumptions for the system being assessed. This approach can lead, in different circumstances, to under-estimation or over-estimation of assessed doses. The review carried out within this study provides information to allow judgements to be made about assumptions of equilibrium within various environmental systems, and thus can be used to support the justification for a specific-activity model.

Development of a computational model has not been undertaken at this stage. However, the literature reviewed provides a comprehensive basis for developing such a model that could be the subject for further collaborative research. Furthermore, the other conceptual models developed in this study provide a basis for developing mathematical and computational models appropriate to a wide variety of ecosystems of potential relevance in post-closure radiological impact assessments. In addition, it is hoped that the output provides the starting point for development of site-specific C-14 dose assessment models, especially for potential post-closure releases from solid waste disposal facilities.

In addition to consideration of modelling C-14 and carbon behaviour in the biosphere, the report includes an appendix discussing selection of dose coefficients appropriate to safety assessment. In particular, the appendix includes a description of developments in recommendations concerning dose coefficients for C-14.



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# 1 Introduction

C-14 is an important radionuclide for safety assessment studies associated with operational discharges as well as radioactive waste disposal. As C-14 has almost identical chemical characteristics to stable carbon, which is a fundamental component of organic molecules, C-14 requires special consideration when assessing potential consequences of releases to the biosphere. Over the past fifteen years, a sequence of BIOPROTA workshops, model-data and model-model comparison exercises have contributed to both improvements to, and helping to build confidence in, the representation of C-14 in assessments covering both short and long-term releases to the biosphere.

In this study, consideration is given to the transport and distribution of C-14 in terrestrial, freshwater and marine environments following its discharge into those environments either in effluents from operating facilities but also, particularly, because of possible releases from disposal facilities for solid radioactive wastes. The latter might occur for very extended periods compared with the period of operational discharges.

Discharges may occur to air, with subsequent deposition in the terrestrial environment, or, more usually for subsurface sources, to surface waters, including springs, streams, rivers and lakes, estuaries and marine waters, to soils, either in solution or in the gas phase, and to wetlands. Following discharge, C-14 is transported through the environment by a wide variety of aeolian, fluvial and biotic processes, and becomes integrated into the biogeochemical cycle of stable carbon. This results in the various carbon 'pools' in the environment exhibiting time-dependent changes in the specific activity (or C-14:C-12 ratio) of their contents. However, in many models used for C-14 dose assessment, this complex pattern of changes is simplified to a model in which the specific activities of various environmental media of relevance are assumed to be the same (or related through fixed ratios) and independent of time. These 'specific activity models' have advantages in simplicity of application. However, they commonly rely on assumptions about equilibrium over temporal and spatial scales and should not be applied without consideration of whether they are valid assumptions for the system being assessed. This approach can lead, in different circumstances, to under-estimation or over-estimation of assessed doses. It also fails to make use of site characterisation information that would typically be available, as needed for assessment of doses from other radionuclides.

The review undertaken within this study provides information to allow judgements to be made about assumptions of equilibrium for various environmental systems. Based on this review, the study then goes on to develop conceptual models of carbon transport in various types of environments and uses the conceptual models for terrestrial agricultural and surface-water environments to demonstrate how such conceptual models can be used in the development or auditing of mathematical models for the environmental transport and distribution of C-12, C-13 and C-14.

## ***Objectives, approach and structure***

The biogeochemical carbon cycle operates at various spatial and temporal scales. Although it is a closed, mass-conserving cycle at the global scale, at regional and local scales the import and export of carbon across the boundaries of the identified domain can be of great significance. Thus, for example, carbon in freshwater systems cannot be discussed in isolation from a consideration of carbon inputs from terrestrial catchments and outputs to the marine environment. Therefore, in this report, a nested approach is taken to reviewing the literature, with the largest spatial and temporal scales considered first to define the overall global cycle. Within this context, regional and local open cycles for different types of environments are then reviewed. In turn, specific aspects of those regional and local cycles of relevance in radiological impact assessments are examined in more detail. This approach permits the review to focus on topics or processes of specific interest, e.g. the turnover of carbon in freshwaters and its transport across the water-atmosphere interface, while ensuring that the relationship of those processes to the broader picture of carbon import, export, storage and recycling is maintained. This is particularly important when situations are considered in which the biogeochemical cycle is out of equilibrium at some spatial or temporal scale due, for example, to changing climate conditions, land-use changes or ecosystem succession.

Chapter 2 provides a review of literature relevant to modelling aspects of the biogeochemical carbon cycle. Specifically, literature relevant to the transport and distribution of C-14 in terrestrial, freshwater, estuarine and marine environments is reviewed. It also includes literature on stable carbon and on C-13:C-12 ratios, since this information can be used to infer how C-14 may be transported and accumulated in various environmental media, including biota.

Chapter 3 builds on the review to provide conceptual models for the transport and distribution of C-14 in the surface environment, with these conceptual models structured to give an appropriate basis for developing mathematical models that can be used to represent the transport of C-14 and its radiological impacts on human health and the environment.

Chapter 4 demonstrates how the conceptualisations can be used as a basis for developing mathematical models by illustrative application to a terrestrial agricultural environment coupled to a surface-water drainage system.

A wide range of environments is considered and how those environments can interact. However, the work presented here is site generic. The intention is that the output provide suitable background material for use in development of site-specific models. The approach to model development considers material presented in the BIOMASS methodology (IAEA 2003) and in its recent review and update (Lindborg et al. 2022).

Conclusions are provided in Chapter 5 and references are provided in Chapter 6.

Selection of dose coefficient appropriate to safety assessment is then discussed in an appendix. In particular, the appendix includes a description of developments in recommendations concerning dose coefficients for C-14.

## 2 Review of the behaviour of carbon in the biosphere

To provide context for the subsequent review, Section 2.1 describes source terms for C-14 to the biosphere relevant to dose assessments. The overall biogeochemical carbon cycle is summarised in Section 2.2. Transport and accumulation of carbon in different ecosystems is then described, covering:

- agricultural systems in Section 2.3,
- forest ecosystems in Section 2.4,
- wetland ecosystems in Section 2.5,
- freshwater ecosystems in Section 2.6, and
- estuarine and marine systems in Section 2.7.

Conclusions from the overall review are then provided in Section 2.8.

### 2.1 Source terms of interest for assessment purposes

Releases of C-14 from a disposal facility may enter the surface environment either dissolved in groundwater or in the gas phase. In respect of the gas phase, it is important to note that the bulk carrier gas may not be the same as the C-14 bearing gas. Potential bulk gases are hydrogen (typically arising from metal corrosion occurring under reducing conditions), carbon dioxide and methane. In respect of C-14 bearing compounds that may be present in either the gaseous or liquid phases, the degradation of cellulosic materials may yield a variety of organic compounds. In aerobic environments, cellulosic materials will largely oxidise to carbon dioxide, but in anaerobic environments carbon dioxide and methane will be produced in approximately equimolar amounts. In cementitious environments, some C-14 bearing isosaccharinic acid could be formed. Methane could also be produced in small amounts from microbial interactions with barium-carbonate sludges (RWM 2015).

A potentially important source of C-14 is graphite wastes that arise mainly from gas-cooled reactors. As discussed by the IAEA (2004), various neutron activation reactions can produce C-14 in nuclear power reactors. In UK Advanced Gas-Cooled (AGR) reactors, the graphite moderator is the main source. Leaching experiments indicate that the main C-14 bearing form released from graphite is carbon dioxide, with small amounts of carbon monoxide and volatile organics (likely dominated by methane) (RWM 2015).

Corrosion of metals can also give rise to C-14 bearing compounds. Thorne and MacKenzie (2005) pointed out that C-14 may be present in metallic uranium as uranium carbide. It has been reported that this compound is hydrolysed to 86 % by volume methane, 11 % hydrogen and 1.8 % ethane, with small quantities of saturated C<sub>3</sub>-C<sub>6</sub> hydrocarbons (Rees et al. 2003). In Magnox cladding, C-14 may be present as magnesium carbide (MgC<sub>2</sub>), and this could react with water to give rise to C-14 bearing acetylene. This acetylene could react with corrosion-derived hydrogen to form ethylene and, possibly, ethane. However, there is no evidence that these various processes would occur in practice (Rees et al. 2003). Thorne (2005) pointed out that the proposal that magnesium carbide might be present assumes that both nitrogen atoms in the impurity MgN<sub>2</sub> are converted to C-14. It seems much more likely that only one would be converted giving magnesium cyanide, Mg(CN)<sub>2</sub>. Under anoxic conditions, this could react with water to give hydrocyanic acid, but under oxidising conditions C-14 bearing (CN)<sub>2</sub> might form.

In respect of iron and steel, Hicks et al. (2003) demonstrated that methane, ethane, propane, propene, butane and C<sub>3</sub> compounds could be produced in iron-water systems. The authors attribute the formation of these compounds to reduction of aqueous carbon dioxide at the metal surface, but production from within the metal is not ruled out.

In a review of published and unpublished literature, Nuclear Technologies plc (2005) found that, in the case of aluminium, C-14 would be released as methane, or another insoluble, reduced gaseous form. In the case of uranium, they concluded that C-14 would be released as hydrocarbons (predominantly methane). For Magnox, Zircaloy and steels, Nuclear Technologies plc (2005) considered that there were insufficient data available on which to draw confident conclusions. Spent ion-exchange resins might give rise to small amounts of C-14 bearing formate, acetate and carbonates (Magnusson 2007).

C-14 may be released to atmosphere from reprocessing (as discussed in Section 2.3.3 for la Hague) or from incineration of graphite wastes. However, off-gases can be treated to capture C-14, yielding, for example, a BaCO<sub>3</sub> sludge that can be further treated to produce a solid waste (IAEA 2004).

More recent information on potential C-14 source terms was presented by Maryna Surkova (FANC) in Limer (2019). This presentation related to the EU-sponsored CAST (CARbon-14 Source Term) Project. The scope of the project was to increase understanding in the potential release mechanisms of C-14 from radioactive waste materials, considering: wastefrom dissolution and C-14 leaching, waste package corrosion, and the long-term evolution of the disposal system. By increasing understanding of these potential release mechanisms, the high-level objectives of the project were to decrease uncertainties in the long-term safety of radioactive waste disposal and increase confidence in safety cases for such disposal.

The project had work packages investigating steels, Zircaloy, ion-exchange resins (IEX) and graphite. For each of these, work was undertaken to better characterise the source term with respect to inventory, release rates and speciation.

With respect to steels, the objectives were to develop analytical techniques for identification and quantification of C-14 species formed during corrosion of irradiated steels under conditions relevant to cement-based repositories. The steels work package team also sought to validate existing activation models by measuring C-14 inventories in irradiated steel and carrying out experiments and modelling to improve the understanding of the speciation and release rate of C-12/C-13 and C-14 from corrosion of steels. The main outcomes from this work package were that the C-14 inventory can be calculated from the nitrogen content of the steel and other metallic minerals together with the neutron flux during reactor operations. The neutron flux and energy spectrum depends on location in the reactor and this affects the production of C-14. This spatially dependent production can be modelled mathematically. However, there are often uncertainties associated with the amount of nitrogen in the steel, which can differ significantly from the amount reported by the manufacturer, and the exact position of the component in the reactor. This can lead to discrepancies of up to 300–400 % between the modelled and measured C-14 inventories. There is limited information on the fate and speciation of C-14 released under disposal conditions. However, Mibus et al. (2018) comment that there is a congruent release from activated steels with corrosion, mainly in the gas phase as C-14-methane.

With respect to Zircaloy, the objectives were to obtain a better understanding of C-14 behaviour in fuel cladding and the speciation of released C-14. In contrast to the situation with steels, there was found to be good agreement between the modelled and measured C-14 inventories in Zircaloy. It was noted that the type of Zircaloy influences the total C-14 inventory and its speciation. The C-14 release is mainly dominated by the oxide. With respect to C-14 speciation, the organic fraction is more important than the inorganic, with the speciation evolving with time. Several methods were used to investigate the corrosion rates of Zircaloy, which found that irradiated Zircaloy generally corroded more rapidly than unirradiated Zircaloy. In contrast, Sakuragi (2017) reported that corrosion rates of activated zirconium were lower than those of non-activated zirconium. However, the underlying processes are still not fully understood, with more work being undertaken.

With respect to ion-exchange resins, it was noted that spent ion-exchange resins are almost always mixed in storage tanks at the nuclear facility. Their ionic load and activity depend on chemical or radiochemical saturation, maximum service lifetime, unusual pressure drop behaviour (clogging); and the fluid quality (becoming insufficient).

The CAST project found that there is little information regarding speciation of C-14 accumulated in spent ion-exchange resins. However, some unexpected partitioning of the C-14 between inorganic (carbonate) and organic fractions (predominantly small molecular weight molecules such as formate, acetate and oxalate) was identified, with most of the C-14 activity arising from the inorganic part. The CAST review noted that there are no published data regarding the leaching of organic molecules from spent ion-exchange resins.

With respect to graphite, the objectives of CAST were to understand the factors determining the release of C-14 from irradiated material. This was achieved by determining the C-14 inventory in graphite and its spatial distribution, measuring the release rate and speciation of C-14 to solution and gas in contact with aqueous solutions, and determining the impact of selected waste treatment options on C-14 release. From this study, it was determined that a substantial fraction of the C-14 in graphite will not be releasable on decadal to centennial timescales. That which can be released will have some quick and some slower release mechanisms, with location in the graphite having the potential to affect the release rates

In summary, the results from CAST do not alter the conclusions that could be drawn from the information previously available. Overall, the two principal C-14 bearing compounds of interest as source terms to the biosphere are carbon dioxide and methane either dissolved in groundwater or carried by bulk gas. Other C-14 bearing compounds that could enter the biosphere in minor amounts include carbon monoxide, ethane, acetylene, various C<sub>1</sub> to C<sub>5</sub> hydrocarbons, compounds containing C-N bonds (e.g. amines and cyanides), formate, acetate and other carboxylates, carbonates, and isosaccharinic acid. In the remainder of this review, only releases of carbon dioxide and methane are addressed in detail, with some supplementary remarks on carbon monoxide because of its potential to be more efficiently taken up by plants than is carbon dioxide.

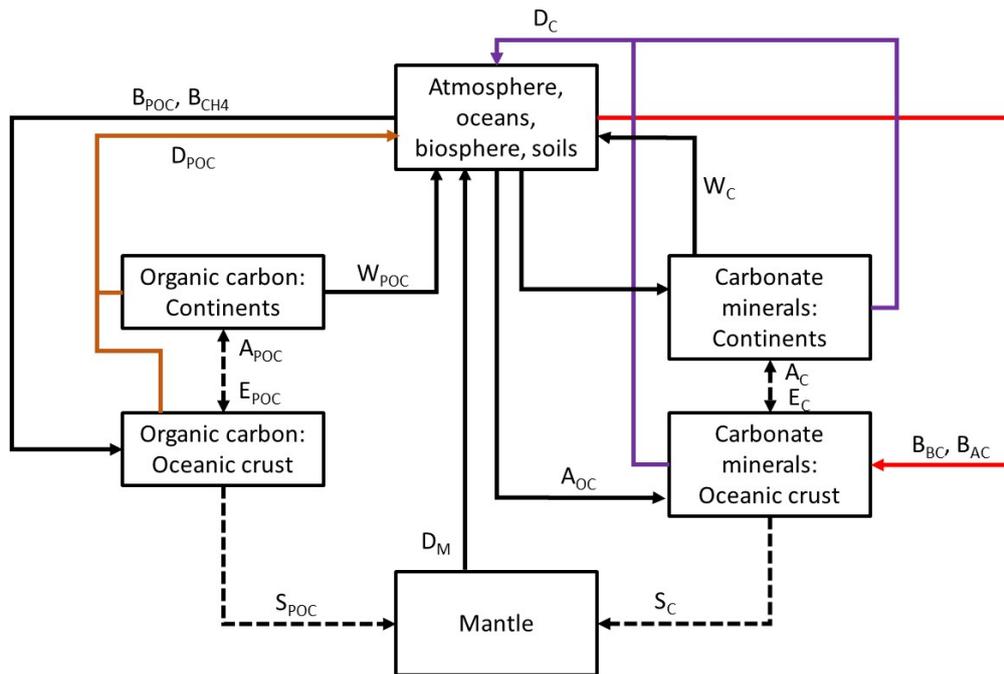
The various physical and chemical forms of C-14 present in waste discharges and solid wastes have implications for the assessment of doses for different discharge and disposal routes. This information is therefore relevant to model development. It also has relevance in the selection of appropriate dose coefficients to be used in different exposure pathways (see Appendix A). Recent revisions to these dose coefficients, based on the latest scientific information, are significant.

## **2.2 The overall global biogeochemical carbon cycle**

### **2.2.1 The combined organic and inorganic carbon cycle**

At the coarsest level, the global biogeochemical carbon cycle can be distinguished into four pools. These comprise carbon in the biosphere, organic carbon present in sediments of the geosphere that can be recycled to the biosphere or may be subducted into the mantle, which comprises the third major pool. The fourth is carbonate minerals derived from carbon released by rock weathering and mainly precipitated and deposited in the deep ocean. These carbonate minerals, like organic carbon may be subducted into the mantle. The carbon cycle is closed by volcanic transfers of carbon from the mantle to the biosphere. This overall cycle has been summarised by Wallmann and Aloisi (2012), as illustrated in Figure 2-1.

Wallmann and Aloisi (2012) estimate the sizes of the various global carbon pools during the Holocene, but before significant perturbations by humans. These pools are listed in Table 2-1.



**Key:**

- $B_{POC}$  – burial of POC in marine sediments,
- $B_{CH4}$  – burial of methane in marine sediments,
- $B_{BC}$  – burial of biogenic carbonates in marine sediments,
- $B_{AC}$  – burial of authigenic carbonates in marine sediments,
- $D_{POC}$  – metamorphic degassing of POC,
- $D_C$  – metamorphic degassing of carbonate minerals.
- $D_M$  – degassing of the mantle,
- $W_{POC}$  – weathering of POC on the continents,
- $W_C$  – weathering of carbonates on the continents,
- $A_{OC}$  – alteration of oceanic crust,
- $E_{POC}$  – erosion and transport of POC to the ocean,
- $A_{POC}$  – accretion of POC during continental collision,
- $E_C$  – erosion and transport of carbonate minerals to the ocean,
- $A_C$  – accretion of carbonate minerals during continental collision,
- $S_{POC}$  – subduction of POC in the mantle,
- $S_C$  – subduction of carbonate minerals in the mantle.

**Figure 2-1.** The geological carbon cycle (redrawn from Wallmann and Aloisi 2012). Fluxes discussed in detail and quantified by them are shown with solid arrows. Note that the weathering of silicate minerals is neglected in this figure.

**Table 2-1. Global pools of carbon (adapted from Wallmann and Aloisi 2012).**

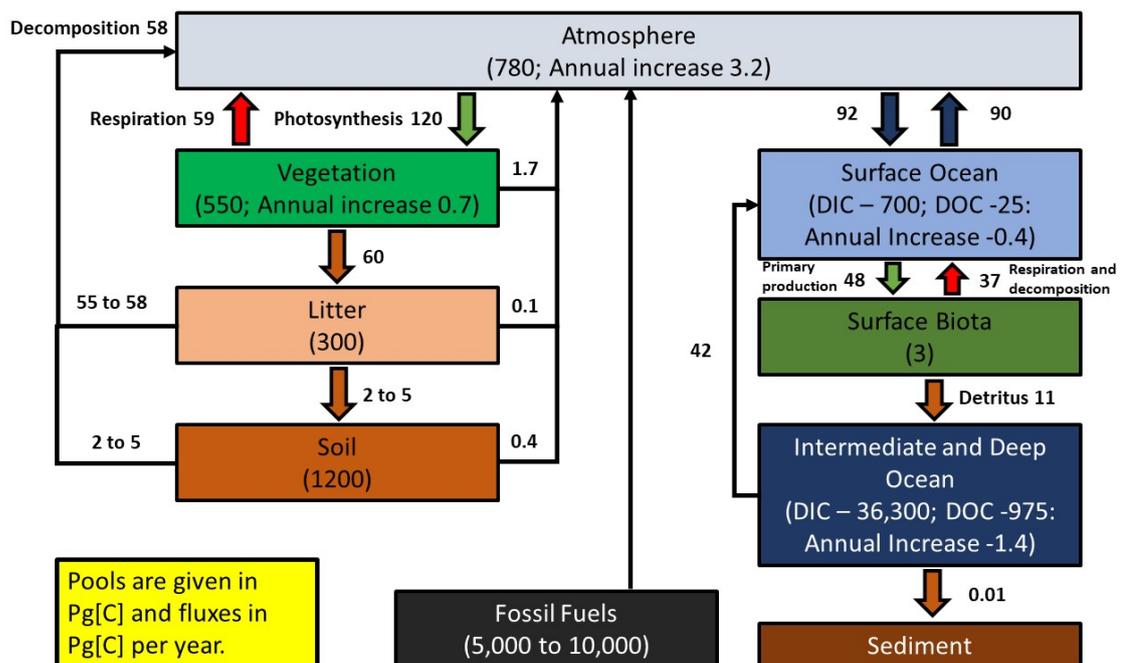
Carbon Pool	Amount (Units of $10^{18}$ mol[C])
Carbonate carbon in rocks	5000
Organic carbon in rocks	1250
Methane in gas hydrates	0.1 to 0.3
Carbon in soil	0.3
Inorganic carbon dissolved in the deep ocean (> 100 m depth)	2.7
Inorganic carbon dissolved in the upper ocean (< 100 m depth)	0.1
Atmospheric carbon dioxide	0.06
Terrestrial biota	0.05
Marine biota	0.0005

Carbon fluxes associated with both organic and inorganic carbon that applied during the earlier part of the Holocene are listed in Table 2-2.

**Table 2-2. Global fluxes of carbon ( $\text{Tmol}[\text{C}] \text{y}^{-1}$ ) during the early Holocene (from Wallmann and Aloisi 2012). Note that sources of carbon dioxide can be sinks for bicarbonate and vice versa, see also Figure 2-3).**

Organic Carbon		Inorganic Carbon	
<i>Land</i>		<b>Carbon dioxide sources</b>	
Carbon dioxide fixation in living biomass	4 700	Mantle degassing ( $D_M$ )	3.1 to 5.5
Accumulation of terrestrial organic carbon in vegetation, soil and detritus	33	Metamorphism of carbonates ( $M_C$ )	2.0 to 4.0
Export of terrestrial organic carbon to the oceans	33	Biogenic carbonate burial ( $B_{BC}$ )	20.0
Weathering of fossil organic carbon ( $W_{POC}$ )	8 to 16	<b>Carbon dioxide sinks</b>	
<i>Oceans</i>		Silicate weathering on land	10 to 11
Carbon dioxide fixation in living biomass	4 040	Silicate weathering in marine sediments	3.3 to 13.3
Export of marine biomass into the deep ocean	800	Carbonate weathering ( $W_C$ )	12.3
Rain of particulate organic matter to the seafloor	190	<b>Bicarbonate sources</b>	
Burial of organic matter in surface sediments	10 to 65	Carbonate weathering ( $W_C$ )	24.6
Burial of organic matter, authigenic carbonate and methane in deep sediments ( $B_{POC}+B_{AC}+B_{CH_4}$ )	5.4 to 27	Silicate weathering on land	10 to 11
		Silicate weathering in marine sediments	3.3 to 13.3
		<b>Bicarbonate sinks</b>	
		Biogenic carbonate burial ( $B_{BC}$ )	40.0
		Alteration of oceanic crust ( $A_{OC}$ )	1.5 to 2.4

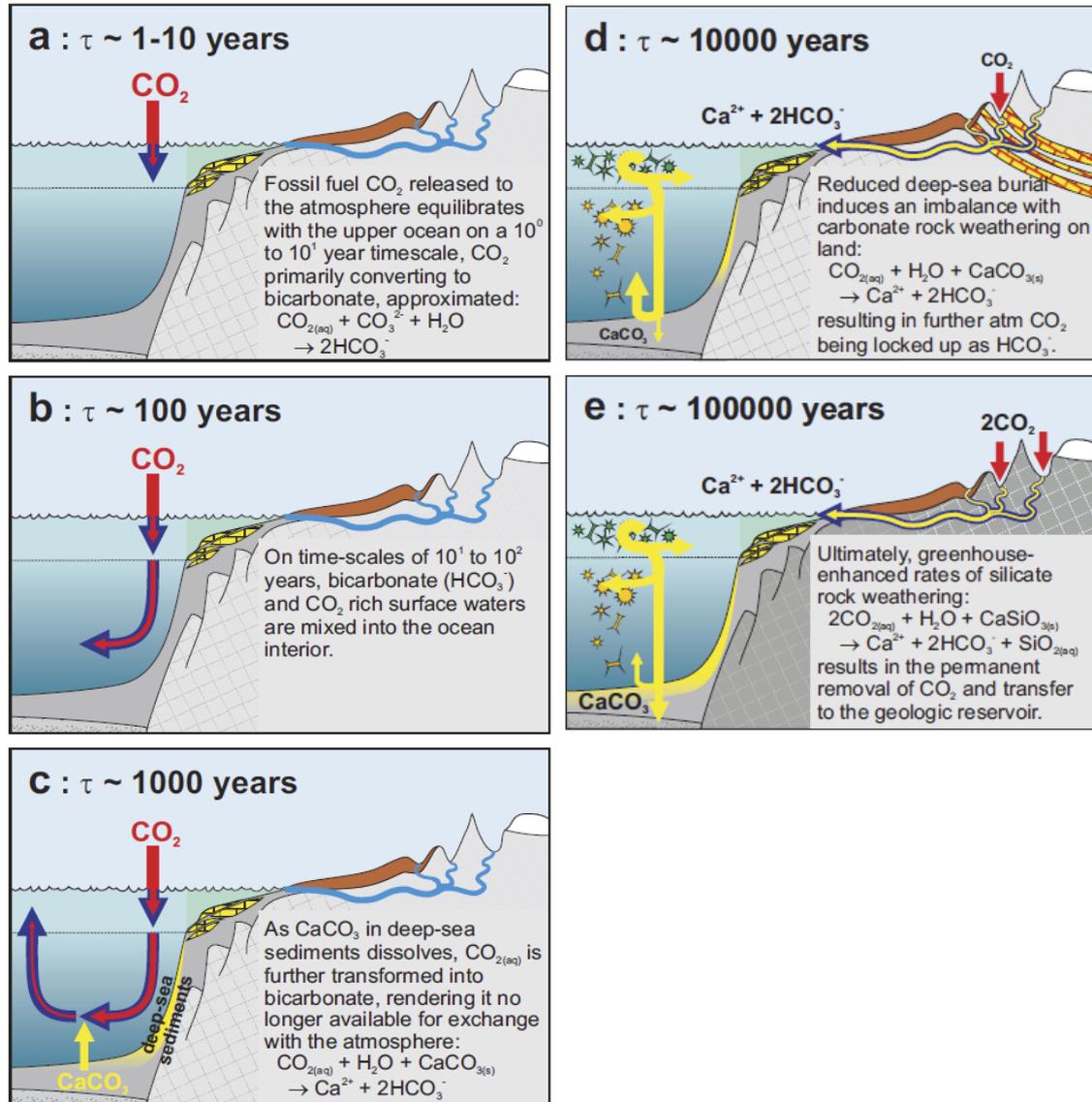
On the timescales of interest in this study, which range up to about a hundred thousand years, depending on the type of disposal system under consideration, the recycling of carbon from the mantle can be neglected. However, anthropogenic inputs of carbon into the surface environment cannot be neglected and result in an overall imbalance of the biogeochemical carbon cycle. That cycle has been characterised quantitatively by Houghton (2005), as illustrated in Figure 2-2.



**Figure 2-2.** The contemporary global carbon cycle (redrawn from Houghton 2005). Note that the minor pools associated with terrestrial animals and the various forms of carbon in freshwaters are not included in this simplified scheme.

The kinetics of the perturbation to the carbon cycle due to anthropogenic releases of carbon dioxide has been illustrated by Lord et al. (2015a, 2015b) and is shown in Figure 2-3. Timescales of relevance range from a few years, for atmosphere-ocean exchanges up to 100 ka or more for processes affected by the rate of weathering of silicate rocks.

Wallmann and Aloisi (2012) reviewed the various carbon fluxes described above and developed a balanced flux applicable over the last one million years. Specifically, they balanced carbon sources against carbon sinks, and balanced carbon dioxide sources against carbon dioxide sinks. Results of their analysis are given in Table 2-3. Note that the individual sources and sinks are largely the same for both carbon and carbon dioxide, but that the balance equations differ.



**Figure 2-3.** The kinetics of transfers of carbon released by anthropogenic processes (Lord et al. 2015b, ©2015 the original authors and reproduced with permission).

**Table 2-3. Mean carbon fluxes over the last one million years (Tmol[C] y<sup>-1</sup>).**

Source or Sink	Symbol	Flux Range	Balanced Flux
<b>Carbon sources</b>			
Mantle degassing	D <sub>M</sub>	3.1 to 5.5	4.3
Metamorphic degassing of carbonate rocks	D <sub>C</sub>	2.0 to 4.0	2.5
Chemical weathering of carbonate rocks	W <sub>C</sub>	10 to 16	11.7
Metamorphic degassing of POC	D <sub>POC</sub>	0.4 to 0.6	0.5
Chemical weathering of POC	W <sub>POC</sub>	8 to 16	9.0
<b>Total</b>			<b>28.0</b>
<b>Carbon sinks</b>			
Burial of biogenic carbonate at the seafloor	B <sub>BC</sub>	14 to 17	16.0
Alteration of oceanic crust	A <sub>OC</sub>	1.5 to 2.4	2.0
Burial of POC, authigenic carbonates and methane in marine sediments	B <sub>POC</sub> +B <sub>AC</sub> +B <sub>CH4</sub>	5.4 to 27	10.0
<b>Total</b>			<b>28.0</b>
<b>Carbon dioxide sources</b>			
Mantle degassing	D <sub>M</sub>	3.1 to 5.5	4.3
Metamorphic degassing of carbonate rocks	D <sub>C</sub>	2.0 to 4.0	2.5
Metamorphic degassing of POC	D <sub>POC</sub>	0.4 to 0.6	0.5
Chemical weathering of POC	W <sub>POC</sub>	8 to 16	9.0
Burial of biogenic carbonate at the seafloor	B <sub>BC</sub>	14 to 17	16.0
<b>Total</b>			<b>32.3</b>
<b>Carbon dioxide sinks</b>			
Chemical weathering of silicate rocks on land	W <sub>CSIL</sub>	6 to 10	7.1
Chemical weathering of silicates in marine sediments	W <sub>OSIL</sub>	3.3 to 13.3	3.5
Chemical weathering of carbonate rocks	W <sub>C</sub>	10 to 16	11.7
Burial of POC, authigenic carbonates and methane in marine sediments	W <sub>POC</sub> +W <sub>AC</sub> +W <sub>CH4</sub>	5.4 to 27	10.0
<b>Total</b>			<b>32.3</b>

### 2.2.2 The carbon cycle in terrestrial ecosystems

Because of their significance in most radiological impact assessment studies, it is appropriate to expand on the simplified representation of the terrestrial environment shown in Figure 2-2. The overall carbon cycle in terrestrial ecosystems has been summarised in detail in Chapter 5 of Schlesinger and Bernhardt (2020). The key considerations are carbon sequestration from atmospheric carbon dioxide by photosynthesis, carbon release from biomass as carbon dioxide by respiration, and carbon release from biomass during its decomposition. Other important considerations are net primary production of biomass and the fate of that net primary production, including accumulation in, and release from, organic matter in soils.

The overall reaction for photosynthesis is  $\text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{CH}_2\text{O} + \text{O}_2$ . This process occurs in two stages. First capture of light energy allows water molecules to be split and high-energy molecules to form. This drives the second reaction, mediated by ribulose biphosphate carboxylase (Rubisco), in which  $\text{CO}_2$  is added to carbohydrates. The overall photosynthesis reaction is the same for C3 and

C4 plants.<sup>1</sup> Although C3 plants predominate in global net primary production, C4 plants may account for up to 23 % of the total. However, because C4 plants are largely warm-climate grasses, they are not woody and their contribution to global biomass is small (Schlesinger and Bernhardt 2020). The availability of water and light are key controls on rates of photosynthesis, but C3 and C4 plants have different water-use efficiencies, as well as different degrees of C-13:C-12 isotopic fractionation.

Water-use efficiency (WUE) in plants is defined as the ratio of moles of CO<sub>2</sub> fixed per mole of H<sub>2</sub>O lost. It typically ranges from 0.86 to 1.5 depending on environmental conditions. WUE depends on the atmospheric concentration of CO<sub>2</sub>, increasing as the concentration increases. It is also correlated with leaf nitrogen content because nitrogen content is a measure of enzyme content and Rubisco accounts for 20 to 30 % of leaf nitrogen (Schlesinger and Bernhardt 2020).

Plant respiration is largely the result of mitochondrial activity and is, therefore, also correlated with the nitrogen content of plant tissues. Photosynthesis masks respiration during the day. Also, for leaf tissues, mitochondrial respiration is lower during the day than at night but is supplemented by photorespiration that eliminates the toxic compound phosphoglycolate produced in a competitive reaction of Rubisco with oxygen (Schlesinger and Bernhardt 2020).

Whereas photosynthesis occurs in above-ground plant parts, respiration occurs in all tissues, including roots. Because of root respiration, carbon dioxide is released into the soil from the roots, and carbon dioxide concentrations in the soil atmosphere are typically much higher than in the above-ground atmosphere. The implications of this for radiological impact assessment calculations is taken up in subsequent sections of this review.

About 50 % of carbon fixation by photosynthesis is respired by plants, and plant respiration tends to increase with increasing temperature, accounting for high rates of respiration in tropical forests. In contrast, rates of photosynthesis are only weakly dependent on temperature (Schlesinger and Bernhardt 2020).

Net primary production is the difference between gross primary production and plant respiration. It is often expressed as kg[dry mass] m<sup>-2</sup> y<sup>-1</sup>. However, as plant dry mass is typically 45 to 50 % carbon, it can also be readily expressed as kg[C] m<sup>-2</sup> y<sup>-1</sup>. Net primary productivity typically increases as a function of intercepted radiation, but only a small fraction of the total energy received in sunlight is used for biomass production (about 1 % in forests and < 5 % for crops). Net primary production is also related to the rate of input of precipitation. Normally, the relationship is  $2 \times 10^{-3}$  kg[C] m<sup>-2</sup> per mm of rainfall, but is lower in barren, arid regions, where a large proportion of the precipitation rapidly evaporates (Schlesinger and Bernhardt 2020).

Allocation of net primary production varies with vegetation type and age. In forests, 25 to 35 % of above-ground production is found in leaves. In shrublands this rises to 35 to 60 % and in grassland communities, essentially all net primary production is found in photosynthesising tissues. Across a wide range of species, the allocation of photosynthate to leaf growth relative to stem growth is 0.53 (Schlesinger and Bernhardt 2020).

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<sup>1</sup> Most crops are C3 plants, i.e. the first carbon compound produced during photosynthesis contains three carbon atoms. However, at high temperatures and under intense light conditions, oxygen has a high affinity for the photosynthetic enzyme Rubisco. Therefore, in these conditions, oxygen can bind to Rubisco instead of carbon dioxide, and through photorespiration, oxygen reduces C3 plant photosynthetic and water-use efficiencies. In environments with high temperatures and light intensities, that tend also to have soil moisture limitations, some plants evolved C4 photosynthesis. A unique leaf anatomy and biochemistry enables C4 plants to bind carbon dioxide when it enters the leaf and to produce a 4-carbon compound that transfers and concentrates carbon dioxide in specific cells around the Rubisco enzyme, significantly improving both photosynthetic and water-use efficiencies. This means that in high temperature and intense light environments, C4 plants tend to be more productive than C3 plants. Examples of C4 plants include corn, sorghum, sugarcane, millet, and switchgrass. However, the C4 anatomical and biochemical adaptations require additional plant energy and resources than C3 photosynthesis, and so in cooler environments, C3 plants are typically more photosynthetically efficient and productive.

Because measurements of roots are difficult, many studies estimate net primary production only for above-ground parts of plants. However, a general rule is that root biomass contributes 20 to 40 % of total biomass, and that root growth uses 15 to 25 % of net primary productivity. In addition, root exudates can account for up to 20 % of net primary productivity in some circumstances (Schlesinger and Bernhardt 2020).

Net primary productivity contributes to the accumulation of biomass, but biomass is, in turn, lost by processes such as leaf fall, grazing and plant death. Net ecosystem production is defined as net primary production less losses to herbivores and decomposers. Net ecosystem production is typically positive in young temperate and boreal forests, ranging from 0.12 to 0.75 kg[C] m<sup>-2</sup> y<sup>-1</sup> (see Table 5.2 of Schlesinger and Bernhardt 2020).

Globally, net primary production is estimated as 60 to 70 Pg[C] y<sup>-1</sup>, with the spatial distribution of that production readily mapped using satellite data. Net primary production and standing biomass for various biomes are given in Table 2-4.

**Table 2-4. Net primary production and standing biomass for various biomes (from Table 5.3 of (Schlesinger and Bernhardt, 2020)).**

Biome	Area (10 <sup>6</sup> km <sup>2</sup> )	Net primary productivity (kg[C] m <sup>-2</sup> y <sup>-1</sup> )	Total net primary productivity (Pg[C] y <sup>-1</sup> )	Biomass (kg[C] m <sup>-2</sup> )	Total plant carbon pool (Pg[C])
Tropical forests	17.5	1.250	20.6	19.400	320
Temperate forests	10.4	0.775	7.6	13.350	130
Boreal forests	13.7	0.190	2.4	4.150	54
Mediterranean shrublands	2.8	0.500	1.3	6.000	16
Tropical savanna/grassland	27.6	0.540	14.0	2.850	74
Temperate grasslands	15.0	0.375	5.3	0.375	6
Deserts	27.7	0.125	3.3	0.350	9
Arctic tundra	5.6	0.090	0.5	0.325	2
Crops	13.5	0.305	3.9	0.305	4
Ice	15.5	-	-	-	-
<b>Total</b>	<b>149.3</b>	-	<b>58.9</b>	-	<b>615</b>

Thus, globally, with a total plant carbon pool of 615 Pg[C] and a total net primary productivity of 58.9 Pg[C] y<sup>-1</sup>, the mean residence time of carbon in the terrestrial plant carbon pool is 615/58.9 = 10.4 years. However, it can be substantially longer in forests, due to localisation of much of the carbon in the wood of trees.

Although grazing, fires and losses of volatile organic compounds from the soil all represent losses of net primary productivity from terrestrial ecosystems, the dominant process is the delivery of dead organic matter to soil as detritus. This is then subject to decomposition leading to the release of CO<sub>2</sub> and the formation of humus that accumulates in the lower soil profile. In many systems, decomposition shows a rapid initial phase with a timescale of less than one year, followed by one or more slower phases, with some material persisting for decades or longer. Decomposition rates are typically determined by temperature, moisture and the chemical composition of the litter. The total global soil carbon pool (excluding soil carbonates) is 2344 Pg[C] and soils from various biomes typically contain 20 to 45 kg[C] m<sup>-2</sup> (see Table 5.4 of Schlesinger and Bernhardt 2020). With a pool size of 2344 Pg[C] and a net primary productivity of 58.9 Pg[C] y<sup>-1</sup>, the mean residence time of carbon in soil is estimated to be 2344/58.9 = 39.8 years.

## 2.3 Transport and accumulation of carbon in agricultural ecosystems

As noted in Section 2.1, releases of C-14 from a disposal facility, as carbon dioxide or methane, may enter the soils of agricultural ecosystems either dissolved in groundwater or in the gas phase. The following sections describe the behaviour of both carbon dioxide and methane in agricultural soils.

### 2.3.1 The solubility of carbon dioxide and methane in soil water

When C-14 bearing carbon dioxide and methane enter the soil, they will partition between soil solution and soil atmosphere in proportions that depend upon their solubilities. Solubilities are expressed in various ways, but here the definition used by Sander (2015) in his compilation is adopted, i.e.

$$H^{\text{cp}} = c_a/P$$

where  $H^{\text{cp}}$  ( $\text{mol m}^{-3} \text{ Pa}^{-1}$ ) is the solubility coefficient,  $c_a$  ( $\text{mol m}^{-3}$ ) is the concentration in the aqueous phase and  $P$  (Pa) is the partial pressure of the compound in the gas phase. However, it is also useful to express the coefficient as a dimensionless ratio between the gas-phase concentration and the aqueous-phase concentration,  $K_{\text{H}}^{\text{cc}}$  (Sander 2015), given by:

$$K_{\text{H}}^{\text{cc}} = 1/H^{\text{cp}}RT$$

where  $R$  is the gas constant ( $8.314 \text{ J mol}^{-1} \text{ K}^{-1}$ ) and  $T$  is the temperature (K).

Values for  $H^{\text{cp}}$  and  $K_{\text{H}}^{\text{cc}}$  for carbon dioxide and methane at temperatures appropriate to soils in various climate zones are given in Table 2-5.

**Table 2-5. Solubilities of carbon dioxide and methane in water (Sander 2015).**

T (K)	$H^{\text{cp}}$ ( $\text{mol m}^{-3} \text{ Pa}^{-1}$ )		$K_{\text{H}}^{\text{cc}}$ (-)	
	Carbon dioxide	Methane	Carbon dioxide	Methane
273.15	6.89E-4	2.36E-5	0.639	18.7
278.15	5.89E-4	2.11E-5	0.735	20.5
283.15	5.05E-4	1.89E-5	0.840	22.4
288.15	4.36E-4	1.71E-5	0.957	24.5
293.15	3.79E-4	1.54E-5	1.08	26.6
298.15	3.30E-4	1.40E-5	1.22	28.8
303.15	2.89E-4	1.27E-5	1.37	31.1
308.15	2.54E-4	1.16E-5	1.54	33.5
313.15	2.24E-4	1.07E-5	1.71	36.1
318.15	1.99E-4	9.78E-6	1.90	38.6
323.15	1.77E-4	9.01E-6	2.10	41.3

From Table 2-5, carbon dioxide has a much higher solubility in water than does methane. Therefore, when carbon dioxide enters the soil zone either in the gas phase or in solution, it will rapidly equilibrate such that the volumetric concentrations in soil solution and the soil atmosphere are similar. In contrast, methane entering the soil zone in the gas phase will largely remain in the gas phase, whereas methane entering the soil zone in solution will have a strong tendency to outgas into the soil atmosphere.

### 2.3.2 The oxidation of methane in soil

The oxidation of methane in soils has been comprehensively reviewed by Shaw and Thorne (2016) and by Murguia-Flores et al. (2018). Oxidation of  $\text{CH}_4$  can occur both in the presence and absence of oxygen, although anaerobic oxidation (which can use various chemical species including sulphate, nitrate, nitrite, iron and manganese as a terminal electron acceptor) appears to be limited to marine sediments (Magonigal et al. 2005). Under aerobic conditions, oxidation of  $\text{CH}_4$  is carried out by a

group of gram-negative bacteria known as the methanotrophs. On a global scale it is estimated that oxidative 'consumption' by soil methanotrophs removes 15 to 45 MT (megatonnes) of CH<sub>4</sub> from the atmosphere each year (Schlesinger 1997).

Methanotrophic bacteria metabolise CH<sub>4</sub> as their sole source of carbon and energy, typically according to the reaction  $\text{CH}_4 + \text{O}_2 \rightarrow \text{CO}_2 + 2\text{H}_2\text{O}$ , though they may also use other oxygen donors such as nitrite or nitrate.

Methane is made available for methanotrophy within aerobic soils by CH<sub>4</sub> production (methanogenesis) in deeper anoxic soil or anoxic micro-niches, such as the centres of wet soil crumbs, by acetotrophic or hydrogenotrophic reactions. Methanogenesis is initiated by the onset of anoxia although, since methanogenic Archaea are obligate anaerobes, their survival during periods in which soils become aerobic is not fully understood (Conrad 1996). Methane in the soil gas is also drawn down from the free atmosphere, if methanotrophic consumption in the aerobic surface soil exceeds methanogenic supply from anaerobic soil. If this supply exceeds consumption, then the soil provides a net source of methane that diffuses out of the soil into the free atmosphere.

As a group, the methanotrophs and methanogens occupy different, but overlapping, ecological niches. Overall, methanotrophy may be most pronounced at the interface of anaerobic and aerobic soils and can be most evident in soils that are subject to periodic anaerobiosis, such as rice paddies (Conrad 1996).

Consumption of methane in aerobic soil is affected by Type I and Type II bacterial oxidation (see below). Type I methanotrophs include the genera *Methylobacter* and *Methylomonas*, whereas Type II includes *Methylosinus* and *Methylocystis*. Both groups of organisms are methylotrophs, i.e. organisms that can oxidise C<sub>1</sub> compounds. A third group of methanotrophs, Type X, includes *Methylococcus*, which uses both the ribulose monophosphate (RuMP) and serine pathways of the Type I and Type II groups. In addition, there are representatives of other phyla that are classified as methanotrophs.

The initial step in methane oxidation involves the enzyme methane monooxygenase (MMO) (related to ammonia monooxygenase, AMO), which catalyses the production of methanol (CH<sub>3</sub>OH), from which formaldehyde (HCHO) is then produced, catalysed by dehydrogenase. Formaldehyde is used in the synthesis of multi-carbon compounds within microbial biomass via either the RuMP or serine pathways. Organisms using the RuMP pathway are known as Type I methanotrophs, whereas those using the serine pathway are the Type II methanotrophs.

Megonigal et al. (2005) have pointed out that the transport pathway of each molecule of methane, from the point of production to the free atmosphere, will dictate the likelihood that it will be oxidised to CO<sub>2</sub> *en route*. The thicker the aerobic layer of soil overlying the anoxic seat of methanogenesis, the greater the degree of oxidation. Even within anoxic soils, thin oxidised layers can exist around plant roots (Armstrong et al. 1994) which transport oxygen from the free atmosphere to the rhizosphere. Methane oxidation can occur within these thin layers but is unlikely to be very efficient due to the short residence time within the oxic rhizosphere. In contrast, thick and porous engineered soil layers above landfills provide an environment in which methane oxidation can be extremely high (Whalen et al. 1990). This is due to the high upward flux of methane from a mature (anoxic, reducing) landfill which both stimulates the existing populations of methane-oxidising microbes and selects for those sub-populations that can make most effective use of high fluxes of methane as a substrate for growth and reproduction.

Continuous artificial supply of methane into the basal layers of oxic soils can also lead to the stimulation of methanotrophic activity to exaggerated levels (Steven et al. 2006) which may be unreflective of the oxidation rates achievable under ambient conditions.

The efficiency of methane oxidation in an oxic region of the soil is controlled by its residence time in that region; hence the degree of oxidation is likely to be inversely related to the rate of diffusion through such layers. Freeman et al. (2002) suggested that diffusion of methane through the unsaturated upper layers of a wetland peat subject to drought was sufficiently rapid to offset any increase in CH<sub>4</sub> oxidation after drainage of the peat. They concluded that the reduction in methane emissions after the onset of drought was due to a reduction in methanogenesis within the peat rather than an increase in oxidation capacity of the peat. Megonigal et al. (2005) have questioned whether such rapid diffusion is likely in mineral soils, with the possible exception of sandy soils.

The oxidation of methane proceeds in four steps, with the formation of three intermediate compounds – methanol, formaldehyde and formic acid – prior to the formation of the final reaction product, CO<sub>2</sub>. Since each step is an enzyme-mediated reaction it is to be expected that the overall oxidation rate ( $v$ ) will be subject to Michaelis-Menten kinetics. Thus, the kinetics of methane oxidation can be characterised using the maximal rate or ‘velocity’ of reaction ( $V_{\max}$ ) and the Michaelis constant ( $K_m$  – the methane concentration giving rise to half  $V_{\max}$ , sometimes referred to as the half saturation constant). Conrad (1996) has pointed out that a bacterium’s ability to oxidise methane at ambient concentrations, which can be low, is limited by the  $K_m$  value – the lower the  $K_m$  the lower the methane concentration at which oxidation can occur at an appreciable rate. Additionally, as the methane concentration decreases below the  $K_m$  value, a threshold may be reached below which oxidation ceases. As discussed below, in practice the threshold is well below ambient atmospheric concentrations and appreciable rates of oxidation (of the order of 0.6 mg m<sup>-2</sup> d<sup>-1</sup>) are measured in field conditions.

Four enzymes are involved in the overall oxidation process – MMO and three dehydrogenases, but it is MMO that appears to have received most attention in the literature (Lieberman, Rosenzweig 2004). MMO is common to both Type I and Type II methanotrophs but exists either as membrane-bound (particulate or pMMO) or soluble (cytoplasmic or sMMO) forms. The latter forms are found only in Type II and Type X methanotrophs. In measurements of  $K_m$  values for the two forms of MMO in bacterial cultures and purified enzyme preparations, pMMO appears to occupy the lower end of a range from 1 – 92 μM (King 1992). Type I methanotrophs that rely solely on pMMO, also demonstrate relatively low  $K_m$  values for methane oxidation, indicating a higher affinity for and lower utilisable range of CH<sub>4</sub> concentrations than applies to Type II organisms.

Conrad (1996) has pointed out a clear discrepancy between  $K_m$  values determined for bacterial cultures and intact upland soils. Methanotrophic bacteria in culture exhibit a range of  $K_m$  values for methane oxidation from approximately 20 to 1 600 ppmv, whereas  $K_m$  values for upland soils range from approximately 0.5 to 5 ppmv. The ambient atmospheric methane concentration of 1.8 ppmv lies within the latter range, whereas the former range is considerably higher than concentrations of methane likely to be encountered by methanotrophs exposed to typical atmospheric concentrations. Conrad (1996) concluded from this discrepancy that the microorganisms responsible for methane oxidation in soils had not yet been isolated and identified. Furthermore, Conrad (1996) suggested that the methanotrophic bacteria that have been isolated and identified occupy niches within the soil that expose them to elevated rather than atmospheric methane concentrations, possibly at the interfaces between zones of methanogenesis and methanotrophy. The highest  $K_m$  values observed are associated with landfill covers, soils that have received experimental manipulations or pre-treatments, and artificial experimental systems (Shaw and Thorne 2016). Contin et al. (2012) have demonstrated that consumption rates are generally higher in grassland than in arable soils for both untreated and sewage-sludge amended soils. A comprehensive table of  $K_m$  and  $V_{\max}$  values in different soils, landfill covers and experimental systems is provided at Table 3.1 of Shaw and Thorne (2016), which is reproduced here as Table 2-6.

In Table 2-6, the data are arranged in order of increasing  $K_m$  value from top to bottom. The units of methane consumption rate given in the final column are normalised on various bases, e.g. per unit of mass of soil present in the experimental apparatus or per unit volume of the soil and are sometimes expressed as the fraction of methane consumed per unit time and sometimes as the mass or volume of methane consumed per unit time. These rates are included for completeness and are of limited usefulness in model development. Rather, they provide an input to the derivation of the  $K_m$ ,  $V_{\max}$  and Threshold values given in other columns of the table.

As the concentration of methane becomes extremely low, methane consumption may stop altogether below a well-defined but finite threshold concentration. Conrad (1996) discussed threshold concentrations below which trace gases such as methane are not consumed by microorganisms. Just as  $K_m$  values for methane oxidation are higher for microorganisms in culture than in soils, the threshold for methane oxidation by methanotrophs in culture (0.024 ppmv) is higher than the reported range of threshold concentrations for upland soils (0.0007 to 0.017 ppmv). This supports the conclusion that the identities of methanotrophs in soils remain unknown and that they operate at a range of methane concentrations below those at which cultured methanotrophs operate. Nevertheless, in either case, the threshold is far below the ambient atmospheric concentration of 1.8 ppmv.

In flask incubation studies, Bender and Conrad (1993) determined two thresholds for methane oxidation as the methane concentration declined in the headspace of the flasks. The first ( $Th_a$ ) was the threshold at which the declining methane concentration versus time curve deviated from linearity, which was interpreted as the point at which oxidation deviated from first-order kinetics. A second threshold at a lower methane concentration ( $Th_m$ ) was reached at which oxidation apparently ceased and the methane concentration remained constant. Values for  $Th_m$  in three cambisols were 0.02 ppmv, 90 × lower than the ambient atmospheric methane concentration of 1.8 ppmv.  $Th_m$  values of 0.1 to 0.4 and 0.5 ppmv have been reported for tundra soil and landfill cover soil, respectively, by Whalen and Reeburgh (1990) and Whalen et al. (1990).

As mentioned above, intermediate metabolites formed during oxidation of methane to  $CO_2$  are methanol, formaldehyde and formic acid. There is no evidence from the literature through to 2014 that the production and subsequent utilisation of these intermediates are rate-controlling steps in the overall methane oxidation process within soils, or that the metabolites can be transported a significant distance from their point of production to their point of utilisation. However, in an interpretation of  $^{13}CH_4$  tracer experiments conducted on behalf of RWM, there appears to be evidence for a significant delay between the oxidation of methane and the subsequent production of  $CO_2$  (Hoch et al. 2014). If this is correct, it may provide scope in future for more detailed analysis of the role of metabolic intermediates in the conversion of  $^{14}CH_4$  to  $^{14}CO_2$  (Shaw and Thorne 2014).

**Table 2-6. Michaelis-Menten kinetics of methane oxidation in soils and other materials (Shaw and Thorne 2016).**

Ecosystem / Soil Type	$K_m$ (ppmv)	$V_{max}$ ( $\mu\text{mol g}^{-1} \text{h}^{-1}$ )	Threshold [ $CH_4$ ] ppmv	Methane consumption rate	Units of methane consumption rate
Sandy podzol planted with Norway spruce	5.2	0.0002		< 0.0001–0.001	$\text{h}^{-1} \text{cm}^{-3}$
Acid brown earth, top of A horizon	6.0	0.0003	0.04	0.03	$\text{L g}^{-1} \text{DW h}^{-1}$
Acid brown earth, top of H horizon	9.0	0.0004		0.04	$\text{L g}^{-1} \text{DW h}^{-1}$
Forest soil / Perlite mixture (50:50)	15	0.0141			
Acid brown earth, top of H horizon	17	0.0007		0.04	$\text{L g}^{-1} \text{DW h}^{-1}$
Forest cambisol	22	0.0036	<sup>†</sup> 2.72/0.02	0.38	$\text{cm}^3 \text{h}^{-1} \text{g}^{-1} \text{DW}$
Acid brown earth, top of H horizon	25	0.0007	0.14	0.02	$\text{L g}^{-1} \text{DW h}^{-1}$
Forest – coniferous – organic horizon	25	0.0037			
Meadow cambisol	28	0.0007	<sup>†</sup> 0.19/0.02	0.29	$\text{cm}^3 \text{h}^{-1} \text{g}^{-1} \text{DW}$
Cultivated cambisol	32	0.0006	<sup>†</sup> 0.28/0.02	0.25	$\text{cm}^3 \text{h}^{-1} \text{g}^{-1} \text{DW}$
Cultivated cambisol	43	0.0008			
Meadow cambisol	46	0.0011			
Acid brown earth, bottom of H horizon	48	0.0130	0.04	0.19	$\text{L g}^{-1} \text{DW h}^{-1}$
Forest – coniferous – mineral horizon	56	0.0028			
Eutric cambisol	144	0.1370			
Haplic podzol	476	0.4430			
Drained peatland planted with spruce and birch	510	0.0062		0.0001–0.0005	$\text{h}^{-1} \text{cm}^{-3}$
Mollic gleysol	738	0.5500			
Landfill cover	1800	0.1560	0.50		
Tank packed with dry quartz sand	2110	-		2.06	$\text{h}^{-1}$
Forest soil / Perlite mixture (50:50)	5000	0.063			
Silty loam landfill cap, 30 cm depth	30600	-		10–15.8	$\text{h}^{-1}$
Pre-incubated humisol	50000	15.32	0.10		

Note: <sup>†</sup>The higher value is  $Th_a$ , the lower value  $Th_m$  (see text below for an explanation).

