
Modelling chronic and accidental releases of carbon 14 to the environment

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Abstract. The TOCATA model simulates ^{14}C transfer within terrestrial and/or continental water ecosystems, in response to chronic and accidental hypothetical emissions of ^{14}C , in a liquid and/or gaseous form. This model has been developed from bibliographical knowledge on C cycling and common hypothesis to existing ^{14}C transfer models, in order to come within the conceptual and mathematical frameworks of the SYMBIOSE project. In this context, the conceptual modelling deals with discretizing the continental biosphere into elementary components likely to be contaminated and identifying interactions or transfer processes of ^{14}C between two components. The mathematical modelling for calculating stocks and fluxes under accidental conditions is based on a system of differential equations expressing conservation of pollutant activity for each component of the conceptual model. It is shown that the analytical solutions obtained when temporal derivatives of the differential equation system are equal to zero correspond to the activities at “steady-state” simulated for chronic contaminations.

1. INTRODUCTION

Radioactive ^{14}C is formed as a by-product of nuclear power generation, and a part of it is released to the environment. It easily becomes incorporated into food webs *via* photosynthesis by primary producing organisms, which gives ^{14}C its radiological importance (Cook et al., 1998). Because of the long radioactive half-life of ^{14}C (5730 years), increased environmental specific activities (i.e., quantity of ^{14}C per gram of carbon) may persist for tens of thousands of years and constitute an incremental radiation exposure to many environmental and anthropogenic systems. In particular, studies of human exposure to ^{14}C have shown that internal radiation following ingestion is the most important exposure pathway (compared to internal radiation following inhalation and external radiation).

In recent years, concern about the quantity of ^{14}C released to the atmosphere or continental waters and its subsequent incorporation into living material in the vicinity of nuclear power plants has prompted a number of modelling approaches to assess ^{14}C transfer in the environment and its radiological impact on man *via* the food chain. Today, most of the ^{14}C transfer models are based, explicitly or implicitly, on the hypothesis of conservation of the isotopic $^{14}\text{C}/^{12}\text{C}$ ratio between the different (eco) systems in concern. However, ^{14}C is usually assumed to be released by the source in a chronic mode, and impact assessments are made from annual average values. Therefore, most existing models can not be used for assessing the impacts of hypothetical accidental releases, as non-equilibrium conditions between ^{14}C and ^{12}C prevail.

The TOCATA (Transfer of Carbon 14 and Tritium in Terrestrial and Aquatic environments) model described in this paper simulates ^{14}C transfer within terrestrial and/or continental water ecosystems, in response to both chronic and accidental hypothetical releases of carbon 14.

The model has the following main characteristics:

- 1) It is based on bibliographical knowledge of C cycling and common hypothesis to existing models of ^{14}C transfer. However, it attempts to overcome some of the shortcomings of existing models by adopting a dynamic ecosystem approach, where both important biological (e.g. photosynthesis, growth, trophic transfer and mineralisation) and environmental processes (e.g. air–water exchange, air or water movement) are included.
- 2) It is implemented in the conceptual and mathematic frameworks of the SYMBIOSE modelling platform [3,4,5]. In this context, the conceptual modelling deals with describing and analysing the structure (potential stocks of contaminant) and functioning (potential fluxes of contaminant) of the biosphere, by defining components and elementary processes of mass transfer. The mathematical modelling is based on a system of differential equations defined for accidental conditions, expressing conservation of ^{14}C activity for each component of the conceptual model.

2. CONCEPTUAL MODELLING

The conceptual modelling of ^{14}C transfer in TOCATTA is based on a similar framework than the one defined in the SYMBIOSE modelling platform [3,4,5]. The biosphere is described as an organised and hierarchical framework of biotic and non biotic components, that are likely to reciprocally exchange radionuclides through biological, physical and chemical mechanisms called elementary processes (or interactions) of mass transfer.

2.1 Interaction matrix

The TOCATTA model is based on the principle of the interaction matrix used in SYMBIOSE to describe the biosphere (Gonze et al., 2002). Three major environmental systems are considered: the atmosphere, the agricultural world and the river (Figure 1). The anthropogenic system interacts with the previous ones, and constitutes their “endpoint”. The source of ^{14}C released to the environment in a liquid or gaseous form is the nuclear plant itself. Anything not considered in these systems is described as the “rest of the world”, that can constitute a source or a sink for the radioactive contamination (e.g. forest ecosystem, geosphere).

In the current version of the model, the main interactions between the environmental systems are the following ones: net primary production (i.e. photosynthesis minus plant respiration), soil microbial respiration, ingestion by man of vegetal and animal products, and fishes (Figure 1).

Source	Gaseous releases		Gaseous or liquid releases		
	Atmosphere	Net Primary Production			
	Soil respiration	Agricultural system		Ingestion	
			River system	Ingestion	
				Anthropogenic system	
					Rest of the world

Figure 1. Global interaction matrix used in the TOCATTA model showing the different systems (diagonal elements) and interaction processes (off-diagonal elements).

Each of the systems defined above is discretized into more detailed components with specific interactions defined between two of them (cf. Le Dizès, 2004). The conceptual modelling approach related to each of the systems and components and elementary interactions are fully described in Le Dizès (2004). The next sections focus briefly on the TOCATTA agricultural module by describing the components and some of the concepts used to model ^{14}C transfer, in response to an accidental release to the atmosphere.

2.2 The TOCATTA agricultural module

The agricultural system defined in TOCATTA has been derived from the ASTRAL code (Calmon & Mourlon, 2002). It contains five main components:

- Prairie (other appellations: pasture, grassland)
- large scale farming
- leafy vegetables
- fruit and root vegetables
- animals.

Each of the previous plant components consist of two sub-components: plant and soil. The plant sub-component is represented by a foliar system or an “edible organ” system. For simplicity, the root system is not considered in the current version of the model. The animals are considered only through their organs. The calculation is made for edible parts, e.g; meat, milk, or eggs.

In the case of accidental emissions of ^{14}C to the atmosphere, a dynamic modelling approach based on plant growth curves is used in TOCATTA. We make the assumption that an isotopic equilibrium condition between plant component and the atmosphere is reached at each time step of the simulation (i.e. 1 day); each step corresponding to a given period of time according to which atmospheric ^{14}C is assumed to be constant. Therefore the quantity of new plant biomass created during a given time step has the same isotopic ratio than the surrounding air. This approach therefore requires the discretization of a given growth curve by using the same time step defined for the time-dependant evolution of atmospheric ^{14}C (Figure 2).

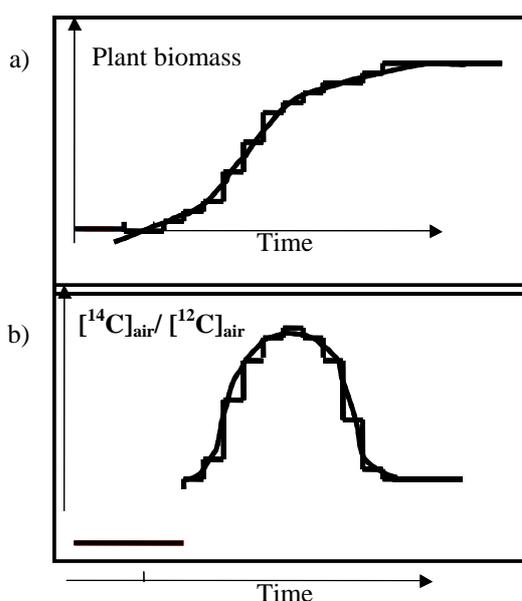


Figure 2. Simultaneous discretization of (a) a given plant growth curve and (b) the temporal evolution of atmospheric isotopic ratio, at a given distance of the emission.

3. MATHEMATICAL MODELLING

Mathematical modelling deals with parametrizing each of the described processes involved in the ^{14}C transfer between components. Here we focus on a particular process, i.e. the incorporation of atmospheric ^{14}C into plants and describe how this process has been formalized (1) in existing models for chronic releases and (2) in the TOCATTA model for accidental releases of ^{14}C to the atmosphere.