In their review, Shaw and Thorne (2016) compiled 91 rates of net methane consumption in soils reported in the literature, together with 13 rates measured for ambient CH<sub>4</sub> during the RWM experimental programme. A subset of 68 of the literature values was used to investigate the frequency distribution of methane oxidation rates for three major ecosystem types of relevance to the UK. These comprised 39 rates for forest (coniferous and deciduous), 7 rates for temperate grassland, and 22 rates for agricultural/arable ecosystems. The overall frequency distribution for all three ecosystem types was approximately log-normally distributed with a geometric mean of 0.57 mg m<sup>-2</sup> d<sup>-1</sup>. The geometric mean flux for forest soils was higher than for grassland soils, but the difference was not statistically significant. However, the geometric mean rate for agricultural and arable soils was lower than for forest and grassland soils, consistent with results from Dobbie et al. (1996) and Contin et al. (2012). Measured rates from the RWM experimental programme relating to arable soils were statistically indistinguishable from literature values for agricultural/arable soils. Shaw and Thorne (2016) concluded from this analysis that agricultural tillage significantly reduces methane oxidation in soils under field conditions, compared with *in situ* oxidation rates in natural ecosystems, especially forests. This agrees with the conclusions of Cha and Parkin (2001), who emphasised that changes in soil nitrogen status due to fertiliser additions may differentially affect the methane-oxidising community, with the effects differing from system to system, but with higher concentrations of NH<sub>4</sub><sup>+</sup> tending to inhibit methane oxidation.

In respect of the vertical distribution of methanogenesis and methanotrophy, in some forest soils, it has been observed that the highest rates of both methanogenesis and methanotrophy are in the organic layers close to the surface (Bradford et al. 2001, Yavitt et al. 1990). However, numerous studies (six are cited by Conrad 1996) have shown that consumption of atmospheric methane occurs between the A and B horizons. This seems counter-intuitive, because the majority of microbial activity is located at the soil surface, which is where oxidation of other trace gases (e.g. H<sub>2</sub> → H<sub>2</sub>O) is known to occur. Fluctuations in soil moisture and higher ammonium concentrations towards the soil surface have been suggested as possible reasons for reduced methane oxidation in this region of the soil profile (Schnell and King 1994, 1996). If the majority of methanotrophic activity occurs at depth in unsaturated soils, the implication is that the organisms responsible are metabolising methane at concentrations considerably below the free atmosphere concentration of 1.8 ppmv and the associated K<sub>m</sub> values should reflect these ‘operational’ concentrations.

Shaw and Thorne (2016) interpreted the data reviewed above in terms of a mathematical model applicable at all concentrations of methane. A modified version of Michaelis-Menten kinetics with a concentration threshold to represent methane metabolism in oxic soils was found to be a suitable generalised basis on which to develop such a model. Assigning values to the model parameters based on the literature review summarised above, it was found that the fraction of methane oxidised in the soil zone after entry from below is likely to be in the range 0.5 to 1.0, except possibly at extremely high fluxes and in the absence of an incubation effect. For a model in which the threshold above which Michaelis-Menten kinetics apply is much less than the ambient concentration of methane in the atmosphere (as is typically observed to be the case), an active methane-oxidising population of microbes will already be present in the soil before any flux from below occurs. Thus, the fraction of methane oxidised in the soil zone after entry from below is likely to be above 0.9, since that flux will encounter, and may further stimulate, an already active microbial population. At low fluxes of methane, the effects of Michaelis-Menten kinetics can generally be neglected and field estimates of ambient atmospheric methane consumption rates can be used directly. The high fraction of methane estimated to be oxidised in the Shaw and Thorne (2016) model is consistent with the conclusion of Hoch (2014) that the characteristic length scale over which methane is oxidised in soils is likely to be no more than a few tens of centimetres and that, provided that the water table is at a depth that is more than a few times this length, the conversion of <sup>14</sup>CH<sub>4</sub> to <sup>14</sup>CO<sub>2</sub> will be essentially complete, regardless of the particular values of the diffusion coefficient and oxidation rate constant that are adopted.

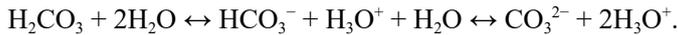
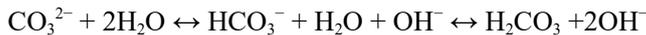
### **2.3.3 The transport of carbon dioxide in soils**

#### ***Chemical speciation and partitioning between phases***

There is competition between methane transport in soils and its oxidation to carbon dioxide, with a large fractional oxidation expected to occur in agricultural soils that are well aerated to a depth of a few tens of centimetres or more. This means that the transport of carbon dioxide in soils is relevant,

even if the C-14 enters the soil system in the form of methane. Furthermore, C-14 entering the soil as carbon dioxide in the gas phase or dissolved in groundwater, will rapidly equilibrate between soil solution and the soil atmosphere, with similar volumetric concentrations in each phase.

In practice, C-14 entering the soil as carbon dioxide, dissolved in groundwater or generated from the oxidation of methane is likely to be present in the soil solution as a mixture of carbonate ( $\text{CO}_3^{2-}$ ), bicarbonate ( $\text{HCO}_3^-$ ), dissolved carbon dioxide and carbonic acid ( $\text{H}_2\text{CO}_3$ ). Furthermore, there are not only exchanges between  $\text{CO}_2$  in the dissolved and gas phases, but there are also kinetic exchanges in the solution phase including the reactions:



Because the hydration equilibrium constant at 25 °C for  $[\text{H}_2\text{CO}_3]/[\text{CO}_2]$  is  $1.7 \times 10^{-3}$  in pure water and  $1.2 \times 10^{-3}$  in seawater, the majority of the carbon dioxide in solution is not converted into carbonic acid and remains as  $\text{CO}_2$  molecules. In the absence of a catalyst, the equilibrium is reached quite slowly. The rate constants are  $0.039 \text{ s}^{-1}$  for the forward reaction ( $\text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{H}_2\text{CO}_3$ ) and  $23 \text{ s}^{-1}$  for the reverse reaction ( $\text{H}_2\text{CO}_3 \rightarrow \text{CO}_2 + \text{H}_2\text{O}$ ). It is these reactions, together with the partitioning of  $\text{CO}_2$  between the liquid and gas phases that determine the overall behaviour of  $\text{CO}_2$  in soils. At pressures up to about 5 atmospheres the solubility follows Henry's Law, i.e.  $[\text{CO}_2] = 0.032P$ , where  $P$  is the partial pressure of  $\text{CO}_2$ . Thus, at an atmospheric concentration of 400 ppmv, as exists at the present day, the concentration in water that is in equilibrium with the atmosphere is  $0.032 \times 4 \times 10^{-4} = 1.3 \times 10^{-5} \text{ M}$  and this governs the exchange of carbon dioxide between soil solution and the soil atmosphere.

Thus, when  $^{14}\text{CO}_2$  enters an unsaturated soil or sediment in solution, the  $^{14}\text{CO}_2$  will rapidly equilibrate between the  $\text{CO}_2$  present in solution and the  $\text{CO}_2$  present in the gas-filled pore space. In vegetated soils and sediments, the  $\text{CO}_2$  concentration in the pore space will generally be higher than in the ambient atmosphere above the soil surface. This is a consequence of root and microbial respiration. However, Henry's law will still apply so the fractions present in solution and in the gas-filled pore space will be independent of the absolute concentration. Thus, if  $\phi$  is the total porosity and  $S$  is the degree of saturation, the gas-filled porosity per unit volume will be  $\phi(1-S)$  and the water-filled porosity will be  $\phi S$ . For a partial pressure of  $P$ , the content in the gas phase per litre of soil or sediment will be  $P\phi(1-S)/22.4$  moles, since 1 mole of gas at Standard Temperature and Pressure (STP) occupies 22.4 litres. The content of the solution phase will be  $0.032\phi SP$  moles. Thus, the fraction in the gas phase will be  $0.0446(1-S)/\{0.0446(1-S) + 0.032S\}$ . Thus, at  $S = 0.5$ , about 60 % of the soil or sediment  $\text{CO}_2$  will be in the gas phase and  $^{14}\text{C}$  entering soil in solution incorporated in  $\text{CO}_2$  will be partitioned similarly.

Carbon dioxide that is present in the gas-filled pore space of unsaturated soils or sediments will rapidly exchange with the above-ground atmosphere. Thus,  $^{14}\text{CO}_2$  entering the unsaturated zone in solution will rapidly equilibrate between the solution and gas phases and will be lost from that mixed 'pool' across the soil surface by gas exchange processes (mainly diffusion, but with a small advective component due to the pumping effect of atmospheric pressure variations). A detailed, diffusion-based model for  $^{14}\text{C}$ -losses from soil in the gas phase has been developed for RWM (RWM 2014).

The  $^{14}\text{C}$  in solution in unsaturated soils and sediments after its entry to those soils and sediments as  $^{14}\text{CO}_2$ , will mainly remain as  $^{14}\text{CO}_2$ . However, small fractions will also be present as carbonate, bicarbonate and carbonic acid (see above). The proportions of each will depend upon the chemical composition of the soil or sediment water.

### ***Uptake by plant roots***

As discussed above,  $^{14}\text{CO}_2$  entering the unsaturated zone of a soil will rapidly be lost across the soil surface, mainly by diffusion. However, the  $^{14}\text{CO}_2$  will also exchange with  $\text{CO}_2$  present in soil solution and will, therefore, potentially be available for uptake by plant roots. The potential for carbon in soil solution to be taken up by plant roots has been extensively discussed, but the relevant literature is rather limited and equivocal. Most of the carbon that is incorporated in plant biomass is obtained

from the atmosphere by photosynthesis, as is most of the carbon that is used as a substrate for plant respiration. However, it seems that a small fraction of plant carbon is obtained by root uptake and this needs to be considered when modelling the behaviour of  $^{14}\text{CO}_2$  in soil-plant systems. If the source term of C-14 into the system of interest is to the atmosphere, then the small uptake from roots would appear to be negligible. If the C-14 source term is from the geosphere, then although the majority carbon source to the plant will still be the atmosphere, the C-14 source via the roots may be of interest since the C-14 seen by the roots will not have been diluted by  $\text{CO}_2$  in the plant canopy and wider atmosphere. The literature on this topic has been reviewed by Hoch (2014). He points out that  $\text{CO}_2$  forms some bicarbonate ( $\text{HCO}_3^-$ ) when it dissolves in water and that root uptake of  $\text{HCO}_3^-$  and its incorporation into plant carbon through the action of phosphoenolpyruvate (PEP) carboxylase is well documented.

As discussed by Hoch (2014), early studies of the effects of carbonated water on crop yields showed that the plants were able to derive carbon from the  $\text{CO}_2$  dissolved in the water. A series of studies using a labelled carbon source showed that potatoes growing in nutrient solution absorbed  $\text{CO}_2$  through their roots and transported it to the site of photosynthesis in the plant leaves (Arteca et al. 1979, Arteca and Poovaiah 1982a). Substantial increases in tuberization, stolon length, number of tubers per stolon and plant dry weight were observed in plants that had their roots exposed to a gas stream consisting of 45 %  $\text{CO}_2$  for twelve hours. Root uptake of  $\text{CO}_2$  was measured also in tomato, eggplant, rice, peas, beans, oats, corn, wheat and citrus (Arteca and Poovaiah 1982b, Baron and Gorske 1986, Bialczyk et al. 1994, Higuchi et al. 1984, Labanauskas et al. 1971, Schafer 1988, Yurgalevitch and Janes 1988).

For most plants, productivity appears not to be affected substantially by the quantity of  $\text{CO}_2$  absorbed. Schafer (1988) found that root uptake of  $\text{HCO}_3^-$  accounted for only 0.0044 to 0.0121 of the total carbon assimilated by wheat shoots. Others have suggested that  $<0.05$  of the  $\text{CO}_2$  fixed by a plant could be absorbed by the root system and it was thought unlikely that crop yields could be increased using carbonated irrigation water (Skok et al. 1962, Stolwijk and Thimann 1957).

Hoch (2014) points out that these observations are consistent with an analysis of root uptake of carbon in rice (Hoch 2013) that suggests that no more than 0.02 of plant carbon is absorbed through the roots.

Hoch (2014) developed a theoretical model for the uptake of carbon by plant roots because of the passive entry of soil water into the plants in the transpiration stream. This is based on use of the transpiration ratio, which is defined as the ratio of the mass of water transpired by a plant during its growing season to the mass of dry matter (usually exclusive of roots) produced. Hoch (2014) cites a range of 450 to 950 kg  $\text{H}_2\text{O}$  transpired per kg dry mass produced for C3 plants (Black 1973), which is much higher than the range of 250 to 350 kg  $\text{H}_2\text{O}$  transpired per kg dry mass for C4 plants and 50 to 125 kg  $\text{H}_2\text{O}$  transpired per kg dry mass for plants adapted to arid conditions that use the CAM photosynthesis pathway. Of the carbon taken-up in the transpiration stream, 0.5 is assumed to be lost in respiration and 0.5 used in biomass production.

Applying this model to results from a field experiment conducted for RWM (Atkinson et al. 2014), Hoch (2014) computed that the fraction of carbon that a plant absorbs through its roots is 0.0086. This is consistent with the observations in the literature reviewed above and indicates that the root uptake of carbon by plants can be fully accounted for by passive uptake in the transpiration stream.

Once  $^{14}\text{CO}_2$  has been released across the soil-atmosphere boundary it is available for foliar uptake by plants and utilisation in photosynthesis. In this context, the key consideration is exchange of carbon dioxide between the plant canopy and the overlying atmosphere. This exchange is affected by the characteristics of the plant canopy, specifically its height and density. For assessment purposes, a resistance-analogue approach to modelling transport through the canopy can be adopted (see Chapter 5 of (Hoch 2014) for the governing equations and relevant parameter values).

### ***Uptake from the above-ground atmosphere***

For uptake via photosynthesis, the specific activity in the biomass of the plant can be equated to the specific activity within the plant canopy, i.e. the small effect of isotopic discrimination between C-14 and C-12 in photosynthesis can be neglected. As discussed in the previous subsection, the specific activity of the canopy atmosphere can be calculated from the concentration of  $^{12}\text{CO}_2$  in the canopy atmosphere and the flux of C-14 from the soil multiplied by the aerodynamic resistance of the plants

to determine the concentration of  $^{14}\text{CO}_2$  in the canopy atmosphere. In the model of Hoch (2014), a small correction is applied to allow for a non-zero C-14 concentration in the atmosphere above the canopy that arises from the C-14 flux through the top of the canopy. This correction depends on the area of the release. Overall, Hoch (2014, Section 5.1.1) demonstrates that the C-14 concentration in the canopy atmosphere,  $C_{\text{canopy}}$  ( $\text{Bq m}^{-3}$ ), is given by:

$$C_{\text{canopy}} = F(\Psi + \Phi)/u_a$$

Where the dimensionless proportionality constant  $\Psi$  depends on the release area and is approximately 10 for a release area of  $1 \times 10^4 \text{ m}^2$ , about 25 for a release area of  $1 \times 10^5 \text{ m}^2$  and about 40 for a release area of  $1 \times 10^6 \text{ m}^2$  (see Table 5.4 of Hoch 2014), bearing in mind that the most frequent atmospheric stability classes in the UK are Categories C and D, see (Clarke 1979).  $\Phi$  is related to the aerodynamic resistance of the canopy and is given a value of 208 in Hoch (2014). The quantity  $u_a$  ( $\text{m s}^{-1}$ ) is the windspeed at a reference height that is chosen to be 2 m above the ground. Note that in this formula,  $F$  has units of  $\text{Bq m}^{-2} \text{ s}^{-1}$ .

Hoch (2014) calculated the numerical value of  $\Phi$  for a pasture (grass) canopy of height  $h = 0.12 \text{ m}$  using the expression  $\{\ln[(z-d)/z_{0m}]\} \{\ln[(z-d)/z_{0c}]\} / \kappa^2$ , where  $z$  (m) is the reference height above the plant canopy at which the windspeed and carbon dioxide concentration are measured,  $d$  (m) is the height of the zero-plane displacement,  $z_{0m}$  (m) is the roughness length controlling the transfer of momentum,  $z_{0c}$  (m) is the roughness length controlling the transfer of water vapour, carbon dioxide and sensible heat, and  $\kappa$  ( $= 0.41$ ) is von Karman's constant (Allen et al. 1998). The calculation assumed that the zero-plane displacement  $d = \frac{2}{3}h$ , the roughness length controlling transfer of momentum  $z_{0m} = 0.123h$ , and the roughness length controlling transfer of carbon dioxide  $z_{0c} = 0.1z_{0m}$ . This expression shows that there is only a weak dependence on the height of the zero-plane displacement, but a somewhat stronger, logarithmic dependence on the roughness length. An increase or decrease in either roughness length by a factor of five<sup>2</sup> would result in a decrease or increase in this aerodynamic resistance by a factor of  $\ln(5) = 1.6$ .

Hoch (2014) estimates the concentration of  $^{12}\text{CO}_2$  in the canopy atmosphere to be about 350 ppmv (converted to  $1.75 \times 10^{-4} \text{ kg[C] m}^{-3}$ ). This is based on an above-canopy value of 395 ppmv (applicable in 2013 AD) and allows for some depletion within the canopy due to the net difference between uptake for photosynthesis and loss through plant respiration. On this basis, the specific activity in the canopy atmosphere,  $A_{\text{canopy}}$  ( $\text{Bq kg[C]}^{-1}$ ) is given by:

$$A_{\text{canopy}} = F(\Psi + \Phi)/1.75 \times 10^{-4} u_a$$

Hoch (2014) takes  $\Psi = 10$  for a release area of  $1 \times 10^4 \text{ m}^2$ ,  $\Phi = 208$  for pasture, and  $u_a = 2 \text{ m s}^{-1}$  to derive a typical relationship:

$$A_{\text{canopy}} = 6.23 \times 10^5 F \quad \text{Bq kg[C]}^{-1}$$

where the flux,  $F$ , has units of  $\text{Bq m}^{-2} \text{ s}^{-1}$ .

Although there is a dependence of  $\Psi$  upon the release area, that dependence is not very strong. Specifically,  $\Psi$  is about 25 for a release area of  $1 \times 10^5 \text{ m}^2$  and about 40 for a release area of  $1 \times 10^6 \text{ m}^2$  (see Table 5.4 of Hoch 2014, bearing in mind that atmospheric stability categories C and D dominate in UK conditions). For comparison, the box model of atmospheric dispersion used by Thorne and Walke (2013) gives a value of  $\Psi$  of 50. This emphasises that the main determinant of the specific activity in the canopy atmosphere is the windspeed  $u_a$  rather than the release area assumed or the atmospheric dispersion model adopted. Thus, Thorne and Walke (2013) adopted a realistic, average windspeed for Category D conditions of  $5 \text{ m s}^{-1}$  (see Figure 10 of their report), whereas Hoch (2014) adopted a cautious estimate of  $2 \text{ m s}^{-1}$ .

For the more realistic windspeed of  $5 \text{ m s}^{-1}$  and for a release area of  $1 \times 10^5 \text{ m}^2$  ( $\Psi = 25$ ),  $A_{\text{canopy}} = 2.66 \times 10^5 F \text{ Bq kg[C]}^{-1}$ .

Consideration also must be given to uptake of C-14 by plants following its atmospheric dispersion. Various models have been developed for this purpose. In the context of solid radioactive waste disposal, the main interest is in chronic releases at relatively uniform rates. Models for the uptake

<sup>2</sup> Covering the approximate range of roughness lengths between short grass and grain in Table 3.1 of Foken (2008).

of C-14 bearing forms of CO<sub>2</sub>, CO and CH<sub>4</sub> for application in this context have been developed by Thorne and Kelly (2015). For CO<sub>2</sub>, they adopt a simple specific activity model and assume that the specific activity of carbon-14 bearing carbon dioxide in plants will be similar to the specific activity in the air to which they are exposed, since most plant carbon is obtained from atmospheric carbon dioxide by photosynthesis.

For carbon monoxide, Thorne and Kelly (2015) make use of Hoch (2014) and Shaw and Thorne (2016) to argue that carbon monoxide is metabolized by plants. In the light, the carbon monoxide is converted mainly to sucrose and proteins. The distribution of carbon-14 amongst the products of metabolism of carbon-14 bearing carbon monoxide suggests that most of the absorbed carbon monoxide is reduced and incorporated into serine and that this is then converted into sucrose. Some carbon monoxide is found to be oxidized to carbon dioxide, part of which may be re-fixed in photosynthesis, but this is not the major pathway of metabolism. In darkness, carbon monoxide is absorbed nearly as fast as in the light but is then almost completely converted to carbon dioxide and released. In the light, the rate of fixation is roughly proportional to the concentration of carbon monoxide at high concentrations of the gas but is unrelated to rates of photosynthesis. The carbon monoxide fixing capacity of vegetation at air concentrations of 200 to 360 ppmv has been calculated to be 12–120 kg[CO] km<sup>-2</sup> d<sup>-1</sup> (12–120 mg[CO] m<sup>-2</sup> d<sup>-1</sup>), which was stated to approach the rate found in soils at much higher concentrations of carbon monoxide than those used in these experiments.

In respect of soils, Thorne and Kelly (2015) observe that carbon monoxide oxidation rates can range from about 10 mg[CO] m<sup>-2</sup> d<sup>-1</sup> up to more than 1 000 mg[CO] m<sup>-2</sup> d<sup>-1</sup>. The reasons for this wide range of variation are not well understood (though major factors may be that they derive from a mix of field measurements and laboratory measurements on small soil samples, and at very different concentrations of carbon monoxide), but it is clear that net oxidation rates are higher than those for methane, which are typically in the range of 0.2 to 3.6 mg[CH<sub>4</sub>] m<sup>-2</sup> d<sup>-1</sup> (Shaw and Thorne 2016). These oxidation rates must be considered in the context of typical atmospheric concentrations of carbon monoxide. Mean monthly atmospheric concentrations of carbon monoxide in the atmosphere range up to 0.3 ppmv and global background concentrations range between 0.05 and 0.12 ppmv. In contrast, in urban traffic environments of large European cities, the 8-hour average carbon monoxide concentrations are generally lower than 17 ppmv with short-lasting peaks below 53 ppmv (Hoch 2014).

As noted above, the carbon monoxide fixation rate of vegetation ranges from about 12 to 120 mg[CO] m<sup>-2</sup> d<sup>-1</sup> at air concentrations of 200 to 360 ppmv. However, the fixation rate is not readily correlated with the air concentration within this range of concentrations. The range of fixation rates corresponds to 5 to 50 mg[C] m<sup>-2</sup> d<sup>-1</sup>. For comparison, the overall yield of plants is typically a few kilogrammes fresh weight per m<sup>2</sup> over their growing season. If plants have a dry weight to fresh weight ratio of 0.1 and the fraction of that dry mass that is carbon is 0.4, then a yield of 5 kg fresh weight per m<sup>2</sup> corresponds to 0.2 kg[C] m<sup>-2</sup>. For a growing season of 120 days, this corresponds to an average carbon accumulation rate of 1 670 mg[C] m<sup>-2</sup> d<sup>-1</sup>. This demonstrates that plants cannot obtain more than about 0.003 to 0.03 of their carbon from carbon monoxide uptake, even in the presence of carbon monoxide concentrations in air that are similar to current carbon dioxide concentrations. However, it is not known to what extent the carbon monoxide assimilation rate would decrease if the carbon monoxide concentration in air was decreased from 200 to 360 ppmv to a more typical ambient value of less than 0.3 ppmv. Thorne and Kelly (2015) make the reasonably cautious assumption that the carbon assimilation rate is the minimum found for an atmospheric concentration of carbon monoxide of 200 to 360 ppmv but note that the actual rate at ambient concentrations could be much lower. Under this cautious assumption, plants could obtain up to 0.003 of their content of carbon by assimilation of carbon monoxide from the air. For comparison, if the assimilation efficiency for carbon monoxide was equal to the assimilation efficiency for carbon dioxide, then the fraction of plant carbon derived from carbon monoxide in air would be in the ratio of the concentrations of the two gases. This would typically be 0.1 ppmv/390 ppmv = 0.0003, i.e. one order of magnitude lower than the cautious assumption adopted by Thorne and Kelly (2015).

In the case of methane, Thorne and Kelly (2015) remark that the consumption rate of methane in soils shows little dependence on the atmospheric methane concentration at low atmospheric concentrations (see also Murguía-Flores et al. 2018). At low concentrations, typical consumption rates are almost

always in the range from 0.12 to 3.6 mg m<sup>-2</sup> d<sup>-1</sup>. If the carbon-14 bearing methane consumed in soil is obtained solely from the overlying atmosphere, then, based on the oxidation rates quoted above, the effective deposition velocity is

$$(0.12 \text{ to } 3.6)/1.29 = 0.093 \text{ to } 2.8 \text{ m d}^{-1} = 1.1 \times 10^{-6} \text{ to } 3.2 \times 10^{-5} \text{ m s}^{-1}.$$

This is all metabolised and is mainly converted to <sup>14</sup>CO<sub>2</sub> in the soil zone (though some is converted directly to biomass by methanotrophs). From this point, the model developed by Hoch (2014) for C-14 bearing carbon dioxide entering soil is applied.

For short-term releases various models are available. These are discussed and compared in the context of simulating releases from the Orano La Hague nuclear fuel reprocessing plant in Northwest France, which has been operating since the late 1960s and has authorised atmospheric discharges of approximately 15 TBq y<sup>-1</sup>. Between September 2006 and July 2008, IRSN monitored C-14 at a site 2 km north of the La Hague site. Data relating to meteorological conditions, source terms, atmospheric concentrations, soil and pasture concentrations were collected. Hourly measurements of Kr-85 in the atmosphere at the monitoring site, combined with monthly measurements of Kr-85 and C-14 in the atmosphere, were used to derive an implied hourly C-14 atmospheric source term at the site. Considerable variation in the C-14 concentration in the atmosphere at the monitoring site was observed.

The field data collected by IRSN provided an opportunity to validate the modelling of C-14 uptake into pasture from the atmosphere. Therefore, as part of a BIOPROTA study, five organisations applied their models to the La Hague scenario (Limer et al. 2017). The models varied in structure and processes explicitly represented.

The models used the same atmospheric source term and aimed to calculate C-14 concentrations that followed the observed C-14 concentrations in the pasture. The models were adapted to accommodate the high-temporal resolution of input data and performed well when compared against the observational data.

The calculated soil concentrations of C-14 were more variable, with a dependency on the assumed initial conditions. However, for an atmospheric source term the dominance of that means that the soil compartment(s) do not significantly affect the key results of crop concentrations.

### ***Uptake by domesticated animals***

The uptake of C-14 by domesticated (farm) animals can be estimated either at equilibrium or from a biokinetic model. However, these two approaches are readily related to each other.

Consider, for example, the behaviour of C-14 in a single compartment system. The mass of this compartment is taken to be  $M$  kg and the stable carbon concentration in it is  $C_c$  kg[C] kg<sup>-1</sup>. Thus, the mass of stable carbon in the compartment is  $MC_c$  kg[C] and this is taken to be independent of time. Initially, at  $t = 0$ , the C-14 content of the compartment is taken to be zero, so the specific activity of carbon in the compartment is also zero. From  $t = 0$  onward, the compartment is supplied with a constant input flux of carbon of  $F$  kg[C] s<sup>-1</sup>. This carbon is taken to have a specific activity of  $A$  Bq kg[C]<sup>-1</sup>. Thus, the flux of C-14 entering the compartment from  $t = 0$  is  $FA$  Bq s<sup>-1</sup>.

As the carbon content of the compartment remains constant, the flux of carbon out of the compartment is equal to the input flux, i.e.  $F$  kg[C] s<sup>-1</sup>. As the mass of carbon in the compartment is  $MC_c$  kg[C], the rate coefficient for loss of either stable carbon or C-14 from the compartment is  $k = F/MC_c$  s<sup>-1</sup>. Thus, neglecting the effects of radioactive decay (because of the long half-life of C-14) and using the standard equation for build-up of activity in a single compartment system with constant input rate (Klos and Thorne 2020), the content of C-14 in the compartment,  $Q_c(t)$  (Bq) is given by:

$$Q_c(t) = (FA/k) \{1 - \exp(-kt)\} = AMC_c \{1 - \exp(-kt)\}$$

However, the mass of carbon in the compartment is  $MC_c$ . Therefore, the specific activity of carbon in the compartment,  $A_c(t) = Q_c(t)/MC_c = A \{1 - \exp(-kt)\}$ , i.e. at equilibrium, the specific activity in the compartment becomes equal to the specific activity in the source. The characteristic timescale for approach to equilibrium is  $MC_c/F$  seconds.

The approach illustrated above for a single compartment can also be applied to multi-compartment systems, with the specific activity of the flux leaving a compartment at any time equal to the specific activity of the material in the compartment at that time.

If the concentration of carbon in the source material to the compartment in the above example is  $C_s \text{ kg[C] kg}^{-1}$ , then the activity concentration in the source material is  $C_s A \text{ Bq kg}^{-1}$ . From the above, the activity concentration in the compartment at any time  $t \geq 0$  is given by:

$$C_c A_c(t) = C_s A \{1 - \exp(-kt)\}$$

Conventionally, uptakes of radionuclides by domesticated animals at equilibrium are estimated using transfer factors,  $TF$ s, where the concentration in an organism is related to the total rate of intake. In the context of the above example, this corresponds to:

$$TF = C_c A \{1 - \exp(-kt)\} / FA = C_s \{1 - \exp(-kt)\} / F$$

Thus, at equilibrium, the  $TF$  is the ratio of the concentration of the stable element in the organism to the rate of intake. For farm animals,  $TF$  values are typically specified with units of  $\text{d kg}^{-1}$  for meat and meat products and  $\text{d L}^{-1}$  for milk. As with  $CR$  values,  $TF$  values are often measured in short-term experiments in which equilibrium will not have been achieved.

The carbon intake of animals is almost entirely from their diet and varies in proportion to energy demand. Although energy demand increases with body mass, the change is not proportional. This is because the surface area of the animal increases more slowly with increasing size than does the body mass. Typically, the energy requirement increases as the 0.75<sup>th</sup> power of body mass (Chapter 11 of Lawrence and Fowler 2002). Therefore, it is a reasonable approximation to set  $F = \alpha M^{0.75}$ , where  $\alpha$  is a calibration coefficient. Because of the fractional power involved, the value of  $\alpha$  is dependent on the units adopted. Here,  $F$  has units of  $\text{kg[C] d}^{-1}$  and  $M$  has units of  $\text{kg[fresh weight]}$ . For adult humans, the International Commission on Radiological Protection (ICRP 1975) gives a reference body mass of  $70 \text{ kg[fresh weight]}$  and a carbon intake rate of  $0.3 \text{ kg[C] d}^{-1}$ . Thus,  $\alpha = 0.3/70^{0.75} = 0.0124$ . Applying this to a dairy cow of  $500 \text{ kg[fresh weight]}$  gives a carbon intake rate of  $0.0124 \times 500^{0.75} = 1.31 \text{ kg[C] d}^{-1}$ . Taking the dry matter content of plants to have a fractional carbon content of 0.475, this corresponds to  $2.76 \text{ kg[dry weight] d}^{-1}$ . This is rather low for dairy cows because it neglects the energy required for milk production. For comparison, Lawrence and Fowler (2002) give the daily metabolisable energy requirement for pigs as  $458 \text{ kJ} \times M(\text{kg})^{0.75}$ . As the standard feed for pigs has an energy content of  $12 \text{ MJ kg}^{-1}$ , this translates into a dietary requirement of  $0.0382 M^{0.75} \text{ kg[feed] d}^{-1}$ . If the energy content of the feed is defined on a dry mass basis (not stated, but implied, by Lawrence and Fowler 2002), this corresponds to  $0.0185 M^{0.75} \text{ kg[C] d}^{-1}$ . For a  $70 \text{ kg}$  pig, this is  $0.448 \text{ kg[C] d}^{-1}$ . This is somewhat larger than the human value. In practice, metabolisable energy requirements are typically increased above maintenance to achieve growth or milk production (see Table 11.8 of Lawrence and Fowler 2002). Here, a reference value for  $\alpha$  of 0.02 is adopted, i.e. the carbon content of diet is calculated as  $0.02 M^{0.75} \text{ kg[C] d}^{-1}$ . For sensitivity studies, a reasonable range for  $\alpha$  is 0.01 to 0.04.

The carbon content of tissues can be based on data for humans. The ICRP (1975) gives a total body content of carbon of  $16 \text{ kg}$  in a total body mass of  $70 \text{ kg}^3$ , corresponding to  $0.229 \text{ kg[C] kg[fresh weight]}^{-1}$ . Thus:

$$TF_{\text{equilibrium}} = 0.229/0.02 M^{0.75} = 11.45/M^{0.75} \text{ d kg[fresh weight]}^{-1}$$

<sup>3</sup> Appendix D of Limer et al. (2011) cites ICRP (1975) as stating that the mass fraction of carbon in muscle is 0.107 on a fresh weight basis. This is based on  $3 \text{ kg}$  of carbon in  $28 \text{ kg}$  of muscle. In contrast, the ICRP (1975) gives a total body content of  $16 \text{ kg}$  of carbon in a body mass of  $70 \text{ kg}$ , i.e. a mass fraction of 0.229. The difference arises because soft tissues other than muscle and, specifically, adipose tissues, have a larger mass fraction of carbon than muscle. As meat and meat products typically include both muscle and other soft tissues, it is here cautiously assumed that the whole-body value applies to the food products of relevance.

For milk, the carbon content is lower than for body tissues. Milk typically comprises 13 % milk solids, with the remainder being water (Lawrence and Fowler 2002). Taking the milk solids to be 50 % carbon<sup>4</sup>, the carbon content of milk is 0.065 kg[C] L<sup>-1</sup>. Thus:

$$TF_{\text{equilibrium}} = 0.065/0.02M^{0.75} = 3.25/M^{0.75} \text{ d L}^{-1}$$

For cattle of 500 kg[fresh weight], this gives a  $TF$  value of 0.031 d L<sup>-1</sup>.

The characteristic timescale for equilibration is  $MC_0/F$  d and this can be rewritten as  $M^{0.25}C_0/\alpha$ . Thus, there is only a weak dependence on body mass. For cattle of 500 kg[fresh weight], the characteristic time is 54 days. Therefore, extended periods of exposure to constant rates of C-14 intake in diet are required for equilibrium  $TF$  values to be applicable.

In applying the above approach, live weights are required for dairy cattle, beef cattle, sheep, goats and chickens.

For dairy cattle, target weights for calving are typically about 500 kg. Slaughter weights for beef steers are typically 450 to 550 kg (Soffe 2003, Chapter 19: Cattle). Therefore, for both beef and dairy cattle  $M$  can be taken as 500 [450, 550] kg. Smaller body weights could be appropriate for some types of intensively reared animals.

For mature sheep and goats, live weights of 40 to 80 kg are typical (Soffe 2003, Chapter 20: Sheep and Goats). However, carcass weights for lambs at slaughter are typically 16 to 25 kg (Soffe 2003, Chapter 20: Sheep and Goats), corresponding to about 30 to 50 kg live weight.

Breeder chickens of age 22 to 30 weeks have a body weight of 2.5 to 3.3 kg (Soffe 2003, Chapter 22: Poultry). However, most chickens have shorter lifetimes and smaller body masses at slaughter.

## 2.4 Transport and accumulation of carbon in forest ecosystems

There is a broad spectrum of various types of forests and woodlands. Here, for simplicity, the focus is on forested areas on aerated or wet soils. That is, specifically, the soils behave in respect of the carbon storage pools and fluxes in a manner like agricultural (surface-aerated) or wetland (surface-wet) ecosystems, see Sections 2.3 and 2.5, respectively. The key differences between these ecosystem types are that the vegetation in forests is taller and the carbon turnover times in the trees are, in part, longer than in the other ecosystems. Also, the canopy structure in forests is often rather heterogeneous (multi-layered). Many forest types also support a humus layer (consisting of partially decomposed litter) on top of the mineral soil layers, resembling aerated top peat layers in wetlands (and some forests may be developed on a peatland).

### 2.4.1 Carbon storages and cycling in forests

Also in forests, carbon assimilation from the atmosphere (photosynthesis) and plant production, root uptake of carbon, carbon release from biomass by respiration, carbon release from biomass during decomposition, and carbon accumulation in soil are the key characteristics of the carbon cycling (cf. Section 2.3).

In boreal forests (Kellomäki 2005), during regeneration after major perturbations (e.g. clear-cut harvesting) the gross production of trees rapidly increases and biomass is quickly accumulated in the ecosystem, but the maintenance of this biomass requires a gradually increasing part of the gross production and the net production slows down; the biomass accumulation ceases when the gross production and respiration are in balance. The biomass of understorey vegetation typically decreases with the increase in tree biomass (Kellomäki 2005). Martin-Benito et al. (2021) have recently studied an old-growth oak forest in northern Spain for the carbon dynamics and longevity of the storage in old trees. Tubiello et al. (2021) present a recent summary of carbon emissions and carbon sequestration in forest ecosystems throughout the globe.

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<sup>4</sup> In Appendix D of Limer et al. (2011), whole milk is taken to comprise about 4 % fats, 3.5 % proteins and 5 % carbohydrates. As fat is about 0.64 carbon, proteins about 0.50 carbon and carbohydrates about 0.4, a weighted average value for milk solids is 0.50. This makes the carbon content of whole milk 6.5 %.

As outlined above, formation of a humus layer from the litter production is a key feature of forests; the litter consists, for example, of foliage, twigs and dead branches, bark, cones, (fine) roots (Kellomäki, 2005). The humus layer and the relatively slow decomposition of it and its leachates supports the very long-term carbon storage functioning of forest soils (cf. Section 2.4.2). If the vegetation is, or turns into, a peat-forming type (i.e. the litter being relatively recalcitrant to decomposition), peat is gradually accumulated, and the site may develop into a wetland (Section 2.5). In such cases the rising groundwater table is often lowered by (enhanced) ditching in commercially managed forests to secure the production of timber.

Deadwood and particularly (standing or fallen) dead trunks may comprise considerable carbon storage and increase carbon turnover time in forests, even if this is spatially sporadic; this is pronounced in boreal forests, where the decomposition is much slower than in the tropics (e.g. Seibold et al. 2021). However, the amount and type of deadwood tends to be heavily affected by forest management practices. Seibold et al. (2021) found that across 55 forest sites on six continents, the deadwood decomposition rates increased with temperature, and the strongest temperature effect is found in high precipitation domains; however, precipitation affects the decomposition rates negatively at low temperatures and positively at high temperatures (Seibold et al. 2021). At least in boreal spruce forests, the decomposition of dead trunks may be coupled to methane oxidation (Mäkipää et al. 2018).

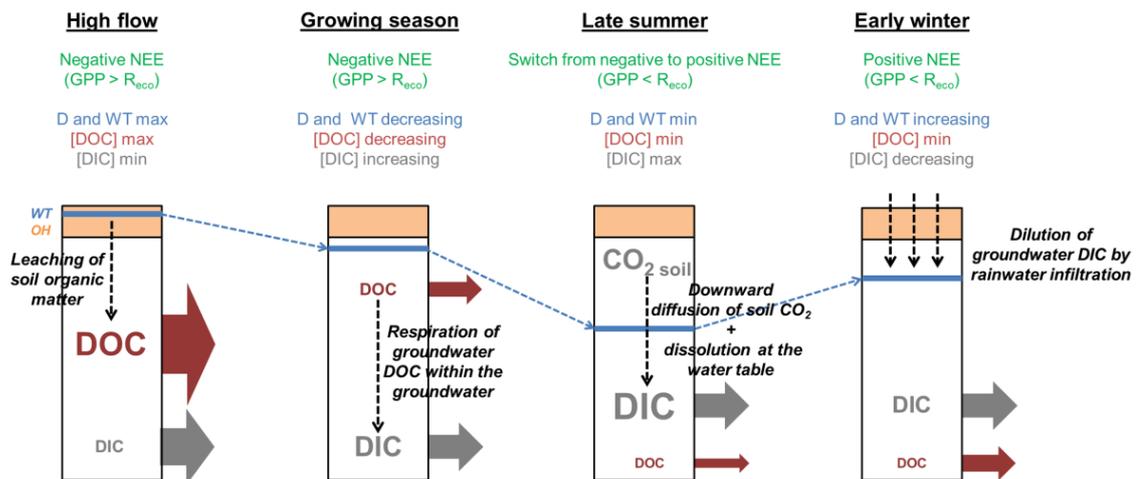
### 2.4.2 Forest soils

As outlined above, the discussion on the soil processes in Section 2.3 is applicable also to forests.

The dissolved organic carbon (DOC) derives from throughfall, stemflow, litter leaching, root exudation and decomposition of root and other litter (Deirmendjian et al. 2018, referring to Bolan et al. 2011). However, a large fraction of DOC in the soil solution may be easily adsorbed and stabilised against biological degradation (Deirmendjian et al. 2018, referring to Kaiser and Guggenberger 2000, Kalbitz et al. 2000, Kalbitz and Kaiser 2008, Sanderman and Amundson 2009, Shen et al. 2015), although it can relatively easily be washed-off by rainfall events and infiltration (Deirmendjian et al. 2018, referring to Kawasaki et al. 2005, Shen et al. 2015). In temperate forests, the highest DOC concentrations are typically found in the uppermost mineral soil horizon, but the concentrations vary widely across sites (Michalzik et al. 2001). The majority of the dissolved inorganic carbon (DIC) in the shallow groundwater derived from the dissolved CO<sub>2</sub> from microbial and root respiration in the soil (Deirmendjian et al. 2018).

Carbon leaching from forests typically represents only a small fraction (a few percent) of the net ecosystem exchange (Deirmendjian et al. 2018, also referring to Shibata et al. 2005, Kindler et al. 2011, Magin et al. 2017), but this relatively minor flow is key to the carbon linkage of the forests to the stream network. In temperate forests, the DOC leaching fluxes correlate positively with the annual precipitation, the throughfall and the soil pH (Michalzik et al. 2001). The highest DOC leaching fluxes derive from the humus layer (Michalzik et al. 2001), which is also the primary source for dissolved organic matter in general due to active microbial degradation (Michalzik et al. 2001, referring to Cronan and Aiken 1985, Qualls et al. 1991, Qualls and Haines 1991, Guggenberger et al. 1994, Currie et al. 1996). For the quality of DOC as a substrate for microbial degradation, see Qualls and Haines (1992). In laboratory conditions, the leaching of DOC from the forest humus layer correlated positively with temperature, soil moisture, sulphate concentrations, C:N ratio of the particulate organic matter and leaching frequency, and negatively with ionic strength and metal saturation of the DOC (Michalzik et al. 2001, referring to Clark and Gilmour 1983, Evans et al. 1988, Vance and David 1989, Christ and David 1996, Gödde et al. 1996, Tipping 1998). In the vertical direction, Michalzik et al. (2001) found that, across 42 deciduous and coniferous temperate forests, the DOC transport from the humus layer to the top mineral soil horizon was on average 17 % (range: 6 to 30 %) of the annual litter input in terms of the carbon content. For the production and transport of DOC in forest soils, Prescott and Vesterdal (2021) have published a recent review. There is also a range of soil decomposition models readily available (see, e.g. Michalzik et al. (2003), Ľupek et al. (2016), Mondini et al. (2017), Huang et al. (2018), and Mao et al. (2019).

The formation of lateral runoff fluxes in a pine forest on a podzolic soil has been studied on a low-slope sandy floodplain in Landes de Gascogne, Gironde, southwest France in a location where no surface runoff was observable (Deirmendjian et al. 2018).<sup>5</sup> The concentrations of DOC and DIC in the groundwater had opposing trends with the water table depth (Figure 2-4): The DOC concentrations were the highest in the winter when the water table reached the organic soil horizon, whereas those of DIC reached the maximum concentrations in late summer, that is, at the low water table. It was also observed that there may have been transformation of DOC into DIC within the groundwater during the spring at a rate of  $60 \text{ mmol m}^{-2} \text{ day}^{-1}$  (Deirmendjian et al. 2018). However, the DOC and DIC trends became decoupled when the ecosystem transformed from autotrophic (here, dominated by the root respiration) to heterotrophic (decomposition-dominated) in late summer. The seasonal trends of DOC and DIC concentrations in the groundwater were followed also by the water in the adjacent headwater streams, although with smaller magnitudes as the leaching and runoff comprised only approximately 2 % of the total  $\text{CO}_2$  sink in the forest and about 75 % of the DIC in the streams quickly degassed to the atmosphere (Deirmendjian et al. 2018). The export of DOC into the streams occurred mostly during the high-flow conditions in the spring (about 90 %) and exhibited sensitivity to the spatial and temporal heterogeneity of the groundwater flow rates, but the DIC export was evenly distributed between the 2–3 months of the high-flow regime and the rest of the year with less heterogeneity (Deirmendjian et al. 2018). However, groundwater storage in the soil column was found to react quickly to the precipitation and evapotranspiration and act as a buffer towards the export to the streams, which may partly explain the rather clear connection of DOC and DIC to water-table variations (Deirmendjian et al. 2018). With catchments having a high contribution of surface runoff, for example due to steeper topography or less permeable soils, the carbon export may be more affected by hydrological events (Deirmendjian et al. 2018, referring to Raymond and Saiers 2010, Wilson et al. 2013).



**Figure 2-4.** Conceptual model of fluxes of dissolved organic and inorganic carbon (DOC and DIC, respectively) in a very low sandy slope in a pine forest soil in southwest France, at a site where there is no observable surface runoff (Deirmendjian et al. 2018). NEE, GPP,  $\text{R}_{\text{eco}}$ , OH, WT and D denote the net ecosystem exchange, the gross primary production, the ecosystem respiration, the organic soil horizon, the water table and the drainage, respectively. Main processes acting are represented by dashed arrows and carbon export by solid arrows, the width of which indicates the magnitude of the flux. A ‘positive NEE’ indicates that the ecosystem is a net  $\text{CO}_2$  source to the atmosphere (reproduced based on the CC BY 3.0 licence pertaining to the original work).

<sup>5</sup> Note that the study site of Deirmendjian et al. (2018) is a forest plot instrumented for continuous measurements of precipitation, evapotranspiration and net ecosystem exchanges of  $\text{CO}_2$  and sensible and latent heat (and belongs to the ICOS network), as well as for groundwater and stream discharge and water quality monitoring; this is a potential site for further model testing, as are several similar sites.

During high groundwater flow conditions, the DOC is often found less aged and less recalcitrant than during base-flow conditions (Deirmendjian et al. 2018, referring to Schiff et al. 1997), particularly if the groundwater table reaches the organic soil horizon and changes its biogeochemical conditions and the DOC retention capacity (Deirmendjian et al. 2018) (Figure 2-4). The bioavailability of the DOC is linked to the content of the low-molecular weight compounds, whereas the compounds of higher molecular weight (such as fulvic and humic acids) are more recalcitrant to microbial decomposition (Deirmendjian et al. 2018), so that the high-flow conditions tend to favour microbial metabolism (Deirmendjian et al. 2018) not unlike a wetland-type situation (Section 2.5).

In subsoils of temperate forests, the DOC leaching is generally controlled by retention in the B horizon rich in extractable aluminium and iron oxides (Deirmendjian et al. 2018, referring to Kaiser et al. 1996, Lundström et al. 2000, Michalzik et al. 2001, Sauer et al. 2007, Kindler et al. 2011). When saturated, reducing conditions prevail in such horizons, resulting in dissolution of the iron oxides that limit the retention of DOC through the organometallic complexation, they give rise to possible DOC releases into the groundwater if the conditions have earlier been oxidising (Deirmendjian et al. 2018, referring to Hagedorn et al. 2000, Camino-Serrano et al. 2014, Fang et al. 2016). However, at least in some subtypes of podzols, the degree of complexation of organic carbon with the Al-Fe oxides increases with depth (Deirmendjian et al. 2018, Achat et al. 2011, Ferro-Vázquez et al. 2014). It was estimated for the French site that only about 1 % of the total soil organic carbon is in the dissolved fraction, and therefore only a small part of the soil organic carbon is available for export to streams (Deirmendjian et al. 2018).

In the French pine forest studied, the groundwater becomes enriched in DIC, relative to DOC, in the spring and summer, resulting from the microbial consumption of DOC after the period of high groundwater flows, followed by heterotrophic ecosystem conditions in the late summer favouring downward diffusion and dissolution of CO<sub>2</sub> from the unsaturated layer (Deirmendjian et al. 2018) (Figure 2-4).

For methane production and consumption, drier forest soils exhibit similar behaviour to that in other unsaturated environments addressed in Section 2.3. Thus, there tends to be a methane flux from the atmosphere to the soil, indicating net oxidation of methane in the soil (Bender and Conrad 1993, Shaw and Thorne 2016, and many others). Forest soils are often found somewhat more active in methane oxidation than grasslands and cultivated soils, but less active than wetland soils (Bender and Conrad 1995, Le Mer and Roger 2001, Shaw and Thorne 2016). This may also be linked to the higher methane production near the groundwater table combined with shorter diffusion distance through the unsaturated layer (Bender and Conrad 1994, Le Mer and Roger 2001, Shaw and Thorne 2016), as forest soils tend to be less intensively drained than agricultural soils. There may be also less desiccation stress at low soil moisture to suppress the methanotrophy (von Fischer et al. 2009) and lower nitrogen levels to reduce the methane uptake in microbes (Hütsch et al. 1994, Dunfield and Knowles 1995, Mosier et al. 1998, Le Mer and Roger 2001, Jang et al. 2006) in forests than in cultivated soils (the occurrence of desiccation stress depending also on the irrigation regime, however). Regina et al. (2007) have also suggested that the lesser degree of disturbance of the soil structure (e.g. by ploughing), particularly of the soil aggregates and pore space structure, may favour methanotrophic activity.

However, depending on the degree and efficiency of natural or manmade drainage, some forest areas may have the groundwater table close to the ground surface for large parts of the year so that the soil and the ecosystem behaves more as in a treed wetland (Section 2.5) with characteristically high methane production that may exceed the oxidation capacity (Le Mer and Roger 2001, p 33). Indeed, excess soil moisture and fresh carbon input are prerequisites for methane production in podzolic soil (Korkiakoski et al. 2021) typical of many forest types. This is linked to the observations of the highest rates of both methane production and its oxidation occurring in the organic (top) horizons of forest soils (Yavitt et al. 1990, Le Mer and Roger 2001, Saari et al. 2004, Shaw and Thorne 2016). There may be also small-scale and/or seasonally wet areas within forests that resemble wetland systems (e.g. vernal pools (Ayayee et al. 2020), drainage channels, and ditches).

In temperate forests, the soil-to-atmosphere CO<sub>2</sub> flux may be much reduced with a decline in soil moisture stressing the roots and microbes and possibly leading to limitation of soil respiration (Deirmendjian et al. 2018, referring to Davidson et al. 1998, Epron et al. 1999). In porous sandy soils, drought may favour downward diffusion of CO<sub>2</sub> and its dissolution into the groundwater (Deirmendjian et al. 2018) (see also Figure 2-4).

### 2.4.3 Carbon uptake by forest plants

For the foliar uptake mechanisms and photosynthesis, the processes in forests are the same as for other terrestrial plants, see Section 2.3.3. In many types of the forests there is also a moss or lichen layer and/or considerable amount of understorey vegetation at ground and field layers below the tree canopies. In some forest types there is a gap occupied only by the tree trunks between the understorey and the bottom of the tree canopies. However, with some trees, the canopy can start almost from the ground. Regardless of the forest type, there is a considerable vertical distribution of the primary productivity (and respiration, e.g. (Launiainen et al. 2015, Kovalets et al. 2018).

Root uptake mechanisms essentially are the same as presented in Section 2.3.3. However, roots of trees typically extend deeper (and wider) than those of forest understorey, grassland or crop plants. It is to be noted that recently, Majlesi et al. (2019) have studied the root uptake of carbon and C-14 by Scots Pine seedlings, although grown on a former peat extraction site. They found that up to 3–5 % of the carbon uptake was through roots, in contrast to the atmospheric source, partly attributable to the ectomycorrhizal fungi inhabiting the root system; however, the soil-derived carbon may remain in the roots and not be translocated into the needles (Majlesi et al. 2019).

### 2.4.4 Air exchange in the forest canopy

Regardless of the heterogeneity of the canopy layer(s) in forests, the general principles of air exchange in forest canopies are the same as presented in Section 2.3.3.

In boreal forests, temporal variations occur at multiple timescales, from seconds (radiation regime inside the canopy, leaf gas exchange, turbulent flow, etc.) to seasonal (vegetation phenology and biomass, annual cycle of functional substances) and longer (age structure, species composition, management activities) (Launiainen et al. 2015). An increase in the canopy foliar area increases the fraction of precipitation intercepted, resulting in a decrease in throughfall and soil infiltration (Launiainen et al. 2015), affecting the supply of DOC from the atmosphere and leached from the canopy and litter and humus layers (Section 2.4.2). Another implication of such a change is that the absorption of solar radiation is enhanced and the carbon uptake and transpiration rates increase, consuming the soil water storage, which may lead to a water deficit in the root zone particularly if the mineral soil is coarse-textured (Launiainen et al. 2015); this can further lead to a number of modifications in various processes related to the vegetation, carbon and water fluxes, and soil processes (Launiainen et al. 2015, pp 385–386).

The largest gradients (and heterogeneities) in vegetation canopies are vertical due to absorption of solar radiation and momentum, and it is a common modelling simplification to assume that the horizontal gradients remain inconsequently small compared with the vertical inhomogeneity across the full canopy height (Launiainen et al. 2015). There are several processes that affect the turbulent transport and the CO<sub>2</sub> and water exchange between leaves, canopy elements and the atmosphere across the canopy volume (Launiainen et al. 2015, p 386).

Launiainen et al. (2015) developed a one-dimensional multi-layer and multi-species soil–forest canopy model (APES) for CO<sub>2</sub>, water and energy flows that accounts for the vertical structure and functional diversity of boreal forests and tested that for field observations at the Hyytiälä forest station, Finland. This model describes the main structural and functional compartments, including those related to the moss layer and the understorey vascular plants and their interactions with the forest soil and tree canopy (Launiainen et al. 2015). With stand and soil characteristics derived from measurements and the functional sub-model parameters from literature and shoot-scale measurements, the model predicted the observed fluxes of CO<sub>2</sub>, water and sensible heat well both for diurnal and seasonal timescales, considering the heterogeneities of the forest stand and uncertainties in the boundary conditions (Launiainen et al. 2015).

Kovalets and Avila (2016), in turn, developed a relatively simple assessment model as well as a more detailed Lagrangian model, the latter of which has been validated using measurement data from the Norunda forest station in Sweden (Kovalets and Avila 2016, Kovalets et al. 2018) and tested with measurement data from the Hyytiälä forest station in Finland (Thorne et al. 2018, Kovalets and Avila 2021). The Finnish test case was based on a previous BIOPROTA project (Thorne et al. 2018), where it can be concluded that the profiles of CO<sub>2</sub> concentrations and isotopic ratios within and

above the forest canopy reasonably agree with the measurements (Thorne et al. 2018, Kovalets and Avila 2021) and that the isotopic signatures arising from the soil are rapidly damped in the first few metres of the atmosphere above the soil (Thorne et al. 2018). For the Swedish site, the modelled wind speeds reasonably agree with the measurements above the canopy, but not within, leading to an overestimation of the mixing and an underestimation of the vertical gradient of wind speed just beneath the top of the canopy (Kovalets et al. 2018). Using an alternative, second-order closure model for neutral stratification situations yields a reasonable agreement with the wind speed measurements at the top of the canopy, but significant deviations from the measurements below the middle part of the canopy (Kovalets et al. 2018). The modelled CO<sub>2</sub> concentrations agreed with measurements reasonably well, although inside the canopy layer daytime observations could be successfully reproduced in the model only if the vertical distribution of the respiration fluxes was adequately considered (Kovalets et al. 2018).

#### **2.4.5 Uptake by animals**

The carbon uptake by animals dwelling in or visiting a forest can be modelled by assuming an equilibrium or utilising a biokinetic model, as described for domesticated animals in Section 2.3.3.

The reasonably common allometric approach (a combination of biokinetics/ecological thermodynamics and equilibrium assumptions) can also be expanded by relating the radionuclide concentration in the animal diet with the radionuclide concentration in the animal body by summing up the contributions of relevant food groups (in forests, for example tree wood (twigs), tree leaves, understory plants and mushrooms) (Avila 2006). This is relatively straightforward for herbivores, but the same approach could be applied for carnivores (and omnivores) and for considering other transfer pathways, such as water ingestion and inhalation (Avila 2006). Further, the food intake rates could be expressed as a function of metabolic rates (Avila 2006, referring to Whicker and Schulz 1982). Organ-specific concentrations could be estimated with the help of more detailed kinetic models or simply by applying empirical measured ratios between concentrations in organs and in the whole body (Avila 2006, referring to Coughtry et al. 1985).

For environmental radiation protection, it needs to be noted that there is a considerable variability in the home range areas of the forest fauna (or forest-visiting species), as well as in seasonality (e.g. migration) (BIOPROTA 2015). The range of species also extends from soil-burrowing to avian species with a range of nesting behaviour (including hibernation in the northern latitudes) (Posiva 2013, and others).

It has also been proposed recently that particularly large herbivores (e.g. ungulates) can considerably impact carbon cycling in boreal forests (Leroux et al. 2020).

### **2.5 Transport and accumulation of carbon in wetland ecosystems**

English terminology related to mires, peatlands and in general wetlands tends to vary with the context. According to a reasonably well accepted set of definitions (Joosten and Clarke 2002, Päivänen and Hännell 2012, Joosten et al. 2017a), a wetland is an area that is inundated or saturated by water relatively often and for long times enough to support vegetation adapted for living in saturated soil conditions and this includes both peatlands and mineral-soil sites. A peatland, then, is “an area with or without vegetation with a naturally accumulated peat layer at the surface”, peat meaning “sedentarily accumulated material of at least 30 % (dry mass) of dead organic material”; a peatland area “where peat is currently being formed” being called a mire (Joosten and Clarke 2002, Päivänen and Hännell 2012).

It is to be noted that the widest definition (‘seasonally wet’) can cover most of the land areas in regions with generally positive water balance. However, here, to differentiate from the other ecosystems addressed, the focus is on land areas with the groundwater table being for considerable parts of the year closer to the surface than a ‘few tens of centimetres’ so that most of the methane is not oxidised in the topsoil (cf. Section 2.3.2). In wetlands, the water level may also be above the ground locally

and temporally (including small ponds), which adds characteristics addressed for aquatic systems in Section 2.6. For treed areas, in turn, the continuum is towards forests addressed in the previous section.

There are wetlands commonly also in the littoral zones of freshwater, estuarine and marine systems as an interface or rather a continuum between the ‘land’ and ‘water’ systems. Indeed, for example in a common Swedish practice, wetland is understood to comprise both land with the groundwater table above or near the ground surface and vegetation-covered water areas with at least half of the vegetation being hydrophilic (Löfroth 1991, Gunnarsson and Löfroth 2009).

Some wetlands have brackish or saline conditions, but these have not been directly addressed here due to the limitations of the project framework. Neither is paddy cultivation directly addressed here, even though part of the literature used (particularly regarding the methane processes) may derive from such conditions. It has become clear that there is ample recent literature addressing greenhouse gas balances of such ecosystems, including consideration of combinations of plant cultivation and aquaculture.

### 2.5.1 General characteristics of wetlands

In terms of the carbon cycling and regarding the ‘definition’ of a wetland above, there is little difference between a non-peated wetland and a very shallow lake (or such an estuary/coastal area). What makes a difference here is the intimate coexistence of wetness and the accumulation of carbon as peat. In general, peat formation requires a positive water balance (surplus of water), low rate of decomposition (helped by anoxia), and supply of poorly degradable organic matter (peat-forming vegetation adapted to the wet and low-oxygen substrate) (Nykänen et al. 1995, Limpens et al. 2008). Even though there is a large variability of peatland ecosystems (with the *Sphagnum* mires dominating the literature), there are a number of unifying characteristics of such ecosystems (Limpens et al. 2008):

- There is a strong but temporally variable vertical structuring in the redox potential, governed by the groundwater table fluctuations.
- The upper peat layer (‘acrotelm’, typically 5–40 cm thick) is mostly unsaturated and oxic during the growing season and it supports most of the biological activity, whereas the layer(s) below are waterlogged and anoxic (the deeper, the more permanently so).<sup>6</sup>
- The roots and the root respiration are mainly restricted to the upper zone with at least intermittently unsaturated and oxic conditions.
- The litter degradability decreases with depth in general, as the organic matter ages and the ongoing decomposition produces increasingly recalcitrant residues, but the input from dead roots, root exudates and litter leachates results in some non-uniformity in this with depth.
- The transport of gases and solutes is slower with depth due to the self-compaction of the peat column decreasing the diffusion coefficients and hydraulic conductivity.
- The concentrations of electron acceptors, particularly oxygen, decrease with their distance from the atmospheric source (i.e. from the surface) and tend to rapidly become depleted in the saturated zone.

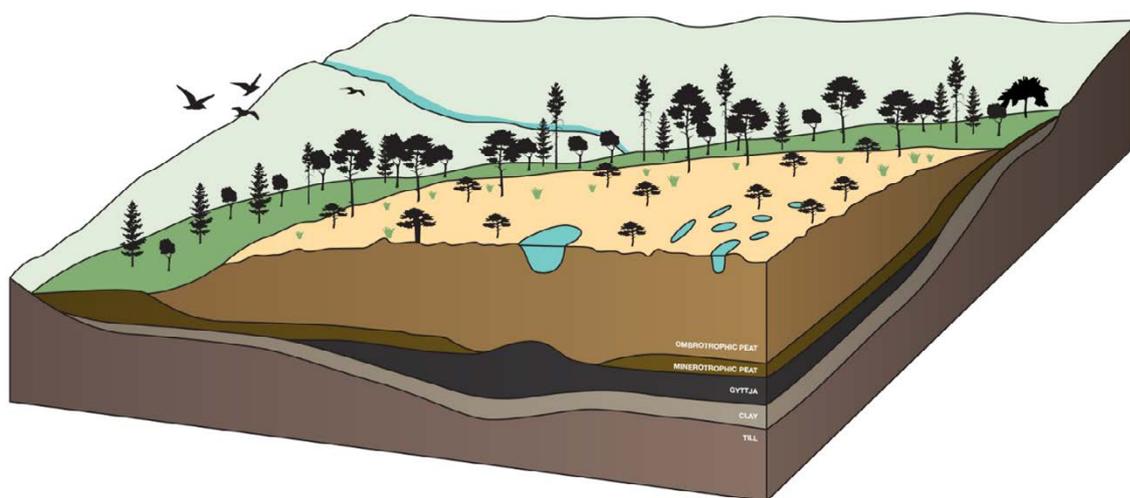
Peatlands can be classified in two main types: bogs (ombrotrophic) and fens (minerotrophic soils/mires). Bogs are fed by rainwater and atmospheric deposition, whereas fens receive ground and surface water (Vitt 2006, Päivänen and Hånell 2012, Joosten et al. 2017a, 2017b, Korrensalo 2017). Bogs typically follow fens in mire development, and therefore there is aged minerotrophic peat underlying the later bog peat (Päivänen and Hånell 2012, Aro et al. 2017, 2018, Joosten et al. 2017a, 2017b). Following from the hydrogeomorphology, bogs have lower nutrient and cation levels, particularly of Ca, and lower pH than fens (Vitt 2006, Laine et al. 2000, Juottonen 2008, Päivänen and Hånell 2012, Joosten et al. 2017a, 2017b, Korrensalo, 2017). The vegetation of the northern bogs is characterised by *Sphagnum* mosses, which further acidify their surroundings, whereas fens typically have more sedges (*Carex* spp.) and other grass-like plants adapted to be rooted in the anoxic peat via their

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<sup>6</sup> For more detailed, yet brief, description on the development and functioning of the redox front linked to continuous peat accumulation, see Section 6.1.1 and the end of Section 6.1.2 in Ikonen (2022).

aerenchyma (i.e. intercellular spaces forming a gas conduit allowing oxygen transport into the roots) (Koncalova 1990, Armstrong et al. 1991, Rydin and Jeglum 2006, Juottonen 2008, Päivänen and Hånell 2012). Drier bog types and subareas may have trees sporadically, often stunted in growth due to nutrient limitation and/or wetness; in some fen types, trees tolerating the wetness and other conditions have a better supply of nutrients and can grow to resemble a mineral-soil forest (Päivänen and Hånell 2012, Posiva 2013, Joosten et al. 2017a, 2017b). However, there are also ‘transition mires’ between the fen and bog types, as well as several different other peatland types such as kettle-hole mires, sloping mires, and spring-water mires (Joosten et al. 2017a, 2017b). It also needs to be recognised that with peat bogs, in addition to the ombrotrophic central parts, there are typically younger lagg and marginal slope areas exhibiting a gradient towards the fen types due to the expansion following the hydrogeomorphic change with the peat accumulation (Päivänen and Hånell 2012, Aro et al. 2017, 2018) (Figure 2-5).

Northern mires are often characterised also by an uneven surface topography (in the lateral scale of about ten metres) intimately linked with the water table and the plant communities. Even though this is more prominent in and more often described for bogs, similar spatial heterogeneity can be observed also in fens, for example with sedge tussocks. In bogs, there are often drier hummocks rising up to 0.1–0.5 m above the mean water table, intermediate lawn surfaces, hollows which have standing water pools in winters and a water table near the surface in summer, and permanent pools (Clymo and Pearce 1995, Korrensalo 2017, Laine et al. 2009, Rydin and Jeglum 2013). Such surface patterning, or microforms, emerge from a self-organising process that provide resilience towards environmental perturbations through the diversity within the plant communities and the variation of physical properties or microhabitats, although being also able to rapidly transform to another quasi-stable state with a stronger perturbation (Belyea and Baird 2006, Dise 2009, Heijmans et al. 2013, Turetsky et al. 2012, Korrensalo 2017). Even though homogeneous plant communities with comparable leaf area are known to function similarly in the ecohydrological sense, they may have different photosynthetic and respiration rates and relationships with and sensitivity to environmental variables, and thus also differ in important characteristics of carbon cycling such as productivity, litter decomposability and association with fungi; in addition, there are many related vegetation–biogeochemistry feedbacks such as the decomposability of litter and the resulting growing substrate quality and its hydrological properties, linking to the redox conditions and microbial activity, partly fuelled also by the root exudates (Riutta et al. 2007a, 2007b, Williams et al. 2006, Limpens et al. 2008, p 1479, Laine et al. 2009).



**Figure 2-5.** A schematic presentation of a typical northern bog complex with bog pools and an underlying, older minerotrophic fen phase extending into younger minerotrophic marginal slope and lagg areas (reproduced from the material of Ikonen et al. (2015) by courtesy of EnviroCase Ltd.).

For the transport of underground releases of C-14, it needs to be noted that the coupling of the surface ecosystem to the groundwater is strong in fens and becomes severed with peat accumulation and consequent hydrogeomorphic change at the transition to the bog phase (Posiva 2013, Ikonen et al. 2015, Aro et al. 2017, 2018). However, the groundwater release and radionuclide accumulation in the deeper peat deposits may continue, and this inventory may become mobilised, or the deposits may become available for example through drainage for forestry or peat extraction (Ikonen et al. 2015, Aro et al. 2018) (or with major changes in the hydrological regime with climate change). The transport in the deeper, permanently waterlogged peat layers is typically diffusive, but it can nevertheless cover considerable distances over centennial and millennial timescales (Clymo and Bryant 2008, Clymo and Williams 2012). In addition, bog pools may considerably shorten the distance between the subsoil and the surface system even in areas with thick peat deposits, and cracks and pipes in the peat may provide preferential transport paths (Limpens et al. 2008, Beadle et al. 2015, Aro et al. 2017, 2018, Ikonen et al. 2020).

In the context of C-14 transport in wetland systems, an extensive data set is available relating to Duke Swamp, Canada. This has become contaminated with C-14 leached in groundwater from an upslope radioactive waste disposal area. Transport of C-14 within the swamp has been modelled using a two-layer approach that distinguishes transport in an upper, metabolically active layer from transport in a lower metabolically inactive layer. With suitable parameterisation, reasonable agreement with field observations has been obtained (Thorne et al. 2018).

## 2.5.2 Carbon storages and cycling in wetlands

As with other ecosystem types, also in wetlands carbon assimilation from atmosphere (photosynthesis) and possibly from the soil, plant production, litterfall and senescence, carbon release from biomass by respiration, decomposition and carbon accumulation in the soil layers are the key aspects of carbon storage and cycling. As outlined in the previous section, carbon accumulation is the most characteristic feature in wetlands, particularly peatlands, but it needs material input from plant production. However, “the peat accumulates under the living mire plants not because of high net primary productivity but rather a slow decomposition rate” (Päivänen and Hånell 2012, p 44). An important factor in the decay is how long the organic matter stays in the oxic surface layer before the rising redox front reaches and engulfs it into the anoxic peat layer (‘catotelm’) (Päivänen and Hånell 2012). However, as a rule of thumb, 5–20 % of the primary production of mires is already initially deposited in catotelm (Päivänen and Hånell 2012). Also, particularly the *Sphagnum* mosses have been found associated with microbial communities that can alleviate the nitrogen limitation typical of peat bogs through fixation of atmospheric nitrogen (Davis et al. 1964, Basilier et al. 1978, Auman et al. 2001, Bodelier and Laanbroek 2004, Raghoebarsing et al. 2005, Larmola et al. 2010, 2013).

Peatlands act as persistent sinks for atmospheric carbon (Päivänen and Hånell 2012, Yu 2012, Rydin and Jeglum 2013, Korrensalo 2017). However, the net *carbon* uptake can be altered or even turned into net release through variations in temperature, moisture and light conditions (Alm et al. 1999, Waddington and Roulet 2000, Lund et al. 2012, Nijp et al. 2015, Korrensalo 2017). However, most peatlands are, despite being net sinks of carbon, net emitters of *methane* originating from the final stage of degradation of soil organic matter (Hoch et al. 2014, and many others). Also, temporal and spatial variability in the methane efflux to the atmosphere has been found to be several orders of magnitude higher than that of carbon dioxide, whereas the cumulative annual sum of the latter seems to be affected more by the climate and physical conditions than internal factors of a mire (Limpens et al. 2008).

Dissolved organic carbon (DOC) compounds that consist of leachates, exudates from roots and recent photosynthates may be transported not only vertically deeper but also laterally to wet depressions, fuelling soil microbial respiration (Boddy et al. 2007, Biasi et al. 2014), or exported through runoff to streams (Section 2.5.3). About 40–70 % of this carbon is relatively quickly released back to the atmosphere as carbon dioxide produced in plant root and shoot respiration (Gifford 2003, Litton et al. 2007, Korrensalo 2017). In the unsaturated surface layer, microbes decompose dead plant material aerobically and produce CO<sub>2</sub> (Section 2.3). The CO<sub>2</sub> is quickly lost to the atmosphere, because the moss canopy is very porous to gases and water, even if rather opaque to visible light (Clymo and Hayward 1982, Hayward and Clymo 1982, 1983, Clymo and Pearce 1995, Clymo and Bryant 2008). Below the water table, anoxic conditions prevail, and CH<sub>4</sub> is produced by anaerobic decomposers

(Section 2.3 and see below), and thereafter oxidised (Section 2.3 and see below), unless it escapes to the atmosphere – in much the same way as in lake and stream bottom sediments (Section 2.6). However, there may be quick back-coupling to the vegetation; of the carbon assimilated by *Sphagnum* mosses, up to 26 % may originate from the methane production, particularly where submerged (Raghoebarsing et al. 2005, Larmola et al. 2010, 2013). The rest of DOC in the system, up to 15 % of the original primary production, remains undecomposed and becomes peat (Clymo 1984, Gorham 1991, Francez and Vasander 1995, Korrensalo 2017).

Water table fluctuations and the spatial heterogeneity of the water table level due to the microforms control the spatial variability of the carbon sink and the proportions of carbon released as CO<sub>2</sub> or CH<sub>4</sub> (Updegraff et al. 2001, Rydin and Jeglum 2013, Korrensalo 2017, Susiluoto et al. 2018, and many others); the lower the water table, the larger proportion of the readily decomposable fresh plant litter is decomposed in aerobic conditions (Korrensalo 2017) and the higher the proportion of CO<sub>2</sub> of the total carbon efflux. The average water table at a location determines, on the other hand, habitats for different ‘eco-hydro-biogeochemical’ plant functional types (Chapin et al. 1996, Frohling et al. 2010, Korrensalo, 2017), which in turn affect both autotrophic and heterotrophic respiration through their root systems and via the DOC quality (Bubier et al. 2003a, Williams et al. 2006, Riutta et al. 2007a, 2007b, Leppälä et al. 2008, Laine et al. 2009, Stráková et al. 2011, Rydin and Jeglum 2013, Korrensalo 2017).

Because plant biomass usually decreases from the dry end of the wetness gradient towards the hollows (Vasander 1982, Moore et al. 2002, Korrensalo 2017), the autotrophic respiration can be expected to follow the same pattern. Regarding the heterotrophic respiration (i.e. decomposition), it is stimulated by a thicker aerobic layer, even though the litter formed in the dry plant communities is more resistant to decomposition (Silvola et al. 1996, Turetsky et al. 2008, Stráková et al. 2011, Korrensalo 2017). In turn, the decomposition of the less recalcitrant litter of plants inhabiting the hollows is hindered by the high water table (Bengtsson et al. 2016, Korrensalo 2017). Combined, this fits to the common observations of the total respiration being the highest in the hummock areas (Alm et al. 1999, Laine et al. 2006, 2007a, Strack et al. 2006). As a rule of thumb, drier bogs, with or without permafrost, seem to have very small, wetter bogs intermediate-level, and fens high or very high methane emissions (Shurpali et al. 1993, Suyker et al. 1996, Christensen et al. 2004, Laine et al. 2007b, Roulet et al. 2007, Limpens et al. 2008).

### **2.5.3 Wetland soils**

#### ***Water-carried fluxes of dissolved carbon***

In peatlands, the water movement and associated transport of dissolved and particulate carbon are dominated by the overland or near-surface flows (Limpens et al. 2008, pp 1480–1481), sensitive to the surface patterning by the vegetation (microforms) and the connectivity of the resulting stream and pool network combined, particularly in older mires, with the gradients in the overall relief across the wetland. The near-surface flow is dominated by the wetter seasons and shorter wet episodes and is focused on the moss canopy and the rather porous surface peat (acrotelm), whereas the groundwater flow in the deeper peat layers is severely limited due to the increasing self-compaction and clogging of the pore space with recalcitrant DOC (Limpens et al. 2008, pp 1480–1481). However, considerable flow conduits (‘peat pipes’) may form particularly in association to bog pools or deformation of the peat deposit with its growth or perturbation (e.g. ground frost, ditching or peat extraction) (Limpens et al. 2008, pp 1480–1481, Dinsmore et al. 2011, Billett and Harvey 2013). The partly apparent downward supply of carbon in the peat layer is typically a combination of diffusion and the growth of the peat column.

#### ***Methane production***

As readily outlined in Section 2.3.2, methane is produced in anoxic soil or anoxic micro-niches by methanogenic microbes. Methane production arises from fermentative degradation of soil organic matter (e.g. dead roots) or photosynthates exuded by plant roots, making plants an important regulating factor of methanogenesis also in wetlands (Lu and Conrad 2005, Conrad 2007, Laanbroek 2010, Bodelier 2011).

The base of the transition zone ('mesotelm') between the oxidised top peat and the anoxic deeper peat is a major site of anoxic decomposition and methane production (Sundh et al. 1994, Belyea 1996, Segers 1998, Kettunen et al. 1999, Eriksson et al. 2010, Clymo and Williams 2012). Even though the main locus of methane production in peatlands is a relatively thin layer at the water table, the CO<sub>2</sub> and CH<sub>4</sub> concentrations increase with depth in the peat, suggesting that there is continued gas production (from the peat decay) in deeper layers (Clymo and Bryant 2008), balanced by the diffusion-dominated slow upward flux. However, the methane production potential peaks a few centimetres or tens of centimetres below the water table and declines with depth (i.e. towards the more decomposed peat) (Williams and Crawford 1984, Sundh et al. 1994, Krumholz et al. 1995, Saarnio et al. 1997, Edwards et al. 1998, Clymo and Bryant 2008, Eriksson et al. 2010, Juottonen 2008]. The location of the peak of the methanogenic potential somewhat below the water table may be due to competition with rapid sulphate reduction (which also converts DOC into a gaseous form) in the periodically oxic transition zone around the average water table (Watson and Nedwell 1998, Vile et al. 2003, Clymo and Bryant 2008).

Somewhat differently from freshwater environments favouring acetate as a precursor of methane, either hydrogenotrophic or acetotrophic methanogenesis may dominate in mires; in permafrost conditions also stimulation of CH<sub>4</sub> production by methanol may occur (Lansdown et al. 1992, Kotsyurbenko et al. 2004, Metje and Frenzel 2005, 2007, Ganzert et al. 2007, Juottonen 2008). However, typically, the acetotrophic methanogenesis is associated with the upper peat layers with abundant labile organic carbon, whereas the H<sub>2</sub>- and CO<sub>2</sub>-dependent methanogenesis predominates in the more recalcitrant deeper peat layers (Hornibrook et al. 1997). Keller and Bridgman (2007), as well as Juottonen (2008, p 6), linked variation in the importance of acetotrophic methanogenesis during the growing season to the presence of root exudates and reported that the acetotrophic pathway dominates methane productions in fens. A similar relationship was reported by Galand et al. (2010), whereas hydrogenotrophic methanogenesis has been reported to be the dominant pathway in bogs (Duddleston et al. 2002, Horn et al. 2003). Thus, the pathway of methanogenesis has been observed to shift from acetotrophic in surface peat, rhizosphere, and sedge fens to hydrogenotrophic in more oligotrophic deeper peat and in *Sphagnum*-dominated bogs – also, seasonal fluctuations are, fittingly, reported more for the acetotrophic production (Juottonen, 2008, p 6). Further information on the methanogenic microbial communities is summarised in Section 6.2 in Ikonen (2022).

The rate of methane production in peat is controlled by the substrate quality, temperature, pH, competition with other microbial pathways, and supply of substrate from the precursor processes; typically, the methane production is limited by the availability of suitable substrates (labile, suitably decomposed but fresh carbohydrates from the plants, opposed to older, more recalcitrant lignin and humic compounds) (Moore and Dalva 1993, Le Mer and Roger 2001, Juottonen, 2008, p 7, Limpens et al. 2008, p 1478, Raivonen et al. 2017, pp 4666–4667). However, it needs to be noted that the aerenchymal plants such as sedges (see below) allocate labile carbon as their root exudates directly into the water-saturated layer and create locally oxic micro-niches (Joabsson et al. 1999, Ström et al. 2003, Juottonen 2008). Fen vegetation may also favour acetotrophic methanogenesis through their root system and root exudate adaptation (Merilä et al. 2006, Juottonen 2008). Also, as already outlined above, the vegetation type influences the litter chemistry and decomposability.

There are also optimal ranges to both temperature (20–35 °C) and pH (6–7) for methanogenesis in peat, although these are rarely met particularly in the northern mires (Juottonen, 2008, p 7). Low microbial activity in bogs as contrast to fens has been related to low pH, low nutrient levels, and recalcitrant or even inhibitory nature of *Sphagnum* residue, making bog peat a poor substrate for microbes. However, comparisons of production rates of CH<sub>4</sub>, CO<sub>2</sub>, and acetate have suggested that the restriction of microbial activity may concern methanogenesis in particular, rather than total anaerobic microbial activity; low pH as such could shape the communities and restrict acetotrophic production in favour of the hydrogenotrophic pathway (Juottonen, 2008, p 28). The limitation of CH<sub>4</sub> production has been particularly severe in bogs at temperatures < 15 °C, but a similar pattern of low CH<sub>4</sub> production potential at ≤ 15 °C and strong increase with temperature was also observed in the Siikaneva fen, which has a low pH that is comparable to the pH of bogs (Juottonen 2008, p 28).

There may also be competition for electron acceptors with other terminal decomposers; for example, with sulphate reduction due to airborne sulphate deposition for H<sub>2</sub>, or with acetogens for CO<sub>2</sub> (Juottonen 2008, p 7). Indeed, there are observations of acetate accumulation in some northern mires with

low methane production levels, even though acetogenesis is considered thermodynamically more unfavourable in peat (Juottonen, 2008, p 7). Couplings and competition with the transformations of other elements have been reported mostly for C, N, Fe and S (Limpens et al. 2008, p 1478). Furthermore, the heterotrophic respiration seems to rely on the removal of the decomposition products in the saturated zone, limiting also the methanogenic capacity at depth; there is thus also a coupling with the water (or gas) residence time (Limpens et al. 2008, p 1478). Methanogenic growth in bog peat could also be limited by lack of required trace elements such as Ni, Fe, and Co (Basiliko and Yavitt 2001, Juottonen 2008).

### ***Methane transport and efflux mechanisms***

Gases, such as O<sub>2</sub>, CO<sub>2</sub>, CH<sub>4</sub>, and H<sub>2</sub> move through the acrotelm (the generally oxic surface layer in peatlands) by three processes: 1) diffusion, 2) ebullition, and 3) through plant roots and stems (Whalen 2005, Clymo and Bryant 2008, Clymo and Williams 2012, Juottonen 2008). For methane, oxidation into CO<sub>2</sub> may alter the transport path due to differences in gas transport properties between CH<sub>4</sub> and CO<sub>2</sub>. In the deep peat, however, only diffusion is plausible, and there CO<sub>2</sub> travels the same path with practically the same rate since CO<sub>2</sub> and CH<sub>4</sub> have nearly the same diffusion coefficients (Clymo and Bryant 2008, Clymo and Williams 2012) even though their solubilities differ considerably.

The general picture of competing transport and oxidation as presented in Section 2.3.2 is valid also here. For lateral and other water-carried fluxes, see above.

### **Diffusive fluxes**

The bulk of methane transported at the catotelm–acrotelm interface seems to be mostly consumed, because the methane oxidation is significantly faster than the upward transport by diffusion (Hornibrook et al. 2009). However, if the layer with potential oxidation is thin and the transport is fast enough, more methane would potentially be emitted, as well as if the other transport pathways contribute; therefore, the depth of the water table regulating the thickness of the oxic acrotelm is an important factor for methane emission also when considering the role of the diffusion path (Sundh et al. 1994).

Properties of the peat column also affect the diffusion of solutes and gases (Raivonen et al. 2017, p 4668): porosity determines the rate of diffusion; the lower it is, the slower is the diffusion. The porosity of peat is generally high (at least 80 %), so therefore peat is relatively easily passable by diffusion, compared with many other soil types. However, diffusion through the surficial peat column is thought to be a minor component in the total methane emissions from a peatland when aerenchymal vegetation (see below) is present at the site, because the diffusion coefficient in water is approximately four orders of magnitude lower than in gas inside the plant roots and stems, and because the probability of the methane becoming oxidised is higher when travelling through peat in contrast to the plant-facilitated bypass mechanisms.

However, considering the long timescales related to groundwater-mediated discharges from the bedrock or subsoil into peat deposits, diffusion distances may be considerable, even if the timescale of diffusive transport is small for the biochemical processes (Clymo and Bryant 2008, Clymo and Williams 2012). The significance of the diffusive transport is enhanced by the severely limited advective groundwater flow in the deep peat layers. However, the thinner the peat is, the more pronounced would the other transport pathways be in this respect.

### **Ebullition**

As outlined in Section 2.3.1, methane tends to reside in the gas phase. However, in waterlogged wetland soils, there is little soil atmosphere available, and bubbling of supersaturated methane from the soil solution is common from surficial peat layers.

The methane in excess of the maximum solubility is transported as gas bubbles to the peatland surface (referred to as ebullition). Estimates of the role of ebullition vary in the range of 0–70 % of the total methane emissions from wetlands, making it possibly a dominant pathway of methane emissions (Glaser et al. 2004, Coulthard et al. 2006, Wania et al. 2010, Xu et al. 2016, Peltola et al. 2018) (but also cf. the aerenchymal transport discussed below).

In peatlands, direct ebullition to the surface is rare since ebullition events usually take place when the water table is below surface and, thus, the ebullitive gas flux is released to the air within the topmost peat layer or moss canopy (Raivonen et al. 2017, Peltola et al. 2018). For the origin of the gas bubbles in respect of the depth in the peat layer, there is observational and theoretical evidence for both deep and surface locations, although for somewhat different reasons; similarly, whether or not the vascular plants hinder the ebullition is under debate (Peltola et al. 2018, pp 946–948). In principle, the ebullition occurs when the concentration of dissolved gases reaches saturation, but in practice, methane ebullition has been observed in wetlands already at concentrations below such a limit (Raivonen et al. 2017). The critical threshold triggering an ebullition event is at around 10–20 % of the total peat volume (Rosenberry et al. 2006, Peltola et al. 2018). In some models, the gas bubble formation is addressed through applying the ideal gas law and Henry’s law, with the gas pressure, temperature and changes in the pore water concentrations regulating the bubble volume (Fechner-Levy and Hemond 1996, Peltola et al. 2018). For triggering an ebullition event, models mostly consider decreasing atmospheric pressure (in some models also linked to wind speed changes), but also water-table changes, peat temperature, changes in the pore-water gas concentration, and total pressure of water-dissolved gases exceeding the ambient pressure are commonly used (Peltola et al. 2018, p 938).

### **Aerenchymal transport**

Differences in methane emissions within and between peatlands, epilittoral wetlands in lakes and rice paddies can largely be attributed to the vascular plant community, associated with 25–97 % of the total methane emissions (Sebacher et al. 1985, Schütz et al. 1989, van der Nat and Middelburg 1998, Käki et al. 2001, Juutinen et al. 2003, Bastviken et al. 2008, Kölbener 2010, Wania et al. 2010, Xu et al. 2016, Peltola et al. 2018, Susiluoto et al. 2018). Similarly to many aquatic plants, almost all vascular plants typical of wetlands have an internal gas transport system (aerenchymae, also referred as the plant bypass or shunt mechanism) regarded as an adaption to low oxygen levels where oxygen is transported downwards and carbon dioxide is transported upwards (Schütz et al. 1991, Cronk and Fennessy 2001, Käki et al. 2001, Juutinen et al. 2004, Brönmark and Hansson 2005), the same system also ventilating out the methane and other gases from the rooting zone (Whiting and Chanton 1992, Clymo and Pearce 1995, Laanbroek 2010, Bodelier 2011). The mechanism is partly based on diffusion due to the gas concentration gradients but mostly on the convective transport caused by pressure, temperature and humidity gradients (Dacey and Klug 1979, Schütz et al. 1991, Chanton et al. 1992, Käki et al. 2001, Juutinen et al. 2004, Brönmark and Hansson 2005, Clymo and Bryant 2008, Clymo and Williams 2012). The methane transported through these plant aerenchyma largely escapes oxidation (Schimel 1995, Juottonen 2008). The abundance of vascular plants therefore is of importance both to the transport of methane, as well as for stimulating methane production through root exudates. Particularly, roots of sedges typical of fens (especially *Carex* species) extend relatively deep and therefore they have a large contact surface with the anoxic peat, especially the fine roots (Raivonen et al. 2017, p 4667). In addition to herbaceous plants, also some trees (e.g. *Alnus* spp.) have aerenchymae (Rusch and Rennenberg 1998, Le Mer and Roger 2001). As outlined above, the plant community is dependent on the type of the wetland and is related to its developmental stage.

With aerenchymal transport, considerable diurnal variability is often reported (Dacey and Klug 1979, Whalen and Reeburgh 1988, Chanton et al. 1992, Mikkilä et al. 1995, Kohl et al. 1996, Thomas et al. 1996, Waddington et al. 1996, Grünfeld and Brix 1999, Käki et al. 2001, Juutinen et al. 2004), which has been attributed either to stomatal opening (*Scirpus* spp.) or to convective transport related to temperature differences (*Phragmites* spp.) (van der Nat et al. 1998, Le Mer and Roger 2001). Also, some observations of the redox potential affecting not only microbial processes but also aerenchymal transport have been reported (Kludze et al. 1993, Kludze and Delaune 1995, Le Mer and Roger 2001). However, some plant types (e.g. *Carex* spp., *Calamagrostis* spp., *Equisetum fluviatile*) do not seem to have the diurnal variability (Moosavi and Crill 1997, Hyvönen et al. 1998, Käki et al. 2001, Juutinen et al. 2004) due to possibly having physiological limitations restricting aerenchymal transport to diffusion only (Juutinen et al. 2004, p 2), but the studies tend to be rather short-term (Milberg et al. 2017). In addition, seasonal variability in the aerenchymal gas-flow resistance has been reported particularly for the taller plants (e.g. *Phragmites australis*) (Grünfeld and Brix 1999, Kohl et al. 1996, Käki et al. 2001, Juutinen et al. 2004, Brönmark and Hansson 2005).

### **Microbial methane consumption and methane oxidation**

Methane oxidation processes have essentially been extensively reviewed by Shaw and Thorne (2016) as summarised in Section 2.3.2, and similar considerations apply to the wetlands (Ikonen 2022). This methanotrophy occurs mostly in the oxic surface moss and peat layers, but also in oxidised micro-niches particularly in the rhizosphere (cf. aerenchymal transport above) (Armstrong et al. 1994, Le Mer and Roger 2001, Shaw and Thorne 2016, Raivonen et al. 2017), even though Shaw and Thorne (2016) consider the latter inefficient due to the short residence time within the thin oxic microlayer. A considerable part of the methane produced in wetlands becomes metabolised by the aerobic methane oxidising bacteria (MOB) before it can escape to the atmosphere. The MOB transform oxygen and methane into carbon dioxide for their energy generation and growth (Conrad 2007, Semrau et al. 2010, Bodelier 2011), which further supports the food webs of soil fauna and beyond.

In peatlands, the methane oxidation potential is typically an order of magnitude higher and less variable by the rate than the methane production potential (Segers 1998, Keller and Bridgman 2007, Galand et al. 2010). In methanogenic environments such as wetlands, oxygen availability is the typical limiting factor to the methane oxidation (Le Mer and Roger 2001). Thus, when the methane passes through the oxic surface layer, depending on the thickness of the layer and tortuosity of the transport path, 60–90 % or even all of the methane produced in anoxic peat may be oxidised into CO<sub>2</sub> by aerobic methanotrophic bacteria (Segers 1998, Frenzel and Karofeld 2000, Le Mer and Roger 2001, Pearce and Clymo 2001, Juottonen 2008, Raivonen et al. 2017, p 4667). In addition, methane oxidation has been reported inside the roots and in the submerged parts of the leaf sheaths of rice plants, as well as in anoxic submerged soils (Le Mer and Roger 2001).

In a literature review of methane transport and fate in the biosphere in the context of radioactive waste disposal, essentially conducted in 2017 (Ikonen 2022), it was concluded that it is difficult to make a balanced estimate of the relative roles of the methane transport pathways and therefore the potential of methane to avoid oxidation, since so many regulating factors are in play at the same time and since there is also potentially considerable spatial and temporal variability involved. Importantly, it was concluded also that particularly taking the large uncertainties inherent to the future landscape development into account, in relation to the release locations from the geosphere, the present common assumption in assessment modelling that all C-14 released from the repository will be transformed into CO<sub>2</sub> and entering food chain through plant uptake is pessimistic but not unjustifiably so, since such situations cannot be completely ruled out in any of the recipient environments (Ikonen 2022).

### **2.5.4 Plant uptake of carbon and canopy processes in wetlands**

In plant communities, both photosynthesis and respiration are dependent on the seasonally varying temperature, moisture and light conditions, as well as the leaf area of the plants themselves (Korrensalo 2017). Furthermore, in peatland communities, seasonal changes in root respiration are controlled by the water table fluctuations and ambient temperature (Fenner and Freeman 2001, Lafleur et al. 2005, Maanaviilja et al. 2011, Korrensalo 2017). In addition, there is also a degree of interannual and successional variability and change in the growing conditions and the responses of the plant communities that are of importance to the carbon balance and long-term carbon accumulation (Waddington and Roulet 2000, Bubier et al. 2003b, Lund et al. 2012, Korrensalo 2017).

For root uptake, see Section 2.3.3, noting the role of aerenchymae with many plant species (Section 2.5.3). For foliar uptake, the discussion in Section 2.3.3 is applicable; for more densely treed wetlands, note also the discussion in Sections 2.4.3 and 2.4.4.

### **2.5.5 Carbon uptake by resident and visiting animals**

For a generally outline, see the discussion on domesticated animals (Section 2.3.3), that on forest dwelling and visiting species (Section 2.4.5), and for species dwelling in peatland hollows and ponds also the discussion on the freshwater species (Section 2.6.3). Biota communities typical to northern bog pools have recently been explored empirically by Kangasniemi et al. (2016) and conceptually in the context of potential radiological exposures by Ikonen et al. (2020).

As a particular coupling between the soil processes and animals in peatland ecosystems, recent studies have explored the role of reindeer droppings increasing methane production on northern fens (Laiho et al. 2017, Fritze et al. 2021). Tentatively, the mechanism is related to the rumen methanotrophs, mainly *Methanobrevibacter* spp., introduced to the soil microbial community (Fritze et al. 2021). There may be similar impact also by other ungulates (ruminants) and possibly other animals even on other types of wetlands and even in drier terrestrial ecosystems.

## **2.6 Transport and accumulation of carbon in freshwater systems**

Freshwaters cover only a small proportion of the Earth's surface area, but are extremely important for the carbon cycle, affecting regional carbon balances (Cole et al. 2007) and acting as intermediate environments that sequester and transform carbon from the atmosphere and terrestrial environment and transport it to coastal environments. Inland waters provide ideal conditions for microbial metabolism of organic matter, and they process disproportionately large amounts of organic carbon (Catalán et al. 2016).

### **2.6.1 Freshwater systems and their terrestrial catchments**

Freshwater systems include springs, streams, rivers and lakes. Within the biosphere they are most conveniently described in terms of drainage networks that route water flows through surface-water catchments. The catchment of a freshwater system is the area of land drained by a well-defined part of the drainage network. Where branched systems occur, e.g. multiple streams draining to a river, a sub-catchment may be defined for each stream, with these sub-catchments comprising components of the overall river catchment. Within a catchment, the movement of water and sediments transfers both nutrients and contaminants within the system and across its boundaries, such that there is a close coupling between terrestrial and aquatic, and biotic and abiotic components. Catchments and sub-catchments are separated by higher ground (interfluves) that delineate the extent of the catchment and constitute flow divides for incoming precipitation (Giller and Malmqvist 2011).

Catchments play a crucial role in shaping streams, rivers and lakes. They not only determine the morphology of the water bodies, but also the chemical composition of the water that, in turn, affects biotic communities and their interactions. Precipitation infiltrates into the catchment and is in contact with soils, subsoils and the underlying rock as it moves downslope, resulting in changes in its chemical composition (Giller and Malmqvist 2011). The degree to which the catchment affects the chemical composition of surface and subsurface water varies according to several factors. For example, catchments subject to high rainfall will exert less influence on the chemical composition of freshwater bodies due to the rapid transit of water through the catchment. Similarly, the transit time for water in small catchments will be low, which may again limit the opportunity for the chemical composition of precipitation to be affected and for carbon and other nutrients be leached. Land use within the catchment will also affect water chemistry. For example, intense agricultural activity can result in high nutrient loading in lakes and eutrophication (Brönmark and Hansson 2017).

In the following subsections, key features and processes associated with freshwater drainage networks and their catchments are briefly described. Whilst freshwater streams, rivers and lakes are often highly interconnected, processes can vary considerably between fluvial and lacustrine systems and are therefore discussed separately.

### **2.6.2 General characteristics and key processes**

#### ***Streams and rivers***

River networks, comprising streams and rivers, are recognised as important components of the global carbon cycle (Deirmendjian and Abril 2018). Streams and rivers are characterised by flowing water within a channel that can vary considerably in size along the long profile of a river. Water flow in fluvial systems is unidirectional, but the rate of flow can vary widely, being largely driven by rainfall, but also influenced by catchment characteristics, including soil type and its permeability and depth, the types of vegetation present and the slope of valleys. Groundwater will also influence water flow. Globally, the mean residence time of water in rivers and streams is 7–14 days (Giller and Malmqvist

2011). Although the pattern of flow in streams and rivers is governed by channel characteristics, it is important to bear in mind that, in the longer term, both the form of the channel, e.g. straight or meandering, and its dimensions are governed by topography, the underlying lithology and the volume of flowing water available.

The rate of flow and chemical characteristics of water are largely driven by rainfall and can therefore vary widely according to rainfall events. However, there is also longitudinal variation in water flow from the headstream to the point at which rivers meet the sea as tributaries merge and additional water percolates through the catchment to the water body. Water flow is also affected by catchment characteristics, including the soil type, its permeability and depth, the slope of valleys and the types of vegetation present.

The geology of the catchment has a large influence on the water chemistry of streams and rivers. For example, rivers and streams flowing through granitic catchments are often characterised as having low levels of dissolved salts and low pH whilst those within catchments overlying sedimentary rocks are more likely to have water that is neutral or alkaline with higher levels of dissolved salts (Giller and Malmqvist 2011). The vegetation in the catchment also influences the water chemistry. This is particularly evident with coniferous trees where rainfall can be acidified as it passes through the canopy (Giller and Malmqvist 2011).

Streams and rivers are spatially variable, consisting of riffles and pools with turbulence being induced by factors including streambed roughness, stream gradient, channel flow and fluctuations in discharge. Oxygen enters rivers primarily via diffusion across the air-water interface, but levels can be affected by turbulence. As such, fast-flowing sections of rivers are likely to be supersaturated with oxygen, whereas slow-flowing pools are more likely to have lower levels. These pools are also likely to be associated with particulate organic matter as the slower flowing water allows for sedimentation to occur. Decomposition of accumulated organic matter by microbes and invertebrate communities may then reduce oxygen levels in pools further because of metabolic processes.

In addition to terrestrial influences on streams and rivers, groundwater exchange also occurs and can influence water flow. Exchange between surface waters and groundwaters in streams and rivers occurs within hyporheic zones, which are areas of porous saturated sediments that act as a transitional area between surface and groundwater hydrological systems. The hyporheic zone is a spatially and temporally variable feature of streams and rivers, expanding and contracting seasonally with changes in surface and subsurface flows. Hyporheic mixing occurs via downwelling and upwelling circulation cells in the porous sediment. The process is driven by spatial and temporal variations in channel characteristics including streambed pressure, bed mobility, alluvial volume and hydraulic conductivity (Tonina and Buffington 2009). Downwelling leads to solutes and oxygen-rich surface water being transferred advectively into sediments. Upwelling groundwater can also be a source of nutrients to the zone. The exchange between groundwaters and surface waters within the hyporheic zone results in physicochemical gradients that influence the type and quality of subsurface habitat available to organisms (Tonina and Buffington 2009).

Vegetation in streams and rivers is heterogeneously distributed, being affected by factors such as substrate, water flow, turbidity and the availability of light. Light levels are affected by factors including season, altitude, shading by trees etc., and water depth and clarity. Between 20 % and 98.5 % of the light striking the surface of a river is reflected, depending on the angle of incidence, which can vary according to time of day, latitude and orientation of the stream or river valley in relation to the sun throughout the day (Giller and Malmqvist 2011). Of the light entering the water, the amount penetrating to depth will be affected by factors such as turbidity and water colour, but generally declines logarithmically with depth, according to Beer's law (Giller and Malmqvist 2011).

The riparian zone (the boundary between the aquatic and terrestrial systems) can have a large influence on fluvial systems, acting as a source of carbon to streams, and controlling bank erosion and sediment inputs. The riparian zone can also affect primary productivity through shading. There is a negative relationship between the light reaching stream autotrophs and riparian canopy development. It is estimated that between 5 % and 10 % of sunlight reaches 1<sup>st</sup> and 2<sup>nd</sup> order streams whereas up to 46 % can reach the bottom of 9<sup>th</sup> order rivers (Giller and Malmqvist 2011).

Heavily shaded or turbid rivers are generally devoid of macrophytes. Where macrophytes are present (e.g. in riparian zones) there can be clear seasonality (Giller and Malmqvist 2011). The presence or absence of primary producers in response to shading etc., also affects the distribution of invertebrate species. In contrast to lakes where many species are pelagic in nature, biota in fluvial systems tend to be more closely associated with the substrate. The physical properties of the substrate are therefore important, providing habitats for benthic invertebrates as well as food sources. In general, substrate particle size increases with the velocity of the water flow such that in rapid turbulent flowing areas of streams and rivers the substrate is likely to be dominated by gravel, pebbles and boulders as smaller particles are transported downstream. In slower-flowing areas there will be greater opportunity for sedimentation of finer particles resulting in finer, more uniform substrates. Nonetheless, even in coarser substrates, there can be pockets of finer sediments, where the coarser particles provide protection for finer sediments, as well as organic matter and detritus from being carried away such that more heterogenic substrates can form. However, the dynamic nature of fluvial systems results in sediment particles and detritus being redistributed with changing water flows, for example due to rainfall events (Giller and Malmqvist 2011). Generally, the more heterogenic the substrate, the greater the diversity and abundance of species due to the greater range of micro habitats and microflow patterns available for species to inhabit. Species abundance and diversity also tend to be higher in areas where detritus gathers such as twigs and leaves, which provide both habitat and food source for organisms.

The carbon cycle is driven by water movement and, as such, streams and rivers play a critical role in the lateral exchange of carbon between land, lakes and oceans (Duarte and Prairie 2005, Ward et al. 2017). Streams and rivers are also associated with strong carbon dioxide and methane fluxes to and from the atmosphere (Butman et al. 2018).

Gas transfer velocities in the lower reaches of large rivers have been shown to be much greater than further upstream due to large channel widths, long fetches, tidally influenced current velocities, and stronger winds closer to the sea. Although CO<sub>2</sub> concentrations may decrease along downstream gradients, enhanced gas transfer velocities can result in equivalent or greater CO<sub>2</sub> evasion rates in the lower parts of rivers compared with upstream (Ward et al. 2017).

## **Lakes**

Brönmark and Hansson (2017) define a lake as “a waterbody where wind-induced turbulence plays a major role in the mixing of the water column”. Lakes form in areas of the landscape that are lower than their surroundings and their morphology is governed by the shape of the terrestrial catchment in which they lie. Inflow to the lake may be from general surface and subsurface drainage across the catchment or as more direct stream or river inputs. However, the flows in those streams and rivers will usually derive, in whole or in part, from general surface or subsurface drainage. Most lakes also have streams or rivers as natural outflows that balance and maintain lake water levels, although endorheic lakes also occur, in which water is lost solely by evaporation or underground seepage (Wetzel 2001).

Lakes are comprised of various habitats. These include the littoral zone (the near-shore part of a lake where light penetrates to the bottom) the pelagic zone (the open water area) that can be divided, depending on the depth of water in the lake, into the photic and aphotic zones, in addition to the benthic habitat that relates to the sediment. The photic zone is the part of the water column where there is a net production of oxygen from photosynthesis and occurs where light intensity is above 1 % of surface light. The depth of the photic zone is affected by the amount of dissolved and suspended substances but, in general, 50 % or more of light energy is absorbed in the first metre of the water column (Brönmark and Hansson 2017). The depth range over which photosynthesis and respiration rates are approximately equal is known as the compensation zone and below this is the aphotic zone.

Temperature is a key environmental factor for freshwater ecosystems, affecting both abiotic and biotic components. Solar energy is largely absorbed in the upper few metres of lakes resulting in heating of the upper layer of water, leading to seasonal thermal stratification between the warmer less dense upper waters and colder deeper waters in lakes with sufficient water depth. Shallower lakes can also become stratified if the water is dark coloured because this results in a steeper gradient in the absorption of solar energy with depth. The stratum between these layers is known as the

thermocline or metalimnion, where there is a steep thermal gradient between the warm oxygen-rich but nutrient-poor surface water (epilimnion) and cold oxygen-depleted, but nutrient-rich deep water (hypolimnion). Stratification and the formation of a thermocline is a key process in the current context, affecting both the structure and function of the lake and the carbon cycle within it.

The typical stratification and circulation cycle for a moderate-sized temperate lake is described in Brönmark and Hansson (2017). During the winter, water temperature will be at its lowest and the water will be at its highest density (at a temperature of approximately 4 °C), with little difference in temperature with depth. At this time, mixing will be at a maximum due to lack of stratification through the water column and the entire water body will circulate in what is termed the spring turnover. As solar radiation increases and the water warms through spring, there is an increased water temperature in the upper part of the water column where light absorption is at a maximum. During warm, calm conditions, a variation in water temperature of only a few degrees can result in sufficient density differences between the upper warmer waters and cooler waters at depth for stratification to occur and a thermocline to develop. The thermocline acts as a barrier to mixing between the epilimnion and hypolimnion throughout the summer such that turbulence-induced water circulation is restricted to the upper epilimnion. During the autumn, solar radiation energy input to the lake reduces, but heat loss through evaporation continues, leading to a reduction in temperature and density within the epilimnion. Once the difference in temperature and density between the epilimnion and hypolimnion is sufficiently reduced, the thermocline 'barrier' is weakened and increased turbulence from autumn winds leads to recirculation of the water column (the autumn turnover) and mixing between the oxygen-rich surface water and nutrient-rich deep water. Continued cooling through the winter then returns lake water to its most dense and an inverse stratification can occur if the surface of the lake cools below 4 °C becoming less dense than the underlying water. The density difference between the layers is small, however, and is easily disrupted by wind turbulence. Should temperatures drop low enough, surface ice can form which then shields the lake from wind energy impacts until temperatures once again increase in the spring leading to ice melt and the restart of the cycle.

During the period of stratification, the lake is distinguished into the epilimnion, where circulation occurs and light intensity and primary productivity are high, and the cold hypolimnion where circulation is minimal, photosynthesis is largely absent and carbon input to the system is largely the result of sedimentation of organisms from the epilimnion. Once they reach the hypolimnion, they decompose, releasing nutrients back to the water. With the lack of mixing between the two layers, nutrient availability in the epilimnion can become a limiting factor for primary productivity.

In addition to triggering lake stratification, temperature also affects oxygenation of the water. The amount of oxygen in solution generally decreases as the water temperature increases but can be affected by physical factors such as turbulence and biological processes such as rates of photosynthesis and respiration. There are two main sources of oxygen to lakes – photosynthetic oxygen and oxygen that diffuses across the atmosphere-water interface. When lakes are fully mixed, oxygenation will be close to 100 % throughout the water column, but with stratification, both photosynthetic oxygen and atmospheric oxygen are largely restricted to the epilimnion where saturation levels above 100 % can occur in well-lit habitats during the daytime because of active photosynthesis. Oxygen concentrations then reduce in the epilimnion during the night as respiration becomes the dominant process for both plants and animals. Without continued diffusion of oxygen to the deeper hypolimnion, where light penetration is insufficient to support photosynthesis, oxygen concentrations become depleted because of respiration by benthic organisms and decomposition of organic matter, and the hypolimnion may become an unsuitable habitat for many aquatic biota until mixing throughout the water column resumes in the autumn. Ice formation in the winter may also affect oxygen levels, preventing gas exchange across the water-atmosphere interface and limiting light penetration. Under such conditions, combined with decomposition, considerable oxygen depletion may occur.

Several factors affect the depth of the epilimnion, the temperature of the hypolimnion, and mixing between them. These include latitude, lake morphology, climate, weather and water clarity. In some lakes where ice formation does not occur, there is one long period of mixing, rather than distinct spring and autumn turnover phases, and in shallow lakes that are not subject to stratification, mixing may continue throughout the year. The extent to which waters are circulated and mixed within lakes will also be affected by the depth of the lake, with bottom waters of very deep lakes potentially being

excluded from mixing. The influence of wind-induced turbulence on the circulation and mixing of lakes will be greater in lakes with a large surface area where fetch (the length of the lake that wind acts on) is greatest (Brönmark and Hansson 2017).

### **2.6.3 Carbon transport in freshwater systems**

Abiotic carbon is transported in freshwater systems both in dissolved form (as carbon dioxide in solution and as bicarbonate) and as particulate material. These two forms are generally described as DIC (dissolved inorganic carbon) and PIC (particulate inorganic carbon), respectively. In contrast, DOC and POC denote dissolved organic carbon and particulate organic carbon, respectively. The transport of these various forms in freshwater systems is governed by processes of water and sediment transport, as described in the previous subsection. Thus, here the focus is on biotic processes involving uptake, retention and loss of these various forms. It is emphasised that the metabolism of organic carbon in streams and rivers is responsible for a large amount of outgassing of carbon dioxide to the atmosphere, particularly from hydrological retention zones that extend the residence time of organic carbon and give microbial processes time to operate (Battin et al. 2008). Cole et al. (2007) estimated that worldwide, inland waters receive about  $1.9 \text{ Pg[C] y}^{-1}$  from the terrestrial landscape, of which about  $0.2 \text{ Pg[C] y}^{-1}$  is buried in aquatic sediments, at least  $0.8 \text{ Pg[C] y}^{-1}$  is returned to the atmosphere by gas exchange and the remainder is delivered to the oceans. Although carbon dioxide is the main gas released, there can also be significant releases of methane produced in anoxic sediments (Crawford and Stanley 2016). Of the carbon transported to oceans, around 25 % is in the form of POC, 25 % is DOC and 45 % is DIC, with the remainder being particulate inorganic carbon (Polsenaere and Abril 2012).

#### ***Carbon sources, transport and cycling in streams and rivers***

Carbon in fluvial systems can be autochthonous or allochthonous. For any given stream, energy inputs can take the form of instream primary production, litterfall and lateral contributions from riparian vegetation, DOC in groundwater, and inputs from upstream. Fluvial systems can therefore be categorised as heterotrophic (dependent on external carbon sources) or autotrophic (primary production exceeds allochthonous input) depending on the ratio between primary production (P) and respiration (R). Systems with a P/R of 0.5 or above are considered autotrophic (Giller and Malmqvist 2011). Changes in whether streams and rivers are autotrophic or heterotrophic can be observed on various temporal and spatial scales. For example, streams may be autotrophic during periods of low rainfall, but with increased flow they become heterotrophic, due to rainfall bringing in more outside carbon. Upstream is also more likely to be autotrophic as compared with downstream where the water flow is larger than it is up-stream and turbulence can affect light penetration. In addition, downstream locations will be more highly influenced by catchment inputs but can be influenced by biotic and abiotic factors. For example, greater flow and turbulence in higher-order rivers can limit light penetration or large-scale grazing may reduce primary productivity (Stanley et al. 2016).

The primary carbon source driving the autochthonous food web in streams and rivers (and in lakes) is dissolved inorganic carbon (DIC) in the form of carbon dioxide or bicarbonate that is fixed by photosynthetic organisms. This DIC may derive from atmospheric exchange of carbon dioxide across the air/water interface, via water exchange (e.g. carbon dioxide or bicarbonate in river or stream water, groundwater or rainwater flow through the surface water catchment area) (Ascough et al. 2010, Brönmark and Hansson 2017). The different reservoirs of DIC supporting streams and rivers have different isotopic signatures that can be used to help distinguish sources in different systems and the variability in sources throughout a system (Polsenaere and Abril 2012, Deirmendjian and Abril 2018). Groundwater can be a particularly important source of DIC, particularly in headstreams, with root and soil microbial respiration contributing to the groundwater DIC levels during periods when the water table is high, allowing interaction with soils (Deirmendjian et al. 2018). The relative contribution of the different sources of DIC can therefore vary considerably, with rainfall and temperature having a large influence (Deirmendjian et al. 2018). Overall, the contribution of groundwater DIC decreases with increasing stream order and water flow, as within stream carbon dioxide production increases (Deirmendjian and Abril 2018).

Due to the flowing nature of fluvial systems and the short residence time of water, phytoplankton are often less abundant in streams and rivers, except in lake outflows. Periphyton and macrophytes are therefore the main primary producers, with macrophytes primarily being associated with low-gradient streams and rivers where finer sediments are present for roots. The rates of primary production vary considerably.

Where atmospheric exchange is a dominant consideration, the availability of carbon dioxide can be a limiting factor in aquatic systems. The diffusion of carbon dioxide in water is around 10 000 times slower than in air and, as macrophytes incorporate carbon dioxide, microzones with low carbon-dioxide concentrations can be formed around the macrophytes due to the low diffusion rate (Brönmark and Hansson 2017). Some aquatic plants, including species within the genera *Isoetes* and *Littorella*, have learned to adapt to these low carbon dioxide concentrations that can occur during the day when photosynthesis is active by using crassulacean acid metabolism (CAM), which allows carbon dioxide to be taken up at night, when respiration rates are highest resulting in carbon dioxide levels being replenished, and transformed into malic acid which is then stored in vacuoles. The malic acid is then converted back to carbon dioxide during the day, when carbon dioxide concentrations in water are limited, and used in photosynthesis. CAM can provide up to 50 % of the total carbon requirement of the plant (Brönmark and Hansson 2017). Emergent macrophytes can access both carbon dioxide in air and dissolved in water.

Allochthonous organic matter, in the form of DOC and both coarse and fine POC, has long been known to fuel stream food webs and can be a major input to the carbon budgets of fluvial systems. One of the first budgets constructed for a forested headwater stream was for Hubbard Brook, New Hampshire (USA), which provided a comprehensive account of inputs, standing stocks, and outputs of organic matter, and demonstrated the overwhelming contribution of allochthonous material to stream-energy budgets (99 % of total inputs) (Stanley et al. 2016). Early budget studies indicated that streams were detritus-based, heterotrophic in nature and therefore highly dependent on watershed-derived organic matter, but later studies then demonstrated that autochthonous production could also be a substantial source of organic matter to headwater streams (Stanley et al. 2016).

Allochthonous DOC from the catchment can account for a significant portion of the total organic carbon input to streams and rivers (Stanley et al. 2016). Sources of allochthonous DOC include transport of terrestrially decomposed organic matter in surface water run-off, rain fall through overhanging canopies, and groundwater (Giller and Malmqvist 2011) and the greatest inputs tend to be associated with the drainage of wetlands and organic soils (Stanley et al. 2016). DOC inputs are influenced by several different factors, including hydrologic flow paths, hydraulic conductivity, stream size, and land use (Stanley et al. 2015). Precipitation and soil type can also be important factors (Deirmendjian et al. 2018). Generally, the largest DOC concentrations are associated with streams draining wetlands and watersheds with organic soils (Stanley et al. 2016). Groundwater can also be an important source of DOC to streams. For example, in a study of first order streams, Deirmendjian et al. (2018) found that of the DOC entering streams in groundwater, 70 % was either degraded or immobilised, preventing downstream export to second-order streams. However, occasional high rainfall or large snow/ice melt events can result in a pulse of terrestrial DOC entering streams that is then rapidly transported downstream to larger rivers. The rapid water flow during such events reduces the residence time of the DOC in headstreams and transports it to larger downstream rivers or is exported to coastal systems, thus altering the downstream gradient in labile terrestrial DOC through the stream-river continuum (Raymond et al. 2016).

The quality of DOC varies according to its origin. In-stream derived DOC, resulting from detrital leaching, exudates from primary producers and animal excreta, tend to be more readily utilised sources (Giller and Malmqvist 2011). Terrestrial DOC tends to be more recalcitrant (Stanley et al. 2016). Labile DOC tends to be utilised in streams and rivers close to its point of entry whereas less labile forms tend to be exported downstream (Stanley et al. 2016). The uptake of DOC into the food web is largely the result of microbial uptake of small labile DOM and subsequent transfer to consumers but can be via flocculation to form fine POC that is then available to filter feeders in addition to microbes (Giller and Malmqvist 2011).

Allochthonous coarse POC (CPOC) is largely derived from the riparian zone in the form of branches, leaves, flowers and pollen from overhanging and surrounding vegetation as well as terrestrial invertebrates falling into the water. Inputs of CPOC are therefore primarily controlled by seasonality and

the vegetation in the riparian zone such that streams in forested catchments can have significantly higher CPOC inputs compared with those in unforested catchments (Stanley et al. 2016). CPOC tends to collect in front of boulders, be snagged on larger debris such as twigs or, in slow-flowing areas, CPOC can sediment out of the water column. Twigs and branches present in streams and rivers can also capture other CPOC, which serves to increase the residence time of collected detritus (Giller and Malmqvist 2011).

Allochthonous Fine Particulate Organic Carbon (FPOC) largely enters streams and rivers because of wind blow, surface water run-off and animal excreta. FPOC can also result from the within stream degradation of allochthonous CPOC or from flocculation of DOC. In contrast to CPOC, the distribution of FPOC in fluvial systems tends to be positively related to stream order (Giller and Malmqvist 2011).

The relative contribution of allochthonous carbon resources can vary considerably throughout the course of the water body. In general, less coarse POC enters open streams compared with forested streams and quantities tend to decrease from low-order streams to higher-order rivers (Giller and Malmqvist 2011). Low-order streams are more likely to be shaded which can limit in-water primary production and allochthonous sources of carbon such as leaves are more likely to dominate the system. For example, energy budget studies have suggested that allochthonous carbon sources can contribute up to 99 % of the carbon requirements of headwater streams draining deciduous forests (Stanley et al. 2016). Higher-order rivers are likely to be much wider and the relative influence of the riparian zone will be lower and, with greater light reaching the water, primary production can increase. Seasonality is also an important consideration. For example, autumn leaf fall can be a major source of CPOC to streams and rivers in temperate fluvial systems. Salmonid spawning events can also introduce seasonal inputs of CPOC to rivers. The retention of CPOC tends to reduce from low- to higher-order rivers (Giller and Malmqvist 2011).

The decay rate of organic carbon in inland waters is significantly and negatively related to the water retention time (Catalán et al. 2016). Across all inland waters (i.e. including lake systems), the mean half-life of organic carbon has been estimated as 2.5 ( $\pm 4.7$ ) years, which is similar to that of soil litter, but significantly lower than for marine waters and sediments (hundreds to thousands of years) (Catalán et al. 2016). The processing of organic matter can result in both carbon dioxide and methane being released to the water column. Methane production can be particularly high on river floodplains (Ward et al. 2017). However, a large proportion of the methane generated in streams and rivers can be rapidly oxidised within the water column in rapidly flowing, turbulent waters (Ward et al. 2017).

Fresh leaf litter entering fluvial systems is not readily consumed by aquatic biota but can be substantially and rapidly modified because of microbial degradation. On entry to streams and rivers, up to 25 % of the initial dry weight of leaves can be lost as DOC within the first 24 hours with continued losses over longer timeframes. Microbial colonisation and degradation of leaves then results in the release of FPOC and up to 75 % of the leaf mass can be degraded to FPOC by the action of microbes over a matter of weeks (Giller and Malmqvist 2011). Microbial metabolism of POC also leads to the production of DIC in the form of both carbon dioxide and methane (Stanley et al. 2016). The action of microbes in processing leaves also serves to condition the leaves making them more digestible by detritivorous micro- and macro-invertebrates. The detritivores then further help in the breakdown of CPOC to FPOC either because of leaf fragmentation during feeding or through the production of faeces. The microbial community responsible for conditioning leaves also acts as a direct food source for detritivores, contributing up to 10 % of a detritivores diet (Giller and Malmqvist 2011). The micro- and macro-invertebrates provide one of the highest quality food resources in fluvial systems, supporting predatory fish. Terrestrial insects can also provide an important food resource for some fish species, such as trout (Giller and Malmqvist 2011).

Spatial cycling of organic matter is an important feature of streams and rivers. The unidirectional flow of water means that as carbon is metabolised by the biotic community, the carbon released as DOC or POC is transported to be utilised by the downstream community such that there is a net downstream loss of carbon with each uptake-release carbon cycle. This 'spiraling' of carbon results in an increased retention of carbon in fluvial systems (Giller and Malmqvist 2011). The length of spirals, i.e. the distance travelled between release of carbon and re-utilisation, depends on the biotic community and velocity of water flow. In low-flowing systems, carbon is likely to travel only short distances before being recaptured.

The hyporheic zone is also a potentially important feature affecting the carbon cycle in streams and rivers. The zone provides a diverse habitat for a range of biota from microorganisms to invertebrates and fish and can contribute up to 50 % of the total ecosystem respiration (Stanley et al. 2016). The primary factors influencing metabolism in streams and rivers include light availability, temperature, nutrient and organic matter supply, which are themselves influenced by local and regional factors (Stanley et al. 2016).

The processing of organic matter in streams and rivers leads to carbon dioxide production through respiration of micro- and macro-fauna. Some of this carbon dioxide will be taken up by primary producers in downstream waters and some will be transported in water to estuaries and coastal waters, but a significant amount can be lost to atmosphere (Butman et al. 2018, Hao et al. 2021) either as methane or carbon dioxide. Streams and rivers are often supersaturated by carbon dioxide compared with concentrations in the above-water atmosphere and, as such, significant rates of degassing can occur (Deirmendjian and Abril 2018). For example, Deirmendjian et al. (2018) estimated that around 75 % of DIC entering first-order streams in groundwater was rapidly lost to atmosphere with rates of degassing being of the order of  $3.1 \pm 2.2 \text{ gC m}^{-2} \text{ y}^{-1}$ . Degassing fluxes can, however, be influenced by various factors, including seasonality, water flow and turbulence and catchment characteristics (Polsenaere and Abril 2012).

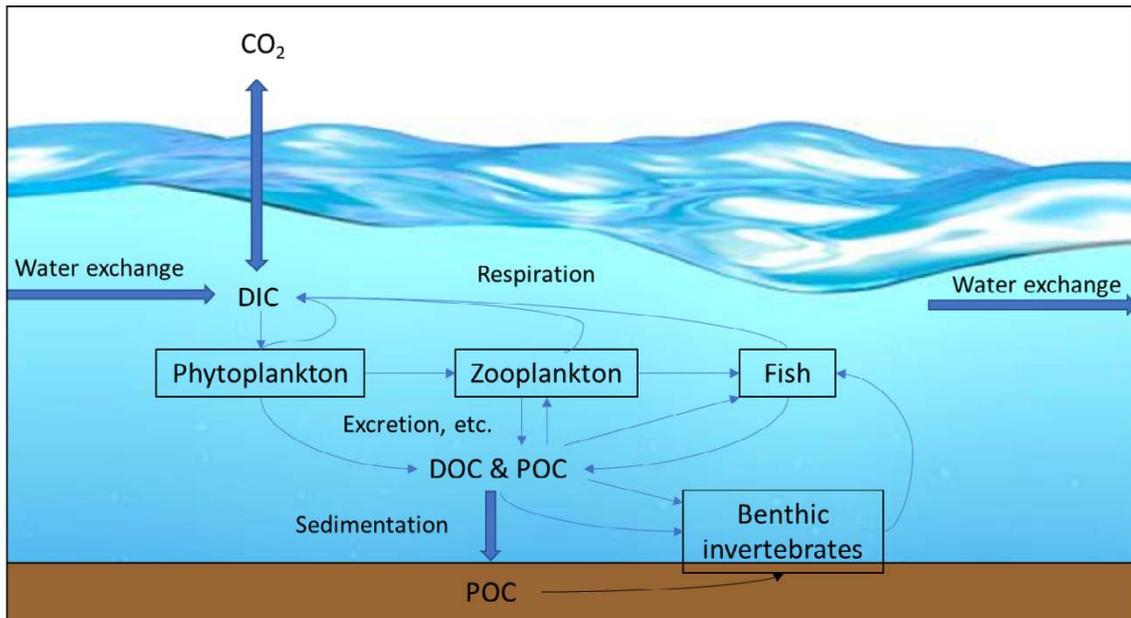
Degassing hotspots can occur that are often linked to turbulent areas (Billett and Harvey 2013) or groundwater upwelling zones (Deirmendjian and Abril 2018, Deirmendjian et al. 2018). Rainfall events that increase turbulence can therefore increase emissions and the presence of waterfalls can lead to higher emissions (Natchimuthu et al. 2017). Deirmendjian et al. (2018) also reported an increase in degassing rates during higher water flow in streams. The quantity of carbon dioxide emitted to atmosphere from streams and rivers also varies diurnally with up to around 30 % more being emitted at night than during the day (Gómez-Gener et al. 2021).

The processing of organic matter in streams and rivers can also result in methane being released to the water column, and many fluvial systems are supersaturated with methane because of the decomposition of organic matter (Stanley et al. 2016). Methane production tends to be particularly high on river floodplains because of the decomposition of organic matter in anoxic sediments (Ward et al. 2017). Methane can occur because of diffusion from the atmosphere, transport in water from upstream, or from groundwater discharge. A large proportion of the methane generated in streams and rivers can be rapidly oxidised within the water column in rapidly flowing, turbulent waters (Ward et al. 2017) or lost to atmosphere (Stanley et al. 2016). Losses to atmosphere may occur because of diffusion, ebullition or plant-mediated transport (i.e. transport through aerenchyma) (Stanley et al. 2016). Global emissions to atmosphere are estimated to be equivalent to around 40 % of the efflux from lakes and 14 % of that from wetlands (Stanley et al. 2016). The oxidation of methane by methanotrophs at anoxic-oxic interfaces provides a means by which methane-derived carbon can enter the food web of streams and rivers.

### ***Carbon sources, transport and cycling in lakes***

Lakes are important in terms of the breakdown and storage of organic matter and often represent areas of enhanced primary production due to high levels of algal and plant production and cycling of carbon (Ward et al. 2017). The traditional understanding of the carbon cycle in lakes (see Figure 2-6) is that DIC is then sequestered into organic matter by phytoplankton where it is then available for transfer throughout the food web that, in its most basic form, can be considered as phytoplankton → zooplankton → small fish → large fish, but is in reality often much more complex, varying according to the abiotic and biotic features of the lake. For example, lake morphology, sediment characteristics, pH, temperature, and nutrient, oxygen and light availability all influence lake food webs as do biotic factors such as competition and predation through their influence on the structure and dynamics of lacustrine communities (Brönmark and Hansson 2017).

In addition to uptake by phytoplankton, DIC can be incorporated into organic matter by macrophytes. The presence of macrophytes tends to be restricted to the shallow littoral zones of lakes where light penetrates to sediment depth. However, some macrophytes can root within the aphotic zone, where they grow tall to access light. Thus, most primary production in lakes occurs either in the photic pelagic zone or the littoral zone. However, in shallower clear-water lakes, benthic primary production can also occur.



**Figure 2-6.** Simple representation of the carbon cycle in a lake, with focus on the pelagic system (from Thorne et al. 2018).

The DIC utilised in photosynthesis enters lakes either from atmospheric exchange of carbon dioxide across the air/water interface, or via water exchange (e.g. carbon dioxide or bicarbonate in river or stream water, groundwater, or rainwater flow through the surface water catchment area) (Ascough et al. 2010, Brönmark and Hansson 2017).

Once incorporated, carbon may be retained within the biomass of phytoplankton and/or macrophytes and be available to primary consumers or be cycled back to the water column in the form of respired DIC, or DOC or POC through processes such as excretion and death. This carbon may then return to the biological cycle through incorporation in bacterial biomass or direct consumption by heterotrophic biota, with respiration then returning carbon for fixation, once again, by primary producers or be lost from the system, e.g. through atmospheric exchange. Depending on the state of mixing within the water column, POC may sediment, providing a carbon source to benthic organisms. The breakdown of macrophytes can also supply a carbon source to detrital feeding biota in littoral and benthic zones.

Although substantial amounts of carbon cycle between the various biotic and abiotic carbon pools within lakes, substantial gains and losses of carbon occur across the boundaries of the lacustrine system. Inputs of carbon derive from the atmosphere, surface waters, groundwater, and erosion of lake sediments, regolith and bedrock. Losses occur to atmosphere, to outflowing streams, to groundwaters and to the deposited sediments underlying the lake. Gains and losses may both occur across the same interface, e.g. between lake waters and the atmosphere.

Lakes act as important regulators for the storage and breakdown of organic matter and are hence important elements in determination of the sizes of carbon ‘pools’ and of the fluxes of carbon between them. These are often areas of enhanced primary production and can provide rivers and streams lying downstream with fresh, autochthonous organic matter. However, much of the organic matter produced in the lacustrine environment may become buried in bottom sediments and hence its carbon content may become relatively unavailable. Such buried organic matter will often be in an anoxic environment where anaerobic mineralisation can occur, making it an important source of methane that is then released to the water column and exchanges with the overlying atmosphere. Lake sediments can accumulate significant amounts of organic matter, particularly where there are large inputs of organic carbon from terrestrial catchments and methanogenesis (the production of methane from organic matter under anoxic conditions) can be an important process in the anoxic organic-matter rich sediments of lakes. Substantial amounts of methane can be produced. For example, it is estimated that 20–59 % of POC reaching sediments is converted to methane (Ward et al. 2017). Key factors governing methane production are temperature and the availability of organic matter (DeIsonetro et al. 2016).

The diffusion of methane from the anoxic sediments in which it was produced to aerobic sediments or to the water column then provides the opportunity for it to be converted to microbial biomass by methane-oxidising bacteria (MOB) present at the interfaces between anoxic and aerobic conditions (Bastviken 2008, Ravinet et al. 2010, Jones and Grey 2011). The presence of such interfaces will largely be governed by lake mixing regimes (Lennon et al. 2006). Methane concentrations in surface waters tends to be positively related to the fraction of the lake volume that is anoxic and negatively related to the DOC concentration in the lake (Bastviken et al. 2004).

The highest densities of MOB occur at oxic-anoxic interfaces and their action can result in a large proportion of methane diffusing from anaerobic lake sediments being oxidised before reaching the air-water interface, depending on factors such as water column depth, temperature, season and stability (i.e. well-mixed vs. stratified) (DelSontro et al. 2016). For example, 51–80 % of the methane produced in the deep sediments of three lakes in Wisconsin, USA, was oxidised in the water column, but most of the methane released from shallow sediment escaped oxidation and reached the atmosphere (Bastviken et al. 2008). The emission of methane to atmosphere and ratio of methane to carbon dioxide emissions increase with seasonal increases in temperature (Yvon-Durocher et al. 2014).

Unlike methane production, which is largely controlled by temperature, methane oxidation is primarily controlled by the concentrations of methane and oxygen (Bastviken et al. 2008). Consequently, increased methane production because of rising temperatures will also lead to increased methane oxidation. Overall, it is estimated that between 30 % and 99 % of the methane produced can be oxidised (Bastviken et al. 2008). Seasonal variations in methane production are therefore observed with the highest rates of methane oxidation occurring in late summer and early autumn (Utsumi et al. 1998).

MOB can be highly productive, and the microbial biomass produced is then available to other aquatic organisms, thus supporting the aquatic food web (Jones and Grey 2011, Brönmark and Hansson 2017). The consumption of MOB is particularly evident in benthic chironomid larvae (Deines et al. 2007, 2009, Jones and Grey 2011, Sanseverino et al. 2012) with methane-derived carbon contributing up to 60 % of chironomid carbon biomass (Jones and Grey 2011). This is particularly significant because chironomid larvae often dominate the benthic fauna of lakes (Deines et al. 2007). In addition to uptake by benthic invertebrates, there is also evidence of methane-based carbon contributing to the biomass of pelagic zooplankton, although this is more likely limited to those zooplankton with vertical migratory behaviour that brings them into the vicinity of oxic-anoxic interfaces where MOB may be active (Jones and Grey 2011).

Stable carbon ratios have shown that much of the carbon in pelagic food webs has a benthic origin, with fish being a strong link between the pelagic and benthic food webs (Brönmark and Hansson 2017). For example, MOB-specific fatty acids have been measured in the tissues of a range of aquatic invertebrates and fish (Sanseverino et al. 2012) and up to 65 % of the diet of lake fish can result either directly or indirectly from the consumption of benthic animals (Brönmark and Hansson 2017). Respiration by MOB consumers will also release carbon dioxide back into the system, allowing methane-derived carbon to be utilised by primary producers, thus further supporting the cycling of methane-based carbon within the food web.

The link between methane production and uptake into consumers has been suggested as being particularly important in lakes with higher terrestrial-DOC inputs, which may favour methane production (Jones et al. 1999). This is contradicted, however, by the findings of Lennon et al. (2006), that terrestrial DOC is recycled to carbon dioxide through heterotrophic respiration within lake ecosystems, thus increasing the lake DIC pool available to aquatic primary producers. More recalcitrant terrestrial POC accumulated in lake sediments could, however, provide an important carbon source for methane production (Keaveney et al. 2015a).

For methane that is not incorporated into the food web, losses to atmosphere can occur. Ebullition is a key process that governs methane losses from lakes to atmosphere, accounting for between 30 % and 96 % of total methane emissions from lakes (Bastviken et al. 2004, DelSontro et al. 2016, Casper et al. 2000). Ebullition can be episodic and linked to air-pressure changes but can give rise to rapid emission events that bypass the oxic zone (Peltola et al. 2018). The majority of ebullition occurs in shallow waters of up to a few metres depth (Bastviken et al. 2004). Storage fluxes can also be a significant pathway for methane losses from lakes, whereby methane ‘stored’ in the hypolimnion during lake stratification is rapidly emitted with water column mixing, which can account for up to

45 % of methane emissions from small lakes, reducing with lake size (Bastviken et al. 2004). Certain plant species can also act as vectors for the transfer of methane from sediments to atmosphere via aerenchyma.

In addition to the carbon sources outlined above, the terrestrial environment can also be an important carbon source for lakes, subsidising the carbon demands of aquatic food webs. It is also possible for terrestrially derived carbon to exceed the carbon demands for primary production in water bodies such that the system becomes net-heterotrophic, with respiration exceeding photosynthesis such that the water body becomes a net producer of carbon dioxide that is then transported out of the system, either through gas exchange with the atmosphere or via water exchange.

Catchments play an important role in influencing the chemical composition of freshwater bodies as rainwater interacts with soils and minerals during its transit through the catchment area. This includes the leaching of humic substances that can then enter lakes and rivers, providing a potential allochthonous carbon source for aquatic organisms (Brönmark and Hansson 2017). Terrestrial leaves and other debris can also provide a source of carbon to aquatic systems that can be metabolised by bacteria.

The carbon entering aquatic systems from the surrounding terrestrial environment may derive from recent photosynthesis, or have been stored in soils (Keaveney et al. 2015a). Climate has been found to be an important factor influencing the transfer of terrestrial carbon to aquatic systems, with warmer climate and higher precipitation resulting in greater DOC mobility through catchments (Vuorenmaa et al. 2006, Keaveney et al. 2015a). Catchment size is also an important factor (Vuorenmaa et al. 2006).

Jones et al. (1999) suggest that the predominant form of terrestrial carbon entering lakes is DOC. Weidel et al. (2008), however, suggests that both DOC and POC are important forms of carbon entering aquatic systems from the surrounding terrestrial environment. This is supported by the findings of Jones et al. (1999) that terrestrial DOC and POC dominated carbon pools of 12 small forest lakes in Finland. Wilkinson et al. (2013) also found that terrestrial DOC and POC were important to the overall aquatic carbon budgets throughout 39 temperate lakes in northern USA; the average contribution of terrestrial POC to the overall POC pool throughout the lakes was approximately 55 %, whereas the DOC pool was strongly dominated by terrestrially derived material with the mean over the same set of lakes being close to 100 %. Only the most eutrophic of the lakes were not dominated by terrestrially derived DOC (Wilkinson et al. 2013). The terrestrial POC contribution is consistent with that reported by Pace et al. (2004), where 40–55 % of POC was attributed to terrestrially derived carbon. Similar results were observed for Loch Ness in Scotland, with the pelagic food web being highly dependent on detrital carbon arising from terrestrial vegetation in the catchment (Jones et al. 1998). The proportional contribution of terrestrial carbon to aquatic food webs is, however, likely to vary seasonally, with maximum contributions coinciding with periods of lower primary productivity. This is supported by the findings of Keaveney et al. (2015a, 2015b) that calanoid copepods switch between autochthonous carbon dietary sources in summer when bio-energetically-rich algal blooms are present and terrestrially derived carbon sources in winter when phytoplankton are fewer.

Terrestrially derived carbon, both DOC and POC, can act as substrates for bacterial production in aquatic systems, with subsequent consumption of bacteria by zooplankton and other aquatic organisms providing an alternative route into the aquatic food web from that of uptake via primary producers (Keaveney et al. 2015a, 2015b). In addition to incorporation of this terrestrially derived carbon into bacterial biomass, respiration of bacteria results in the release of DIC to the water column, that can then be utilised by phytoplankton (Jones et al. 1999). Trophic transfer of this terrestrially derived carbon then occurs via consumption by heterotrophs of phytoplankton and bacterial biomass, or through direct grazing on terrestrial POC. The role of bacteria in the carbon cycle of lakes can be particularly pronounced in the summer when algal blooms result in the release of carbon by senescent algae. Up to 50 % of carbon fixed by phytoplankton can be exuded and used by bacteria (Brönmark and Hansson 2017).

In terms of DOC, research suggests that algal DOC is, on the whole, more labile and energetically rich than terrestrially derived DOC and is, therefore, utilised more rapidly (Guillemette et al. 2013, Keaveney et al. 2015b). Terrestrial DOC utilisation is, however, more variable with labile and recalcitrant forms being present that are utilised over short and long timeframes, respectively, with the more labile terrestrial DOC being utilised in parallel with algal DOC (Guillemette et al. 2013,

Keaveney et al. 2015a). Lennon et al. (2006) suggest that of the terrestrially derived DOC entering lakes, up to 90 % may be biologically recalcitrant with the remainder being consumed primarily by aquatic bacteria, that then respire between 35 % and 99 % of the DOC as CO<sub>2</sub>. This would enable the labile terrestrial DOC to be available to aquatic primary producers, with that taken up into bacterial biomass being available to consumers. Keaveney et al. (2015a) suggest that the recalcitrant terrestrial carbon is more likely to enter lake sediments. This is consistent with the findings of Caraco et al. (2010) that, whilst both old (recalcitrant) and new (labile) terrestrial carbon were incorporated within aquatic food webs, the proportionate uptake into zooplankton biomass was lower for the older, recalcitrant carbon. With algal DOC being more energetically rich than the majority of terrestrial DOC, eutrophic lakes, with high algal productivity, are more likely to have reduced terrestrial carbon accumulated within the food web than oligotrophic lakes.

Carbon inputs from the terrestrial system therefore enable the abundance of consumer in aquatic food webs to increase beyond that which could be supported by aquatic productivity alone (Brönmark and Hansson 2017). However, the reverse is also the case where aquatic systems contribute to terrestrial carbon budgets. For example, freshwater bodies provide food and drinking water for terrestrial birds and mammals and many terrestrial insect species also have aquatic life stages such that juvenile growth is supported by aquatic carbon that is then transferred to the terrestrial system with the adult form.

## **2.7 Transport and accumulation of carbon in estuarine and marine systems**

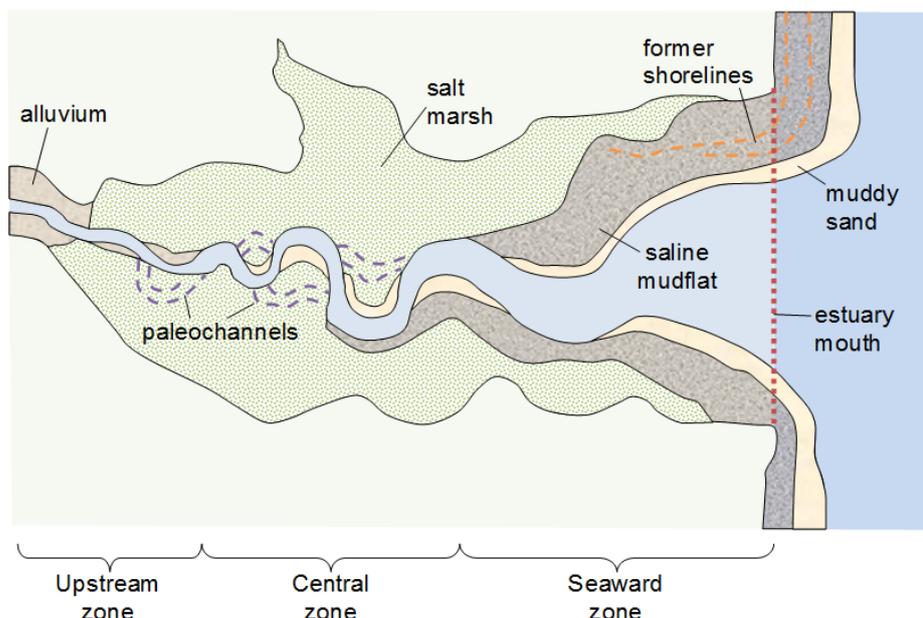
### **2.7.1 General characteristics, processes and carbon cycling in estuaries**

Estuaries represent transitional zones between rivers and oceans and are highly dynamic ecosystems with salinity gradients from fully freshwater to fully saline where water flow is influenced diurnally by tides (Ward et al. 2017). They are often associated with strong currents and turbid waters. The combination of river flow and tidal forces lead to considerable variation in water flow dynamics and sediment scour, resulting in high suspended sediment loads (Little 2009). During tidal flood periods, sediments can be transported upstream such that sediments can be traps, not only for fluvial sediments and, but also for oceanic sediments and organic matter, with sediments being realigned with each tidal cycle (Little 2009).

Estuaries are highly diverse, ranging from river-dominated systems, such as rivers flowing to the Baltic Sea where there is little tidal influence, to tidally dominated systems, such as the Severn Estuary in the UK where tidal ingress extends several kilometres inland (Little 2009). The mixing regimes for freshwater and marine water within estuaries is also diverse, ranging from salt-wedge systems that are highly stratified with less dense freshwater floating above seawater, through partially mixed/moderately stratified where tidal influences are larger giving rise to greater turbulent mixing of fresh and sea water to fully mixed, non-stratified systems where tidal forces dominate (Little 2009). The tidal regime not only affects stratification in the water column, but also regulates, to a large extent, the residence time of freshwater (Borges and Avril 2011).

In contrast to lakes where turbulence tends to be wind generated and streams and rivers where water depth and flow drive turbulence, turbulence in estuaries is often driven by both wind and boundary friction resulting from tidal currents (Abril et al. 2009). Where wind and currents work in opposite directions, turbulence can be increased.

Compared with coastal environments, estuaries tend to have high habitat diversity (e.g. regions of mud, sand, gravel, rock and saltmarsh) that all interact. As a result, both biodiversity and productivity can be high (Little 2009, Ward et al. 2017). Many estuaries are associated with tidal wetlands or saltmarshes that can be highly productive and provide DOC and POC to estuarine and coastal systems (Bauer et al. 2013).



**Figure 2-7.** Components of the estuarine environment (from Walke et al. 2013, reproduced with permission of the Nuclear Decommissioning Authority).

The cycling of carbon within the estuary is strongly influenced by the physical characteristics of the estuary, particularly the stratification regime and residence time of water that are themselves regulated to a large extent by the tidal regime (Borges and Abril 2011). Large amounts of allochthonous carbon enters estuaries from rivers and most estuaries are net heterotrophic systems (i.e. they are supported by external sources of carbon) (Chen and Borges 2009, Borges and Abril 2011). The fact that many estuaries are net heterotrophic results from large quantities of labile organic matter being transported by rivers, combined with high turbidity that minimises light penetration (Borges and Abril 2011).

DOC and POC in estuaries can be derived from terrestrial, marine and/or estuarine primary production with different sources having unique reactivity and residence times that affect their degradation within estuaries and export to coastal waters. Significant losses of DOC and POC can occur within estuaries because of microbial degradation, photochemical oxidation, scavenging, sedimentation, and salinity induced flocculation (Bauer et al. 2013). Large amounts of carbon can also be exported from estuaries; for example, Bauer et al. (2013) estimate that estuaries export about 10 % more carbon to continental shelves than they import from rivers, thus augmenting the supply of DIC and organic carbon to coastal systems.

The processing of carbon both in sediments and water within estuaries produces both carbon dioxide and methane, depending on the mixing regime (Borges and Abril 2011). For example, in well-mixed systems both methane and carbon dioxide can be produced in response to the large amount of allochthonous carbon entering the system via rivers, but in stratified systems, anoxic conditions are more likely to arise in bottom layers, promoting methanogenesis in sediments and the diffusion of methane back to surface waters, some of which will be oxidised to carbon dioxide (Borges and Abril, 2011). On average,  $32 \text{ mol C m}^{-2} \text{ y}^{-1}$  can be released from estuaries in the form of carbon dioxide (Chen and Borges 2009). The carbon dioxide and methane are either reutilised within the estuary, lost to atmosphere or exported to the coastal environment.

The flux of methane to atmosphere depends on factors such as methane production and oxidation rates and gas transport that are themselves affected by depth and mixing within the water column and temperature with factors such as the presence of vegetation and turbidity leading to temporal and spatial variability (Borges and Abril 2011). In general, methane fluxes tend to decrease with distance toward the sea, which inversely relates to a general increase in sulphate concentrations from the riverine to seaward ends of estuaries and reduction in labile organic carbon concentrations.

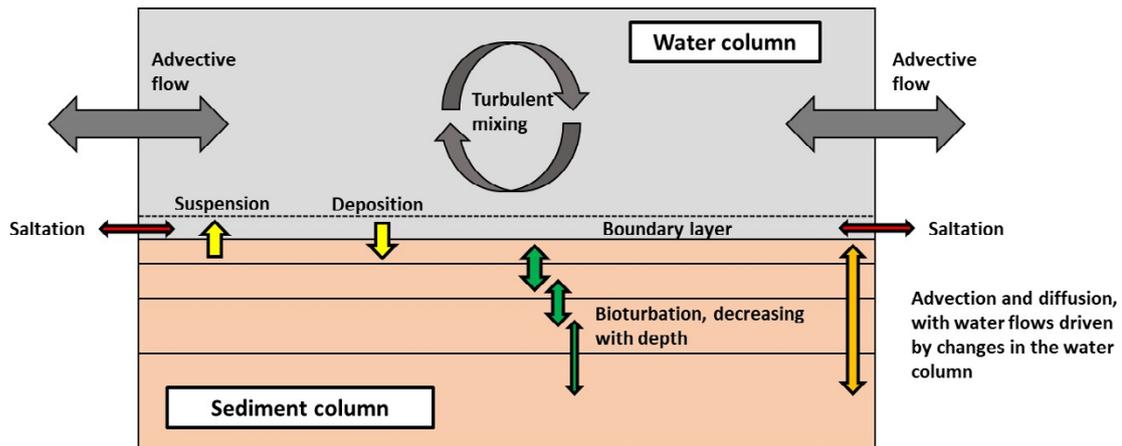
Within the upper estuary, as concentrations of methane increase in sediments because of methanogenesis, gas bubbles can form that transfer methane directly to the atmosphere (ebullition). Ebullition tends to occur where gas bubbles form in the upper 10–20 cm of sediment and occurs primarily during low tide, due to high tides stabilising hydrostatic pressure (Borges and Abril 2011). Methane can also diffuse through sediments, although the majority ( $\geq 90\%$ ) tends to be oxidised at the sediment surface. Gas transport through plants in marsh areas can also promote release from sediments to atmosphere. Where plants are present, plant-mediated transport of methane to atmosphere tends to dominate whereas ebullition dominates in the absence of plants (Borges and Abril 2011). However, plant roots can also increase oxygen concentrations in sediments, leading to an increase in methane oxidation by MOB. Tidal pumping can also occur, leading to a lateral flux of methane (and DIC) from pore waters of intertidal sediments, particularly in marsh areas, to creeks and channels during the tidal ebb. Methane loss from sediments because of tidal pumping can be 2–4 times greater than diffusive losses to atmosphere during low tides (Kelley et al. 1995). Toward the seaward end of an estuary, rates of methanogenesis tend to be lower and water depth and mixing are greater such that oxidation rates increase and methane fluxes to atmosphere are reduced due to oxygenation. Overall methane oxidation rates in estuaries range from around  $8 \times 10^{-5}$  to  $0.7 \text{ d}^{-1}$  although rates are affected by factors such as salinity and wind speeds, which affects the residence time of methane in water (Borges and Abril 2011).

The partial pressure of carbon dioxide ( $p\text{CO}_2$ ) in estuaries is highly variable both spatially and seasonally, reflecting the variability of organic carbon production and degradation; the extent of variation being considerably greater than in other coastal environments (Borges and Abril 2011). For example, within estuaries, DOC tends to decrease linearly with increasing salinity and autochthonous POC from the marine coastal environment can have a seasonal influence on the carbon budget of estuaries, being most notable in spring and summer (Abril et al. 2002). Factors influencing carbon dioxide dynamics in estuaries include water exchange and the carbon dioxide content of exchanged water (riverine water tending to have high  $p\text{CO}_2$  values), water residence time, and stratification that can lead to decoupling between organic carbon production and degradation across the water column (Borges et al. 2006 in Borges and Abril 2011). Overall, there tends to be a positive relationship between  $p\text{CO}_2$  values in estuaries and DOC concentrations, arising from either largely terrestrially derived riverine inputs or lateral inputs from terrestrial soils due to groundwater flow or surface water runoff. Other important factors include biogeochemical processes such as calcium carbonate precipitation and dissolution and oxygen utilisation through processes such as nitrification (Borges and Abril 2011).

### **2.7.2 Considerations for modelling C-14 in estuarine systems**

Estuaries are typically developed in soft sediment and the spatial distribution of that sediment changes rapidly with time, affecting the patterns of water flow, which themselves vary diurnally with the tidal cycle and on longer timescales. Therefore, realistic assessment modelling C-14 transport through an estuary implies the need to simulate sediment dynamics, water flows and the transport of DIC, DOC, PIC and POC in an integrated way. In the context of modelling of C-14 transport in the environment, detailed modelling of estuaries has been as a neglected topic.

A convenient simplification for compartmental modelling distinguishes an estuary into upper, middle and lower components. The upper estuary is of low salinity and is strongly conditioned by unidirectional input from the river. In contrast, the lower estuary is of high salinity and is dominated by the influx and efflux of tidal waters. The middle estuary comprises a mixing zone dominated by river flows at low tide and tidal flows at around high tide. Typically, estuaries developed in soft sediments are funnel shaped, implying that the largest exchanges of water are in the lower estuary, where the area of the estuary is largest, and the tidal range is greatest. The sediments underlying the estuary can often be simplified and represented as a stratified column. As illustrated in Figure 2-8, transport in this column can be due to advection or diffusion in the pore water but can also be due to bioturbation due to sediment-inhabiting organisms. Transfers between the upper layer of sediment and the water column are due to suspension and deposition of particles, but exchanges between materials sorbed to solids and present in solution mean that transfers occur both in the solid and liquid phases. In the water column, transport is largely due to advection in the water flows, with this advective transport giving rise to turbulent mixing in the different areas of the estuary. Particles can also be transported in the boundary layer at the bottom of the water column by the process of saltation in which the particle moves in a discontinuous manner with suspension followed by short-distance transport and deposition, with the process repeating as flows in the boundary layer change. A quantitative compartmental model for radionuclide transport in the estuarine environment is described by Walke et al. (2013).



**Figure 2-8.** Transport processes in an estuarine environment.

Estuaries are characterised by dynamic environmental conditions expressed along evolving ecotones that provide an abundance of unique ecosystem niches driving high levels of biodiversity and productivity and provide a range of ecosystem services including nutrient/pollutant cycling and sediment and carbon storage as well as fisheries production (Ward et al. 2017).

### 2.7.3 The marine environment

Coastal and near-shore systems occupy around 7 % of the seafloor and less than 0.5 % of the ocean volume yet play a significant role in the biogeochemical cycling of carbon (Bauer et al. 2013, Chen and Borges 2009). For example, it is estimated that coastal systems contribute between 10 and 30 % of the overall primary production in oceans (Bauer et al. 2013, Ward et al. 2017), 30–50 % of inorganic carbon and 80 % of organic carbon burial in sediments and could contribute up to about 50 % of the organic carbon supplied to the deep open ocean (Bauer et al. 2013).

Coastal waters are dynamic open systems where there is continuous exchange of carbon across the various interfaces with the land, open sea, and atmosphere, as well as between seawater and sediments (Ducklow and McCallister 2004). Coastal systems are geochemically and biologically active areas with high rates of exchange of matter and energy with the open ocean (Gattuso et al. 1998). The considerable inputs of carbon to coastal and nearshore systems from current upwelling and riverine inputs supports high biological activity as compared with wider oceans (Chen and Borges 2009).

Many processes in coastal systems are like those in lakes, such as the transformation of DIC to DOC and POC through primary production and subsequent reverse transformation to DIC by respiration and the potential for thermal stratification to occur. However, some important aspects of the carbon cycle differ. For example, the relationship between dissolved gases and the atmosphere differs along with the role of organisms in carbonate mineral formation (Myrbo 2012). For example, some organisms precipitate internal or external body structures of calcium carbonate that can either dissolve in seawater or become incorporated in sediments (Fennel et al. 2019). The carbonate buffering system in seawater leads to carbon dioxide reacting with seawater to form carbonic acid which then dissociates into bicarbonate and carbonate ions, resulting in over 95 % of DIC in seawater being in the form of bicarbonate (Ducklow and McCallister 2004).

Carbon sources and sinks in coastal waters are highly complex (Ward et al. 2017). Sources include diffusion across the air-water interface, terrestrial carbon inputs via rivers and estuaries, as well as more diffuse terrestrial inputs. In some coastal waters, carbon dynamics are driven by ocean circulation but in others they are controlled by riverine inputs (Bauer et al. 2013). Carbon fluxes from rivers are generally well correlated with river discharge but can vary, for example, in regions where high erosion rates in watersheds occur (Bauer et al. 2013). As such, regions with high precipitation rates are likely to see greater inputs of DOC and POC to coastal waters than for drier regions.

The high inputs of DOC and POC to coastal waters from estuaries and subsequent cycling can result in coastal seas close to large estuaries acting as sources of atmospheric carbon dioxide (Chen and Borges 2009). However, the extent to which organic matter inputs contribute to the local carbon balance depends on the biochemical lability of the imported carbon and its residence time in coastal waters (Ducklow and McCallister 2004) and there is growing consensus that much of the terrestrial DOC entering coastal waters becomes chemically altered rather than being oxidised to carbon dioxide (Bauer et al. 2013). Some estuaries do however provide high concentrations of DIC directly to coastal waters, but the ability for this to be incorporated into organic matter through primary production can be limited because of high particulate loads that limit light penetration through the water column (Bauer et al. 2013). High-energy beaches and low-energy tidal flats can also be important in remineralising organic matter deposited during tidal inundation that can then re-enter the coastal waters via transport in rainwater or during the next tidal inundation (Brooks Avery et al. 2012).

Overall, there is a positive metabolic balance in continental shelf when considered on a global scale such that produce slightly more carbon than they respire, with primary productivity being dominated by phytoplankton in many instances (Gattuso et al. 1998, Ducklow and McCallister 2004) although in shallow coastal shelf zones where riverine input is low, light penetration may be sufficient to support significant benthic primary production (Bauer et al. 2013). Whilst globally, coastal regions can be considered carbon dioxide sinks, inner waters close to land tend to be sources of carbon dioxide to atmosphere because of metabolism of the high inputs of terrestrial and estuarine organic carbon and high particulate loads that can limit primary productivity (Bauer et al. 2013). With distance offshore, particulate loads tend to reduce, such that primary productivity is no longer light limited and can be stimulated by the upwelling of nutrients such that waters become a relatively long-term sink for atmospheric carbon dioxide (Walsh 1991).

Zooplankton play an important role in the marine foodchain, grazing around 75 % of primary production daily (Schmoker et al. 2013). Metabolism of phytoplankton by zooplankton then returns carbon back to the water as carbon dioxide or as DOC/POC.

Within the water column, POC can settle out and become incorporated into sediments where it can be metabolised to carbon dioxide or methane, depending upon sediment redox conditions, and released back to the water column, or buried and stored for extended periods (Fennel et al. 2019). Coastal sediments can store large amounts of organic matter, with marine sediments typically having 0.5–5 % organic carbon by weight (Ward et al. 2017). Typically, methane is produced beneath the zone of sulphate reduction (Capone and Kiene 1988). As such, methane concentrations are typically low in the upper layers of sediments and increase with depth to the point where sulphate concentrations reach zero (Borges and Abril 2011).

## 2.8 Conclusions from the review

The biogeochemical carbon cycle has been extensively described at a wide range of spatial and temporal scales. The global carbon cycle has been extensively investigated in the context of modelling climate change at timescales ranging from decades to hundreds of thousands of years. The main carbon ‘pools’ and fluxes between them have been quantified, both in respect of the natural cycle and in terms of the perturbations to that cycle arising from anthropogenic emissions. Key aspects include atmospheric mixing, atmosphere-ocean exchanges, oceanic mixing, calcium carbonate precipitation and dissolution, and continental weathering. This global understanding provides a context in which more local biogeochemical cycling can be embedded and understood.

In respect of source terms of C-14 relevant to the disposal of solid radioactive wastes, a review of the various materials likely to be disposed and releases from them indicates that C-14 bearing carbon dioxide and methane, either in gaseous form or dissolved in groundwater, are likely to be of greatest importance, though C-14 bearing small organic molecules may also require consideration in some contexts. Transport to the biosphere can be in solution or carried by bulk gas, likely to be hydrogen, methane or a mixture of the two.

When C-14 enters the biosphere, it will again be transported in solution and in the gas phase. However, uptake in biota will also be important. A key issue is the extent to which the C-14 in carbon that is released to the biosphere is diluted by mixing with other carbon before exposure of humans and/or relevant populations of non-human biota. Reviews of carbon behaviour in terrestrial and aquatic ecosystems emphasise the importance of considering dissolved and particulate inorganic and organic carbon (DIC, PIC, DOC and POC) and conversions between them. Photosynthesis, respiration, organic matter decomposition and mineralisation are all of significance, and the review has emphasised the importance of understanding microbially mediated reduction and oxidation in all the environments considered, as well as the wide diversity of physical, chemical and biological processes that influence the sizes of carbon pools and the fluxes between them. The literature review does not propose models for C-14 transport in these environments. However, it demonstrates that there is a deep understanding of the various processes of relevance and that there exist extensive datasets that can be used in model formulation and parameterisation.

Although the review presented in the previous subsections includes an extensive reference list, it is emphasised that this comprises only a small fraction of the information available. There are many more secondary (review) references available and these each reference an immense diversity of primary literature. Rather than attempting to review the primary literature, the emphasis has been on providing a high-level explanation of the key processes operating in the various environments considered. This provides the basis for developing broadly agreed conceptual models for the behaviour of carbon and C-14 in these environments. In turn, these conceptual models can be simplified and adapted to make them appropriate to specific situations of interest. From this point on, the primary literature can be referenced in translating a conceptual model appropriate to a specific context into a mathematical model and in providing parameter values or distributions for use with this mathematical model. These steps are addressed in the remainder of this report, with reference to specific test cases of interest.



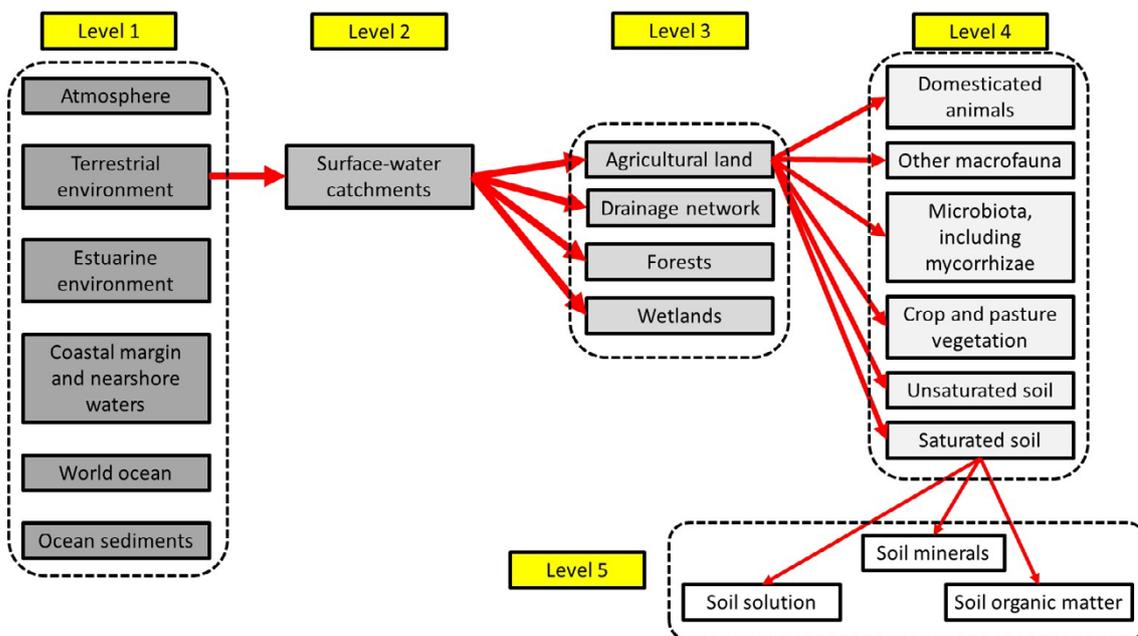
### 3 Conceptual models for C-14 and carbon in the biosphere

#### 3.1 Introduction

The review presented in earlier chapters of this report gives an overview of the biogeochemical carbon cycle. This considers carbon transport in specific environments that could be of relevance in radiological assessment studies. These environments comprise agricultural systems, forests and wetlands, estuaries, coastal regimes and the marine environment. This chapter uses the review material to develop conceptual models of C-14 transport within and between these various environments. The approach adopted to developing these conceptual models is described below. Later subsections then describe the conceptual models developed for the global biogeochemical cycle and for each of the embedded environments, including the links between them.

#### 3.2 Objectives and approach to conceptualisation

The biogeochemical carbon cycle operates at various spatial and temporal scales. Although it is a closed, mass-conserving cycle at the global scale, at regional and local scales the import and export of carbon across the boundaries of the identified domain can be of great significance. Thus, for example, carbon in freshwater systems cannot be discussed in isolation from a consideration of carbon inputs from terrestrial catchments and outputs to the marine environment. Therefore, a nested approach has been taken to developing conceptual models, with the largest spatial and temporal scales considered first to define the overall global cycle within which the conceptual models for specific environments are embedded. In Section 3.3, conceptualisation of the overall global cycle is developed. This recognises terrestrial, estuarine, coastal and deep ocean environments as distinct, and detailed conceptual models for these environments, or components of them, are described in subsequent sections. The overall approach is illustrated in Figure 3-1.



**Figure 3-1.** System disaggregation scheme. The arrows indicate the direction of disaggregation, with Level 1 components disaggregated into Level 2 components etc. The choice of components at each level is not unique, e.g. the world ocean and ocean sediments could be combined at Level 1 and disaggregated at Level 2. Only a few examples of disaggregation are shown here. Comprehensive decompositions of the various high-level components of the environment relevant to C-14 and carbon are provided in the later parts of this chapter.

The disaggregation scheme adopted uses multiple levels. In Figure 3-1, the top five levels are illustrated. However, a greater degree of disaggregation can be adopted, as appropriate. It will be seen that disaggregation can have a spatial basis (Level 2), an ecosystem basis (Level 3) or a functional basis (Level 4). The type of aggregation adopted can differ for the same level depending upon the route to that level. For example, Level 2 disaggregation under ‘world ocean’ does not have to be spatial just because a spatial disaggregation has been adopted for the terrestrial environment.

Disaggregation, as illustrated in Figure 3-1, provides an approach to ensuring that the features of the environment of interest are comprehensively identified. Furthermore, development of the scheme to lower levels permits the user to achieve a level of disaggregation of sufficient detail to support development of a mathematical model or models that can be readily related to the conceptual structure of the system. However, it remains with the user to judge when a sufficient degree of disaggregation has been achieved.

The disaggregation process outlined above addresses only features of the system under consideration. To develop an assessment model, it is necessary to augment this information with details of the processes determining the flows of carbon and C-14 between different features of the system. This can most readily be done using influence diagrams or interaction matrices. An incomplete, high-level influence diagram is illustrated in Figure 3-2.

In Figure 3-2, an agricultural ecosystem is decomposed into a mixture of spatially defined and functional components. This mixture arises because the selected features of the system are drawn from different levels of decomposition. Thus, ‘atmosphere’ is a Level 1 component, whereas ‘saturated soil’ and ‘unsaturated soil’ are Level 4. These could be further decomposed to Level 5, if it was thought desirable to distinguish soil solution, soil minerals and soil organic matter for modelling purposes. Having defined these features, they can be linked by processes that move carbon and C-14 between them. Note that ‘influence’ is used in the restricted sense of a process that transports carbon. Factors that affect rates of transport are considered in mathematical modelling and are treated as variables. Values of these variables are required for the mathematical model and are used there in analytical expressions to estimate rates of transport processes. In turn, these rates are used to model time-dependent sizes of ‘pools’ of carbon and C-14 in the compartments of the mathematical model. Feedbacks can occur with the sizes of carbon ‘pools’ influencing the values of variables and hence rates of transport. Some illustrative, significant carbon transport processes are included in Figure 3-2.

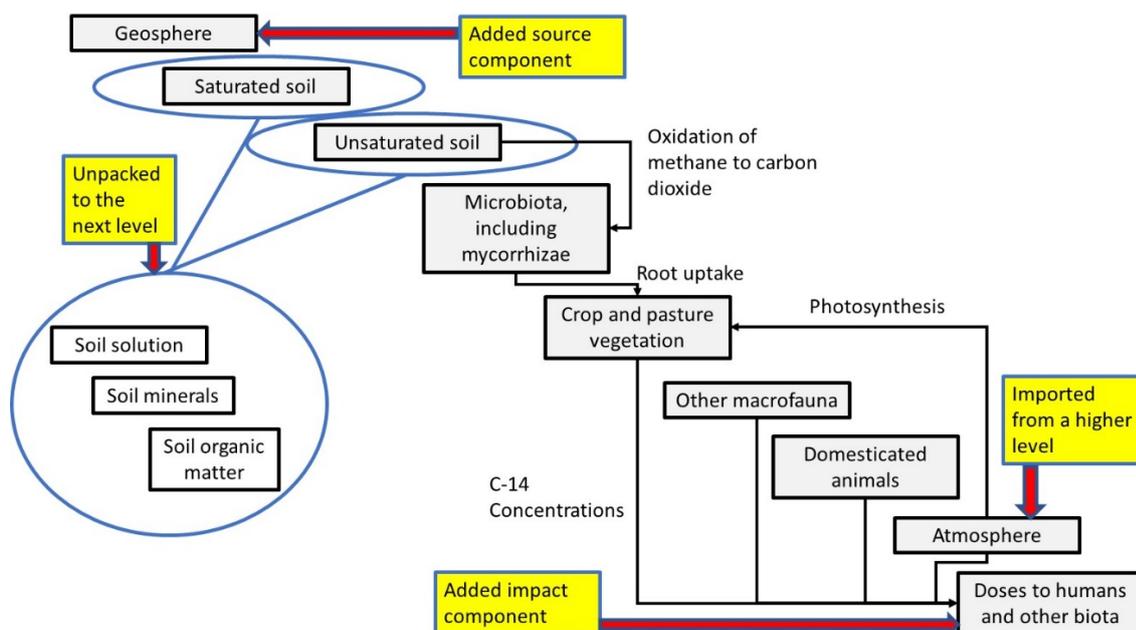


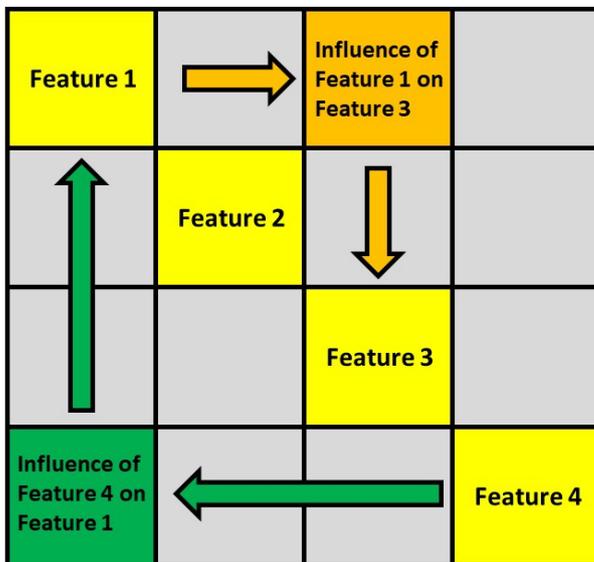
Figure 3-2. Illustrative, incomplete, high-level influence diagram.

Figure 3-2 is extended by adding a source of carbon or C-14. The characteristics of this source determine the degree of disaggregation employed and the way in which the features of the system and the interactions between them should be interpreted. Similar control is exerted by the added impact component that determines how the results from the model are to be interpreted in terms of adverse impacts on human health and the environment.

Although influence diagrams are useful in illustrating features of the system and how they are related by processes and events, they do not provide a well-structured tool for ensuring that a comprehensive identification of those events and processes has been performed. For this purpose, interaction matrices are preferred. In an interaction matrix, the features of the system are listed on the lead diagonal and processes directly relating those features are listed in the off-diagonal elements, with the matrix being read in clockwise fashion, as illustrated in Figure 3-3. Note that in the present context, 'influence' should be interpreted as meaning a process that results in a flux of carbon and/or C-14.

Interaction matrices for C-14 have been developed in previous studies (see Figure 3-4), providing guidance on processes and events that should be addressed in developing the interaction matrices provided in this report.

It is important to recognise that interaction matrices should show only the direct interactions between components on the principal diagonal. Thus, for example, where foliage, litter layer and microbiota are represented on the principal diagonal, foliage provides a direct input to the litter layer by leaf fall and the litter layer provides a direct input to microbiota through decomposition processes. However, there is no direct connection between the foliage and the microbiota in the litter layer. Although this example is straightforward, there are examples where careful thought is required to decide whether a connection is direct or indirect. Also, sometimes simplifications have been made. For example, in the above, the transfer could have been written as foliage to atmosphere to litter layer to microbiota. However, this level of spatial discrimination is not expected to be necessary for the types of radiological assessment under consideration and inclusion would be unnecessarily burdensome. However, where simplifications have been made and are not self-evident, comments have been included in the various tables. Provision of such comments is appropriate for transparency of decisions and for auditing purposes.



*Figure 3-3. Structure of a generalised interaction matrix.*



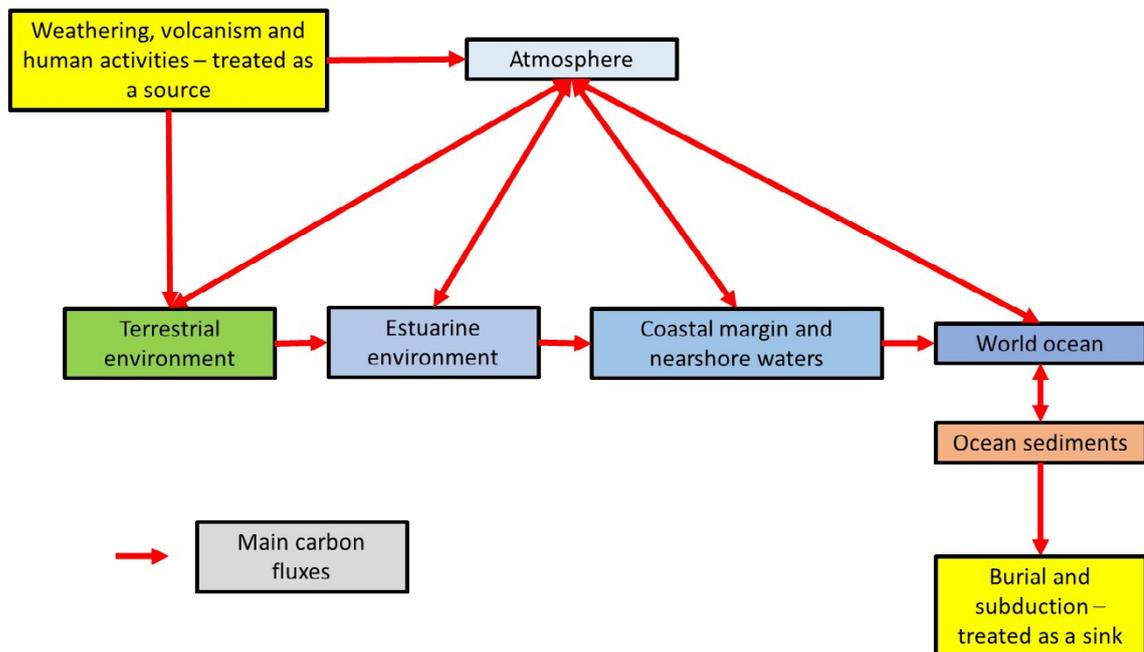
### 3.3 Conceptual model of the overall global biogeochemical carbon cycle

At the coarsest level, the global biogeochemical carbon cycle can be distinguished into four pools. These comprise carbon in the biosphere, organic carbon present in sediments of the geosphere that can be recycled to the biosphere or may be subducted into the mantle, which comprises the third major pool. The fourth is carbonate minerals derived from carbon released by rock weathering and mainly precipitated and deposited in the deep ocean. These carbonate minerals, like organic carbon may be subducted into the mantle. The carbon cycle is closed by volcanic transfers of carbon from the mantle to the biosphere. This overall cycle has been summarised by Wallmann and Aloisi (2012), as illustrated in Figure 2-1.

On the timescales of relevance in this study, the recycling of carbon from the mantle can be neglected. However, anthropogenic inputs of carbon into the surface environment cannot be neglected as this would result in an overall imbalance of the biogeochemical carbon cycle. That cycle has been characterised quantitatively by Houghton (2005), as illustrated in Figure 2-2.

Whereas a representation in this form is appropriate to support modelling of changes in atmospheric carbon dioxide concentrations over various timescales and for different patterns of anthropological emissions, it is less appropriate to modelling the radiological impacts of releases of C-14. This is because it has a focus on functional ‘pools’ of carbon and the fluxes between them, recognising that this is what determines long-term changes in the global atmospheric concentration of carbon dioxide. This, in turn, reflects the consideration that the atmosphere can be treated as a single well-mixed carbon pool on the timescales of interest.

In contrast, for modelling radiological impacts of releases of C-14, situations of interest typically involve a localised release that results in decreasing C-14 concentrations in environmental media with distance from the area of release. Therefore, it is appropriate to adopt a representation that explicitly includes spatial distinctions. At the highest level, these distinctions can be between the broad types of environmental system characterised in the review included in Chapter 2. This leads to a high-level representation of the biosphere as shown in Figure 3-5.



**Figure 3-5.** Level 1 structural elements of the biosphere reproduced from Figure 3-1 showing the main flows of carbon (note that wetlands and the terrestrial drainage network are included within the overall terrestrial environment, as shown in Figure 3-1).

Long-term carbon sources to the biosphere arise from the weathering of rocks and from volcanic events. However, since the industrial revolution, there have been increasing inputs from human activities, notably the use of fossil fuels. These inputs are mainly either direct to atmosphere or to the terrestrial environment. Thereafter, carbon is mainly transported from the terrestrial environment through the surface drainage network to the marine environment. This transport includes dissolved inorganic carbon (DIC), dissolved organic carbon (DOC), particulate inorganic carbon (PIC) and particulate organic carbon (POC). In the ocean, carbon becomes incorporated in living organisms and settlement of their remains results in deposition in ocean sediments and burial of those sediments in combination with plate tectonic movements eventually results in carbon recycling via the upper mantle. Although these carbon fluxes are shown as unidirectional, this relates only to the net or predominant direction of transport. Reverse fluxes can, and do, occur, e.g. dissolution of carbonates present in ocean sediments. Throughout transport from the terrestrial environment to the world ocean, extensive bi-directional exchanges between the surface and atmosphere occur. These exchanges can be at open-water surfaces, associated with photosynthesis and respiration in macroflora and macrofauna, or because of microbiological and chemical processes in soils, subsoils and sediments. It is these exchanges that would result in a gradual decline in atmospheric carbon dioxide concentrations if anthropogenic carbon dioxide emissions were eliminated or much reduced.

In subsequent subsections of this report, the various environments shown in Figure 3-5 are distinguished into more detailed components. It is at this more detailed level that it is appropriate to define both the features of each environment and the events and processes that transport carbon between those features, into the environment from other environments and sources external to the system, or out of the environment to other environments or sinks.

### **3.4 Conceptual models of the transport and accumulation of carbon in agricultural ecosystems**

Agricultural ecosystems are connected to the underlying geosphere, the above-canopy atmosphere and the surface-drainage network. These are all considered as sources and/or sinks of C-14 in relation to the agricultural environment. Where a surface-water catchment includes forests and/or wetlands, these can also comprise sources and/or sinks, both by the surface and subsurface transport of dissolved and particulate carbon and by the conversion of one type of system to another, e.g. through the drainage of wetland to produce agricultural land. In this section, consideration is first given to the internal structure of a conceptual model for agricultural land because this provides a context for identifying linkages with these various other types of environments.

Although agricultural land can become waterlogged to the surface (and may be deliberately flooded in some contexts, e.g. in rice cultivation), its usual condition comprises an unsaturated soil layer overlying a saturated layer. There exists a time-varying boundary zone between the two often described as the capillary fringe. In general, the degree of saturation increases downward, being lowest at the surface and approaching full saturation below the capillary fringe. However, this gradient can be reversed during and after precipitation events, as a pulse of meteoric water infiltrates into the system. Where topographic gradients are present, the degree of saturation may be influenced by lateral flows of water through the surface layers of the soil. i.e. downslope flow, and these lateral flows may discharge to the surface-water drainage network.

The gradients in water saturation in the soil are associated also with geochemical gradients. In this context, it is useful to distinguish between a generally oxic surface layer mapping onto the unsaturated soil and upper part of the capillary fringe and an anoxic zone in the lower part of the capillary fringe and the saturated zone. However, in practice, the situation can be more complex with anoxic micro-environments in the oxic zone. Also, the dynamics of precipitation events can be too rapid for equilibrium to prevail, resulting in transient conditions where the variation in redox potential with depth does not exhibit a simple relationship to the variation of soil saturation with depth.

In describing the variations in saturation and redox potential with depth, no mention has yet been made of the influence of soil composition and soil texture. In relation to water flows and degree of saturation, the relationship between hydraulic conductivity and degree of saturation is significant, whereas in terms of carbon storage and turnover, the presence of organically rich layers may be significant.

Although many agricultural soils used for cropping are mixed to depth by ploughing on an annual basis, this is not a universal practice and direct drilling into untilled land may be practiced. Furthermore, both temporary and, particularly, long-term pastures may have soils that remain undisturbed for times ranging from a few years to decades or more. Therefore, although the vertical structure of agricultural soils is often only a secondary consideration, it cannot be neglected.

The degree of saturation of the soil and the redox conditions that are present have a strong influence on microbiological processes. Specifically, methane may be produced under anoxic conditions and oxidised, via various short-lived intermediate compounds, to carbon dioxide under oxic conditions. Plant roots are typically localised mainly in the oxic, unsaturated zone, but temporal variations in soil water content may result in some parts of the root system penetrating the anoxic zone. Therefore, root uptake of carbon, although generally of limited importance in terms of the overall uptake of carbon by plants, will largely occur from the unsaturated zone, but there may be a small contribution also from the saturated zone. This root uptake can be significant for C-14 that enters the system from the base of the soil column and exhibits much higher concentrations in soil solution and the soil atmosphere than in the overlying canopy atmosphere where exchanges with plants by photosynthesis and respiration largely occur.

Conceptually, plants comprise a root system, an overlying support structure (stems and branches) and tissues specialised for the gas exchanges involved in photosynthesis (foliage). These distinctions are not absolute, e.g. for grasses the structural and photosynthetic elements are combined. The root system can have an intimate connection to a specialised mycorrhizal community, and it can be helpful to distinguish this from the general microbiological community of the soil zone. For agricultural crops and pasture, it is generally sufficient to consider the above-ground parts of the plant as comprising a single-layer canopy. However, there are some agricultural systems where an under-crop is used to complement the main crop and a more complex representation of the canopy is appropriate. However, this issue tends to be of greater significance in forest systems where it may be useful to distinguish the main tree canopy from the understorey vegetation.

The air within the canopy may be considered as a single layer. However, there are distinctions between air close to the ground surface that is less subject to turbulent mixing than air in the upper part of the canopy, which is subject to greater stresses from the wind-field present above the canopy.

Agricultural systems may include animals. These can graze pasture but can also be fed crops grown specifically for that purpose or discarded material from crops grown for human consumption. Excreta from domesticated animals and waste products arising after slaughter may be returned to agricultural systems in the form of fertilisers.

Finally, humans need to be included in the system as consumers of plant and animal products. For C-14, external exposure is of negligible significance and inhalation of C-14 gases is generally of minor importance compared with ingestion of C-14 in foods.

It should be noted that only cultivated plants and domesticated animals are included in this conceptual model. However, other organisms are present in agricultural systems (e.g. uncultivated plants, macroscopic fungi (mushrooms), and a wide variety of mammals, birds, reptiles and invertebrates). These are of potential relevance as components in minor food chain pathways to humans and in assessments of radiological impacts on non-human biota. However, in respect of these considerations, they are of greater potential significance in forest ecosystems, and they are covered under that heading.

Overall, the above discussion suggests the following features to be included in a general conceptual model of C-14 transport and distribution in an agricultural ecosystem:

- saturated, anoxic soil,
- capillary fringe of a varying degree of anoxia,
- unsaturated, oxic soil,
- soil microbial community,
- mycorrhizal associations,
- plant roots,

- plant support system,
- plant foliage,
- lower canopy atmosphere,
- upper canopy atmosphere,
- domesticated animals, and
- humans.

These components interface with the underlying geosphere, from which fluxes of carbon and C-14 both in solution and in gaseous form may be received; with the above-canopy atmosphere, primarily through fluxes of carbon dioxide and methane; and with the surface-water drainage network, primarily in solution. In the longer-term there are also exchanges with other terrestrial environments (forests and wetlands) due to conversions from one type of system to another. These longer-term changes are not addressed in the conceptual model outlined below.

Following from the above, the interactions that need to be considered in a conceptual model of terrestrial ecosystems relate to the bullet point list given above plus geosphere, above-canopy atmosphere and surface-water drainage network. These interactions are summarised below in a set of fifteen tables in standard format. Each table sets out the processes that result in mass transfers of carbon and C-14 from one component of the system to another. Self-interactions are also included (e.g. above-canopy atmosphere to above-canopy atmosphere). These are interactions that result in the redistribution of carbon or C-14 within a component. Redistribution can be a spatial mixing, e.g. turbulence in air, transport between two phases, e.g. between soil solution and soil atmosphere, or a biogeochemical transformation, such as the microbial oxidation of methane to carbon dioxide, or the formation of the products of photosynthesis from carbon dioxide. It is emphasised that these principal components and their interactions could be decomposed further. However, such further decomposition is thought to be appropriate at the stage of developing conceptual and mathematical models for a specific agricultural context rather than in the generic context discussed here. That site-specific modelling will be strongly dependent on site understanding and site modelling, including consideration of landscape evolution and other system changes (Lindborg et al. 2021, 2022).

**Table 3-1. Processes of carbon transfer from the geosphere.**

Receiving Component	Processes of Transfer
Geosphere	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas as bubbles or continuous flow.
Surface-water drainage network	Advection or diffusion in the liquid phase across the beds and banks of water bodies. Advection of gaseous forms in bulk gas across the beds and banks of water bodies.
Saturated, anoxic soil	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas as bubbles or continuous flow.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	Irrigation with well water.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system	Irrigation with well water.
Plant foliage	Irrigation with well water.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	Outgassing from irrigation water.
Domesticated animals	Consumption of drinking water abstracted from wells
Humans	Consumption of drinking water abstracted from wells

**Table 3-2. Processes of carbon transfer from the surface-water drainage network.**

Receiving Component	Processes of Transfer
Geosphere	Infiltration of water into the beds and banks of water bodies.
Surface-water drainage network	Advection and dispersion in flowing or circulating water. Incorporation and decorporation into solids. Exchange between gaseous and solution phases.
Saturated, anoxic soil	Infiltration into banks and flooding in riparian areas.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	Irrigation with abstracted surface water.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system	Irrigation with abstracted surface water.
Plant foliage	Irrigation with abstracted surface water.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	Outgassing from the surfaces of water bodies and from irrigation water.
Domesticated animals	Consumption of drinking water abstracted from surface-water bodies.
Humans	Consumption of drinking water abstracted from surface-water bodies.

**Table 3-3. Processes of carbon transfer from saturated, anoxic soil.**

Receiving Component	Processes of Transfer
Geosphere	Infiltration and downward diffusion.
Surface-water drainage network	Advection or diffusion in the liquid phase across the beds and banks of water bodies.
Saturated, anoxic soil	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas originating from the geosphere or arising from microbial processes as bubbles or continuous flow.
Capillary fringe of a varying degree of anoxia	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas.
Unsaturated, oxic soil	No direct connection.
Soil microbial community	Uptake for use in metabolism. A key process is the production of methane.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection. Plant roots are taken not to penetrate the permanently saturated soil zone in agricultural systems, though such penetration may occur in wetlands.
Plant support system	No direct connection.
Plant foliage	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Domesticated animals	No direct connection. Water abstraction is taken to be from the geosphere or from surface waters.
Humans	No direct connection. Water abstraction is taken to be from the geosphere or from surface waters.

**Table 3-4. Processes of carbon transfer from the capillary fringe.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Transport by interflow during and after precipitation events.
Saturated, anoxic soil	Advection and dispersion, including effects arising from a time-varying position of the water table.
Capillary fringe of a varying degree of anoxia	Advection and dispersion, including effects arising from a time-varying position of the water table. Exchanges between soil solution and soil atmosphere. Oxidation and reduction of carbon compounds, with the direction of reaction depending on local characteristics (oxic or anoxic). Mineralisation of organic matter originating from plant and animal material. Ploughing and other forms of soil disturbance. Bioturbation.
Unsaturated, oxic soil	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Ploughing and other forms of soil disturbance. Bioturbation.
Soil microbial community	Uptake of methane and other organic compounds for use in metabolism. Microbially mediated processes may involve both oxidation and reduction reactions.
Mycorrhizal associations	Uptake of methane and other organic compounds for use in metabolism.
Plant roots	Uptake of carbon compounds present in solution by both active and passive processes. Uptake of gaseous forms for onward transmittal via aerenchyma (though this is likely to be of little significance in agricultural systems, being mainly of relevance in saturated, wetland systems).
Plant support system	No direct connection.
Plant foliage	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Domesticated animals	No direct connection.
Humans	No direct connection.

**Table 3-5. Processes of carbon transfer from unsaturated oxic soil.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Mixing with, and transport by runoff and interflow during and after precipitation events.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Ploughing and other forms of soil disturbance. Bioturbation.
Unsaturated, oxic soil	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Oxidation of organic forms, particularly methane. Mineralisation of organic matter originating from plant and animal material. Ploughing and other forms of soil disturbance. Bioturbation.
Soil microbial community	Uptake for use in metabolic processes.
Mycorrhizal associations	Uptake for use in metabolic processes.
Plant roots	Uptake of carbon compounds present in solution by both active and passive processes.
Plant support system	No direct connection.
Plant foliage	No direct connection.
Lower canopy atmosphere	Gas exchange across the soil surface.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Domesticated animals	Adventitious soil consumption.
Humans	Adventitious soil consumption.

**Table 3-6. Processes of carbon transfer from the soil microbial community.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Capillary fringe of a varying degree of anoxia	Release of products of metabolism to soil solution and soil atmosphere. Death and decay.
Unsaturated, oxic soil	Release of products of metabolism to soil solution and soil atmosphere. Death and decay.
Soil microbial community	Exchange of products of metabolism between organisms within the microbial community.
Mycorrhizal associations	No direct connection. Both communities are microbial, but it is reasonable to treat them as only indirectly coupled.
Plant roots	No direct connection.
Plant support system	No direct connection.
Plant foliage	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Domesticated animals	No direct connection.
Humans	No direct connection.

**Table 3-7. Processes of carbon transfer from mycorrhizal associations.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	Release of products of metabolism to soil solution and soil atmosphere. Death and decay.
Unsaturated, oxic soil	Release of products of metabolism to soil solution and soil atmosphere. Death and decay.
Soil microbial community	No direct connection. Both communities are microbial, but it is reasonable to treat them as only indirectly coupled.
Mycorrhizal associations	Internal transfers between interfaces with the soil and interfaces with plant roots.
Plant roots	Active transfers of products of metabolism.
Plant support system	No direct connection.
Plant foliage	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Domesticated animals	No direct connection.
Humans	No direct connection.

**Table 3-8. Processes of carbon transfer from plant roots.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	Root exudation. Death and decay generating organic matter that is subject to mineralisation on various timescales.
Unsaturated, oxic soil	Root exudation. Death and decay generating organic matter that is subject to mineralisation on various timescales.
Soil microbial community	No direct connection.
Mycorrhizal associations	Active transfers of products of metabolism.
Plant roots	Transport in solution from locations of absorption. Transport in the gas phase through aerenchyma.
Plant support system	Export to the above-ground parts of the plant in solution and in the gas phase.
Plant foliage	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Domesticated animals	Consumption of root crops.
Humans	Consumption of root crops.

**Table 3-9. Processes of carbon transfer from the plant support system.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	Senescence, death and decay generate organic matter that is added to the upper layers of the soil.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	Exchange of nutrients.
Plant support system	Transport of nutrients in solution and of gas via aerenchyma.
Plant foliage	Transport of nutrients in solution and of gas via aerenchyma.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Domesticated animals	Consumption of above-ground plant parts.
Humans	Consumption of above-ground plant parts.

**Table 3-10. Processes of carbon transfer from the plant foliage.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Loss of foliage by leaf fall resulting in deposition on surface-water bodies (noting that strictly this occurs via the atmosphere and that it is a much less significant process for crop plants than for forest vegetation).
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	Senescence, death and decay generate organic matter that is added to the upper layers of the soil.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system	Exchange of nutrients.
Plant foliage	Internal redistribution of nutrients. Uptake of carbon dioxide from the atmosphere and release to the atmosphere in the processes of photosynthesis and respiration. Generation of nutrients used in the production of biomass.
Lower canopy atmosphere	Transfer of carbon dioxide originating from respiration.
Upper canopy atmosphere	Transfer of carbon dioxide originating from respiration.
Above-canopy atmosphere	Transfer of carbon dioxide originating from respiration (from the upper surfaces of foliage at the boundary of the canopy).
Domesticated animals	Consumption of above-ground plant parts.
Humans	Consumption of above-ground plant parts.

**Table 3-11. Processes of carbon transfer from the lower-canopy atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	Gas exchange with the soil atmosphere by advection and dispersion, including the effects of pumping due to pressure variations.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system	No direct connection.
Plant foliage	Uptake of carbon dioxide for use in photosynthesis. There may also be uptake of other gases that can be metabolised (e.g. carbon monoxide).
Lower canopy atmosphere	Advective and diffusive/dispersive transport resulting in mixing.
Upper canopy atmosphere	Advective and diffusive/dispersive transport.
Above-canopy atmosphere	No direct connection.
Domesticated animals	No direct connection.
Humans	No direct connection.

**Table 3-12. Processes of carbon transfer from the upper-canopy atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system	No direct connection.
Plant foliage	Uptake of carbon dioxide for use in photosynthesis. There may also be uptake of other gases that can be metabolised (e.g. carbon monoxide).
Lower canopy atmosphere	Advective and diffusive/dispersive transport.
Upper canopy atmosphere	Advective and diffusive/dispersive transport resulting in mixing.
Above-canopy atmosphere	Advective and diffusive/dispersive transport.
Domesticated animals	No direct connection.
Humans	No direct connection.

**Table 3-13. Processes of carbon transfer from the above-canopy atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	As exchange across the surfaces of water bodies.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system	No direct connection.
Plant foliage	Limited uptake of carbon dioxide for use in photosynthesis at the upper boundary of the plant canopy.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	Advective and diffusive/dispersive transport.
Above-canopy atmosphere	Advective and diffusive/dispersive transport resulting in mixing. Note that this may include both mixing within a local area (e.g. the release area for C-14) and mixing of local air into the larger-scale atmospheric circulation.
Domesticated animals	Inhalation. Assumes that inhalation occurs from above the plant canopy in agricultural contexts.
Humans	Inhalation. Assumes that inhalation occurs from above the plant canopy in agricultural contexts.

**Table 3-14. Processes of carbon transfer from domesticated animals.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Excreta may be discharged to surface waters or washed into surface waters in runoff.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	Excreta may be discharged to surface soils, either directly or after collection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system	Excreta may be discharged onto pasture and retained on plant surfaces.
Plant foliage	Excreta may be discharged onto pasture and retained on plant surfaces.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	Respiration and eructation releasing carbon dioxide and methane.
Domesticated animals	Internal redistribution, noting that carbon is relatively uniformly distributed throughout all the soft tissues of the body, but that biological half-lives differ substantially between different tissues and organs.
Humans	Consumption of animal products.

**Table 3-15. Processes of carbon transfer from humans.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system	No direct connection.
Plant foliage	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	Respiration.
Domesticated animals	No direct connection.
Humans	Internal redistribution, noting that carbon is relatively uniformly distributed throughout all the soft tissues of the body, but that biological half-lives differ substantially between different tissues and organs.

Note: Human excreta are assumed not to be discharged to agricultural systems or local surface waters.

These fifteen tables can be rearranged as an interaction matrix. This is done, in summary form, in Figure 3-6. In this figure, details of the individual processes governing carbon and C-14 fluxes between the various components are not shown. Instead, the off-diagonal elements are colour coded to show the broad classes of processes of relevance.



Consumption by domesticated animals and humans includes drinking water, adventitious ingestion of soil, ingestion of plants, and, for humans, ingestion of animal products. Excretion and respiration apply to both domesticated animals and humans, but human excreta are assumed not to enter the local agricultural environment. Gas exchanges between soils, plants and the atmosphere constitute a major transport pathway and include photosynthesis, respiration, methanogenesis and the oxidation of methane to carbon dioxide. Plants and mycorrhizal associations are intimately related and require consideration of microbial metabolism. However, this is also considered separately for the overall soil microbial community. Irrigation leads to carbon and C-14 transfers in abstracted groundwater or surface water to soils and plant but can also result in outgassing from the abstracted water. Gas exchanges can also arise across the interfaces between water bodies and the atmosphere. Water and gas transport in soils play a major part in carbon and C-14 transport and are best considered together because of the rapid exchanges that occur between the soil atmosphere and soil solution. Loss of plant parts is an important mechanism for transferring organic matter to soils (and secondarily to water bodies) where it is subject to decomposition and mineralisation, releasing the carbon.

The interaction matrix shown in Figure 3-6 is naturally decomposed into a set of interlinked conceptual models, as shown in Figure 3-7. These comprise a groundwater-based model (outlined in yellow), a soil-plant model (outlined in red), a plant-atmosphere model (outlined in blue), an irrigation model (outlined in purple), an intake model (outlined in green) and an excretion model (outlined in brown). Interactions are indicated by overlaps in the model domains and by the few interactions that lie outside these boundaries (green cells). These include losses of plant foliage directly to surface-water bodies, methanogenesis in the microbial community in saturated, anoxic soils, and exchanges of gas across water-atmosphere interfaces and soil-atmosphere interfaces.

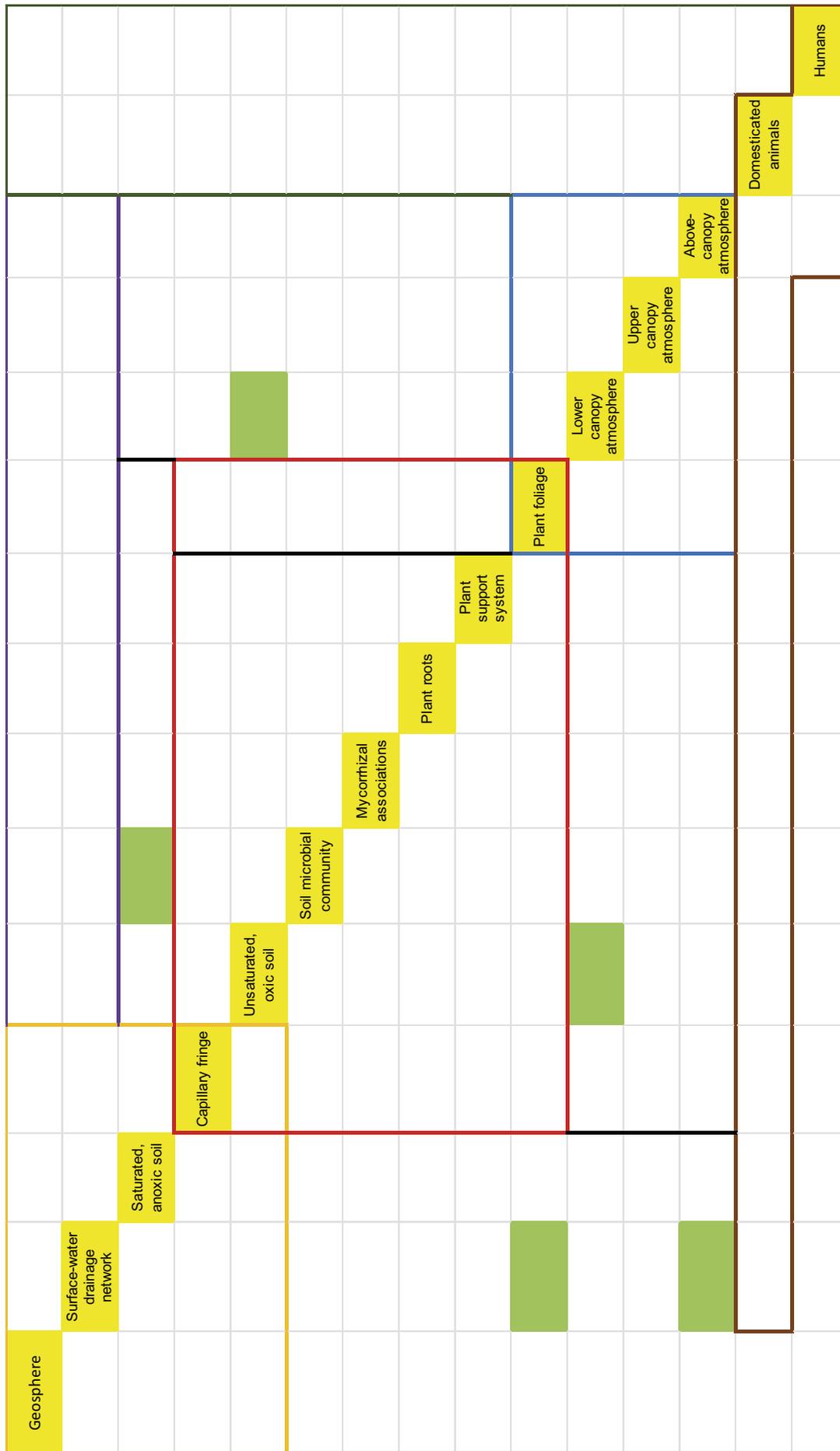


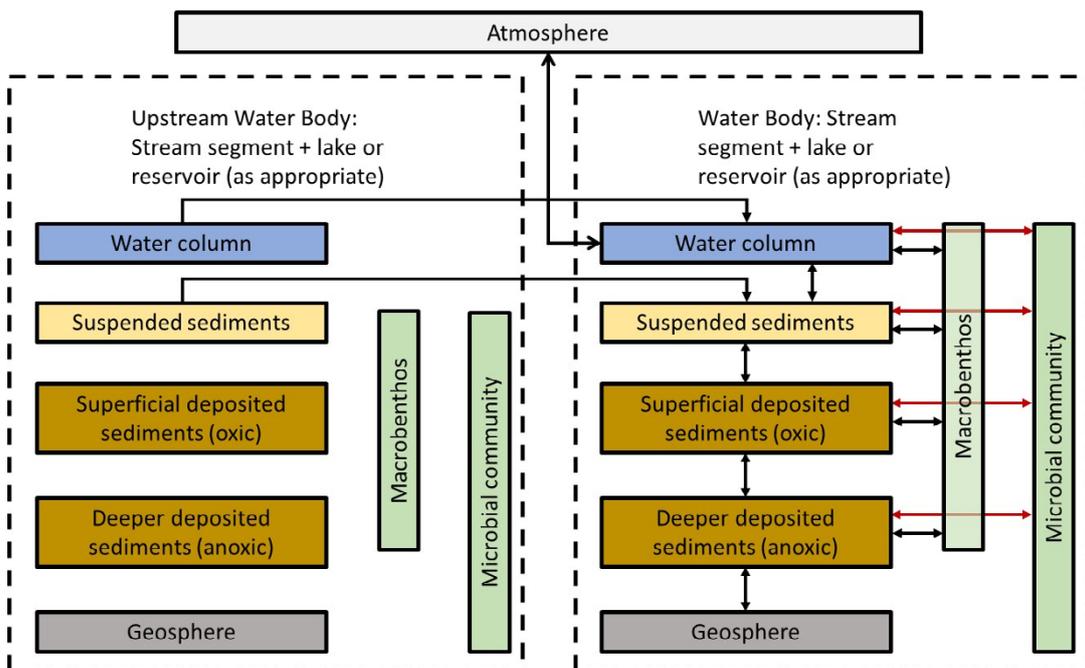
Figure 3-7. Distinction of the interaction matrix for an agricultural system into a set of conceptual models.

### 3.5 Conceptual models of the transport and accumulation of carbon in a surface-water drainage network

The surface-water drainage network comprises streams, rivers, lakes, reservoirs and other water bodies between the origins of streams, e.g. in springs and seeps, and the discharge of those streams and rivers at the coast either directly or via lagoons and estuaries. The seaward end of the drainage network is here defined as the boundary where the water becomes significantly saline, i.e. where tidal mixing with seawater becomes an important consideration in modelling. Overland flows to and from adjacent terrestrial areas are considered in the context of processes mediating C-14 transport between the surface-drainage network and the terrestrial environment but are not defined as principal components.

In general, the surface-water drainage network is dendritic in nature, with smaller streams feeding into larger streams and rivers. Thus, streams and rivers can be distinguished by numbering them in terms of confluences downstream of the specified stream segment, while lakes and reservoirs can be associated with the stream segment at which they are located. In defining a conceptual model, the focus is on a single stream segment that may or may not include a lake or reservoir. Water inputs to this segment comprise flow from the adjacent one or two upstream elements (two if the segment boundary is defined at a confluence), groundwater discharge, overland flow, interflow and direct precipitation. If precipitation water percolates to the water table and reaches the stream segment, this is taken to be indirectly as groundwater discharge or interflow. Direct precipitation is that which falls onto the surface of the water body (stream segment and lake/reservoir, as appropriate). Water outputs from the stream segment comprise groundwater recharge, overland flow, downstream discharge and interflow. In this case, overland flow results from overbanking events. Note that interflow can be either to or from the stream segment. It includes subsurface flows from upstream and to downstream, but also infiltration into and drainage from bank soils and sediments.

Within a stream segment, the principal components are the water bodies, the suspended sediments that they contain, the bottom sediments, and the macroscopic primary producers (macrophytes, i.e. larger plants, here generally described as the macrobenthos). The bottom sediments can be further distinguished as being oxic or anoxic, and, in either case, will include a substantial microbial community. A further distinction can be made between superficial sediments that are susceptible to resuspension and deeper sediments that are subject to burial or exhumation depending upon whether the flow regime is erosive or depositional. Overall, the characteristics of the stream segment and its relationship to the wider drainage network are shown in Figure 3-8.



**Figure 3-8.** General structure of a segment of the surface drainage network ('macrobenthos' refers to all macrophyte primary producers and the details of internal pathways are not shown for the upstream segment).

The upstream water body will supply C-14 as DIC, DOC, PIC and POC, hence the main transport pathways are in the water column and suspended sediments. Similar transport pathways arise downstream from the segment of interest. Exchanges occur also with the local atmosphere. For stable carbon, these will be bidirectional. However, for C-14 released via groundwater, they are likely to be mainly losses to atmosphere, because the released C-14 will be rapidly diluted and dispersed. Chemical and biochemical reactions will couple carbon dissolved in the water column to carbon incorporated in suspended particles. These particles will be coupled to superficial deposited sediments by processes of deposition and resuspension. In turn, the superficial sediments will be coupled to deeper sediments by burial and exhumation. In Figure 3-8, the deeper sediments are shown as anoxic. However, they could comprise both oxic and anoxic zones with transitional regions between them. The macrobenthos will use carbon from the water column and sediments. This will mainly be from the oxic zone. In contrast, the general microbial community will use carbon under both oxic and anoxic conditions, as described in Section 2.3 in the context of agricultural systems. Contributions to carbon transport from both the oxic and anoxic zones may be of particular importance in lakes and reservoirs. For clarity, biota other than primary producers are not shown in Figure 3-8. These will comprise both benthic and pelagic organisms plus others, such as birds, that are not confined to the surface-water network. Also, minor pathways, such as sub-horizontal flow in deposited sediments or saltation at the base of the water column are not shown explicitly.

Based on Figure 3-8 and the above discussion, the following principal elements of the surface water interaction matrix are defined:

- geosphere,
- atmosphere,
- upstream surface water,
- upstream suspended sediment,
- riparian zone,
- surface water,
- suspended sediment,
- superficial oxic deposited sediment,
- deeper oxic deposited sediment,
- deeper anoxic deposited sediment,
- macrobenthos,
- microbial community,
- consumers and detritivores, and
- humans.

Humans are included explicitly for dose assessment purposes, though they also have a role in shaping this and other environments. The riparian zone is included to allow interactions of the stream with its banks to be represented, providing a link with the terrestrial environment. Downstream losses are not shown explicitly, but mirror gains from upstream. The following tables detail the interactions between these principal components.

**Table 3-16. Processes of carbon transfer from the geosphere.**

Receiving Component	Processes of Transfer
Geosphere	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas as bubbles or continuous flow.
Atmosphere	No direct connection.
Upstream surface water	No direct connection (only addressing the component of the geosphere in direct contact to the surface water segment under consideration).
Upstream suspended sediment	No direct connection.
Riparian zone	No direct connection.
Surface water	Advection or diffusion in the liquid phase across the beds and banks of water bodies. Advection of gaseous forms in bulk gas across the beds and banks of water bodies.
Suspended sediment	Advection as particles across the beds and banks of water bodies. Likely to be a minor pathway.
Superficial oxic deposited sediment	No direct connection.
Deeper oxic deposited sediment	Advection or diffusion in the liquid phase if the deeper oxic sediment is in direct contact with the geosphere.
Deeper anoxic deposited sediment	Advection or diffusion in the liquid phase – generally applicable as the anoxic sediment is often taken to lie beneath the oxic sediment.
Macrobenthos	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Humans	No direct connection.

**Table 3-17. Processes of carbon transfer from the atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Advection and dispersion of carbon, mainly in gaseous form but potentially also as particulates.
Upstream surface water	No direct connection (only addressing the component of the atmosphere in direct contact to the surface water segment under consideration).
Upstream suspended sediment	No direct connection.
Riparian zone	No direct connection.
Surface water	Gaseous exchanges across the surface of the water body.
Suspended sediment	No direct connection.
Superficial oxic deposited sediment	No direct connection.
Deeper oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Macrobenthos	Photosynthesis and respiration where the upper parts of plants are exposed above the water surface.
Microbial community	No direct connection.
Consumers and detritivores	Inhalation.
Humans	Inhalation.

**Table 3-18. Processes of carbon transfer from upstream surface water.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection (only addressing the component of the atmosphere in direct contact to the surface water segment under consideration).
Upstream surface water	Advection and dispersion within the flowing water.
Upstream suspended sediment	No direct connection (not modelling transport within the upstream segment in this context).
Riparian zone	No direct connection.
Surface water	Flow of upstream surface water into the segment that is being modelled.
Suspended sediment	No direct connection.
Superficial oxic deposited sediment	No direct connection.
Deeper oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Macrobenthos	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Humans	No direct connection.

**Table 3-19. Processes of carbon transfer from upstream suspended sediment.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection (only addressing the component of the atmosphere in direct contact to the surface water segment under consideration).
Upstream surface water	No direct connection (not modelling transport within the upstream segment in this context).
Upstream suspended sediment	Advection and dispersion within the flowing water.
Riparian zone	No direct connection.
Surface water	No direct connection.
Suspended sediment	Flow of upstream surface water into the segment that is being modelled carries suspended sediment with it
Superficial oxic deposited sediment	No direct connection.
Deeper oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Macrobenthos	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Humans	No direct connection.

**Table 3-20. Processes of carbon transfer from the riparian zone.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection (only addressing the component of the atmosphere in direct contact to the surface water segment under consideration).
Upstream surface water	No direct connection
Upstream suspended sediment	No direct connection
Riparian zone	Advection and dispersion within the pore water of the beds and banks of streams. Chemical and biochemical transformations between DIC, DOC, PIC and POC.
Surface water	Seepage through the bed and banks. Drainage of surface water after overbanking events.
Suspended sediment	Erosion of bank and bed leading to particle release.
Superficial oxic deposited sediment	Changes in channel morphology converting bank materials to bottom sediments <i>in situ</i> (i.e. not by erosion and redeposition, which is an indirect process mediated by suspended sediment).
Deeper oxic deposited sediment	Changes in channel morphology converting bank materials to bottom sediments <i>in situ</i>
Deeper anoxic deposited sediment	Changes in channel morphology converting bank materials to bottom sediments <i>in situ</i>
Macrobenthos	No direct connection.
Microbial community	Introduction of new microbial populations by bank erosion.
Consumers and detritivores	No direct connection.
Humans	No direct connection.

**Table 3-21. Processes of carbon transfer from surface water.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Degassing from water surfaces.
Upstream surface water	No direct connection (opposite the direction of flow).
Upstream suspended sediment	No direct connection.
Riparian zone	Infiltration into the banks and bed. Overbank flooding.
Surface water	Advection and dispersion in the flow.
Suspended sediment	Adsorption and incorporation in particles, e.g. by microbially mediated interactions.
Superficial oxic deposited sediment	Dispersion of flow into the sediments.
Deeper oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Macrobenthos	Passive and active uptake from solution.
Microbial community	Use of dissolved carbon in microbial metabolism.
Consumers and detritivores	Ingestion of water.
Humans	Ingestion of water.

**Table 3-22. Processes of carbon transfer from suspended sediment.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection (opposite the direction of flow).
Upstream suspended sediment	No direct connection.
Riparian zone	Deposition onto the bed and banks due to changes in channel morphology. Transport of sediment in overbank flooding.
Surface water	Desorption and other release from particles, e.g. by microbially mediated interactions.
Suspended sediment	Advection and dispersion in the flow.
Superficial oxic deposited sediment	Particle settling.
Deeper oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Macrobenthos	Particle trapping on surfaces.
Microbial community	Use of particulate carbon in microbial metabolism.
Consumers and detritivores	Ingestion of water.
Humans	Ingestion of water (decreased in significance if the water is filtered).

**Table 3-23. Processes of carbon transfer from superficial oxic deposited sediment.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream suspended sediment	No direct connection.
Riparian zone	Deposition onto the bed and banks due to changes in channel morphology.
Surface water	No direct connection.
Suspended sediment	Resuspension of particles.
Superficial oxic deposited sediment	Burial and bioturbation.
Deeper oxic deposited sediment	Burial, bioturbation and diffusion/dispersion across the interface.
Deeper anoxic deposited sediment	No direct connection unless the deeper oxic deposited sediment is not present or is bypassed. In these circumstances, burial, bioturbation and diffusion/dispersion across the interface all apply.
Macrobenthos	Active or passive uptake by plant roots.
Microbial community	Use in microbial metabolism.
Consumers and detritivores	Ingestion of sediment by benthic organisms.
Humans	No direct connection.

**Table 3-24. Processes of carbon transfer from deeper oxic deposited sediment.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection unless the deeper anoxic deposited sediment is not present or is bypassed. In these circumstances, diffusion/dispersion across the interface applies.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream suspended sediment	No direct connection.
Riparian zone	Deposition onto the bed and banks due to changes in channel morphology.
Surface water	No direct connection.
Suspended sediment	No direct connection.
Superficial oxic deposited sediment	Exhumation and bioturbation.
Deeper oxic deposited sediment	Burial or exhumation, bioturbation
Deeper anoxic deposited sediment	Burial, bioturbation and diffusion/dispersion across the interface all apply.
Macrobenthos	Active or passive uptake by plant roots.
Microbial community	Use in microbial metabolism.
Consumers and detritivores	Ingestion of sediment by benthic organisms present in the sediments rather than on their upper surfaces.
Humans	No direct connection.

**Table 3-25. Processes of carbon transfer from deeper anoxic deposited sediment.**

Receiving Component	Processes of Transfer
Geosphere	Diffusion/dispersion across the interface.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream suspended sediment	No direct connection.
Riparian zone	Deposition onto the bed and banks due to changes in channel morphology.
Surface water	No direct connection.
Suspended sediment	No direct connection.
Superficial oxic deposited sediment	No direct connection.
Deeper oxic deposited sediment	Exhumation, bioturbation and diffusion/dispersion across the interface all apply.
Deeper anoxic deposited sediment	Burial or exhumation. Bioturbation is assumed not to be significant in anoxic conditions.
Macrobenthos	No direct connection. Plant roots are assumed not to extend a considerable distance into the anoxic zone. However, even limited contact could be significant, so this pathway may need further consideration.
Microbial community	Use in microbial metabolism.
Consumers and detritivores	No direct connection.
Humans	No direct connection.

**Table 3-26. Processes of carbon transfer from the macrobenthos.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream suspended sediment	No direct connection.
Riparian zone	Terrestrialisation due to vegetation ingrowth.
Surface water	Release of carbon as gas or in solution.
Suspended sediment	Release of plant detritus, e.g. through weathering, loss of plant parts and death.
Superficial oxic deposited sediment	Exudation in solution and release of plant detritus.
Deeper oxic deposited sediment	Exudation in solution and release of plant detritus.
Deeper anoxic deposited sediment	No direct connection. Plant roots are assumed not to extend significantly into anoxic sediments.
Macrobenthos	Transport and distribution within plants.
Microbial community	Symbiotic associations and exchanges of organic compounds between macrobiota and the microbial community.
Consumers and detritivores	Ingestion
Humans	Ingestion of a few special plant types.

**Table 3-27. Processes of carbon transfer from the microbial community.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream suspended sediment	No direct connection.
Riparian zone	No direct connection.
Surface water	Release of carbon as gas or in solution.
Suspended sediment	Release of microbes as particles of organic matter.
Superficial oxic deposited sediment	Release of carbon as gas or in solution.
Deeper oxic deposited sediment	Release of carbon as gas or in solution.
Deeper anoxic deposited sediment	Release of carbon as gas or in solution.
Macrobenthos	Symbiotic associations and exchanges of organic compounds between macrobiota and the microbial community
Microbial community	Microbially mediated oxidation and reduction reactions depending on the geochemical context, biomass production and mineralisation.
Consumers and detritivores	Ingestion
Humans	No direct connection.

**Table 3-28. Processes of carbon transfer from consumers and detritivores.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Inhalation.
Upstream surface water	No direct connection.
Upstream suspended sediment	No direct connection.
Riparian zone	No direct connection (terrestrial food chain pathways are addressed in other contexts).
Surface water	Excretion and gas transfer (respiration)
Suspended sediment	Excretion, loss of detritus, death and decomposition.
Superficial oxic deposited sediment	Excretion, loss of detritus, death and decomposition.
Deeper oxic deposited sediment	Excretion, loss of detritus, death and decomposition.
Deeper anoxic deposited sediment	No direct connection. Consumers and detritivores are assumed not to be present to a significant extent in anoxic sediments.
Macrobenthos	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	Internal transport and distribution.
Humans	Consumption of animal products.

**Table 3-29. Processes of carbon transfer from humans.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Inhalation.
Upstream surface water	No direct connection.
Upstream suspended sediment	No direct connection.
Riparian zone	No direct connection (terrestrial food chain pathways are addressed in other contexts).
Surface water	Excretion (typically via a sewage system).
Suspended sediment	Excretion (typically via a sewage system).
Superficial oxic deposited sediment	No direct connection.
Deeper oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Macrobenthos	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Humans	Internal transport and distribution.

The corresponding interaction matrix is shown in Figure 3-9, which is set out similarly to that for agricultural ecosystems in Figure 3-6. However, note that the colour key is not identical, reflecting the different dominant processes. The above tables should be consulted for more detail of the processes considered.



In Figure 3-9, the processes are seen to cluster. Inputs and losses to the stream segment are dominated by advection and dispersion/diffusion in the liquid phase (both in solution and in suspended particulates). However, within the stream segment, mechanical processes of mixing, erosion, sedimentation and morphological change have the main role. Plant-related processes occur largely in the macrobenthos that interacts strongly with the water and with the microbial community. Higher trophic levels (consumers, detritivores and humans) have intakes by inhalation and consumption of drinking water and food (including organic detritus), as well as sediment ingested adventitiously. The various types of process are separated into distinct blocks in the interaction matrix, indicating the potential to distinguish the overall conceptual model into four weakly coupled sub-models relating to fluid flow and gas exchange, sediment transport, microbial and plant metabolism, and uptake, loss and biochemical transformation processes at higher trophic levels.

### **3.6 Conceptual models of the transport and accumulation of carbon in forest ecosystems**

There is a broad spectrum of various types of forests and woodlands. Here, for simplicity, the focus is on forested areas on mainly aerated soils. That is, specifically, the soils behave in respect of the carbon storage pools and fluxes in a manner like agricultural (surface-aerated) ecosystems. For forests on mostly wet soils, the soils behave in respect of the carbon storage pools and fluxes in a manner like wetlands, and the reader is referred to the conceptual model for wetland (surface-wet) ecosystems. The other main consideration is that forests are generally characterised by a well-defined litter layer, and understorey vegetation that is distinct from the main canopy. Based on these considerations, the principal components of a forest ecosystem are defined as:

- geosphere,
- surface water drainage network,
- saturated, anoxic soil,
- capillary fringe of a varying degree of anoxia,
- unsaturated, oxic soil,
- litter layer,
- soil microbial community,
- mycorrhizal associations,
- plant roots,
- plant support system for understorey vegetation,
- plant foliage and berries for understorey vegetation,
- plant support system for trees,
- plant foliage for trees,
- understorey canopy atmosphere,
- tree canopy atmosphere,
- above-canopy atmosphere,
- animals (both wild and, in some cases, domesticated<sup>7</sup>), and
- humans.

This results in an interaction matrix for forest ecosystems that is like that for agricultural systems, although a little larger. The interactions of forest ecosystems are summarised below in a set of eighteen tables, in the standard format adopted in the previous subsections. The main differences between models for agricultural ecosystems and forest are the distinctions between litter layer, understorey

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<sup>7</sup> For example, in some cases, this may include sheep or cattle herding in forest pastures.

and main canopy noted above, and the inclusion of wild plants and animals. Whereas many of the interactions related to the litter layer are like those related to the unsaturated soil, the material properties and carbon turnover rates typically differ, thus the distinction. The principal components and interactions are relevant both to biota dose assessments and to human food-chain pathways involving wild foods. In mathematical modelling, it may be appropriate to adopt a multi-layer model for the understory and main canopy to reflect vertical gradients in gaseous carbon (and C-14) concentrations, biomass density, and rates of photosynthesis and plant respiration. Pathways involving wells are included to allow for domestic and agricultural wells being constructed in clearings of the forest occupied by one or a few homesteads.

**Table 3-30. Processes of carbon transfer from the geosphere.**

Receiving Component	Processes of Transfer
Geosphere	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas as bubbles or continuous flow.
Surface-water drainage network	Advection or diffusion in the liquid phase across the beds and banks of water bodies. Advection of gaseous forms in bulk gas across the beds and banks of water bodies.
Saturated, anoxic soil	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas as bubbles or continuous flow.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for understory vegetation	No direct connection.
Plant foliage and berries for understory vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	No direct connection. (Except in cases with livestock or other domesticated animals, consumption of drinking water abstracted from wells.)
Humans	Consumption of drinking water abstracted from wells.

**Table 3-31. Processes of carbon transfer from the surface-water drainage network.**

Receiving Component	Processes of Transfer
Geosphere	Infiltration of water into the beds and banks of water bodies.
Surface-water drainage network	Advection and dispersion in flowing or circulating water. Incorporation and decorporation into solids. Exchange between gaseous and solution phases.
Saturated, anoxic soil	Infiltration into banks and flooding in riparian areas.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for understory vegetation	No direct connection.
Plant foliage and berries for understory vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	As exchange across the surfaces of water bodies.
Animals	Consumption of drinking water abstracted from surface-water bodies.
Humans	Consumption of drinking water abstracted from surface-water bodies.

**Table 3-32. Processes of carbon transfer from the saturated anoxic soil.**

Receiving Component	Processes of Transfer
Geosphere	Infiltration and downward diffusion.
Surface-water drainage network	Advection or diffusion in the liquid phase across the beds and banks of water bodies.
Saturated, anoxic soil	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas originating from the geosphere or arising from microbial processes as bubbles or continuous flow.
Capillary fringe of a varying degree of anoxia	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas.
Unsaturated, oxic soil	No direct connection.
Litter layer	No direct connection.
Soil microbial community	Uptake for use in metabolism. A key process is the production of methane.
Mycorrhizal associations	Uptake of methane and other organic compounds for use in metabolism.
Plant roots	No direct connection. Plant roots are taken not to penetrate the permanently saturated soil zone in upland forest systems, though such penetration may occur in wetlands (wet-soil forests).
Plant support system for understory vegetation	No direct connection.
Plant foliage and berries for understory vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	No direct connection. (Except in cases with livestock or other domesticated animals, consumption of drinking water abstracted from wells.)
Humans	Consumption of drinking water abstracted from wells.

**Table 3-33. Processes of carbon transfer from the capillary fringe.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Transport by interflow during and after precipitation events.
Saturated, anoxic soil	Advection and dispersion, including effects arising from a time-varying position of the water table.
Capillary fringe of a varying degree of anoxia	Advection and dispersion, including effects arising from a time-varying position of the water table. Exchanges between soil solution and soil atmosphere. Oxidation and reduction of carbon compounds, with the direction of reaction depending on local characteristics (oxic or anoxic). Mineralisation of organic matter originating from plant and animal material. Silvicultural soil preparation and other forms of soil disturbance. Bioturbation.
Unsaturated, oxic soil	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Silvicultural soil preparation and other forms of soil disturbance. Bioturbation.
Litter layer	No direct connection.
Soil microbial community	Uptake of methane and other organic compounds for use in metabolism. Microbially mediated processes may involve both oxidation and reduction reactions.
Mycorrhizal associations	Uptake of methane and other organic compounds for use in metabolism.
Plant roots	Uptake of carbon compounds present in solution by both active and passive processes. Uptake of gaseous forms for onward transmittal via aerenchyma (though this is likely to be of little significance in upland forest systems, being mainly of relevance in wet-soil forest and wetland systems).
Plant support system for understorey vegetation	No direct connection.
Plant foliage and berries for understorey vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	No direct connection.
Humans	No direct connection.

**Table 3-34. Processes of carbon transfer from the unsaturated oxic soil.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Mixing with, and transport by runoff and interflow during and after precipitation events.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Silvicultural soil preparation and other forms of soil disturbance. Bioturbation.
Unsaturated, oxic soil	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Oxidation of organic forms, particularly methane. Mineralisation of organic matter originating from plant and animal material. Silvicultural soil preparation and other forms of soil disturbance. Bioturbation.
Litter layer	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Silvicultural soil preparation and other forms of soil disturbance. Bioturbation.
Soil microbial community	Uptake for use in metabolic processes.
Mycorrhizal associations	Uptake for use in metabolic processes.
Plant roots	Uptake of carbon compounds present in solution by both active and passive processes.
Plant support system for understorey vegetation	No direct connection.
Plant foliage and berries for understorey vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Adventitious soil consumption.
Humans	Adventitious soil consumption.

**Table 3-35. Processes of carbon transfer from the litter layer.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Mixing with, and transport by runoff and interflow during and after precipitation events.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Silvicultural soil preparation and other forms of soil disturbance. Bioturbation.
Litter layer	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Oxidation of organic forms, particularly methane. Mineralisation of organic matter originating from plant and animal material. Silvicultural soil preparation and other forms of soil disturbance. Bioturbation.
Soil microbial community	Uptake for use in metabolic processes.
Mycorrhizal associations	Uptake for use in metabolic processes.
Plant roots	Uptake of carbon compounds present in solution by both active and passive processes.
Plant support system for understorey vegetation	No direct connection.
Plant foliage and berries for understorey vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	Gas exchange across the litter-layer surface.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Uptake for use in metabolic processes (detritivores). Adventitious soil consumption.
Humans	Adventitious soil consumption.

**Table 3-36. Processes of carbon transfer from the soil microbial community.**

<b>Receiving Component</b>	<b>Processes of Transfer</b>
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Capillary fringe of a varying degree of anoxia	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Unsaturated, oxic soil	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Litter layer	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Soil microbial community	Exchange of products of metabolism between organisms within the microbial community.
Mycorrhizal associations	No direct connection. Both communities are microbial, but it is reasonable to treat them as only indirectly coupled.
Plant roots	No direct connection.
Plant support system for understorey vegetation	No direct connection.
Plant foliage and berries for understorey vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption.
Humans	No direct connection.

**Table 3-37. Processes of carbon transfer from the mycorrhizal associations.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Unsaturated, oxic soil	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Litter layer	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Soil microbial community	No direct connection. Both communities are microbial, but it is reasonable to treat them as only indirectly coupled.
Mycorrhizal associations	Internal transfers between interfaces with the soil and interfaces with plant roots.
Plant roots	Active transfers of products of metabolism.
Plant support system for understorey vegetation	No direct connection.
Plant foliage and berries for understorey vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption (including mushrooms).
Humans	Consumption of mushrooms.

**Table 3-38. Processes of carbon transfer from the plant roots.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	Root exudation. Death and decay generating organic matter that is subject to mineralisation on various timescales.
Unsaturated, oxic soil	Root exudation. Death and decay generating organic matter that is subject to mineralisation on various timescales.
Litter layer	Root exudation. Death and decay generating organic matter that is subject to mineralisation on various timescales.
Soil microbial community	No direct connection.
Mycorrhizal associations	Active transfers of products of metabolism.
Plant roots	Transport in solution from locations of absorption. Transport in the gas phase through aerenchyma.
Plant support system for understorey vegetation	Export to the above-ground parts of the plant in solution and in the gas phase.
Plant foliage and berries for understorey vegetation	No direct connection.
Plant support system for trees	Export to the above-ground parts of the plant in solution and in the gas phase.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption of plant roots.
Humans	Harvest and use of biomass (typically stumps and large roots).

**Table 3-39. Processes of carbon transfer from the plant support system for understorey.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	Senescence, death and decay generate organic matter that is added to the litter layer prior to subsequent decay, decomposition and mixing with the litter layer.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	Transport of nutrients in solution and of gas via aerenchyma.
Plant support system for understorey vegetation	Transport of nutrients in solution and of gas via aerenchyma.
Plant foliage and berries for understorey vegetation	Transport of nutrients in solution and of gas via aerenchyma.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption of twigs, branches and stems (herbivory).
Humans	Harvest and use of biomass.

**Table 3-40. Processes of carbon transfer from the plant foliage and berries for understorey.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Loss of foliage by litterfall resulting in deposition on surface-water bodies (noting that strictly this occurs via the atmosphere).
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	Senescence, death and decay generate organic matter that is added to the litter layer prior to subsequent decay, decomposition and mixing with the litter layer.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	Transport of nutrients in solution and of gas via aerenchyma.
Plant support system for understorey vegetation	Transport of nutrients in solution and of gas via aerenchyma.
Plant foliage and berries for understorey vegetation	Transport of nutrients in solution and of gas via aerenchyma.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	Transfer of carbon dioxide originating from respiration.
Tree canopy atmosphere	Transfer of carbon dioxide originating from respiration (from the upper surfaces of understorey at the boundary of the understorey canopy atmosphere).
Above-canopy atmosphere	No direct connection.
Animals	Consumption of berries and other above-ground plant parts.
Humans	Consumption of berries and other wild plant foods. Less typically, harvest and use of needles/leaves for various products.

**Table 3-41. Processes of carbon transfer from the plant support system for trees.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Loss of twigs and branches by litterfall resulting in deposition on surface-water bodies (noting that strictly this occurs via the atmosphere, and generally is of a minor mass flux compared to that from the Tree foliage).
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	Senescence, death and decay generate organic matter that is added to the litter layer prior to subsequent decay, decomposition and mixing with the litter layer. Harvest residues.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	Transport of nutrients in solution and of gas via aerenchyma.
Plant support system for understorey vegetation	No direct connection.
Plant foliage and berries for understorey vegetation	No direct connection.
Plant support system for trees	Transport of nutrients in solution and of gas via aerenchyma.
Plant foliage for trees	Transport of nutrients in solution and of gas via aerenchyma.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption of twigs, branches and stems (herbivory).
Humans	Harvest and use of logs, firewood and biomass.

**Table 3-42. Processes of carbon transfer from the plant foliage for trees.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Loss of foliage by litterfall resulting in deposition on surface-water bodies (noting that strictly this occurs via the atmosphere).
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	Senescence, death and decay generate organic matter that is added to the litter layer prior to subsequent decay, decomposition and mixing with the litter layer. Harvest residues.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	Transport of nutrients in solution and of gas via aerenchyma.
Plant support system for understorey vegetation	No direct connection.
Plant foliage and berries for understorey vegetation	No direct connection.
Plant support system for trees	Transport of nutrients in solution and of gas via aerenchyma.
Plant foliage for trees	Transport of nutrients in solution and of gas via aerenchyma.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	Transfer of carbon dioxide originating from respiration.
Above-canopy atmosphere	Transfer of carbon dioxide originating from respiration (from the upper surfaces of foliage at the boundary of the tree canopy atmosphere).
Animals	Consumption of needles and leaves (herbivory).
Humans	Harvest and use of biomass. Less typically, harvest and use of needles/leaves for various products.

**Table 3-43. Processes of carbon transfer from the understory canopy atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	Gas exchange with the soil atmosphere by advection and dispersion, including the effects of pumping due to pressure variations.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for understory vegetation	No direct connection.
Plant foliage and berries for understory vegetation	Uptake of carbon dioxide for use in photosynthesis. There may also be uptake of other gases that can be metabolised (e.g. carbon monoxide).
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	Advective and diffusive/dispersive transport resulting in mixing.
Tree canopy atmosphere	Advective and diffusive/dispersive transport.
Above-canopy atmosphere	No direct connection.
Animals	Inhalation.
Humans	Inhalation (typically mainly children; a common identifier of trees from understory plants is that trees are at 'breast height (1.3 m) or taller).

**Table 3-44. Processes of carbon transfer from the tree canopy atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for understory vegetation	No direct connection.
Plant foliage and berries for understory vegetation	Limited uptake of carbon dioxide for use in photosynthesis at the upper boundary of the tree canopy.
Plant support system for trees	No direct connection.
Plant foliage for trees	Uptake of carbon dioxide for use in photosynthesis. There may also be uptake of other gases that can be metabolised (e.g. carbon monoxide).
Understorey canopy atmosphere	Advective and diffusive/dispersive transport.
Tree canopy atmosphere	Advective and diffusive/dispersive transport resulting in mixing.
Above-canopy atmosphere	Advective and diffusive/dispersive transport.
Animals	Inhalation.
Humans	Inhalation.

**Table 3-45. Processes of carbon transfer from the above-canopy atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	As exchange across the surfaces of water bodies.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for understory vegetation	No direct connection.
Plant foliage and berries for understory vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	Limited uptake of carbon dioxide for use in photosynthesis at the upper boundary of the tree canopy.
Understorey canopy atmosphere	No direct connection.
Tree canopy atmosphere	Advective and diffusive/dispersive transport.
Above-canopy atmosphere	Advective and diffusive/dispersive transport resulting in mixing. Note that this may include both mixing within a local area (e.g. the release area for C-14) and mixing of local air into the larger-scale atmospheric circulation.
Animals	Inhalation (mainly avian species).
Humans	No direct connection.

**Table 3-46. Processes of carbon transfer from animals.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Excreta may be discharged to surface waters or washed into surface waters in runoff.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	Excretion. Death and decay of carcasses.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for understory vegetation	Excreta may be discharged onto and retained on plant surfaces.
Plant foliage and berries for understory vegetation	Excreta may be discharged onto and retained on plant surfaces.
Plant support system for trees	Excreta may be discharged onto and retained on plant surfaces.
Plant foliage for trees	Excreta may be discharged onto and retained on plant surfaces.
Understorey canopy atmosphere	Respiration and eructation releasing carbon dioxide and methane.
Tree canopy atmosphere	Respiration and eructation releasing carbon dioxide and methane.
Above-canopy atmosphere	Respiration and eructation releasing carbon dioxide and methane (mainly avian species).
Animals	Internal redistribution, noting that carbon is relatively uniformly distributed throughout all the soft tissues of the body, but that biological half-lives differ substantially between different tissues and organs. Predation among animal populations.
Humans	Consumption (and other uses) of animal products.

**Table 3-47. Processes of carbon transfer from humans.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Saturated, anoxic soil	No direct connection.
Capillary fringe of a varying degree of anoxia	No direct connection.
Unsaturated, oxic soil	No direct connection.
Litter layer	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for understorey vegetation	No direct connection.
Plant foliage and berries for understorey vegetation	No direct connection.
Plant support system for trees	No direct connection.
Plant foliage for trees	No direct connection.
Understorey canopy atmosphere	Respiration (typically mainly children; cf. Inhalation from the Understorey canopy atmosphere).
Tree canopy atmosphere	Respiration.
Above-canopy atmosphere	No direct connection.
Animals	Population control through hunting for food and game/environmental management. Habitat alteration (forest management). These interactions have not been marked in the interaction matrix, however, since they do not directly affect carbon transport modelling (cf. Consumption of animal products from Animals to Humans).
Humans	Internal redistribution, noting that carbon is relatively uniformly distributed throughout all the soft tissues of the body, but that biological half-lives differ substantially between different tissues and organs.

The corresponding interaction matrix is illustrated in Figure 3-10 for which the following colour key applies to reflect dominant processes. It should be noted that not all processes will be relevant to every situation, as indicated with a circular marker in the interaction matrix and the text in the above tables.

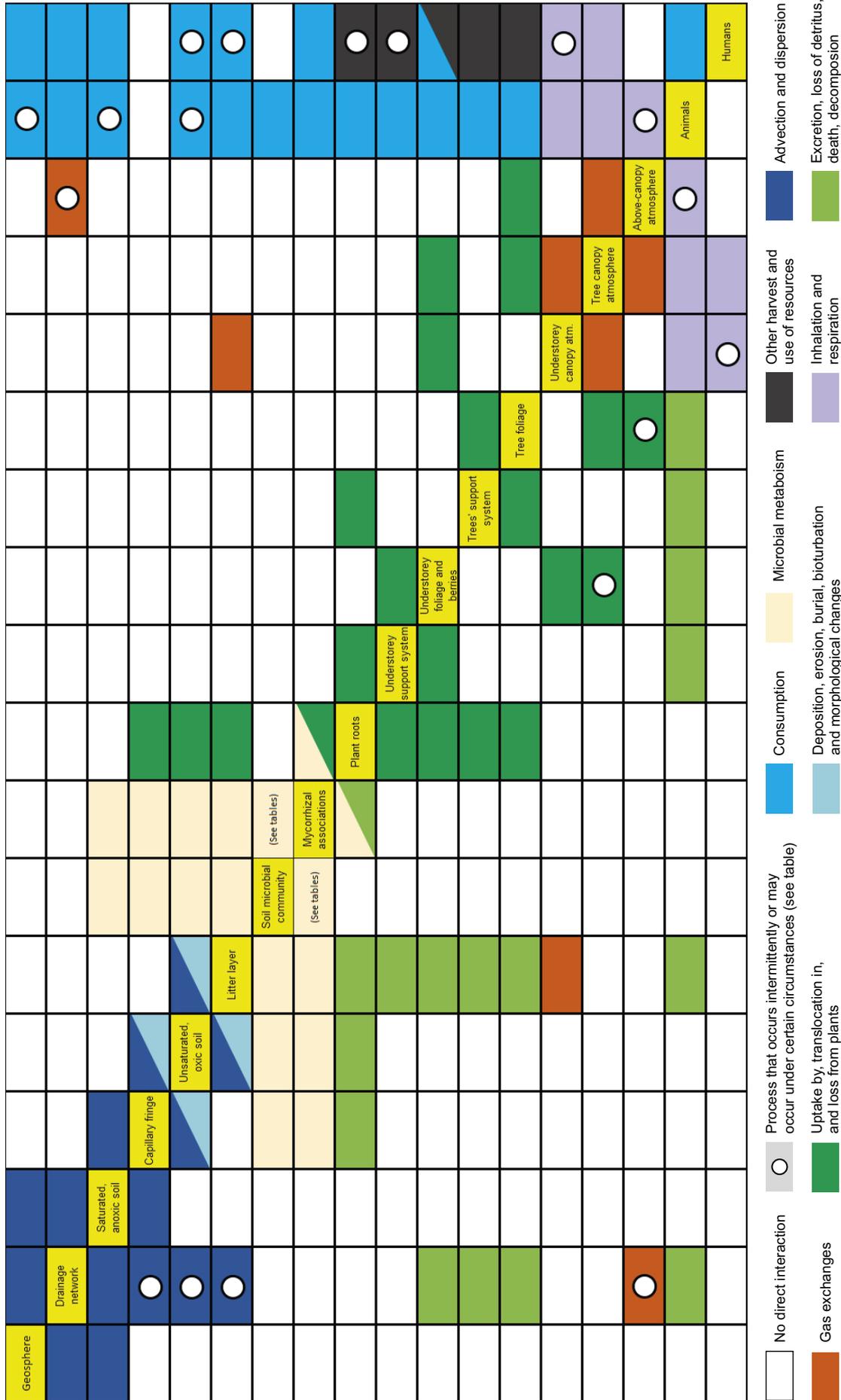


Figure 3-10. Interaction matrix for a forest.

### 3.7 Conceptual models of the transport and accumulation of carbon in wetland ecosystems

Here, the focus is on land areas where the groundwater table is, for considerable parts of the year, close to the surface (i.e., within a ‘few tens of centimetres’) so that most of the methane is not oxidised in the topsoil. In wetlands, the water level may also be above the ground locally and temporally, which adds characteristics addressed for aquatic systems; and for treed areas, in turn, the continuum is towards forests. There are wetlands commonly also in the littoral zones of freshwater, estuarine and marine systems as an interface or rather a continuum between the ‘land’ and ‘water’ systems. These landscape continuities and establishment of interfaces between ecosystem-specific conceptual models tends, therefore, be specific to the case under assessment.

As discussed in the literature review and in the context of this subsection, the distinction of wetlands from the other types of ecosystems is the intimate coexistence of wetness and the accumulation of organic matter (typically peat). In general, this requires a positive water balance (surplus of water), low rate of decomposition (helped by anoxia), and supply of poorly degradable organic matter (vegetation adapted to the wet and low-oxygen substrate and producing a specific type of litter). Such ecosystems are characterised by:

- A strong but temporally variable vertical structuring in the redox potential, governed by the groundwater table fluctuations (sometimes distinguished as the ‘mesotelm’ region, sometimes being the moving interface between acrotelm and catotelm described below; here, the former practice is adopted to support appropriate consideration of the dynamics in mathematical models),
- The upper soil layer (‘acrotelm’ in case of peatlands) is mostly unsaturated and oxic during the growing season and it supports most of the biological activity, whereas the layers below (‘catotelm’) are waterlogged and anoxic (the deeper, the more strictly this is accurate),
- The roots and the root respiration are mainly restricted to the upper zone with at least intermittently unsaturated and oxic conditions,
- The litter degradability decreases with depth in general, as the organic matter ages and the ongoing decomposition produces increasingly recalcitrant residues, but the input from dead roots, root exudates and litter leachates results in some non-uniformity in this pattern of degradability,
- The transport of gases and solutes is slower with depth due to the self-compaction of the soil column decreasing the diffusion coefficients and hydraulic conductivity,
- The concentrations of electron acceptors, particularly oxygen, decreases with their distance from the atmospheric source (i.e. from the surface) and tends to rapidly become depleted in the saturated zone.

As outlined above, the transport in the deeper, permanently waterlogged catotelm is typically diffusive, but it can nevertheless cover considerable distances over centennial and millennial timescales. However, the mineral sub-soil underlying the organic deposits may host more considerable groundwater flow also laterally, in some cases forming a hyporheic linkage to the drainage network in a manner that is largely decoupled from the surface circulation of the actual wetland system above. In addition, in the catotelm, cracks and pipes in the peat may provide preferential transport paths for solutes and gases over the otherwise diffusion-dominated system.

On the surface, a system of permanently flooded hollows and pools is like, and therefore readily addressed by, the conceptual model for a surface-water drainage network. Its formation and connectivity has recently been described and illustrated by Fenton (2021): The formation is often linked to inclined collapse at the acrotelm bottom and to the consequent slip or creep of the acrotelm ‘mattress’ upon the considerably more stationary catotelm, resulting in lateral compression and wrinkling of the acrotelm that may result in the bottoms of the wrinkle waves becoming permanently flooded, which initiates further wetting and gradual inter-connection of the wet locations. Such formation of patterned systems of wet hollows and pools seems typical to a range of gentler slopes, although not always occurring (possibly due to the lack of the initiating perturbation, so far). The deeper pools developing in somewhat longer term may also gradually expand at the cost of loss of their peripheral peat deposits, resulting in gradual mass transfer (collapse) from the wetland to the drainage network system. It is also to be noted that bog pools may considerably shorten the distance between the subsoil and the surface system even in areas with thick peat deposits, depending on their depth that may be linked to the timing of their initiation relative to the peat accumulation.

Depending on the characteristics of the wetland there may be also trees growing at least on drier (slightly elevated) areas. Depending on their density and configuration, they can act as windbreaks, potentially providing a sheltering effect. Their presence also tends to increase turbulence. Also, similarly to a forest ecosystem, trees have a different carbon storage and turnover pattern compared with other vegetation. As well as living individuals of some tree species facilitating aerenchymal gas exchange along their stems, dead trees may also act as gas-exchange channels in and out the soil (Martinez et al. 2022), bypassing the lower vegetation canopy.

Considering animals in wetlands there is a wide range of inhabitants from microfauna in soil layers (and sediments in surface-water drainage networks) to visiting birds and mammals that may, depending on the landscape settings and overall environmental conditions, use wetland areas as a considerable source of resources, even if typically residing mostly elsewhere (e.g., insectivorous birds and bats; grazing moose and other deer). As discussed in the literature review, animals may have a fertilising effect on microbial metabolism and interactions. Furthermore, trampling by larger animals (or humans) may trigger ebullition events releasing dissolved gases from the soil layers.

Humans may utilise wetlands for hunting and picking berries and mushrooms, or harvesting some natural materials (e.g. ornamental, cosmetic, medicinal or fibre plants). Wetlands may be used also to harvest hay or other fodder for livestock typically residing elsewhere (infield–outland agriculture; best represented at the landscape-level conceptual models connecting across ecosystems). Other uses possibly leading to exposure of people to radioactivity in a wetland ecosystem include extraction of peat either on a small scale (typically within a farm, e.g. for absorbent materials in animal husbandry, burning for heat, or organic soil amendment or growth medium in gardens) or peat extraction for fuel on a larger scale that typically becomes destructive to the ecosystem (whereas smaller-scale peat extraction may result only in local disturbances). Such exposure modes may involve radioactivity accumulated over long timescales also in deeper deposits. Peat extraction also typically results in release of part of the accumulated radioactivity into the surface-water drainage network due to artificial lowering of the water table and other disturbances. In some regions, treed wetlands may also be subject to forestry, i.e. harvest of logs, firewood or biomass; this is often associated with artificial lowering of the water table through intensified surface-water drainage.

Based on these considerations, the principal components of a wetland ecosystem are defined as:

- geosphere,
- surface-water drainage network,
- saturated, anoxic, mineral sub-soil,
- catotelm,
- mesotelm of a varying degree of anoxia,
- acrotelm,
- soil microbial community,
- mycorrhizal associations,
- plant roots,
- plant support system for non-tree vegetation,
- plant foliage and berries for non-tree vegetation,
- plant support system for trees (if present),
- plant foliage for trees (if present),
- lower canopy atmosphere (directly associated with the non-tree vegetation),
- upper canopy atmosphere (may include more or less sporadic tree canopy atmosphere),
- above-canopy atmosphere,
- animals, and
- humans.

This results in an interaction matrix for wetland ecosystems that is like that for forest systems, although with some differences in the identity of the principal components (structurally, the mineral sub-soil has been added and the distinct litter layer merged with the oxic soil layer as compared with the forest ecosystem in the previous subsection) and more distinct differences in the interactions as outlined above. The interactions of wetland ecosystems are summarised below in a set of eighteen tables, in the standard format adopted in the previous subsections. The principal components and interactions are relevant both to biota dose assessments and to human food-chain pathways involving wild foods. In mathematical modelling, it may be appropriate to pay attention to appropriately representing the vegetation structure (particularly related to the density of trees) to reflect vertical gradients in gaseous carbon (and C-14) concentrations, biomass density, and rates of photosynthesis and plant respiration.

**Table 3-48. Processes of carbon transfer from the geosphere.**

Receiving Component	Processes of Transfer
Geosphere	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas as bubbles or continuous flow.
Surface-water drainage network	Advection or diffusion in the liquid phase across the beds and banks of water bodies. Advection of gaseous forms in bulk gas across the beds and banks of water bodies.
Mineral sub-soil (saturated, anoxic)	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas as bubbles or continuous flow.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	No direct connection.
Humans	Consumption of drinking water abstracted from wells (relatively unlikely in wetlands, however, given easier construction on drier land).

**Table 3-49. Processes of carbon transfer from the surface-water drainage network.**

Receiving Component	Processes of Transfer
Geosphere	Infiltration of water into the beds and banks of water bodies.
Surface-water drainage network	Advection and dispersion in flowing or circulating water. Incorporation and decorporation into solids. Exchange between gaseous and solution phases.
Mineral sub-soil (saturated, anoxic)	Infiltration into the banks.
Catotelm (saturated, anoxic)	Infiltration into the banks and flooding in riparian areas.
Mesotelm (varying degree of anoxia)	No direct connection (as considered physically thin layer, cf. the preceding and following rows in this table).
Acrotelm (oxic)	Flooding in riparian areas.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	As exchange across the surfaces of water bodies.
Animals	Consumption of drinking water abstracted from surface-water bodies.
Humans	Consumption of drinking water abstracted from surface-water bodies.

**Table 3-50. Processes of carbon transfer from the mineral sub-soil.**

Receiving Component	Processes of Transfer
Geosphere	Infiltration and downward diffusion.
Surface-water drainage network	Advection or diffusion in the liquid phase across the beds and banks of water bodies
Mineral sub-soil (saturated, anoxic)	Advection and dispersion of dissolved carbon. Advection of gaseous forms.
Catotelm (saturated, anoxic)	Advection and dispersion of dissolved carbon. Advection of gaseous forms.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	No direct connection.
Soil microbial community	Uptake for use in metabolic processes.
Mycorrhizal associations	Uptake for use in metabolic processes.
Plant roots	Uptake of carbon compounds present in solution by both active and passive processes. Uptake of gaseous forms for onward transmittal via aerenchymae.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	No direct connection.
Humans	Consumption of drinking water abstracted from wells (relatively unlikely in wetlands, however, given easier construction on drier land).

**Table 3-51. Processes of carbon transfer from the catotelm.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Advection or diffusion in the liquid phase across the beds and banks of water bodies. Mechanical collapse associated with pool expansion (erosive action).
Mineral sub-soil (saturated, anoxic)	Advection and dispersion of dissolved carbon. Advection of gaseous forms.
Catotelm (saturated, anoxic)	Diffusion, advection and dispersion of dissolved carbon. Diffusion and advection of gaseous forms.
Mesotelm (varying degree of anoxia)	Advection and dispersion of dissolved carbon. Advection of gaseous forms.
Acrotelm (oxic)	No direct connection.
Soil microbial community	Uptake for use in metabolism. A key process is the production of methane.
Mycorrhizal associations	Uptake for use in metabolic processes.
Plant roots	Uptake of carbon compounds present in solution by both active and passive processes. Uptake of gaseous forms for onward transmittal via aerenchymae.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	No direct connection.
Humans	Peat extraction and use.

**Table 3-52. Processes of carbon transfer from the mesotelm.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Transport by interflow during and after precipitation events.
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	Burial (mediation of matter input from acrotelm). Advection and dispersion, including effects arising from a time-varying position of the water table.
Mesotelm (varying degree of anoxia)	Advection and dispersion, including effects arising from a time-varying position of the water table. Exchanges between soil solution and soil atmosphere. Oxidation and reduction of carbon compounds, with the direction of reaction depending on local characteristics (oxic or anoxic). Mineralisation of organic matter originating from plant and animal material. Bioturbation.
Acrotelm (oxic)	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Bioturbation.
Soil microbial community	Uptake of methane and other organic compounds for use in metabolism. Microbially mediated processes may involve both oxidation and reduction reactions.
Mycorrhizal associations	Uptake of methane and other organic compounds for use in metabolism.
Plant roots	Uptake of carbon compounds present in solution by both active and passive processes. Uptake of gaseous forms for onward transmittal via aerenchymae.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	No direct connection.
Humans	Peat extraction and use.

**Table 3-53. Processes of carbon transfer from the acrotelm.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Mixing with, and transport by runoff and interflow during and after precipitation events.
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	Burial mass flux (raise of water table resulting from added organic matter). Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Bioturbation (that may include also tramping, possibly further yielding in an ebullitive event or compression locally increasing the mass transfer to mesotelm/ catotelm).
Acrotelm (oxic)	Advection and dispersion in solution. Advection and dispersion in the soil atmosphere. Oxidation of organic forms, particularly methane. Mineralisation of organic matter originating from plant and animal material. Mechanical deformation (sloping creep, lateral compaction, wrinkling). Bioturbation.
Soil microbial community	Uptake for use in metabolic processes.
Mycorrhizal associations	Uptake for use in metabolic processes.
Plant roots	Uptake of carbon compounds present in solution by both active and passive processes.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	Gas exchange across the acrotelm surface (diffusive, advective and ebullitive fluxes).
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Uptake for use in metabolic processes (detritivory). Adventitious soil consumption.
Humans	Adventitious soil consumption. Peat extraction and use.

**Table 3-54. Processes of carbon transfer from the soil microbial community.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Mineral sub-soil (saturated, anoxic)	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Catotelm (saturated, anoxic)	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Mesotelm (varying degree of anoxia)	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Acrotelm (oxic)	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Soil microbial community	Exchange of products of metabolism between organisms within the microbial community.
Mycorrhizal associations	No direct connection. Both communities are microbial, but it is reasonable to treat them as only indirectly coupled.
Plant roots	No direct connection.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption.
Humans	No direct connection.

**Table 3-55. Processes of carbon transfer from the mycorrhizal associations.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Mineral sub-soil (saturated, anoxic)	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Catotelm (saturated, anoxic)	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Mesotelm (varying degree of anoxia)	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Acrotelm (oxic)	Release of products of metabolism both in solution and as gaseous forms. Death and decay.
Soil microbial community	No direct connection. Both communities are microbial, but it is reasonable to treat them as only indirectly coupled.
Mycorrhizal associations	Internal transfers between interfaces with the soil and interfaces with plant roots.
Plant roots	Active transfers of products of metabolism.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption (including mushrooms).
Humans	Consumption of mushrooms.

**Table 3-56. Processes of carbon transfer from the plant roots.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Mineral sub-soil (saturated, anoxic)	Root exudation. Death and decay generating organic matter that is subject to mineralisation on various timescales.
Catotelm (saturated, anoxic)	Root exudation. Death and decay generating organic matter that is subject to mineralisation on various timescales.
Mesotelm (varying degree of anoxia)	Root exudation. Death and decay generating organic matter that is subject to mineralisation on various timescales.
Acrotelm (oxic)	Root exudation. Death and decay generating organic matter that is subject to mineralisation on various timescales.
Soil microbial community	No direct connection.
Mycorrhizal associations	Active transfers of products of metabolism.
Plant roots	Transport in solution from locations of absorption. Transport in the gas phase through aerenchyma.
Plant support system for non-tree vegetation	Export to the above-ground parts of the plant in solution and in the gas phase.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	Export to the above-ground parts of the plant in solution and in the gas phase.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption of plant roots.
Humans	Harvest and use of biomass.

**Table 3-57. Processes of carbon transfer from the plant support system for non-tree vegetation.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	Senescence, death and decay generate organic matter that is added to the acrotelm prior to subsequent decay, decomposition and mixing.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	Transport of nutrients in solution and of gas via aerenchymae.
Plant support system for non-tree vegetation	Transport of nutrients in solution and of gas via aerenchymae.
Plant foliage and berries for non-tree vegetation	Transport of nutrients in solution and of gas via aerenchymae.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption of twigs, branches and stems (herbivory).
Humans	Harvest and use of biomass.

**Table 3-58. Processes of carbon transfer from the plant foliage and berries for non-tree vegetation.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Loss of foliage by litterfall resulting in deposition on surface-water bodies (noting that strictly this occurs via the atmosphere).
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	Senescence, death and decay generate organic matter that is added to the acrotelm prior to subsequent decay, decomposition and mixing.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	Transport of nutrients in solution and of gas via aerenchymae.
Plant support system for non-tree vegetation	Transport of nutrients in solution and of gas via aerenchymae.
Plant foliage and berries for non-tree vegetation	Transport of nutrients in solution and of gas via aerenchymae.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	Transfer of carbon dioxide originating from respiration.
Upper canopy atmosphere	Transfer of carbon dioxide originating from respiration (from the upper surfaces of understorey at the boundary of the Upper canopy atmosphere).
Above-canopy atmosphere	No direct connection.
Animals	Consumption of berries and other above-ground plant parts.
Humans	Consumption of berries and other wild plant foods. Less typically, harvest and use of needles/leaves.

**Table 3-59. Processes of carbon transfer from the plant support system for trees.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Loss of twigs and branches by litterfall resulting in deposition on surface-water bodies (noting that strictly this occurs via the atmosphere, and generally is of a minor mass flux compared to that from the Tree foliage).
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	Senescence, death and decay generate organic matter that is added to the acrotelm prior to subsequent decay, decomposition and mixing. (Harvest residues if a treed wetland is under forestry).
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	Transport of nutrients in solution and of gas via aerenchyma.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	Transport of nutrients in solution and of gas via aerenchyma.
Plant foliage for trees (if present)	Transport of nutrients in solution and of gas via aerenchyma.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	No direct connection.
Above-canopy atmosphere	No direct connection.
Animals	Consumption of twigs, branches and stems (herbivory).
Humans	Harvest and use of logs, firewood and biomass.

**Table 3-60. Processes of carbon transfer from the plant foliage for trees.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	Loss of foliage by litterfall resulting in deposition on surface-water bodies (noting that strictly this occurs via the atmosphere).
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	Senescence, death and decay generate organic matter that is added to the acrotelm prior to subsequent decay, decomposition and mixing. (Harvest residues if a treed wetland is under forestry).
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	Transport of nutrients in solution and of gas via aerenchyma.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	Transport of nutrients in solution and of gas via aerenchyma.
Plant foliage for trees (if present)	Transport of nutrients in solution and of gas via aerenchyma.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	Transfer of carbon dioxide originating from respiration.
Above-canopy atmosphere	Transfer of carbon dioxide originating from respiration (from the upper surfaces of foliage at the boundary of the tree canopy atmosphere).
Animals	Consumption of needles and leaves (herbivory).
Humans	Harvest and use of biomass. Less typically, harvest and use of needles/leaves.

**Table 3-61. Processes of carbon transfer from the lower canopy atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	Gas exchange with the soil atmosphere by advection and dispersion, including the effects of pumping due to pressure variations.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	Uptake of carbon dioxide for use in photosynthesis. There may also be uptake of other gases that can be metabolised (e.g. carbon monoxide).
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	Advective and diffusive/dispersive transport resulting in mixing.
Upper canopy atmosphere	Advective and diffusive/dispersive transport.
Above-canopy atmosphere	No direct connection.
Animals	Inhalation.
Humans	Inhalation (typically mainly children; cf. the conceptual model for forests).

**Table 3-62. Processes of carbon transfer from the upper canopy atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	Uptake of carbon dioxide for use in photosynthesis. There may also be uptake of other gases that can be metabolised (e.g. carbon monoxide).
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	Uptake of carbon dioxide for use in photosynthesis. There may also be uptake of other gases that can be metabolised (e.g. carbon monoxide).
Lower canopy atmosphere	Advective and diffusive/dispersive transport.
Upper canopy atmosphere	Advective and diffusive/dispersive transport resulting in mixing.
Above-canopy atmosphere	Advective and diffusive/dispersive transport.
Animals	Inhalation.
Humans	Inhalation.

**Table 3-63. Processes of carbon transfer from the above-canopy atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Surface-water drainage network	As exchange across the surfaces of water bodies.
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	Limited uptake of carbon dioxide for use in photosynthesis at the upper boundary of the Upper canopy atmosphere.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	Limited uptake of carbon dioxide for use in photosynthesis at the upper boundary of the Upper canopy atmosphere.
Lower canopy atmosphere	No direct connection.
Upper canopy atmosphere	Advective and diffusive/dispersive transport.
Above-canopy atmosphere	Advective and diffusive/dispersive transport resulting in mixing. Note that this may include both mixing within a local area (e.g. the release area for C-14) and mixing of local air into the larger-scale atmospheric circulation.
Animals	Inhalation.
Humans	No direct connection.

**Table 3-64. Processes of carbon transfer from animals.**

<b>Receiving Component</b>	<b>Processes of Transfer</b>
Geosphere	No direct connection.
Surface-water drainage network	Excreta may be discharged to surface waters or washed into surface waters in runoff.
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	Excretion (note that may also indirectly promote microbial activity further in the deeper layers as indicated through the other interactions included in this conceptual model). Death and decay of carcasses.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for non-tree vegetation	Excreta may be discharged onto and retained on plant surfaces.
Plant foliage and berries for non-tree vegetation	Excreta may be discharged onto and retained on plant surfaces.
Plant support system for trees (if present)	Excreta may be discharged onto and retained on plant surfaces.
Plant foliage for trees (if present)	Excreta may be discharged onto and retained on plant surfaces.
Lower canopy atmosphere	Respiration and eructation releasing carbon dioxide and methane.
Upper canopy atmosphere	Respiration and eructation releasing carbon dioxide and methane.
Above-canopy atmosphere	Respiration and eructation releasing carbon dioxide and methane.
Animals	Internal redistribution, noting that carbon is relatively uniformly distributed throughout all the soft tissues of the body, but that biological half-lives differ substantially between different tissues and organs. Predation among animal populations.
Humans	Consumption (and other uses) of animal products.

**Table 3-65. Processes of carbon transfer from humans.**

<b>Receiving Component</b>	<b>Processes of Transfer</b>
Geosphere	No direct connection.
Surface-water drainage network	No direct connection.
Mineral sub-soil (saturated, anoxic)	No direct connection.
Catotelm (saturated, anoxic)	No direct connection.
Mesotelm (varying degree of anoxia)	No direct connection.
Acrotelm (oxic)	No direct connection.
Soil microbial community	No direct connection.
Mycorrhizal associations	No direct connection.
Plant roots	No direct connection.
Plant support system for non-tree vegetation	No direct connection.
Plant foliage and berries for non-tree vegetation	No direct connection.
Plant support system for trees (if present)	No direct connection.
Plant foliage for trees (if present)	No direct connection.
Lower canopy atmosphere	Respiration (typically mainly children; cf. the conceptual model for forests).
Upper canopy atmosphere	Respiration.
Above-canopy atmosphere	No direct connection.
Animals	Control of populations through hunting for food and game/ environmental management. Habitat alteration (forest management). These interactions have not been marked in the interaction matrix, however, since they do not directly affect carbon transport modelling (cf. Consumption of animal products from Animals to Humans).
Humans	Internal redistribution, noting that carbon is relatively uniformly distributed throughout all the soft tissues of the body, but that biological half-lives differ substantially between different tissues and organs.

The corresponding interaction matrix is illustrated in Figure 3-11 for which the following colour key applies to reflect dominant processes. It should be noted that not all processes will be relevant to every situation, as indicated with a circular marker in the interaction matrix and the text in the above tables.

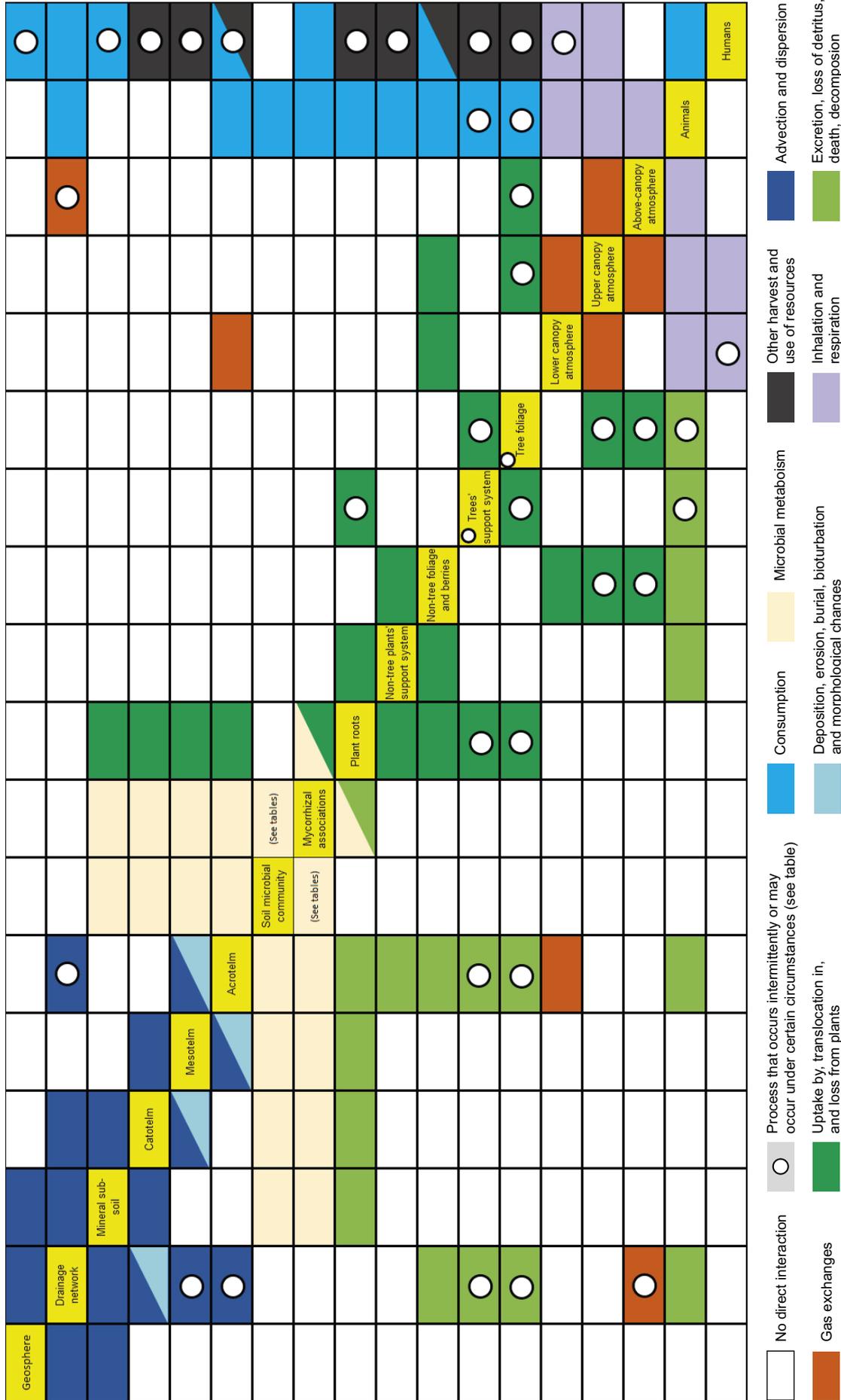


Figure 3-11. Interaction matrix for a wetland.

## 3.8 Conceptual models of transport and accumulation of carbon in estuarine and marine systems

Conceptual models of the transport of carbon in estuaries and coastal marine systems are presented in Sections 3.8.1 and 3.8.2, respectively.

The conceptual model for estuaries is focussed on open estuaries, rather than coastal lagoons (defined as shallow water bodies that are separated from another larger body of water (e.g. the coast) by a narrow barrier) and are generally less influenced by river and tidal flows. This is due to the greater prevalence of estuaries around coastlines as compared with coastal lagoons.

In the case of coastal marine systems, the focus is on foreshore and inshore waters and the interactions between these and the terrestrial-coastal transition zone (e.g. beach) and estuaries (where they are present). Open sea is included only as a sink for carbon in the marine system due to the high level of dilution that would apply to any C-14 entering this system.

### 3.8.1 Estuaries

Estuaries represent diverse and dynamic transitional systems that occur between the surface-water drainage network and the coast and where tidal mixing with seawater becomes an important consideration in modelling. Tidal mixing may be full (i.e. freshwater and seawater are fully mixed during the full period of tidal ingress into the estuary) or limited as a result of transient salinity stratification. This will in part be driven by the tidal range and the bathymetry of the system.

For conceptual model development, a linear system is considered whereby an upstream river flows into an estuary where mixing with coastal waters occurs during tidal ingress. However, it should be noted that systems may be more complex with the confluence of multiple river systems. The dynamics of the system will vary considerably depending on the features of the system (e.g., river flow, extent of tidal inundation, depth etc.).

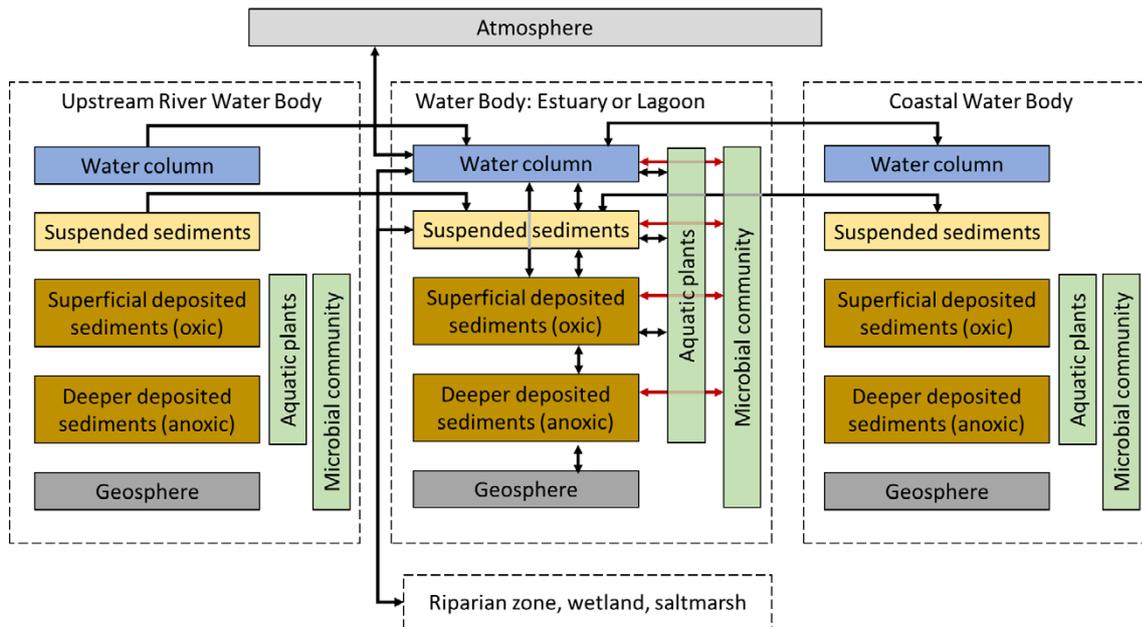
The tidal ingress of coastal waters can have a considerable effect on the overall system, depending upon the dominance of the tidal flow regime and, in some instances (i.e. in macrotidal estuaries), tidal ingress can be considerable, extending several km inland. In such instances it may be appropriate to consider the estuarine system in terms of an upper estuary that is above the limit of saline intrusion but is still subject to the physical influence of tides, a transitional zone that includes the limit of saline intrusion and those areas that are physically influenced by both river flow and tides, and an outer estuary that is of predominantly marine salinity. Further subdivision of the water column may also be appropriate to consider differences between the photic and aphotic zones, depending on the system of interest (e.g., to represent systems with deep water columns).

Consistent with the surface water drainage network, water inputs to estuarine systems comprise flow from the adjacent upstream river, groundwater discharge, overland flow<sup>8</sup>, interflow and direct precipitation, but also include tidal ingress of saline coastal waters (i.e., flood tides). If precipitation water percolates to the water table and reaches the stream segment, this is taken to be indirectly as groundwater discharge or interflow and direct precipitation is then that which falls onto the surface of the water body (stream segment and lake/reservoir, as appropriate). Water outputs comprise (again consistent with the surface water drainage network), groundwater recharge, interflow and overland flow, but also include tidal egress (i.e., ebb tides). Overland flow may occur occasionally (e.g., associated with flooding events driven by high levels of precipitation), or may be more regular because of tidal cycles (e.g., occurring during high spring tides). Overall, there is a net efflux of water from estuaries to the coastal system (considered in Section 2.6.2).

Within an estuary, the principal components are the water bodies, the suspended sediments that they contain, the bottom sediments, and the (macroscopic) aquatic plants. Bottom sediments can be further distinguished as being oxic or anoxic, and, in either case, will include a substantial microbial community. A further distinction can be made between superficial sediments that are more susceptible to resuspension and deeper sediments that are may be less susceptible, depending upon the flow regime. The characteristics of an estuary, in terms of a segment within a broadly linear conceptual model, and its relationship to the wider hydrological system are shown in Figure 3-12.

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<sup>8</sup> Overland flows to and from adjacent terrestrial areas are considered only in the context of processes mediating C-14 transport between estuaries and the terrestrial environment but are not defined as principal components.



**Figure 3-12.** General structure of an estuary in the context of the wider hydrological system. Whilst not illustrated, bi-directional transport of aquatic plants (phytoplankton), microfauna (zooplankton), microbial communities and macrofauna (e.g. fish) may also occur with tidal inundation.

Upstream water bodies will supply carbon as DIC, DOC, PIC and POC, hence the main transport pathways are in the water column and suspended sediments. Coastal waters will also supply carbon as DIC, DOC, PIC and POC, but exchanges will be bi-directional, depending on the state of the tide. Whilst not depicted in Figure 3-12, it should be noted that microflora (i.e., phytoplankton) and microfauna (zooplankton) will be transferred with tides to provide an additional source of carbon to these systems and macrofauna may also migrate to and from estuaries with the tide. Surrounding riparian areas, as well as wetlands and saltmarshes can also be important with respect to the exchange of carbon to and from the system. Exchanges also occur with the local atmosphere. Such exchanges of stable carbon with the atmosphere will be bi-directional, but for C-14 released via groundwater, they are likely to be mainly losses to atmosphere. Within the estuary, chemical and biochemical reactions will couple carbon dissolved in the water column to carbon incorporated in suspended particles. These particles will be coupled to superficial deposited sediments by processes of deposition and resuspension. In turn, superficial sediments will be coupled to deeper sediments by burial and exhumation although the spatial pattern of sediments may be highly dynamic, affecting the presence of benthic macroflora. Consistent with Figure 3-12, deeper sediments are shown as anoxic, but could comprise both oxic and anoxic zones with transitional regions between them. Benthic micro- and macroflora will use carbon from the water column and sediments (mainly from the oxic zone) whereas the microbial community will use carbon under both oxic and anoxic conditions, as described in the context of agricultural systems. Biota other than primary producers are not shown in Figure 3-12, but will comprise both benthic and pelagic organisms plus others, such as birds, that are not confined to the system, but can be particularly abundant in estuaries and may, therefore, be important in the overall carbon cycle in such systems.

Within the model for estuaries, the main elements therefore comprise:

- geosphere,
- atmosphere,
- upstream surface water,
- upstream suspended particulate matter (SPM), inclusive of all organic carbon transported in the water column to the estuary (e.g. organic detritus),

- transition zones (e.g. Riparian zone, salt marshes, mudflats and wetlands<sup>9</sup> etc.),
- water column,
- SPM,
- vegetated oxic deposited sediments,
- unvegetated oxic deposited sediments,
- anoxic deeper deposited sediments,
- macroscopic primary producers (i.e. aquatic plants),
- microbial communities,
- consumers and detritivores,
- coastal water,
- coastal SPM,
- coastal microflora and fauna (e.g. phytoplankton, zooplankton), and
- humans.

Humans are included for dose assessment purposes and the riparian zone, wetlands and saltmarshes are included to allow interactions of the estuary with its immediate terrestrial environment to be represented. The following tables detail the interactions between the principal components. Note that human actions such as estuarine dredging have not been included, but may be important pathways by which carbon could be removed from the system (e.g. where sediments are dredged and disposed inland or to the coast/open sea).

**Table 3-66. Processes of carbon transfer from geosphere.**

Receiving Component	Processes of Transfer
Geosphere	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas as bubbles or continuous flow.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection.
Water column	Advection or diffusion in the liquid phase across the beds and banks of water bodies. Advection of gaseous forms in bulk gas across the beds and banks of water bodies.
SPM	No direct connection.
Vegetated oxic deposited sediment	Advection or diffusion in the liquid phase and/or advection of gaseous forms in bulk gas (in the absence of deeper anoxic deposited sediment).
Unvegetated oxic deposited sediment	Advection or diffusion in the liquid phase and/or advection of gaseous forms in bulk gas (in the absence of deeper anoxic deposited sediment).
Deeper anoxic deposited sediment	Advection or diffusion in the liquid phase and/or advection of gaseous forms in bulk gas.
Primary producers	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Coastal water	No direct connection (focus is on the estuary).
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

<sup>9</sup> Wetlands are considered separately and hence the focus here is on interactions between those wetlands and estuarine systems.

**Table 3-67. Processes of carbon transfer from atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Advection and dispersion of carbon, mainly in gaseous form but potentially also as particulates.
Upstream surface water	No direct connection (only addressing the component of the atmosphere in contact with the estuary).
Upstream SPM	No direct connection.
Transition zones	No direct connection (only addressing the component of the atmosphere in contact with the estuary).
Water column	Gaseous exchange across the surface of the water body. Potential for particulate and liquid aerosol exchange in turbid conditions.
SPM	No direct connection.
Vegetated oxic deposited sediment	No direct connection.
Unvegetated oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Primary producers	Photosynthesis and respiration of emergent vegetation.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Coastal water	No direct connection (only addressing the component of the atmosphere in contact with the estuary).
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	Inhalation.

**Table 3-68. Processes of carbon transfer from upstream surface water.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	Advection and dispersion within flowing water.
Upstream SPM	No direct connection (not modelling transport within the upstream segment).
Transition zones	No direct connection.
Water column	Flow of water into the estuary being modelled.
SPM	No direct connection.
Vegetated oxic deposited sediment	No direct connection.
Unvegetated oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Primary producers	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-69. Processes of carbon transfer from upstream suspended sediment.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection (not modelling transport within the upstream segment).
Upstream SPM	Advection and dispersion within flowing water.
Transition zones	No direct connection.
Water column	No direct connection.
SPM	Flow of upstream surface water into the segment that is being modelled carries suspended sediment with it, which will mix with suspended sediment in the estuary.
Vegetated oxic deposited sediment	No direct connection.
Unvegetated oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Primary producers	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-70. Processes of carbon transfer from riparian zone, saltmarshes, wetlands, mudflats etc. (i.e. transition zones).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Gas exchange.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	Advection and dispersion within the pore water of the beds and banks of the estuary and within sediments and soils of saltmarshes and wetlands. Chemical and biochemical transformations between DIC, DOC, PIC and POC.
Water column	Seepage through banks and sediments/soils and drainage of water following overbanking events and tidal inundation/egress of saltmarshes).
SPM	Release of particles (including organic detritus) from estuary bank and wetland erosion (e.g., from river and/or tidal scouring) and from tidal ingress and egress on saltmarshes. Deposition of plant materials (leaves, pollen etc.).
Vegetated oxic deposited sediment	Changes in estuary morphology converting bank materials to bottom sediments <i>in situ</i> .
Unvegetated oxic deposited sediment	Changes in estuary morphology converting bank materials to bottom sediments <i>in situ</i> .
Deeper anoxic deposited sediment	Changes in estuary morphology converting bank materials to bottom sediments <i>in situ</i> .
Primary producers	No direct connection.
Microbial community	Introduction of new microbial populations by bank/sediment/soil erosion.
Consumers and detritivores	No direct connection.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-71. Processes of carbon transfer from estuary water column.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Degassing across water surface. Potential for particulate and liquid aerosol exchange in turbid conditions.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	Overbanking / inundation during flood events and tidal ingress and infiltration to soils and sediments.
Water column	Advection and dispersion in flow. Chemical transformations / cation competition due to pH and salinity change, potentially affecting DOC/DIC.
SPM	Adsorption and incorporation in particles, e.g. by microbially mediated interactions and chemical transformations / cation competition due to pH and salinity change, potentially resulting in coagulation and flocculation.
Vegetated oxic deposited sediment	Dispersion of flow into the sediments. Adsorption and incorporation in particles.
Unvegetated oxic deposited sediment	Dispersion of flow into the sediments. Adsorption and incorporation in particles.
Deeper anoxic deposited sediment	No direct connection.
Primary producers	Passive and active uptake from solution.
Microbial community	Use of dissolved carbon in microbial metabolism.
Consumers and detritivores	Ingestion of water. Respiration
Coastal water	Advection and dispersion in flow during tidal flow.
Coastal SPM	No direct connection.
Coastal microflora and fauna	Consumption and respiration.
Humans	No direct connection.

**Table 3-72. Processes of carbon transfer from estuary suspended particulate matter.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	Entrapment and deposition during overbanking/tidal inundation. Deposition due to changes in channel morphology.
Water column	Desorption and other release from particles, e.g. by microbially mediated interactions and as a result of changing salinity and pH with saline and freshwater mixing.
SPM	Advection and dispersion in the flow. Coagulation and flocculation of POC/PIC due to saline and freshwater mixing.
Vegetated oxic deposited sediment	Particle settling. Settling of plant organic matter from riparian zone, wetlands and saltmarshes (leaves, twigs, pollen etc.).
Unvegetated oxic deposited sediment	Particle settling. Settling of plant organic matter from riparian zone, wetlands and saltmarshes (leaves, twigs, pollen etc.).
Deeper anoxic deposited sediment	No direct connection.
Primary producers	Particle trapping on surfaces of plants.
Microbial community	Use of particulate carbon in microbial metabolism.
Consumers and detritivores	Ingestion of particulate carbon.
Coastal water	Desorption and other release from particles, e.g. by microbially mediated interactions and as a result of changing salinity and pH with saline and freshwater mixing during tidal ingress.
Coastal SPM	Advection and dispersion in the flow during tidal ingress. Coagulation and flocculation of POC/PIC due to saline and freshwater mixing.
Coastal microflora and fauna	Consumption by zooplankton etc.
Humans	No direct connection.

**Table 3-73. Processes of carbon transfer from vegetated deposited sediment (oxic).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection.
Water column	Leaching of DIC/DOC by infiltrating water. Gas diffusion across the interface.
SPM	Resuspension of particles.
Vegetated oxic deposited sediment	Burial and bioturbation.
Unvegetated oxic deposited sediment	Bed flow.
Deeper anoxic deposited sediment	Burial, bioturbation and diffusion/dispersion across the interface.
Primary producers	Active or passive uptake by plant roots.
Microbial community	Use in microbial metabolism.
Consumers and detritivores	Ingestion of sediment by benthic organisms.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-74. Processes of carbon transfer from unvegetated deposited sediment (oxic).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection.
Water column	Leaching of DIC/DOC by infiltrating water. Gas diffusion across the interface.
SPM	Resuspension of particles.
Vegetated oxic deposited sediment	Bed flow.
Unvegetated oxic deposited sediment	Burial and bioturbation. Bed flow.
Deeper anoxic deposited sediment	Burial, bioturbation and diffusion/dispersion across the interface.
Primary producers	No direct connection.
Microbial community	Use in microbial metabolism.
Consumers and detritivores	Ingestion of sediment by benthic organisms.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-75. Processes of carbon transfer from deeper deposited sediment (anoxic).**

Receiving Component	Processes of Transfer
Geosphere	Diffusion/dispersion across the interface.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection.
Water column	No direct connection.
SPM	No direct connection.
Vegetated oxic deposited sediment	Exhumation, bioturbation and diffusion/dispersion (including ebullition) across the interface.
Unvegetated oxic deposited sediment	Exhumation, bioturbation and diffusion/dispersion (including ebullition) across the interface.
Deeper anoxic deposited sediment	Burial, exhumation and bioturbation.
Primary producers	No direct connection. Plant roots are assumed not to extend significantly into anoxic sediments.
Microbial community	Use in microbial metabolism.
Consumers and detritivores	No direct connection.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-76. Processes of carbon transfer from aquatic plants (macrophytes, phytoplankton and epiplankton).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Respiration by emergent vegetation.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection.
Water column	Release of carbon as gas or in solution.
SPM	Release of plant detritus, e.g. through weathering, loss of plant parts and death (including phytoplankton)
Vegetated oxic deposited sediment	Exudation in solution and release of plant detritus.
Unvegetated oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection. Plant roots are assumed not to extend significantly into anoxic sediments.
Primary producers	Transport and distribution within plants.
Microbial community	Symbiotic associations and exchanges of organic compounds between aquatic plants and the microbial community.
Consumers and detritivores	Ingestion.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	Ingestion of a few special plant types.

**Table 3-77. Processes of carbon transfer from microbial community.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection.
Water column	Release of carbon as gas or in solution.
SPM	Release of microbes as particles of organic matter.
Vegetated oxic deposited sediment	Release of carbon as gas or in solution.
Unvegetated oxic deposited sediment	Release of carbon as gas or in solution.
Deeper anoxic deposited sediment	Release of carbon as gas or in solution.
Primary producers	Symbiotic associations and exchanges of organic compounds.
Microbial community	Microbially mediated oxidation and reduction reactions depending on the geochemical context, biomass production and mineralisation.
Consumers and detritivores	Ingestion.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-78. Processes of carbon transfer from consumers and detritivores.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection.
Water column	Excretion and gas transfer (respiration).
SPM	Excretion, loss of detritus, death and decomposition.
Vegetated oxic deposited sediment	Excretion, loss of detritus, death and decomposition.
Unvegetated oxic deposited sediment	Excretion, loss of detritus, death and decomposition.
Deeper anoxic deposited sediment	No direct connection. Consumers and detritivores are assumed not to be present to a significant extent in anoxic sediments.
Primary producers	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	Internal transport and distribution, biomass production.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	Consumption of animal products.

**Table 3-79. Processes of carbon transfer from coastal water (during tidal inundation).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Degassing from water surface. Potential for particulate and liquid aerosol formation in turbid water.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	Advection and dispersion in saltmarsh sediments, mudflats etc.
Water column	Advection and dispersion in flow. Chemical transformations/cation competition due to pH and salinity change, potentially affecting DOC/DIC.
SPM	Desorption and other release from particles, e.g. by microbially mediated interactions and as a result of changing salinity and pH with saline and freshwater mixing during tidal ingress.
Vegetated oxic deposited sediment	No direct connection.
Unvegetated oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Primary producers	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Coastal water	Advection and dispersion in flow during tidal ingress. Chemical transformations / cation competition due to pH and salinity change, potentially affecting DOC/DIC.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-80. Processes of carbon transfer from coastal suspended particulate matter (during tidal inundation).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection.
Water column	Desorption and other release from particles, e.g. by microbially mediated interactions and as a result of changing salinity and pH with saline and freshwater mixing during tidal ingress.
SPM	Advection and dispersion in the flow.
Vegetated oxic deposited sediment	No direct connection.
Unvegetated oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Primary producers	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Coastal water	No direct connection.
Coastal SPM	Advection and dispersion in the flow. Coagulation and flocculation of POC/PIC due to saline and freshwater mixing.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-81. Processes of carbon transfer from coastal pelagic microflora and microfauna (during tidal inundation).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection.
Water column	Excretion and respiration.
SPM	Excretion, loss of detritus, death and decomposition.
Vegetated oxic deposited sediment	No direct connection.
Unvegetated oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Primary producers	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	Ingestion.
Coastal water	Excretion and respiration.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	No direct connection.

**Table 3-82. Processes of carbon transfer from humans.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Respiration.
Upstream surface water	No direct connection.
Upstream SPM	No direct connection.
Transition zones	No direct connection. Wetlands and terrestrial pathways are addressed separately.
Water column	Excretion (typically via a sewage system).
SPM	Excretion (typically via a sewage system).
Vegetated oxic deposited sediment	No direct connection.
Unvegetated oxic deposited sediment	No direct connection.
Deeper anoxic deposited sediment	No direct connection.
Primary producers	No direct connection.
Microbial community	No direct connection.
Consumers and detritivores	No direct connection.
Coastal water	No direct connection.
Coastal SPM	No direct connection.
Coastal microflora and fauna	No direct connection.
Humans	Internal transport and distribution.

The corresponding interaction matrix is illustrated in Figure 3-13 for which the following colour key applies to reflect dominant processes. It should be noted that not all processes will be relevant to every situation. For example, the type of estuary will determine the extent to which coastal waters interact with transition zones etc. The above tables should be consulted for more detail of the processes considered.

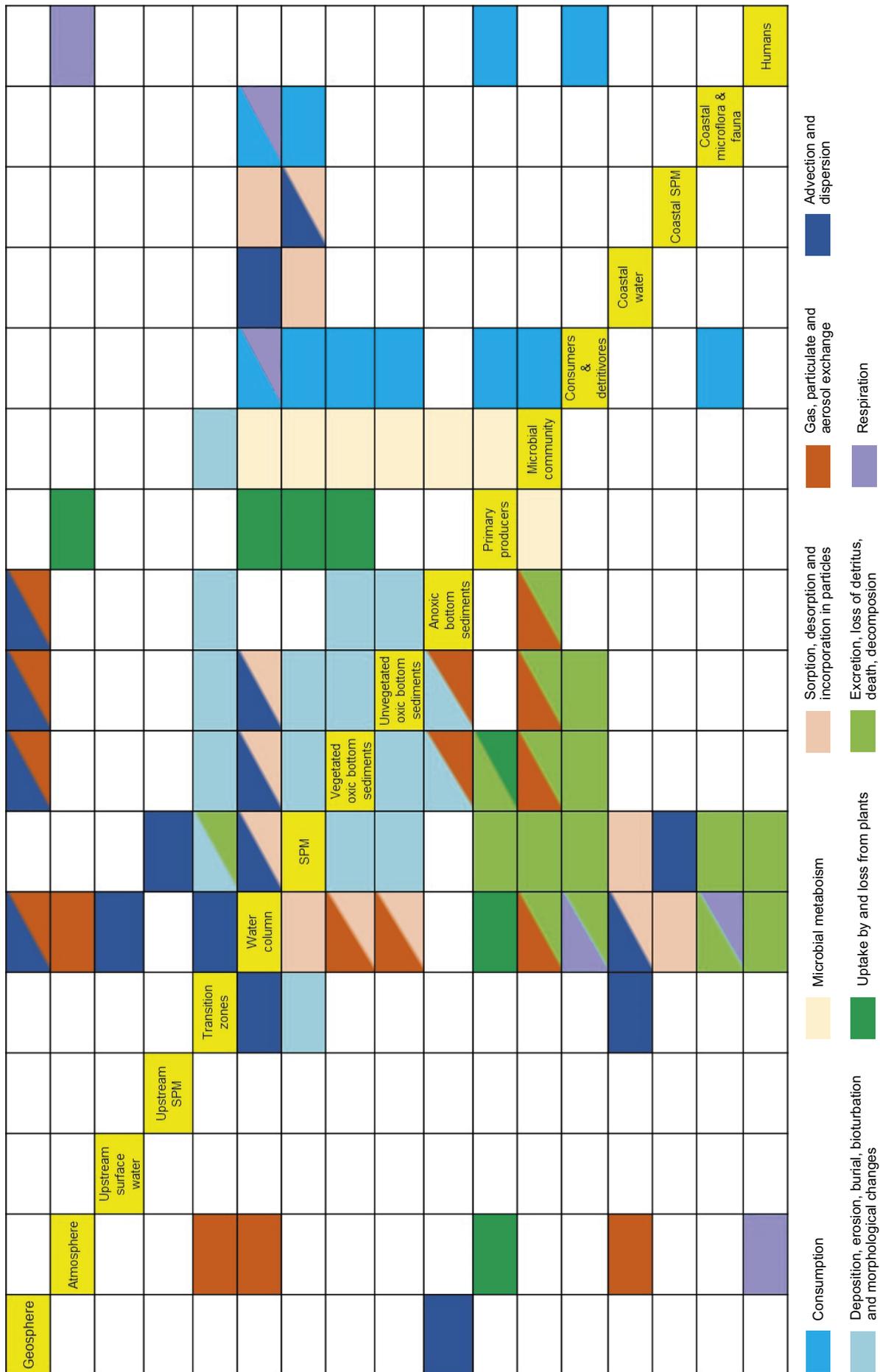


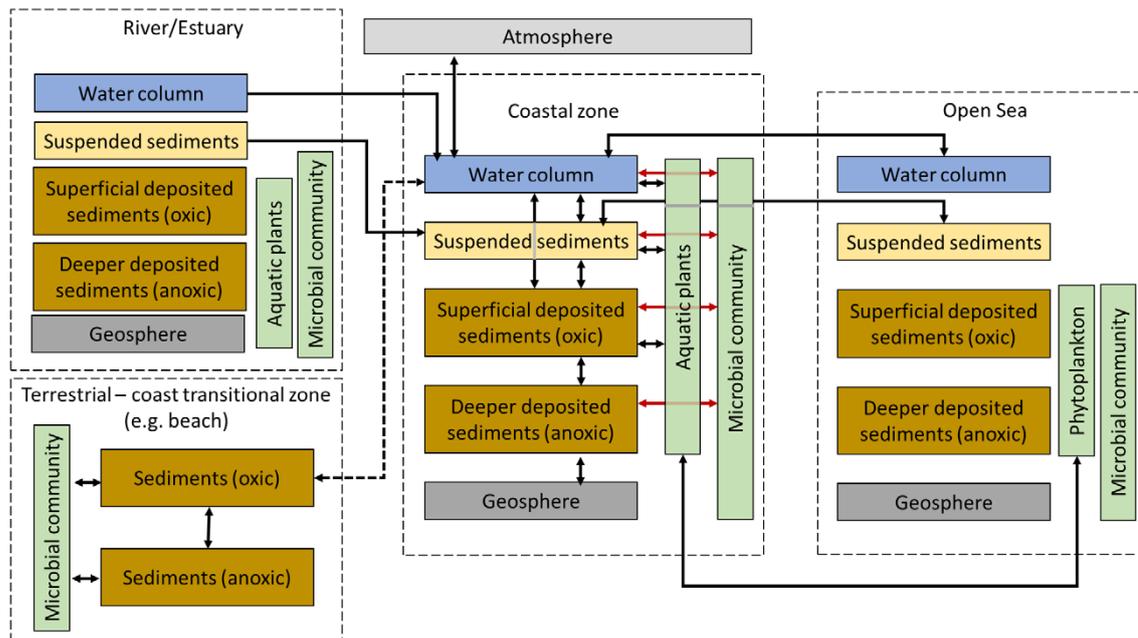
Figure 3-13. Interaction matrix for an estuarine system.

### 3.8.2 Coastal waters

As with estuaries, coastal waters can also be diverse and dynamic systems. The coastal zone can range from an extended shallow foreshore area that is subject to turbulent wave and tidal mixing and where large intertidal areas are exposed during low water, to one where the bathymetry drops off rapidly. Tidal influences and their effect on water-mediated carbon transport will also depend upon the tidal range and whether it is a micro-, meso- or macrotidal system.

The coastal zone may or may not have direct interaction with the surface water drainage system which, depending on the tidal range and bathymetry etc., may include an estuary which may give rise to transient variations in water inputs (freshwater or brackish) and suspended sediment loads. The area influenced by the discharge will vary depending on several factors, including the volume and velocity of the flow, the geomorphology of the system, water circulation (currents), turbulent mixing and differences in temperature and salinity. The action of tides will also influence interactions. For example, estuarine water may have direct interactions with foreshore sediments during the intertidal phase. More discrete input from the terrestrial system may also occur through precipitation and run-off across the transitional zone of the coastline (e.g. beach). In addition, advection and dispersion by currents will transport water along the coast and exchange with the open ocean will occur.

The principal components of coastal systems therefore comprise the water bodies (that may be subdivided into inshore and offshore photic and aphotic zones), suspended sediments, oxic and anoxic coastal sediments, the foreshore (encompassing the intertidal zone) and the back beach that represents the intermediate zone between the coastal and terrestrial systems. Potential characteristics of a coastal zone are illustrated in Figure 3-14.



**Figure 3-14.** General structure of a coastal zone. Note that water and sediment flux from a river/estuary is shown as unidirectional to represent the net efflux from the system but will be bi-directional for estuaries and may also include bidirectional transport of aquatic plants (phytoplankton), microfauna (zooplankton) and microbial communities. Dashed arrow illustrates sea-to-land transfer (sea spray and/or flooding).

Rivers and estuaries discharging to the coastal zone will supply carbon as DIC, DOC, PIC and POC, with water and suspended sediments being the main transport pathways, noting that for estuaries the transport may be bidirectional. Particulate carbon may also enter the coastal water column from the terrestrial-coast transition zone (e.g. beach) through wind blow and dissolved and particulate carbon can be transported as a result of surface water run-off and infiltration and seepage as a result of precipitation and/or coastal flooding events. Such events will also lead to the transport of dissolved and particulate carbon from the coastal zone to the transitional zone. Additional exchanges will occur with the local atmosphere and with the open sea via the water column, suspended sediments and the bidirectional transport of phytoplankton, zooplankton, microbial communities and both benthic and pelagic fauna (not illustrated in Figure 3-14). Within the water column, carbon can be sequestered into organic matter through photosynthesis in the photic zone (both by phytoplankton and macrophytes where the photic zone extends sufficiently close to bottom sediments) and the action of microbial communities and filter-feeding micro- and macrofauna in both photic and aphotic zones with excretion, death and decay then returning carbon back to the water column and/or to sediments. Chemical and biochemical reactions will couple carbon dissolved in the water column to carbon incorporated in suspended particles. Deposition and resuspension will couple particles to the superficial deposited sediments. Superficial sediments will then be coupled to deeper sediments (shown as anoxic but may comprise both oxic and anoxic zones with transitional regions between) by burial and exhumation. Microbial communities in both oxic and anoxic sediments can also metabolise organic matter to release carbon dioxide and methane and provide biomass that may be consumed by benthic micro and macrofauna.

Whilst the coastal zone is depicted as a single component in Figure 3-14, it may be appropriate to subdivide this zone into foreshore (encompassing the intertidal zone that may be subject to mixing throughout the depth of the water column with wave and tidal action giving rise to turbulent mixing zone with sediment resuspension and redistribution) and offshore coastal area.

Within the model for the coastal zone, the main elements therefore comprise:

- geosphere,
- atmosphere,
- estuary/river surface water,
- estuary/river suspended particulate matter (SPM),
- terrestrial-coastal transition zone (e.g. beach),
- foreshore water column,
- foreshore SPM,
- foreshore bottom sediments (oxic),
- foreshore deeper bottom sediments (anoxic),
- offshore water column (photic),
- offshore water column (aphotic),
- offshore SPM,
- offshore oxic bottom sediments,
- offshore deeper anoxic bottom sediments,
- microbial communities,
- (macroscopic) primary producers (i.e. aquatic plants),
- consumers and detritivores,
- open sea, and
- humans.

The terrestrial-coastal transition zone is included to provide a link between the coastal zone and with the terrestrial environment and to allow linkages between these two systems to be represented (in addition to the linkage provided by estuaries/rivers). The open sea is also included as an ultimate sink for carbon. Interactions between the open sea and the offshore area will include the exchange of water and SPM which can settle to the bottom sediments. Consumers and detritivores may also move between the two regions. The foreshore is taken to be the intertidal zone between the high and low water marks. As with previous sections, humans are included for dose assessment purposes.

The following tables detail the interactions between the principal components detailed above. Consistent with Section 3.8.1, human activities that may affect the movement of carbon between components (e.g., dredging of estuarine sediments with subsequent release to banks/coastal zone etc. have not been explicitly detailed but may be relevant in a specific assessment context. Movement of carbon from the coastal zone to the transitional zone by macrofauna has also not been explicitly included (e.g. feeding by wading birds from the intertidal zone) as this is likely to be a minor pathway, but could be an important pathway if considering the exposure of non-human biota.

**Table 3-83. Processes of carbon transfer from geosphere.**

Receiving Component	Processes of Transfer
Geosphere	Advection and dispersion of dissolved carbon. Advection of gaseous forms in bulk gas as bubbles or continuous flow.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	Groundwater upwelling with advection or diffusion in the liquid phase to bottom sediments etc. and/or advection of gaseous forms in bulk gas.
Foreshore water column	No direct connection.
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	Advection and diffusion in the liquid phase. Advection of gaseous forms in bulk gas.
Offshore water column (photic)	No direct connection.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	Advection and diffusion in the liquid phase. Advection of gaseous forms in bulk gas.
Primary producers	No direct connection.
Microbial communities	No direct connection.
Consumers and detritivores	No direct connection.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-84. Processes of carbon transfer from atmosphere.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Advection and dispersion of carbon, mainly in gaseous form but also as particulates and aerosols.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	Gaseous exchange across the surface-atmosphere interface. Deposition of aerosols and particulates.
Foreshore water column	Gaseous exchange across the surface of the water body. Deposition of particles (e.g. transported from the terrestrial-coastal transition zone).
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	Gaseous exchange across the surface of the water body. Deposition of particles (e.g. transported from the terrestrial-coastal transition zone).
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	No direct connection.
Microbial communities	No direct connection.
Consumers and detritivores	Inhalation (minor pathway).
Open sea	No direct connection (focus is on the coastal zone).
Humans	Inhalation.

**Table 3-85. Processes of carbon transfer from estuary water.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Estuary surface water	Advection and dispersion in flowing water.
Estuary SPM	No direct connection (not modelling transport in the upstream section).
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	Flow of water into segment being modelled. Chemical transformations/cation competition due to pH and salinity change, potentially affecting DOC/DIC.
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	Flow of water across intertidal sediments during low tide and dispersion into the sediments and adsorption and incorporation in particles.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	No direct connection.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	No direct connection.
Microbial communities	No direct connection.
Consumers and detritivores	No direct connection.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-86. Processes of carbon transfer from estuary suspended particulate matter.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	Advection and dispersion in flowing water.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	No direct connection.
Foreshore SPM	Flow of upstream surface water into the segment that is being modelled carries SPM with it. Advection and dispersion in the flow. Coagulation and flocculation of POC/PIC due to saline and freshwater mixing.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	No direct connection.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	No direct connection.
Microbial communities	No direct connection.
Consumers and detritivores	No direct connection.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-87. Processes of carbon transfer from terrestrial-coastal transition zone.**

Receiving Component	Processes of Transfer
Geosphere	Diffusion / dispersion across the interface.
Atmosphere	Gaseous exchange. Particle suspension due to wind action.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	Advection and dispersion within sediment pore water. Chemical and biochemical transformations between DIC, DOC, PIC and POC.
Foreshore water column	Drainage following tidal flooding events.
Foreshore SPM	Erosion of particulates during tidal flooding events.
Foreshore bottom sediments (oxic)	Changes in coastal morphology. Sea level rise.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	No direct connection.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	No direct connection.
Microbial communities	No direct connection.
Consumers and detritivores	No direct connection.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-88. Processes of carbon transfer from foreshore water column.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Degassing from water surface. Aerosols/foam generated by wave action.
Estuary surface water	No direct connection (not modelling transport in the upstream section in this context).
Estuary SPM	No direct connection (not modelling transport in the upstream section in this context).
Terrestrial-coastal transition zone	Infiltration during tidal flooding events.
Foreshore water column	Advection and dispersion. Chemical transformation (carbon dioxide to bicarbonate).
Foreshore SPM	Adsorption and incorporation in particles.
Foreshore bottom sediments (oxic)	Dispersion of flow into sediments.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	Advection and dispersion.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	Uptake from solution and incorporation via photosynthesis.
Microbial communities	Use of dissolved carbon in microbial metabolism.
Consumers and detritivores	Ingestion of water, gas exchange (respiration).
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-89. Processes of carbon transfer from foreshore suspended particulate matter.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Release of small particulates in aerosols by wave action.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection (not modelling transport in the estuary in this context).
Terrestrial-coastal transition zone	Entrapment and deposition during tidal flooding events. Deposition due to changes in coastline, including sea level rise.
Foreshore water column	Desorption and other release from particles, e.g. by microbially mediated interactions.
Foreshore SPM	Advection and dispersion. Coagulation and flocculation of POC/PIC.
Foreshore bottom sediments (oxic)	Flocculation and deposition of particles.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	No direct connection.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	Advection and dispersion.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	Particle trapping on surfaces of macroalgae (minor process).
Microbial communities	Use of particulate carbon in microbial metabolism.
Consumers and detritivores	Ingestion.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-90. Processes of carbon transfer from foreshore bottom sediments (oxic).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Gas exchange when sediments exposed.
Estuary surface water	Leaching by infiltrating water and gas exchange during intertidal phase.
Estuary SPM	Scouring and resuspension and scouring when sediments exposed.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	Advection and dispersion and gas diffusion across the interface.
Foreshore SPM	Erosion and resuspension of particles.
Foreshore bottom sediments (oxic)	Bed transport. Burial and bioturbation.
Foreshore deeper bottom sediments (anoxic)	Burial and bioturbation.
Offshore water column (photic)	No direct connection.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	No direct connection.
Microbial communities	Use in microbial metabolism.
Consumers and detritivores	Ingestion of particulate carbon.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-91. Processes of carbon transfer from foreshore deeper bottom sediments (anoxic).**

Receiving Component	Processes of Transfer
Geosphere	Diffusion/dispersion across the interface.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	No direct connection.
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	Exhumation, bioturbation and diffusion/dispersion across the interface.
Foreshore deeper bottom sediments (anoxic)	Burial, exhumation and bioturbation.
Offshore water column (photic)	No direct connection.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	No direct connection.
Microbial communities	Use in microbial metabolism.
Consumers and detritivores	No direct connection.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-92. Processes of carbon transfer from offshore water column (photic).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Degassing from water surface. Aerosols/foam generated by wave action.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	Advection and dispersion.
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	Advection and dispersion. Chemical transformation (carbon dioxide to bicarbonate).
Offshore water column (aphotic)	Advection and diffusion.
Offshore SPM	Adsorption and incorporation in particles.
Offshore oxic bottom sediments	Dispersion into sediments (in absence of aphotic water column).
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	Uptake from solution and incorporation via photosynthesis.
Microbial communities	Use of dissolved carbon in microbial metabolism.
Consumers and detritivores	Ingestion of water, gas exchange.
Open sea	Water exchange.
Humans	No direct connection.

**Table 3-93. Processes of carbon transfer from offshore water column (aphotic).**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	No direct connection.
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	Water exchange.
Offshore water column (aphotic)	Advection and dispersion.
Offshore SPM	Adsorption and incorporation in particles.
Offshore oxic bottom sediments	Dispersion into sediments
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	No direct connection.
Microbial communities	Use of dissolved carbon in microbial metabolism.
Consumers and detritivores	Ingestion of water, gas exchange.
Open sea	Water exchange.
Humans	No direct connection.

**Table 3-94. Processes of carbon transfer from offshore SPM.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	No direct connection.
Foreshore SPM	Water exchange and advection/dispersion.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	Desorption and other release from particles, e.g. by microbially mediated interactions
Offshore water column (aphotic)	Desorption and other release from particles, e.g. by microbially mediated interactions
Offshore SPM	Advection and dispersion.
Offshore oxic bottom sediments	Particle settling.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	No direct connection.
Microbial communities	Use of particulate carbon in microbial metabolism.
Consumers and detritivores	Ingestion of particulate carbon.
Open sea water column	Advection/dispersion with open sea SPM
Humans	No direct connection.

**Table 3-95. Processes of carbon transfer from offshore oxic bottom sediments.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	No direct connection.
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	In absence of aphotic zone: Leaching of DIC/DOC by infiltrating water. Gas diffusion across the interface.
Offshore water column (aphotic)	Leaching of DIC/DOC by infiltrating water. Gas diffusion across the interface.
Offshore SPM	Resuspension of particles.
Offshore oxic bottom sediments	Burial and bioturbation.
Offshore deeper anoxic bottom sediments	Burial, bioturbation and diffusion/dispersion across the interface.
Primary producers	No direct connection.
Microbial communities	Use in microbial metabolism.
Consumers and detritivores	Ingestion of particulate carbon.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-96. Processes of carbon transfer from offshore deeper anoxic bottom sediments.**

Receiving Component	Processes of Transfer
Geosphere	Diffusion/dispersion across the interface.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	No direct connection.
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	No direct connection.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	Exhumation, bioturbation and diffusion/dispersion across the interface.
Offshore deeper anoxic bottom sediments	Burial, exhumation and bioturbation.
Primary producers	No direct connection.
Microbial communities	Use in microbial metabolism.
Consumers and detritivores	No direct connection.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-97. Processes of carbon transfer from microbial communities.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	Release of carbon as gas or in solution, death and decomposition.
Foreshore water column	Release of carbon as gas or in solution.
Foreshore SPM	Adhesion to particles.
Foreshore bottom sediments (oxic)	Release of carbon as gas or in solution, death and decomposition.
Foreshore deeper bottom sediments (anoxic)	Release of carbon as gas or in solution, death and decomposition.
Offshore water column (photic)	Release of carbon as gas or in solution.
Offshore water column (aphotic)	Release of carbon as gas or in solution.
Offshore SPM	Adhesion to particles.
Offshore oxic bottom sediments	Release of carbon as gas or in solution.
Offshore deeper anoxic bottom sediments	Release of carbon as gas or in solution.
Primary producers	No direct connection.
Microbial communities	Microbially mediated oxidation and reduction reactions depending on the geochemical context, biomass production and mineralisation.
Consumers and detritivores	Ingestion.
Open sea	No direct connection.
Humans	No direct connection.

**Table 3-98. Processes of carbon transfer from primary producers.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Potential for gas exchange during intertidal phase (e.g. rocky shore coastlines)
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	Release of carbon as gas or in solution.
Foreshore SPM	Release of plant detritus, e.g. through weathering, loss of plant parts and death (including phytoplankton)
Foreshore bottom sediments (oxic)	Release of plant detritus, e.g. through weathering, loss of plant parts and death/decomposition and burial.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	Release of carbon as gas or in solution (phytoplankton).
Offshore water column (aphotic)	No direct connection.
Offshore SPM	Release as particles of organic matter (phytoplankton).
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	Transport and distribution in plants (macrophytes). Advection and dispersion (phytoplankton)
Microbial communities	No direct connection.
Consumers and detritivores	Ingestion.
Open sea	Advection and dispersion (phytoplankton)
Humans	Ingestion (seaweed).

**Table 3-99. Processes of carbon transfer from consumers and detritivores.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	Excretion and gas exchange.
Foreshore SPM	Excretion, loss of detritus, death and decomposition.
Foreshore bottom sediments (oxic)	Excretion, loss of detritus, death and decomposition.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	Excretion and gas transfer (respiration).
Offshore water column (aphotic)	Excretion and gas transfer (respiration).
Offshore SPM	Excretion, loss of detritus, death and decomposition.
Offshore oxic bottom sediments	Excretion, loss of detritus, death and decomposition.
Offshore deeper anoxic bottom sediments	Excretion, loss of detritus, death and decomposition.
Primary producers	No direct connection.
Microbial communities	No direct connection.
Consumers and detritivores	Internal transport and distribution, biomass production.
Open sea	Migration
Humans	Ingestion.

**Table 3-100. Processes of carbon transfer from open sea.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	No direct connection.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	No direct connection.
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	Water exchange.
Offshore water column (aphotic)	Water exchange.
Offshore SPM	Advection and dispersion of SPM
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Primary producers	Advection and dispersion of phytoplankton
Microbial communities	No direct connection.
Consumers and detritivores	Migration
Open sea	Advection and dispersion. Chemical transformation (carbon dioxide to bicarbonate).
Humans	No direct connection.

**Table 3-101. Processes of carbon transfer from humans.**

Receiving Component	Processes of Transfer
Geosphere	No direct connection.
Atmosphere	Respiration.
Estuary surface water	No direct connection.
Estuary SPM	No direct connection.
Terrestrial-coastal transition zone	No direct connection.
Foreshore water column	Excretion (typically via a sewage system).
Foreshore SPM	No direct connection.
Foreshore bottom sediments (oxic)	No direct connection.
Foreshore deeper bottom sediments (anoxic)	No direct connection.
Offshore water column (photic)	No direct connection.
Offshore water column (aphotic)	No direct connection.
Offshore SPM	No direct connection.
Offshore oxic bottom sediments	No direct connection.
Offshore deeper anoxic bottom sediments	No direct connection.
Offshore primary producers	No direct connection.
Offshore microbial communities	No direct connection.
Offshore consumers and detritivores	No direct connection.
Open sea	No direct connection.
Humans	Internal transport and distribution.

The corresponding interaction matrix is shown in Figure 3-15, for which the following colour key applies to reflect dominant processes. It should be noted that not all processes will be relevant to every situation. For example, the type of shoreline will affect carbon transfers from the transition zone. Furthermore, some processes may only occur intermittently (e.g. during the intertidal phase). The above tables should be consulted for more detail of the processes considered.

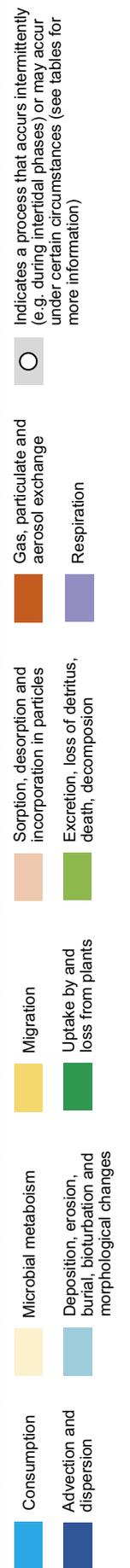
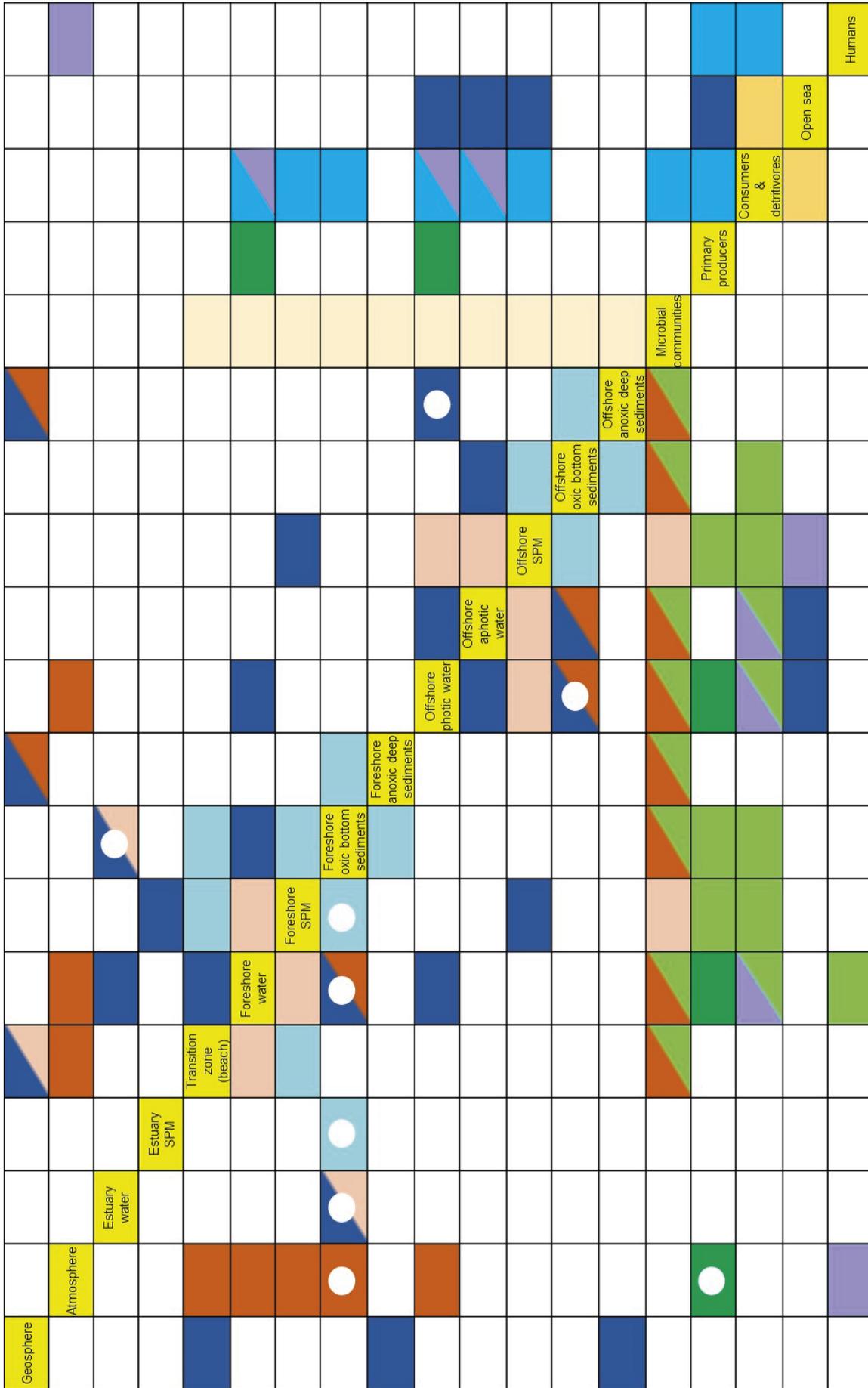


Figure 3-15. Interaction matrix for coastal waters.

### 3.9 Conclusions from conceptual modelling

Subsections 3.3 to 3.8 present site-generic conceptual models for the transport and distribution of carbon and C-14 in the environment. These models demonstrate how the overall biogeochemical carbon cycle can be reconceptualised in relation to specific types of environments, and how conceptual models of those types of environments can be developed that are well-adapted to application in modelling the radiological impacts of discharges of C-14.

The methodology follows the approach and terminology in the original BIOMASS reference biospheres methodology in terms of organising and using information for conceptual model development (IAEA 2003) as reviewed and enhanced in the IAEA's MODARIA II programme (see Lindborg et al. 2022).

For each environment:

- a list of principal components is defined,
- a set of tables is developed setting out explicitly the processes resulting in carbon fluxes between the principal components,
- an interaction matrix is developed in which the principal components are shown on the lead diagonal and the processes linking those components are displayed using a simple colour coding scheme to characterise processes into broad classes, and
- the interaction matrix is examined to identify connected regions that could be treated as sub-models that could be loosely coupled to other sub-models.

It is recognised that carbon transfers also occur between the various types of environments. This is reflected by the inclusion of interfaces between the different environments, in the interaction matrices. These interfaces may be indicated by inclusion of the same part of two ecosystems in the interaction matrices of the two systems. This occurs with riparian zones. Alternatively, the interaction can be represented by inclusion of an external element, such as the atmosphere, which can be both a source and sink in several environments and thus mediate carbon transfers between them. Thus, once conceptual models for the individual ecosystems have been formulated, it is relatively straightforward to address their combination without developing a composite interaction matrix for the larger, multi-ecosystem case. Also, if mathematical models for each ecosystem are developed separately, an integrated model can be formulated by requiring inputs and outputs of carbon to be matched across the identified boundaries between the conceptual models of the various ecosystems.

Not addressed in these sections is the next stage in modelling. Typically, this would be to take a sub-model of relevance and apply it in a specific context, decomposing the principal elements further, as appropriate, and then transforming the expanded sub-model into a mathematical model using methods similar to those outlined by Klos and Thorne (2020). This process is illustrated for a terrestrial agricultural catchment in the following chapter.

## 4 Mathematical model development

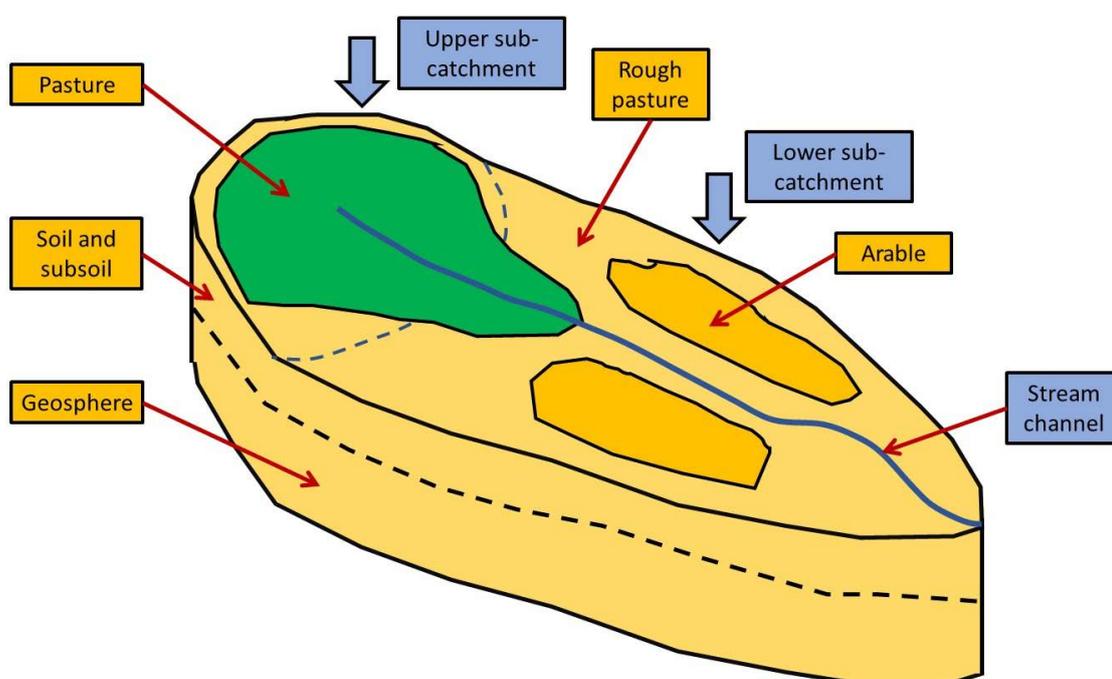
### 4.1 Introduction

In the current BIOPROTA study, consideration has been given to the transport and distribution of C-14 in terrestrial, freshwater and marine environments following its discharge into those environments in effluents from operating facilities but also, particularly, because of possible releases from disposal facilities for solid radioactive wastes, which are projected to occur over longer timeframes.

This chapter makes use of two of the conceptual models described in Chapter 3 to propose a general approach to mathematical model development. It presents the overall structure of the proposed model and develops the associated mathematical structure and equations.

### 4.2 Overall model conceptualisation

Because of the scale of the problem, it has not been possible to address all the different types of environments considered in the development of conceptual models reported in Chapter 3. Therefore, due to their general significance in terms of radiological impact from C-14, it was decided to focus on agricultural ecosystems and, specifically, their links with the surface water drainage network. To this end, the system to be addressed was defined as a lowland, terrestrial catchment used for arable crops and pasture and drained by a single stream channel. To facilitate studies of lateral transport, as appropriate the catchment was distinguished into a sub-catchment located in the upper part of the model domain and draining into the main part of the overall catchment. In an actual catchment, sub-catchments would typically be defined where one or more tributary streams flow into a single stream or river draining the overall catchment. However, in this stylised example, a single stream is adopted, and the catchment is arbitrarily distinguished into an area draining to the upper part of the stream and an area draining to the lower part. Different types of land use are included to illustrate how they may interact, but it is emphasised that their distribution has been selected arbitrarily. The schematic geometry of the system is illustrated in Figure 4-1.



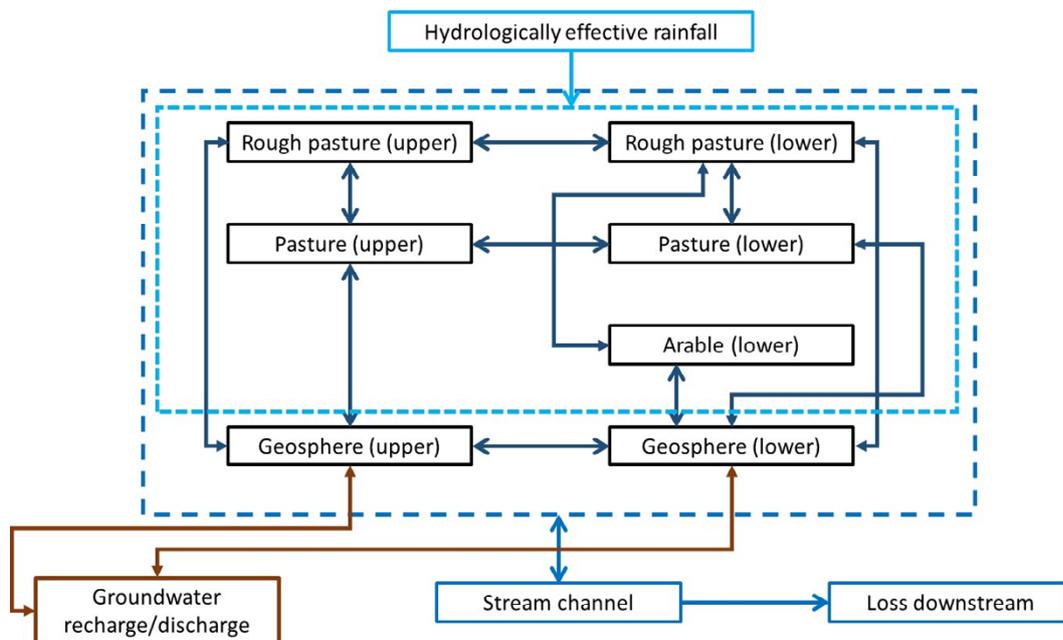
**Figure 4-1.** Illustrative catchment geometry. The broken blue line represents the boundary of the upper sub-catchment, and the broken black line represents the interface between the biosphere and the geosphere, often termed to geosphere-biosphere interface.

### 4.3 Hydrological modelling

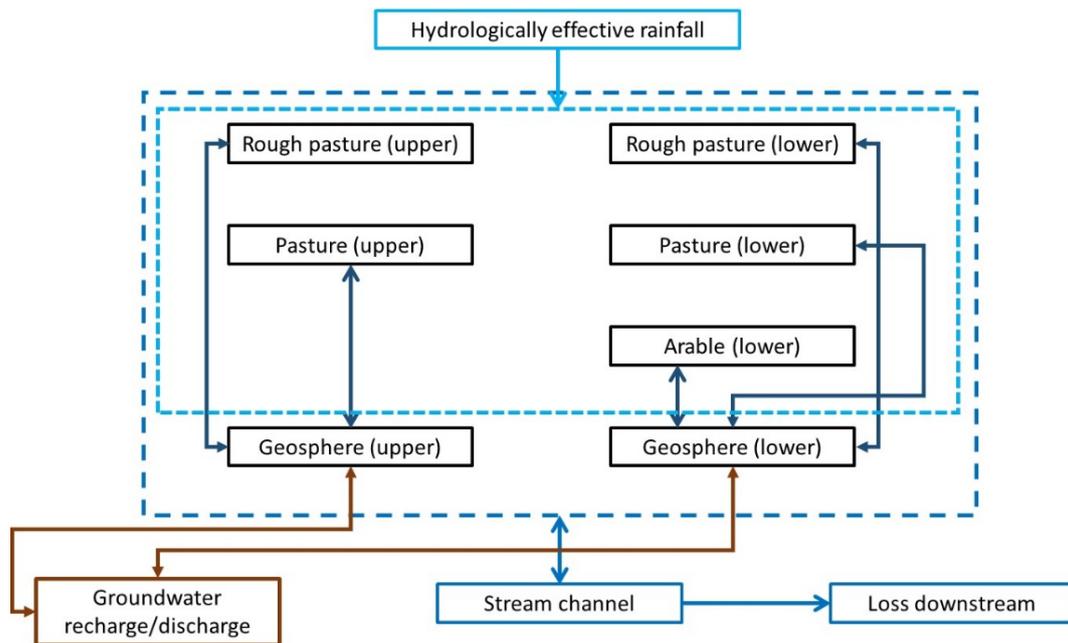
By considerations of symmetry, it is sufficient to model half of this catchment, considering vertical planes below the stream channel and at the interfluves of the main catchment to be no-flow boundaries. The boundary at the base of the catchment may be taken as a prescribed water flux (which may be spatially variable over the base) and the boundary at the top of the catchment may also be taken as a prescribed water flux (the hydrologically effective rainfall, HER, i.e. precipitation minus actual evapotranspiration). Thus, the water balance of the system is as illustrated schematically in Figure 4-2.

The no-flow boundaries limit inputs from upslope and losses downslope. Therefore, inputs of HER plus or minus a contribution from groundwater recharge or discharge is balanced against losses downstream in the stream channel. To simplify the modelling further, lateral transfers between the upper sub-catchment and the lower sub-catchment are neglected, leading to the simplified model shown in Figure 4-3. Such lateral transfers could be reintroduced at a later stage of model development, if preliminary results and other factors, e.g. data arising from site characterisation, suggest that this is appropriate.

Note that the HER is distributed over all the pasture and arable components of the catchment. In contrast, groundwater recharge/discharge exchanges only with the geosphere in the upper and lower sub-catchments. If the stream channel is incised into the geosphere, it can exchange water with all the soil/subsoil components (except for arable land, which is taken to be upslope of the channel banks and separated from the channel by rough pasture in the riparian zone) and with the groundwater in the upper and lower geosphere components of the modelled system.



**Figure 4-2.** Water balance for the illustrative catchment. Hydrologically effective rainfall is defined per unit area and is multiplied by the area of each type of land use identified in the inner dashed blue line box to give the total water input used in water balance calculations. The geosphere, as defined in Figure 4-1, is here distinguished into components associated with the upper and lower catchments, because flows across the geosphere-biosphere interface will differ between these two areas.

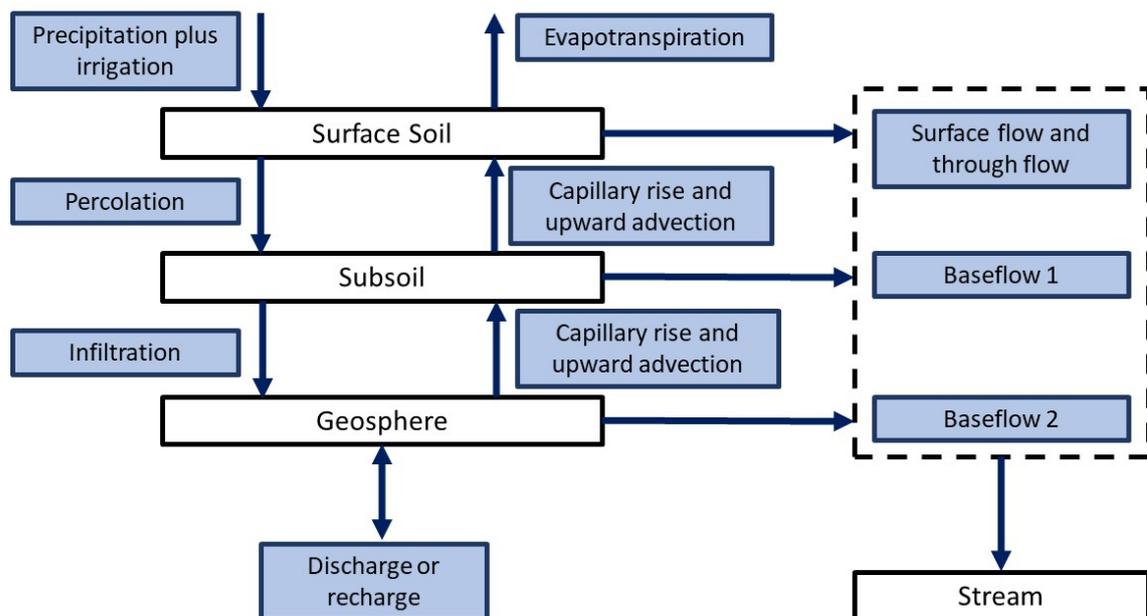


**Figure 4-3.** Simplified water balance for the illustrative catchment.

It is proposed that water flows included in the model should be taken from a simplified water-balance approach. This is illustrated in Figure 4-4.

In this model, a well-drained soil is assumed to be present and there are no flows from upslope. Similarly, there are no flows downslope to other terrestrial areas, so all such flows are directed to the stream. However, in the case of arable land, there is no direct connection with the stream, so all downslope flows are set to zero, requiring the water balance to be satisfied by geosphere recharge (adopting the case that precipitation plus irrigation exceeds evapotranspiration).

The various flows used in this model can be taken from reports such as: Institute of Hydrology (1995).



**Figure 4-4.** Simplified hydrological model of soil and subsoil applicable to both pasture and arable land. Evapotranspiration is shown from surface soil only, but there may be a contribution arising from root uptake of water in the subsoil.

For modelling, it is necessary to define appropriate areas for the upper and lower sub-catchments. Although this report does not go as far as developing a specific mathematical model, it may be useful to note that, typically, gauged surface-water catchments in the UK have areas of a few square kilometres. With HER values of around  $0.3 \text{ m a}^{-1}$  and neglecting groundwater discharge/recharge, this implies stream flow rates of about  $1 \times 10^6 \text{ m}^3 \text{ a}^{-1}$ , which is in line with observations.

### 4.4 Modelling of carbon transport

The above discussion relates only to the hydrological characterisation of the model domain. However, this needs to be overlaid by a representation of carbon transport. For this, consideration needs to be given to the interaction matrices for agricultural and surface-water ecosystems developed under this project. These are illustrated in Figure 3-6 and Figure 3-9.

In respect of the surface-drainage network, it is appropriate also to consider the structural form of the conceptual model adopted. This is illustrated in Figure 4-5.

Overall, based on these figures, the conceptual model for carbon transport in the agricultural catchment under consideration needs to include the principal components listed in Table 4-1.

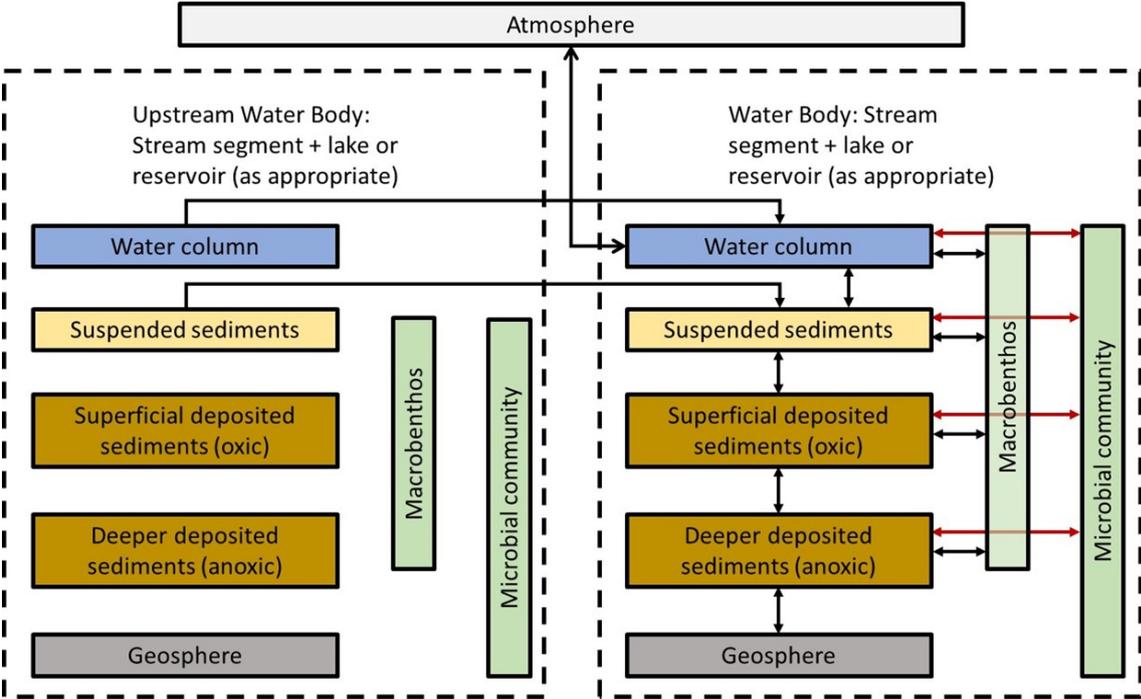


Figure 4-5. Structure of the conceptual model of carbon transport in the surface-drainage network (internal fluxes in the upstream water body have been omitted).

**Table 4-1. Principal components of a carbon transport model. Each major component is distinguished into sub-components and each sub-component is then distinguished into the listed items. Thus, for example, 'Pasture' is a sub-component of 'Lower sub-catchment' and is distinguished into 11 distinct items.**

Major component	Sub-components	Items in each sub-component
Above-canopy atmosphere	None	Not applicable
Upper sub-catchment	Rough pasture	Upper canopy atmosphere
	Pasture	Lower canopy atmosphere
		Unsaturated oxic soil
		Capillary fringe
		Saturated anoxic soil
		Soil microbial community
		Mycorrhizal associations
		Plant roots
		Plant support system
		Plant foliage
Domesticated animals		
Lower sub-catchment	Rough pasture	Upper canopy atmosphere
	Pasture	Lower canopy atmosphere
	Arable	Unsaturated oxic soil
		Capillary fringe
		Saturated anoxic soil
		Soil microbial community
		Mycorrhizal associations
		Plant roots
		Plant support system
		Plant foliage
Domesticated animals		
Surface drainage network	Upper sub-catchment	Water column
	Lower sub-catchment	Suspended sediments
		Superficial oxic deposited sediment
		Deep oxic deposited sediment
		Deeper anoxic deposited sediment
		Macrobenthos (macrophytes)
		Microbial community
Consumers and detritivores		
Geosphere	Upper sub-catchment	None
	Lower sub-catchment	None
Humans	None	Not applicable

For developing a mathematical model, some simplifications can be introduced. Specifically:

- The carbon content of plants can be treated as a single pool. Thus, plant roots, plant support system and plant foliage can be aggregated,
- The soil can be treated as well-oxygenated to depth, so the capillary fringe and deeper anoxic soil can be excluded from consideration and methane production in anoxic soil layers is neglected,
- The soil microbial community and mycorrhizal associations can be aggregated as soil biota,
- The stream bed can be considered to comprise only superficial oxic deposited sediments, because deeper oxic and anoxic sediments are more relevant to lakes,
- The stream macrobenthos (or macrophytes) and microbial community can be aggregated as net stream primary producers (taking into account both production and consumption by the microbial community).

These simplifications give the reduced set of components shown in Table 4-2.

**Table 4-2. Reduced set of principal components of a carbon transport model.**

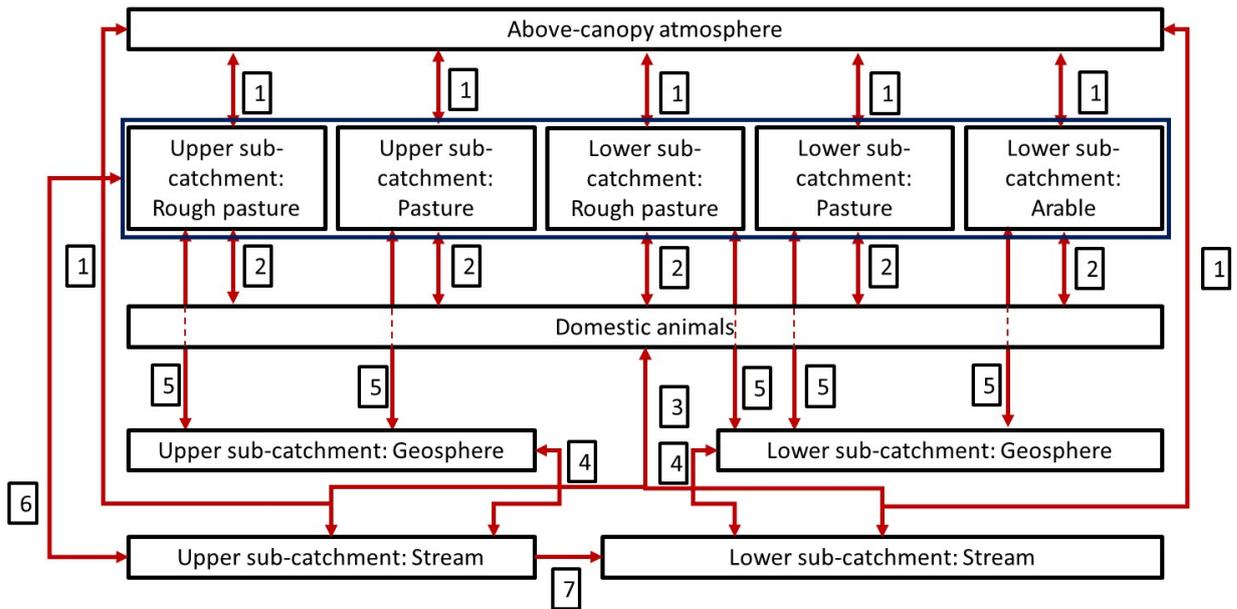
Major component	Sub-components	Items in each sub-component
Above-canopy atmosphere	None	Not applicable
Upper sub-catchment	Rough pasture	Canopy atmosphere
	Pasture	Unsaturated oxic soil
		Soil biota
		Plants
Domesticated animals		
Lower sub-catchment	Rough pasture	Canopy atmosphere
	Pasture	Unsaturated oxic soil
	Arable	Soil biota
		Plants
		Domesticated animals
Surface drainage network	Upper sub-catchment	Water column
	Lower sub-catchment	Suspended sediments
		Superficial oxic deposited sediment
		Stream primary producers
		Consumers and detritivores
Geosphere	Upper sub-catchment	None
	Lower sub-catchment	None
Humans	None	Not applicable

These components and the major carbon fluxes between them are illustrated in Figure 4-6.

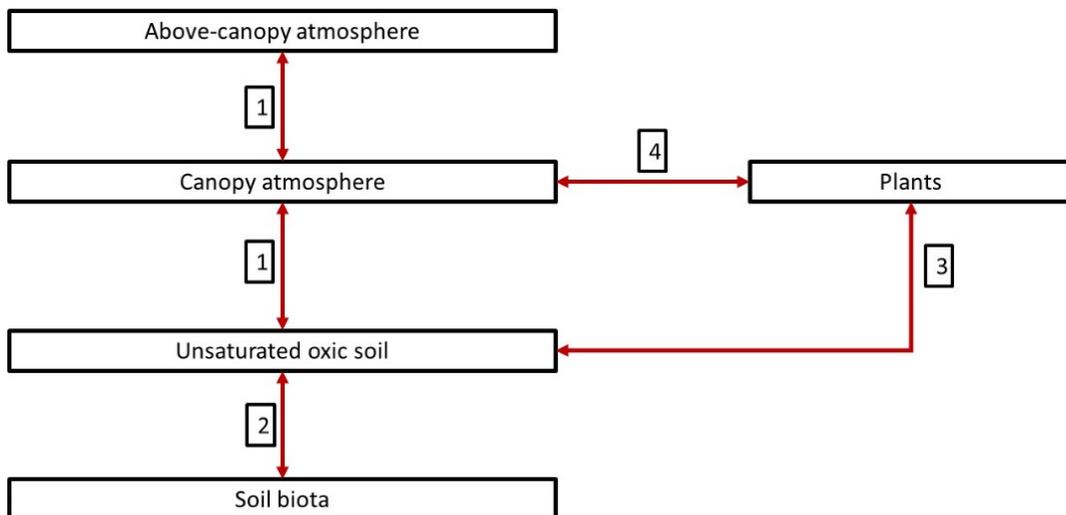
The main fluxes for each sub-catchment and land use are shown in Figure 4-7.

Processes within a stream segment are shown in Figure 4-8.

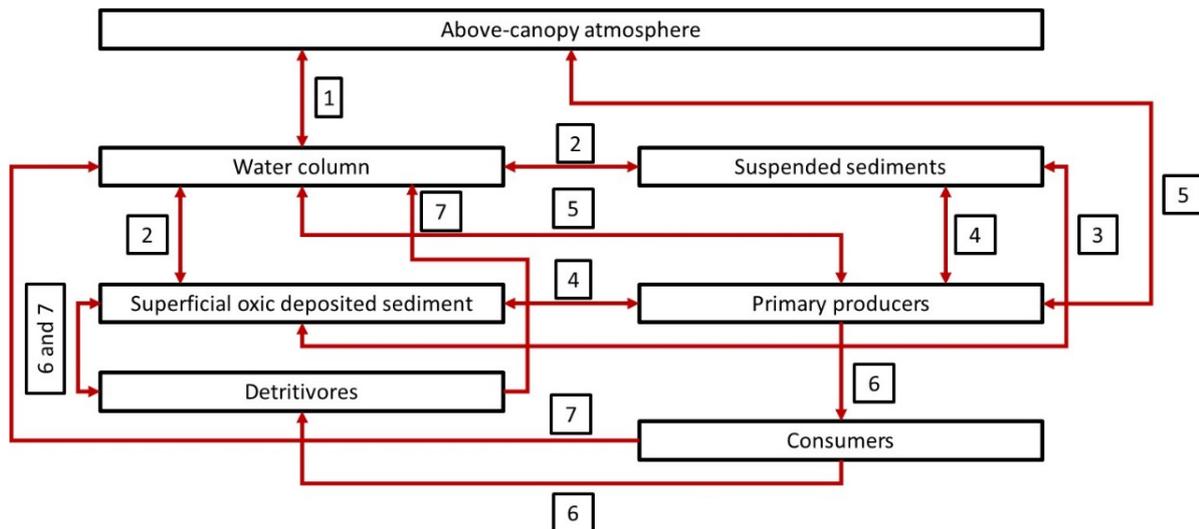
The model has been formulated so that C-14 fluxes can enter the system in solution or in gaseous form from the underlying geosphere. However, it could also be used to represent other fluxes entering the system. Specifically, it could be used to assess the longer-term cumulative impact of operational releases of C-14 to either atmosphere or the surface-water system.



**Figure 4-6.** Main fluxes between components (1 = gas exchange with the sub-canopy atmosphere and the surfaces of open water bodies; 2 = food consumption and excretion; 3 = drinking water consumption and excretion; 4 = water exchange between the geosphere and the stream; 5 = water exchange between the soil and the geosphere; 6 = water exchange between the soil and the stream; 7 = downstream flow; consumption of plants and animals by humans are not shown, nor are internal exchanges within plant-soil systems or within a stream segment).



**Figure 4-7.** Fluxes within the plant-soil system including the sub-canopy and above-canopy atmosphere (1 = gas exchange; 2 = biochemical transformations of chemical form; 3 = active and passive root uptake; 4 = photosynthesis and respiration)



**Figure 4-8.** Processes within a Stream Segment (1 = gas exchange, 2 = sorption/desorption, 3 = deposition/resuspension, 4 = uptake/excretion, 5 = respiration/photosynthesis, 6 = consumption, 7 = excretion)

#### 4.5 Mathematical equations for the hydrological model

The water-balance equations are developed for the full hydrogeological system shown in Figure 4-2. These are then reduced to the equations appropriate to the simplified system shown in Figure 4-3 by setting the omitted flows to zero. The water content of compartment  $i$  is designated  $Q_i$  and flow from compartment  $i$  to compartment  $j$  is designated  $F_{i,j}$ . The subscripts are defined in Table 4-3.

**Table 4-3. Subscripts adopted in the hydrological model.**

Compartment	Subscript
Rough pasture (Upper)	RU
Rough pasture (Lower)	RL
Pasture (Upper)	PU
Pasture (Lower)	PL
Arable (Lower)	AL
Geosphere (Upper)	GU
Geosphere (Lower)	GL
Stream channel (Upper)	SU
Stream channel (Lower)	SL

Water flows into the pasture and arable compartments are specified as  $F_{IN,j}$ , where  $j$  is the receiving compartment. Defining the area of each compartment as  $A_j$  and the hydrologically effective rainfall per unit area as  $H$ , it follows that:

$$F_{IN,j} = A_j H$$

For the geosphere compartments,  $H$  is replaced by  $U$ , where  $U$  is the rate of groundwater discharge or recharge per unit area.  $U$  is positive for discharge and negative for recharge.

Flows out of the system are specified as  $F_{i,OUT}$ , where  $i$  is the discharging compartment.

Each compartment is characterised by an area,  $A_i$ , a depth,  $d_i$ , a total connected porosity,  $\theta_i$ , and a degree of connected water saturation,  $S_i$ . Thus:

$$Q_i = A_i d_i \theta_i S_i$$

The units convention adopted is that  $Q$  has units of  $m^3$ ,  $F$  has units of  $m^3 s^{-1}$ ,  $H$  and  $U$  have units of  $m s^{-1}$ ,  $A$  has units of  $m^2$ ,  $d$  has units of  $m$ , and  $\theta$  and  $S$  are dimensionless.

If the system was being modelled dynamically, a set of equations would be developed expressing  $dQ_i/dt$  in terms of the water flows into and out of the compartment. However, in the modelling approach adopted, those flows are taken to be in balance. Therefore,  $dQ_i/dt = 0$  for all  $i$ .

This leads to:

$$\begin{aligned}
0 &= F_{IN,RU} + F_{SU,RU} + F_{PU,RU} + F_{GU,RU} + F_{RL,RU} - F_{RU,SU} - F_{RU,PU} - F_{RU,GU} - F_{RU,RL} \\
0 &= F_{IN,RL} + F_{SL,RL} + F_{RU,RL} + F_{PL,RL} + F_{AL,RL} + F_{GL,RL} - F_{RL,SL} - F_{RL,RU} - F_{RL,PL} - F_{RL,AL} - F_{RL,GL} \\
0 &= F_{IN,PU} + F_{SU,PU} + F_{RU,PU} + F_{PL,PU} + F_{GU,PU} - F_{PU,SU} - F_{PU,RU} - F_{PU,PL} - F_{PU,GU} \\
0 &= F_{IN,PL} + F_{SL,PL} + F_{RL,PL} + F_{GL,PL} + F_{PU,PL} - F_{PL,SL} - F_{PL,RL} - F_{PL,GL} - F_{PL,PU} \\
0 &= F_{IN,AL} + F_{RL,AL} + F_{GL,AL} + F_{SL,AL} - F_{AL,RL} - F_{AL,GL} - F_{AL,SL} \\
0 &= F_{IN,SU} + F_{RU,SU} + F_{PU,SU} + F_{GU,SU} - F_{SU,RU} - F_{SU,PU} - F_{SU,GU} - F_{SU,SL} \\
0 &= F_{IN,SL} + F_{RL,SL} + F_{PL,SL} + F_{AL,SL} + F_{GL,SL} + F_{SU,SL} - F_{SL,RL} - F_{SL,PL} - F_{SL,AL} - F_{SL,GL} - F_{SL,OUT} \\
0 &= F_{IN,GU} + F_{RU,GU} + F_{PU,GU} + F_{GL,GU} + F_{SU,GU} - F_{GU,RU} - F_{GU,PU} - F_{GU,GL} - F_{GU,SU} \\
0 &= F_{IN,GL} + F_{RL,GL} + F_{PL,GL} + F_{AL,GL} + F_{GU,GL} + F_{SL,GL} - F_{GL,RL} - F_{GL,PL} - F_{GL,AL} - F_{GL,GU} - F_{GL,SL}
\end{aligned}$$

The sum of these expressions is:

$$F_{IN,RU} + F_{IN,RL} + F_{IN,PU} + F_{IN,PL} + F_{IN,AL} + F_{IN,SU} + F_{IN,SL} + F_{IN,GU} + F_{IN,GL} = F_{SL,OUT}$$

Thus, the sum of the hydrologically effective rainfall over the whole catchment plus the net discharge of groundwater into the catchment balances the output in streamflow at the lower end of the catchment.

The simplified model shown in Figure 4-3 removes flows between the upper and lower catchment, except via the stream, and removes flows between different land uses. This leads to the following reduced set of equations.

$$\begin{aligned}
0 &= \mathbf{F}_{IN,RU} + F_{SU,RU} + F_{GU,RU} - F_{RU,SU} - F_{RU,GU} \\
0 &= \mathbf{F}_{IN,RL} + F_{SL,RL} + F_{GL,RL} - F_{RL,SL} - F_{RL,GL} \\
0 &= \mathbf{F}_{IN,PU} + F_{SU,PU} + F_{GU,PU} - F_{PU,SU} - F_{PU,GU} \\
0 &= \mathbf{F}_{IN,PL} + F_{SL,PL} + F_{GL,PL} - F_{PL,SL} - F_{PL,GL} \\
0 &= \mathbf{F}_{IN,AL} + F_{GL,AL} + F_{SL,AL} - F_{AL,GL} - F_{AL,SL} \\
0 &= \mathbf{F}_{IN,SU} + F_{RU,SU} + F_{PU,SU} + F_{GU,SU} - F_{SU,RU} - F_{SU,PU} - F_{SU,GU} - F_{SU,SL} \\
0 &= \mathbf{F}_{IN,SL} + F_{RL,SL} + F_{PL,SL} + F_{AL,SL} + F_{GL,SL} + F_{SU,SL} - F_{SL,RL} - F_{SL,PL} - F_{SL,AL} - F_{SL,GL} - \mathbf{F}_{SL,OUT} \\
0 &= \mathbf{F}_{IN,GU} + F_{RU,GU} + F_{PU,GU} + F_{SU,GU} - F_{GU,RU} - F_{GU,PU} - F_{GU,SU} \\
0 &= \mathbf{F}_{IN,GL} + F_{RL,GL} + F_{PL,GL} + F_{AL,GL} + F_{SU,GL} - F_{GL,RL} - F_{GL,PL} - F_{GL,AL} - F_{GL,SL}
\end{aligned}$$

Input and output quantities are shown in bold. The input quantities are specified by the user and the output quantity is obtained directly as the sum of the input quantities (see above). Also, the first five equations are independent of each other, so they can be satisfied separately. These solutions can be substituted into the final four equations.

To simplify the nomenclature, pairs of terms can be defined as commutators, i.e. we define  $G_{ij} = F_{ij} - F_{ji}$ . This yields:

$$\begin{aligned}
 0 &= F_{IN,RU} + G_{SU,RU} + G_{GU,RU} \\
 0 &= F_{IN,RL} + G_{SL,RL} + G_{GL,RL} \\
 0 &= F_{IN,PU} + G_{SU,PU} + G_{GU,PU} \\
 0 &= F_{IN,PL} + G_{SL,PL} + G_{GL,PL} \\
 0 &= F_{IN,AL} + G_{GL,AL} + G_{SL,AL} \\
 0 &= F_{IN,SU} - G_{SU,RU} - G_{SU,PU} - G_{SU,GU} - F_{SU,SL} \\
 0 &= F_{IN,SL} + F_{SU,SL} - G_{SL,RL} - G_{SL,PL} - G_{SL,AL} - G_{SL,GL} - F_{SL,OUT} \\
 0 &= F_{IN,GU} - G_{GU,RU} - G_{GU,PU} - G_{GU,SU} \\
 0 &= F_{IN,GL} - G_{GL,RL} - G_{GL,PL} - G_{GL,AL} - G_{GL,SL}
 \end{aligned}$$

This condensed form makes it clear that there is an ambiguity in defining values for  $F_{ij}$ . The individual values can be selected freely, subject only to the constraint that for each pair the relationship  $G_{ij} = F_{ij} - F_{ji}$  is satisfied. Incidentally, in rewriting equations in convenient form, it can be useful to use the general relationship  $G_{ij} = -G_{ji}$ .

Thus, in application, 1D hydrological models can be used to calculate solutions to the first five of the above equations. The sixth equation can then be solved to yield  $G_{SU,GU} + F_{SU,SL}$ , but hydrological modelling, or other arguments, must be used to partition the net loss from the upper part of the catchment between recharge of groundwater and stream flow from the upper part of the catchment to the lower part. Similarly, the seventh equation can then be solved to yield  $G_{SL,GL} + F_{SL,OUT}$ . This then gives all the quantities needed to solve the last two equations.

In terms of a suitable hydrological model, the form shown in Figure 4-4 is adopted for each of the areas of interest. The flow into the system is given by the hydrologically effective rainfall and is designated  $F_{IN,SS}$  and by geosphere discharge, designated  $F_{IN,G}$  (which can be negative to indicate recharge of groundwater). Thus, water-balance considerations give:

$$\begin{aligned}
 0 &= F_{IN,SS} + F_{SB,SS} - F_{SS,SB} - F_{SS,ST} \\
 0 &= F_{SS,SB} + F_{G,SB} - F_{SB,SS} - F_{SB,G} - F_{SB,ST} \\
 0 &= F_{SB,G} + F_{IN,G} - F_{G,ST}
 \end{aligned}$$

Using the nomenclature adopted in Figure 4-4, these various flows can be identified as listed in Table 4-4. As above, the convention is adopted that  $F$  has units of  $m^3 s^{-1}$ .

**Table 4-4. Flows in the 1D hydrological model.**

Flow	Description
$F_{IN,SS}$	Input of hydrologically effective rainfall
$F_{SS,SB}$	Percolation
$F_{SB,SS}$	Capillary rise and upward advection
$F_{G,SB}$	Capillary rise and upward advection
$F_{SB,G}$	Infiltration
$F_{IN,G}$	Groundwater discharge (recharge if negative)
$F_{SS,ST}$	Surface flow and throughflow
$F_{SB,ST}$	Baseflow 1
$F_{G,ST}$	Baseflow 2

## 4.6 Mathematical equations for the carbon model

### 4.6.1 General approach

In principle, three sets of equations are required, relating to stable carbon (C-12), its heavier stable isotope (C-13) and radiocarbon (C-14). However, these three sets of equations have a common form and can be represented by a single set of equations by introducing discrimination factors between the isotopes. Specifically, in a compartment representation:

$$dM_{i,X}/dt = F_{i,G,X} + \sum_j k_{j,i,X}M_{j,X} - \sum_j k_{i,j,X}M_{i,X} - k_{i,L,X}M_{i,X} - \delta_{X,C-14}k_{i,R}$$

where  $M_{i,C-12}$ ,  $M_{i,C-13}$ ,  $M_{i,C-14}$  (mol) are the amounts of C-12, C-13 and C-14 in compartment  $i$ , respectively, and  $\delta_{X,C-14}$  is the Kronecker delta (1 if  $X = C-14$  and 0 otherwise) and  $i = j$  is excluded from the summations. Use of the Kronecker delta permits the rate constant for radioactive decay ( $k_{i,R}$ ) to be incorporated into the general set of equations.

The rate constants for transport between the compartments ( $k_{i,j,X}$ ) differ somewhat because of isotopic effects. The flux of C-12 from compartment  $i$  to compartment  $j$  is denoted  $F_{i,j,C-12}$  (mol a<sup>-1</sup>).

Similarly, losses of C-12 from compartment  $i$  to outside the system are  $F_{i,L,C-12}$  (mol s<sup>-1</sup>) and gains to compartment  $i$  from outside the system are  $F_{i,G,C-12}$  (mol s<sup>-1</sup>).

This provides rate coefficients for transfers of C-12:  $k_{i,j,C-12} = F_{i,j,C-12}/M_{i,C-12}$  and, similarly,  $k_{i,L,C-12} = F_{i,L,C-12}/M_{i,C-12}$

The equations for C-12 must be solved first because the time-dependent amounts of C-12 in the various compartments and the C-12 fluxes between the compartments and out of the system are needed to calculate the rate coefficients applicable to C-13 and C-14.

The corresponding rate coefficients for the heavier isotopes include discrimination factors that depend on both the donor and receiver compartment. These are denoted  $D_{i,j,C-13}$  and  $D_{i,j,C-14}$ . Thus:

$$k_{i,j,C-13} = D_{i,j,C-13}k_{i,j,C-12}$$

$$k_{i,j,C-14} = D_{i,j,C-14}k_{i,j,C-12}$$

These discrimination factors are close to 1.0 and it may be convenient to write them in perturbation form, i.e.

$$D_{i,j,C-13} = 1 + \Delta_{i,j,C-13}$$

$$D_{i,j,C-14} = 1 + \Delta_{i,j,C-14}$$

Where the  $\Delta$  values are typically  $< 0.01$ .

This approach also works for losses from the system but involves the assumption that the discrimination factor depends only on the donor compartment. Thus:

$$k_{i,L,C-13} = D_{i,L,C-13}k_{i,L,C-12}$$

$$k_{i,L,C-14} = D_{i,L,C-14}k_{i,L,C-12}$$

The approach set out above cannot be applied directly to gains from outside the system, because the isotopic composition of the source or sources must be specified by the user. This applies particularly to simulations of experimental studies where the sources may comprise carbon isotopically enriched in C-13 or labelled with C-14. The C-12, C-13 and C-14 input fluxes are most conveniently specified directly.

### 4.6.2 Application to the illustrative catchment

From Figure 4-6, Figure 4-7 and Figure 4-8, the carbon pools that characterise the system can be identified. These are set out in Table 4-5. Note that losses from the system by mass transport or radioactive decay are transferred to two loss compartments. This allows mass and activity conservation to be checked when the model is implemented mathematically.

Transfers can occur between all these compartments. The rate coefficients of these transfers are designated  $k_{X,Y}$ , where X and Y are subscripts taken from Table 4-5. These rate coefficients depend on the isotope of carbon under consideration. However, to avoid over-complicating the nomenclature, this is to be taken as implicit rather than being represented explicitly through an additional subscript.

**Table 4-5. Carbon pools characterising the catchment.**

Major Component	Sub-component	Subscript Identifier
Above-canopy atmosphere	Above-canopy atmosphere	ACA
Upper Sub-catchment: Rough pasture	Canopy atmosphere	URA
	Unsaturated oxic soil	URS
	Soil biota	URB
	Plants	URP
Upper Sub-catchment: Pasture	Canopy atmosphere	UPA
	Unsaturated oxic soil	UPS
	Soil biota	UPB
	Plants	UPP
Lower Sub-catchment: Rough pasture	Canopy atmosphere	LRA
	Unsaturated oxic soil	LRS
	Soil biota	LRB
	Plants	LRP
Lower Sub-catchment: Pasture	Canopy atmosphere	LPA
	Unsaturated oxic soil	LPS
	Soil biota	LPB
	Plants	LPP
Lower Sub-catchment: Arable	Canopy atmosphere	LAA
	Unsaturated oxic soil	LAS
	Soil biota	LAB
	Plants	LAP
Domestic Animals	Domestic Animals	DAN
Upper Sub-catchment: Geosphere	Geosphere	USG
Lower Sub-catchment: Geosphere	Geosphere	LSG
Upper Sub-catchment: Stream	Water Column	USW
	Superficial Oxic Deposited Sediment	USO
	Suspended sediments	USS
	Primary Producers	USP
	Consumers	USC
	Detritivores	USD
Lower Sub-catchment: Stream	Water Column	LSW
	Superficial Oxic Deposited Sediment	LSO
	Suspended sediments	LSS
	Primary Producers	LSP
	Consumers	LSC
	Detritivores	LSD
Losses from the System	Mass Losses	LTM
	Radioactive Decay	LTR

The transfer matrix between the compartments is illustrated in Figure 4-9. In this figure, compartments between which no transfers are postulated are shaded out. For compartments between which transfers occur a colour code is used to indicate the processes involved. The following general points should be noted.

1. Elements on the principal diagonal are shaded out, i.e. internal mixing processes within a compartment are not represented explicitly.
2. All returns from the sink compartments to the system are shaded out.
3. The losses by radioactive decay apply only to C-14.
4. The column of abbreviations at the left lists the compartment originating the transfer, whereas the row at the top lists the compartment receiving the transfer.

The key to the colours used in Figure 4-9 is shown below.

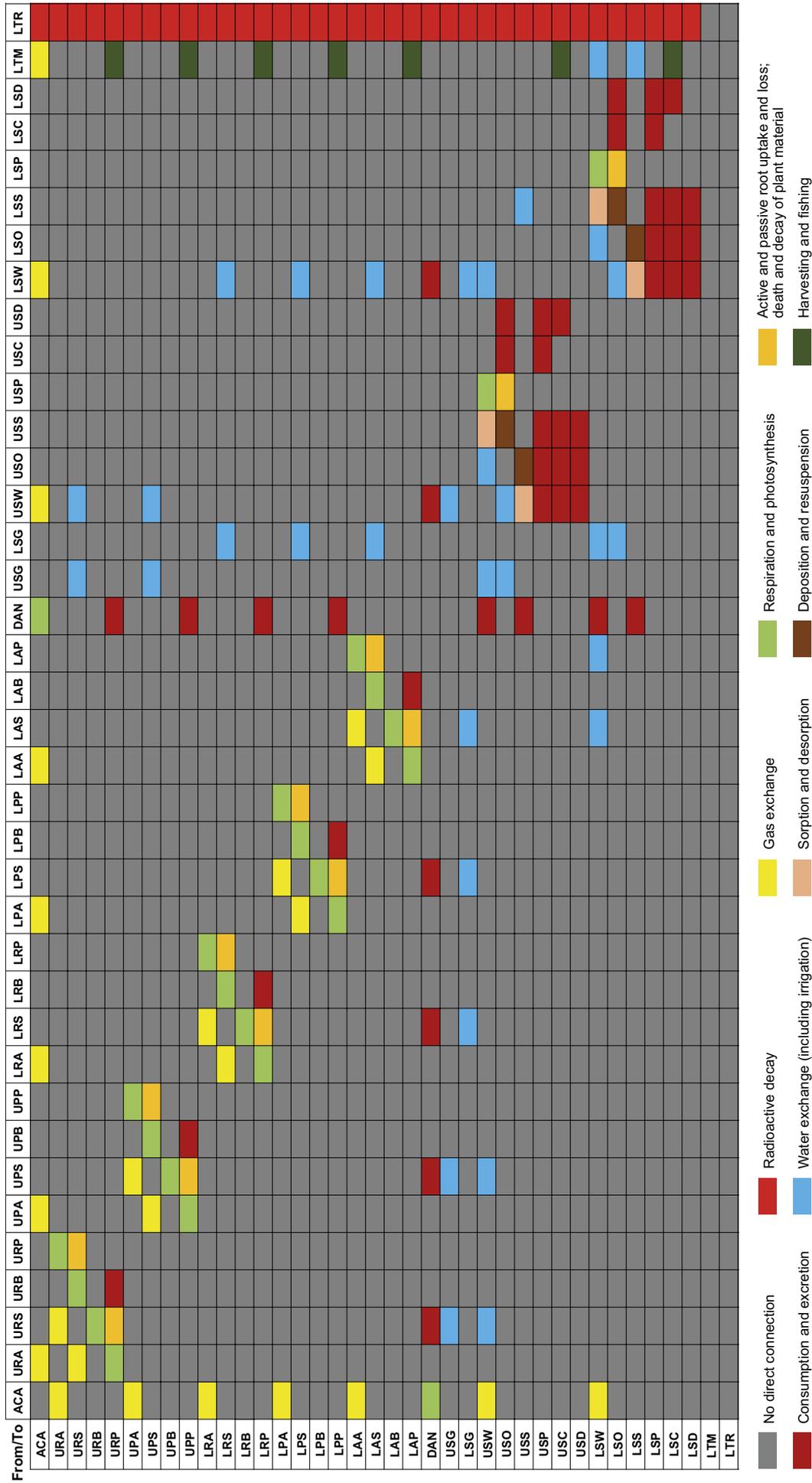


Figure 4-9. Transfer matrix for carbon in an agricultural catchment.

Some comments on the various types of transfer are appropriate. These are given below by category.

**Radioactive decay:** This only applies to C-14 but can be specified for all three radioisotopes using the Kronecker Delta technique described above.

**Gas exchange:** This is typically represented for the soil-plant-atmosphere system using a resistance analogue approach. However, it can also be represented using the advection-dispersion equation, with the diffusive term augmented to take account of the dispersive effects of atmospheric turbulence. For free-water surfaces, the advection-dispersion equation may be used, but an alternative is to use a 'piston velocity', where the velocity corresponds to the depth of a water layer that is completely cleared of dissolved carbon in unit time.

**Respiration and photosynthesis:** For plants, it is often sufficient to estimate the net rate of biomass production and to treat this as the difference in mass flux between total photosynthesis and respiration. For animals, respiration rates can be estimated directly, with the mass balance of carbon calculated from the carbon dioxide concentrations in inspired and expired air. For animals, losses of methane need also to be considered,

**Active and passive root uptake and loss; death and decay of plant material:** Passive root uptake can be calculated from the flux of transpiration water through plants and the dissolved carbon content of the water of transpiration. Active root uptake is indicated in some studies and total (active plus passive) root uptake can be estimated from experimental studies on the fraction of plant carbon obtained via the roots. Death and decay of plants returns carbon to the soil or to surface waters. Rates can be estimated from the rate of loss of plant material to these systems and the carbon content of that material.

**Consumption and excretion:** This consideration applies in both terrestrial and aquatic systems. In the terrestrial system, the main aspects are ingestion of plant material and drinking water (including its suspended sediment load) and excretion to soil by domestic animals. In the model structure shown in Figure 4-9, the minor pathways of ingestion of soil and the contamination of plants by excretion are neglected. In the aquatic system, consumption of water, sediment and primary producers is addressed, with detritivores consuming both primary producers and consumers of primary production. Excretion in aquatic systems is mainly to the water column or to bottom sediments, depending on the habitat preferences of the organisms involved. Direct estimates of the mass of material consumed or excreted and the carbon content of that material can be used to estimate rate coefficients.

**Water exchange (including irrigation):** Water flows can be taken from the hydrological model outlined above. Mass fluxes of different forms of carbon (DIC, DOC, PIC and POC) can be estimated from these flows and measured concentrations of these different forms of carbon. Irrigation is taken to apply only to arable land and mass fluxes between the terrestrial and aquatic ecosystems due to bulk transfers of organic material, e.g. leaf fall into streams and rivers, are neglected. In the simplified model adopted, sub-horizontal water flows between different areas of land use are neglected.

**Sorption and desorption:** These processes are explicitly considered for the water column and suspended sediments in the aquatic system. Although these processes apply also between the base of the water column and the underlying deposited sediments, it is considered that deposition and resuspension of particulate material are the predominant processes in that context. In the terrestrial environment, sorption and desorption will occur within the unsaturated oxic soil. However, these are processes internal to the compartment and are not represented explicitly.

**Deposition and resuspension:** These processes are primarily relevant to suspended and deposited sediments within the aquatic system. Deposition is often estimated using a settling velocity approach and resuspension by using an erosion rate.

**Harvesting and fishing:** Human interactions with the system relate primarily to the removal of material. This includes harvesting of arable crops, cutting of pasture for hay and silage, culling of animals, use of animal products such as milk, and taking of fish and other aquatic organisms. The culling of animals, use of animal products and taking of aquatic organisms implicitly result in harvesting of primary production in both terrestrial and aquatic environments.

Finally, in respect of Figure 4-9, it is noted how the removal of sub-horizontal transfers has resulted in a relatively sparse transport matrix, with well-defined sub-model domains. Furthermore, this presentation provides a transparent method of auditing transfers that may have been omitted and for incorporating them in an enhanced version of the model.



## 5 Conclusions

The importance of using the best available scientific information in the development of assessment models for key radionuclides, such as C-14, has obvious advantages for the optimised design of radioactive waste management programmes. The avoidance of inappropriate caution in model assumptions has been highlighted throughout the work of the BIOPROTA Forum<sup>10</sup> and has been an important driver in this study.

Understanding of physical and chemical forms of C-14 in relevant source terms was a key starting point considered in this study. Significant among the waste streams are those waste streams for which it is not clear which disposal option is most appropriate, such as graphite waste.

The importance of physical and chemical form then runs through the entire process of conceptual and mathematical model development, right up to the point of exposure. Noting this, an Appendix has been provided on the selection of appropriate dose coefficients that considers the latest scientific information relevant to the biokinetic models applied by the ICRP.

A review of the secondary literature on the distribution and transport of isotopes of carbon in the environment has provided the basis for development of a set of site-generic conceptual models of carbon distribution and transport in terrestrial and aquatic ecosystems. In turn, it is shown how the conceptual models for agricultural and freshwater ecosystems can be combined and used as the basis for developing a mathematical model that can be used to develop and parameterise a computational model of the environment that may be used for assessment purposes. The approach to model development has followed the approach suggested in IAEA (2003), as reviewed and enhanced in Lindborg et al. (2022).

The amount of information on behaviour of carbon in the environment is enormous. To achieve a practical model solution, it is necessary to make simplifying assumptions. Simplifications are appropriate when the level of temporal or spatial discrimination is sufficient to meet the purposes of the assessment, as set out in the assessment context. However, where simplifications have been made and are not self-evident, comments have been included in the tables describing the models. Provision of such comments supports transparency of decisions on model assumptions. It also supports the independent auditing of the model and revision of the model as may be necessary if and when the assessment context changes.

In many assessment models used for C-14, the specific activities of various environmental media of relevance are assumed to be the same (or related through fixed ratios) and independent of time. These so-called specific activity models have advantages in simplicity of application. However, they commonly rely on assumptions about equilibrium over temporal and spatial scales and can be applied without consideration of whether they are appropriate assumptions for the system being assessed. This approach can lead, in different circumstances, to under-estimation or over-estimation of assessed doses. It also fails to make use of site characterisation information that would typically be available, as needed for assessment of doses from other radionuclides. The review carried out within this study provides information to allow judgements to be made about assumptions for equilibrium within various environmental systems, and thus can be used to support the justification for a specific-activity model.

Development of a computational model has not been undertaken at this stage. However, the literature reviewed provides an appropriate basis for developing such a model in the future that could be the subject for further collaborative research. Furthermore, the other conceptual models developed provide a basis for developing mathematical and computational models appropriate to a wide variety of ecosystems of potential relevance in post-closure radiological impact assessments. In addition, it is hoped that the output provides the starting point for development of site-specific C-14 dose assessment models, especially for potential post-closure releases from solid waste disposal facilities.

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<sup>10</sup> See the wide range of reports at [www.bioprota.org](http://www.bioprota.org)



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## Selection of appropriate dose coefficients based on system understanding for C-14 and other potentially key radionuclides

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### A1 Background

C-14 has been recognised as a significant radionuclide in management of solid, liquid and gaseous radioactive waste for decades (UKAEA 1983, NCRP 1985, EPRI 1995). This significance arises because C-14 is projected to be among of the radionuclides giving rise to the highest individual doses following discharge to the environment or disposal. Examples of site-specific assessments for solid waste disposal include:

- The UK's LLW Repository Ltd in the case of near-surface disposal of low-level waste (NRPB 1988, LLWR 2011). In the older assessment, C-14 waste from animal research was a significant source term; other waste streams had become more important by the time of the later assessment, with very different characteristics.
- SFR1 in Sweden, in the case of low and intermediate-level waste at intermediate depth. Here, C-14 in reactor operating waste was the dominant dose contributor for the base scenario and realistic biosphere evolution (SKI/SSI 2004).
- The repository under construction at Olkiluoto, Finland, in the case of high-level waste disposal. Ingestion of C-14 in drinking water from a contaminated well was among the dominant exposure pathways for the reference release scenario (POSIVA 2013).

C-14 is also identified as a major contributor to radiation doses due to releases from nuclear facility operations (EPRI 2014). While the timeframes of main interest here are very different from those for disposal in a repository, some aspects of dosimetry and modelling of C-14 behaviour in the biosphere are similarly relevant (Smith 2015a). As well as annual individual doses, C-14 is identified as the major contributor to collective doses to the public from operation of the nuclear power plants and reprocessing plants (UNSCEAR 2000).

C-14 has therefore attracted significant research attention to address uncertainties in the source term, including amounts and chemical form of C-14, e.g., see SSI (2002), as well as migration and retention in a repository near-field and surrounding geosphere, e.g., see Hesbol et al. (1990). A more recent substantial international program concerned treatment and disposal of irradiated graphite and other carbonaceous waste in the CARBOWASTE project.<sup>11</sup>

C-14 has also attracted research to generate improvements in dose modelling for short- and long-term operational releases to the biosphere, e.g. Watkins et al. (1998), Sheppard and Stephensen (2002) and Akata et al. (2013), as well as for releases in the far future from radioactive waste repositories, notably the extensive review in BIOPROTA (2006). This latter work later formed the basis for further exchange of data and modelling expertise in the BIOPROTA programme<sup>12</sup>, as reported in BIOPROTA (2013), Smith and Smith (2014), Smith (2015b), Limer et al. (2009, 2017), SSM (2011) and Limer (2017, 2019). This work represents and references a substantial amount of site-generic and site-specific research and assessment shared openly within the international community.

An interesting example of the need for such work arises concerning the near-surface or deeper disposal of graphite. In both cases, the major post-disposal impacts are likely to be from eventual releases to the biosphere of C-14 and Cl-36. Limer et al. (2010) documented a modelling exercise to determine acceptable release rates to the biosphere, thereby setting provisional containment targets on the combined near field and geosphere system. The exercise used results from international cooperation, both for C-14, as above, but also for another key radionuclide, Cl-36, see (BIOPROTA 2006) and ANDRA (2008). It is interesting that results show that previous assumptions about

<sup>11</sup> <https://igdtp.eu/activity/carbowaste/>

<sup>12</sup> Reports available at [www.bioprot.org](http://www.bioprot.org)

long-term elemental behaviour, as used in so-called specific-activity models, can disguise relevant sub-processes. The work on disposal assessments has benefitted from improved understanding of shorter-term dynamics of the environmental behaviour of C-14, e.g., Aulagnier et al. (2013) and Avila and Kovalets (2016), and Thiry (2022) in relation to Cl-36.

While these and similar investigations have addressed a wide range of factors that could influence the migration and accumulation of radionuclides in the environment, and how they have consequences for intake into the body, little consideration has been given to the implications for selection of dose coefficients.<sup>13</sup>

The example mentioned above, of ingestion of C-14 in drinking water being the dominant exposure pathway, prompted further thought. The International Commission on Radiological Protection (ICRP) biokinetic model for ingested C-14 is based on the assumption that ingested C-14 is in the form of a labelled organic compound of which 100 % transfers across the gut into the bloodstream in a form that can then be readily metabolised and incorporated in body tissues and organs. The question of whether such an assumption would be appropriate in the case of C-14 dissolved in drinking water was therefore raised in Smith and Thorne (2015). At the same time, Smith and Thorne (2015) raised the issue of the lack of coherence between assumptions for consumption of foods containing chlorine, and the assumption used to calculate the reference values of dose coefficients for Cl-36.

This Appendix discusses the factors relevant to the selection of dose coefficients generally, and then gives closer attention to values for C-14. Regulatory factors and international guidance are briefly reviewed, in the context of present day and far future releases as may be relevant to solid waste disposal assessments. Based on experience in applying that guidance, the need to take account of the prevailing circumstances, physical and chemical form at the time of intake, is discussed. Values of dose coefficient for C-14 are presented and the circumstances under which they are appropriate are set out, considering both the older and the most recent ICRP documents, as well as other publications. Consideration is then given to the broader issue of selection of appropriate values of dose coefficients for other radionuclides and discussed in the context of supporting compliance with regulatory requirements, optimisation in waste management and the coherent treatment of uncertainties.

## **A2 Factors affecting the selection of dose coefficients**

### **Basic issues**

The factors affecting the selection of dose coefficients for C-14 are primarily based on the mode of exposure, ingestion or inhalation being most relevant for C-14, and the physical and chemical form at the point of intake. The latter are in turn affected significantly by a wider range of assumptions that would need to be made in conducting a radiological assessment. These factors typically include:

- the temporal dynamics of the release,
- the physico-chemical form of release into an accessible environment,
- the prevailing and potentially changing environmental conditions that might affect that form prior to intake,
- details of the exposure pathway, such as the type of material ingested or inhaled, and
- the assumptions for habits of the potentially exposed group of interest.

Such factors will be of general relevance but are especially relevant for C-14, because of its significance in terms of assessed impacts and because of the role of carbon in biological systems and the foodchain.

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<sup>13</sup> An exception was the BIOPROTA investigation of doses from human intrusion into geological disposal systems (POSIVA 2012) that considered the physical and chemical form of radionuclides that might be inadvertently inhaled. Accordingly, account was taken of the recommendations of the International Commission on Radiological Protection (ICRP) on how dose coefficients can vary according to particle size and mobility from the lung, taking account of realistic data on dust in air in different working conditions.

## Regulatory factors and international guidance

Regulatory requirements, often based on interpretation of international documents, may also constrain assessment assumptions, including the selection of dose coefficients.

In discussing verification of compliance with dose limits, the International Basic Safety Standards (IBSS) issued by the International Atomic Energy Agency (IAEA 2014) say that “the doses per unit intake (dose coefficients) for the estimation of the committed effective dose for ingestion and inhalation of radionuclides *are given in {following} tables*”, and provide data referenced to ICRP (1998, 2012). There is a clear implication that the tabulated values are to be used when applying the IBSS. The data in the tables (IAEA 2014) include values of gut transfer factors and assumptions denoting fast, moderate and slow absorption from the lung, for different compounds of the elements, and the corresponding dose coefficients for ingestion and inhalation for members of the public for these different compounds. There is no discussion of the particle size assumed in the case of inhalation, nor any discussion of scope to use better scientifically based data if they are available, or otherwise appropriate assumptions should they be appropriate in the circumstances.

The ICRP has for many years provided values of dose coefficients both for workers and for members of the public. The values for members of the public were most recently compiled from various previous references, including those describing biokinetic models and data, in ICRP Publication 119 (ICRP 2012), as referenced by the IAEA. The referenced sources for the compiled data take account of the accumulation of improved scientific information concerning the biokinetics and other factors affecting dose coefficients but are based on the definition of dose given in ICRP Publication 60 which dates from 1991 (ICRP 1981) and biokinetic models that are even older. Furthermore, Publication 119 did not take account of revisions to weighting factors that were recommended in ICRP Publication 103 (ICRP 2007). Instead, it was noted that the coefficients tabulated “will be superseded in due course by values based on the Publication 103 recommendations”. These revisions have still not been formally published as regards dose coefficients for members of the public, 15 years since changes to the recommendations on weighting factors were made.<sup>14</sup>

The dose coefficients provided by ICRP are referred to by ICRP as reference values, e.g., at paragraphs 137 and 142 of ICRP Publication 103 (ICRP 2007). Being described as reference values, and noting the comments in the IBSS, to the effect that these are the data to be used in addressing compliance with dose limits, they are commonly used in radiological assessments and there could be a reluctance to consider the use of alternatives, even to take into account the ICRPs recommended changes.

However, it should also be noted from paragraph 146 of ICRP (2007) that the dose coefficients are:

“specified by the Commission with no departure from the anatomical, physiological, and biokinetic characteristics of the Reference Male and the Reference Female (ICRP 2002). Account may be taken of the physical and chemical characteristics of the intake, including the activity median aerodynamic diameter (AMAD) of the inhaled aerosol and the chemical form of the particulate matter to which the specified radionuclide is attached.”

In other words, if the assessor has reliable alternative or additional information, this may be taken into account. In fact, using the appropriate data in the prevailing circumstances has long been recognised by ICRP and others, and applied in practice. See, for example, the assumption for gut transfer factors for isotopes of plutonium and americium in shellfish obtained near to the Sellafield site in northwest England as discussed in Hunt et al. (1986, 1988).

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<sup>14</sup> During the period that this report was under review, the ICRP issued a draft document for consultation setting out updated values, and the bases for them, of dose coefficients for members of the public; the document is available at <https://www.icrp.org/consultations.asp>. The draft for consultation does not identify the changes from previous versions, but the discussion and revised values proposed for C-14 are largely consistent with the information set out in Section A3 herein.

## Accounting for the prevailing circumstances

The case made by Hunt and colleagues was that data on gut transfer factors for environmental forms of radionuclides are essential for estimates of public radiation exposures following ingestion, and therefore in decisions on controlling waste discharges to the environment. In the case discussed by Hunt et al. (1988), it was noted that dose estimates for transuranic nuclides were particularly sensitive to uncertainties stemming from gut transfer data being related to non-environmental forms and/or derived from animal experiments. The main parameter in question at the time was the fraction of intake reaching human body fluids following ingestion, as features in the model of the gastro-intestinal tract used by the ICRP. The ICRP had recently reviewed the metabolism of plutonium and related elements. Data for gut transfer had been derived from animal data; limited verification was provided and the only human data then available was based on the low levels of fallout in foodstuffs. The ICRP proposed a cautious value of  $1 \times 10^{-3}$  for unknown or mixed compounds of Pu and for other actinides. However, it was also indicated that if a different value more suitable to the specific situation can be justified, it should be employed. The argument used by Hunt and his colleagues, to use a lower value of gut transfer factor, and hence, a lower value of dose coefficient than the reference value given by ICRP, was based on direct measurements reported in Hunt et al. (1986) as discussed in Hunt et al. (1988) and further supported in further site-specific field measurements (Hunt et al. 1990).

## Assessment of doses in the far future

The assessment of radiation doses in the far future, as relevant to releases from radioactive waste repositories, presents several additional challenges. These include the assumptions made in relation to natural environmental change, human behaviour that may also affect the biosphere system and the human activities that lead directly to exposure. These factors were discussed in detail in a report setting out the IAEA's so-called BIOMASS reference biospheres methodology IAEA (2003). Here it is suggested that databases on existing or historic local, regional or national dietary and other habits may be relevant to the assessment. When the assessment context dictates that future environmental change should be taken into account, it may also be appropriate to represent human behaviour on the basis of historical or present-day practices from analogue locations having biosphere conditions similar to the projected future biosphere conditions at the site of interest.

This approach was reflected in subsequent advice from ICRP on radiological protection in geological disposal of long-lived solid radioactive waste (ICRP 2013). Here it is said (paragraphs 93 and 94) that it should be assumed that the representative person is located at the time and place of the maximum concentration of radionuclides in the accessible biosphere, with due regard to the assumed climatic conditions for the release scenario being considered. **A representative person and biosphere should be defined using either a site-specific approach based on site- or region-specific information, or a stylised approach based on more general habits and conditions.** The use of stylised approaches will become more appropriate for longer time scales.

Experience with adopting the above recommendations and guidance has been documented in the IAEA MODARIA programme report on environmental change in post-closure safety assessment of solid radioactive waste repositories (IAEA 2016) and in Lindborg et al. (2022)<sup>15</sup> and Griffault et al. (2022).<sup>16</sup> In short, the international guidance has been very effective in leading to the adoption of a common approach to address the above challenges. However, differing conditions at different specific sites and wider specific assessment context issues can lead to corresponding differences in model assumptions.

This adaptive approach to the use of international guidance has been used widely in various disposal assessments, e.g., SKB (2014) and others referenced in Section A1, and in corresponding regulatory guidance, e.g., (EA and NIEA 2009). Here, it is said that it is necessary to show that the biosphere is characterised, understood and capable of analysis to the extent necessary to support the environmental safety case. This may involve consideration of, for example, topography, soils, surface water

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<sup>15</sup> Describing experience in the application of IAEA (2003) and related adaptations and enhancements.

<sup>16</sup> Describing experience and examples of the definition of potentially exposed groups of humans and potentially exposed populations of biota in the context of solid radioactive waste disposal assessment.

systems, flora and fauna distributions and human settlement patterns and activities. It is also noted that a cautiously low guidance value of risk has been selected, so that:

“It is not necessary when expressing the {assessed} aggregate risk for comparison with the risk guidance level to include an additional conservative bias.”

This last aspect of the guidance could be applied to the selection of dose coefficients as much as any other factor.

### A3 Values of dose coefficients for C-14

Noting the above, it is interesting to consider the exposure arising from ingestion of C-14 dissolved in groundwater or well water in which the presence of C-14 is due to release from a radioactive waste repository, as was significantly the case in POSIVA (2013).

As noted in Smith and Thorne (2015), the ICRP dose coefficient used in this case was based on the assumption that the C-14 is in the form of a labelled organic compound of which 100 % transfers across the gut into the bloodstream in a form that can then be readily metabolised and incorporated in body tissues and organs. This assumption is appropriate in the case of C-14 ingested in foods in which the C-14 is incorporated in the substrates of metabolism (sugars and other carbohydrates, proteins and fats). However, in the case of drinking water, the carbon will be predominantly present as dissolved carbon dioxide or dissolved bicarbonate or carbonate (see, for example, Ander et al. 2004). Although transfer across the gut may be close to complete, in these forms almost all the C-14 will be lost by exhalation as CO<sub>2</sub> without first having been metabolised and incorporated in body tissues. This has been demonstrated in human subjects intravenously injected with sodium bicarbonate (Winchell et al. 1970), and as discussed in ICRP (1981). In this case, the dose coefficient would be substantially lower than the value typically used.

No formal advice has been given by ICRP as to the appropriate dose coefficient to be used in the above circumstances. However, Harrison and Leggett (2016) confirmed the ICRP position that site-specific data other than those considered by ICRP may be used in calculations if this can be justified. They also note that ICRP Publication 103 (para 320) (ICRP 2007) says the equivalent dose in an organ and the effective dose:

“must be inferred with the aid of models, usually involving environmental, metabolic, and dosimetric components. Ideally, these models and the values chosen for their parameters should be realistic, so that the results they give can be described as ‘best estimates’”.

This advice appears consistent with the guidance in EA and NIEA (2009).

Harrison and Leggett (2016) go on to say that ICRP was {as of 2016} in the process of updating all dose coefficients for workers and members of the public, including revision of radiation and tissue weighting factors made in ICRP (2007), as well as implementing updates of biokinetic and dosimetric models used in the calculation of dose coefficients for inhalation and ingestion of radionuclides (ICRP 2015). Significantly, they added that,

“Where updated information allows, conservative biokinetic models such as the current generic model for carbon are being replaced with more realistic models intended as central estimators. The updated generic model for carbon absorbed to blood is less conservative than the assumption of a retention half-time of 40 d, while still accounting for the possibility that a dosimetrically significant portion of absorbed radiocarbon may be retained in the body for an extended period. The model being applied to carbon inhaled as CO<sub>2</sub>, and, more generally, to carbon reaching blood as carbon dioxide or bicarbonate, is based on a sizable dataset on the fate of carbon absorbed as bicarbonate, carbonate, or carbon dioxide in human subjects and laboratory animals. **The dose coefficient for ingested C-14 based on the updated generic model for carbon will be about 30 % of the current value. A dose coefficient to be provided specifically for C-14 ingested as bicarbonate will be about 2.5 % of the current value for ingested C-14.**”

Since then, the ICRP has issued updated values of dose coefficients for occupational intakes of radionuclides, in Publication 134 (ICRP 2016). This includes new dose coefficients for isotopes of carbon accounting for the updated biokinetic models, but only for workers, not for members of the public. These new worker values given in Table A-1 alongside the values for an adult given in the Publication 119 compendium (ICRP 2012). The differences between the dose coefficients for C-14 for a worker and for the adult member of the public, when considered as a representative person, are thought to be negligible. The ratios for new to old dose coefficients, are also provided, with the ratio value suggested by Harrison and Leggett (2016) in the case of ingestion  $\text{HCO}_3$  dissolved in water, in the absence of advice from ICRP.<sup>17</sup>

**Table A-1. Values of dose coefficients for C-14.**

C-14 Intake and form	Dose Coefficient, Sv/Bq		
	Publication 119 ICRP (2012), public adult	Publication 134 ICRP (2016), worker	Ratio 134:119
<b>Inhalation</b>			
CO <sub>2</sub>	6.50E-12	1.30E-11	2.00E+00
CH <sub>4</sub>	2.90E-12	5.10E-14	1.76E-02
vapours	5.80E-10	1.70E-10	2.93E-01
<b>Ingestion</b>			
All chemical forms	5.80E-10	1.60E-10	2.76E-01
			<b>Ratio Harrison and Leggett (2016):119</b>
HCO <sub>3</sub> dissolved in water	Not considered	Not presented, but the relevant modelling assumptions are provided <sup>a</sup>	circa 2.5E-2

<sup>a</sup> In particular, the model for carbon reaching the blood as carbon dioxide or bicarbonate.

## A4 Discussion

The changes in the ICRP reference values for dose coefficients for workers include a doubling for inhalation of CO<sub>2</sub>, and a more than 50-fold reduction for CH<sub>4</sub>. This highlights the significance of accounting for the chemical form at the time of inhalation, e.g., it being grossly conservative to assume 100 % CO<sub>2</sub> if it is known not to be.

Knowing the chemical form of atmospheric discharges is also relevant in the context of ingestion, given that methane needs to be converted to carbon dioxide for photosynthesis to occur leading to exposure via ingestion. In any case, the reduction in dose coefficient for ingestion by almost a factor of 4 is also important, as well as for liquid discharges to water bodies, both freshwater and marine. For freshwater bodies that are consumed directly, the reduction is substantially larger for dissolved HCO<sub>3</sub>, and, from the scientific discussion, should be similarly reduced for dissolved CO<sub>2</sub>.

If these changes are translated into the corresponding quantities for the public, as would appear both likely and overdue<sup>18</sup>, they may have significant implications for the optimisation of the management C-14 in reactor effluent and solid wastes such as graphite. As seen from above references, C-14 can be a significant contributor to prospective doses from near surface and deeper solid waste disposal facilities which can lead to limitations on disposal capacity. An appropriately optimized balance

<sup>17</sup> Discussion in Publication 134 and Leggett (2017) suggests that dissolved CO<sub>2</sub> in the bloodstream would behave bio-kinetically as HCO<sub>3</sub>, following either inhalation or ingestion. The model used in Publication 134 for carbon reaching the blood as carbon dioxide or bicarbonate is a modification of a model proposed by Leggett (2004). However, no dose coefficient is presented for ingestion of either HCO<sub>3</sub> or CO<sub>2</sub> if ingested when dissolved in drinking water.

<sup>18</sup> It is interesting that the fundamental information that supports the changes to the biokinetic involves research that was published 18 or more years ago.

between discharge and disposal relies on understanding of many factors, but they should include the selection of the most up to date and assessment relevant dose coefficients. This in turn relies on knowledge of latest developments in biokinetic modelling as well as application of source term and/or system understanding<sup>19</sup>, to determine the relevant form at the point of intake.

While the provision of ICRP advice on biokinetic models is of huge value, it would not be practical for ICRP to provide advice relevant in all possible site-specific circumstances. The expertise needed to apply relevant revisions to ICRP models is only a relatively minor technical burden compared with the overall need for system understanding that is generally required for any major facility. The relevant models are well described by ICRP and others. For example, Manger (2011) proposes a generic biokinetic model for C-14 designed so that the user may adjust the level of dosimetric conservatism to fit the information at hand and may adjust parameter values for consistency with subject-specific or site-specific bioassay data.

The discussion here applies not just to C-14 but applies to selection of dose coefficients for other key radionuclides such as Cl-36. In the latter case the issue is to achieve consistency between assumptions for consumption of foods containing chlorine and the ICRP model assumption for intake of table chlorine.

As noted at Section A1, ICRP has always been clear in their advice to use the best available information to inform selection of dose coefficients, as well as noting where it has applied caution, including in the case of C-14.<sup>20</sup> ICRP Publication 147 (ICRP 2021) has recently reiterated the point, noting that:

“Reference dose coefficients are provided for particular circumstances of exposure, including specific chemical and physical forms of ingested and inhaled radionuclides. Site-specific information on the exposure should be used if available and if the level of exposure warrants more precise estimation of dose.”

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<sup>19</sup> See as discussed in Lindborg et al. (2021, 2022).

<sup>20</sup> As noted by Manger (2011), “The generic biokinetic model currently recommended by the International Commission on Radiological Protection for the treatment of systemic radiocarbon assumes uniform distribution of activity in tissues and a biological half-time of 40 d. This model is intended to generate cautiously high estimates of dose per unit intake of C-14 and, in fact, generally predicts a much higher effective dose than systemic models that have been developed on the basis of biokinetic studies of specific carbon compounds.”