3.1 Existing models

Most existing models of transfer of ^{14}C simulate the incorporation of atmospheric $^{14}\text{CO}_2$ into plants *via* photosynthesis in response to chronic releases. The analysis of some of these transfer models reveals a general consensus, all the equations being based on the hypothesis of conservation of the isotopic $^{14}\text{C}/^{12}\text{C}$ ratio between plants and the atmosphere. Transfer of atmospheric $^{14}\text{CO}_2$ to plants is therefore always modelled in the same way, with only minor differences lying in the choice of parameter values or in the way parameters are clarified (Zach, 1978; Killough, 1984; Quinault, 1995; NCRP, 1996; Tamponnet, 2002). The following equation is often used:

$$[^{14}\text{C}]_{\text{veg}} = f_{\text{veg}}^c \cdot \frac{[^{14}\text{C}]_{\text{air}}}{[^{12}\text{C}]_{\text{air}}} \quad (1)$$

$[^{14}\text{C}]_{\text{veg}}$: Concentration of carbon 14 in plants [$\text{Bq}\cdot\text{kg}^{-1}$]

$[^{14}\text{C}]_{\text{air}}$: Concentration of carbon 14 in the air in the vicinity of plants [$\text{Bq}\cdot\text{m}^{-3}$]

$[^{12}\text{C}]_{\text{air}}$: Concentration of carbon 12 in the air [$\text{kg}\cdot\text{m}^{-3}$]

f_{veg}^c : Plant C proportion [$\text{kg}\cdot\text{kg}^{-1}$]

3.2 The TOCATTA model

3.2.1 Net primary production

The TOCATTA model simulates the assimilation of atmospheric $^{14}\text{CO}_2$ into plants *via* net primary production (*npp*) in response to accidental releases. This transfer process is explicitly described in the model as follows ($TM_{\text{air,veg}}^{\text{npp}}$, in $\text{Bq}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$):

$$TM_{\text{air,veg}}^{\text{npp}} = \left[\frac{dM_{\text{veg}}}{dt} \right]^{gro} \cdot f_{\text{veg}}^c \cdot \frac{pm \cdot [^{14}\text{C}]_{\text{air}}}{[^{12}\text{C}_{\text{CO}_2}]_{\text{air}}} \quad (2)$$

$\left[\frac{dM_{\text{veg}}}{dt} \right]^{gro}$: Plant biomass increment defined in Figure 2a for each time step [$\text{kg}\cdot\text{d}^{-1}$].

$[^{14}\text{C}]_{\text{air}}$: Concentration of carbon 14 in the air in the vicinity of plants [$\text{Bq}\cdot\text{m}^{-3}$]

$[^{12}\text{C}_{\text{CO}_2}]_{\text{air}}$: Concentration of carbon 12 in the air in the form of CO_2 [$\text{kg}\cdot\text{m}^{-3}$]

f_{veg}^c : Plant C proportion [$\text{kg}\cdot\text{kg}^{-1}$]

pm : proportion of carbon 14 released to the atmosphere in the form of CO_2 (input data)

3.2.2 Grazing and deposition

These two processes are simulated for the prairie, whose foliar system is assumed to (1) be continuously eaten by herbivores (grazing) and (2) regularly fall onto soils (deposition). The ^{14}C transfer rates associated with these two processes are respectively defined by ($TM_{veg,ani}^{gra}$ for grazing and $TM_{veg,soil}^{dep}$ for deposition, expressed in $\text{Bq}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$):

$$TM_{veg,ani}^{gra} = -[^{14}\text{C}]_{veg} \left[\frac{dM_{veg}}{dt} \right]^{ali} \quad (3)$$

$$TM_{veg,soil}^{dep} = -[^{14}\text{C}]_{veg} \left[\frac{dM_{veg}}{dt} \right]^{dep} \quad (4)$$

where $[^{14}\text{C}]_{veg}$ is the mass activity of the plant foliar system ($\text{Bq}\cdot\text{kg}^{-1}$), $\left[\frac{dM_{veg}}{dt} \right]^{ali}$ and $\left[\frac{dM_{veg}}{dt} \right]^{dep}$ the loss of foliar biomass associated with grazing and deposition, respectively ($\text{kg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)

3.2.3 Processes associated with biomass cycles

Modelling processes associated with biomass cycles is a prerequisite for assessing transfer fluxes within plant components as it provides the evolution of plant biomass through time.

For prairie, foliar biomass is constant through time as we make the assumption that processes of grazing and deposition compensate exactly biomass growth at each time step. Therefore, foliar biomass can be described by the following equation:

$$\left[\frac{dM_{veg}}{dt} \right] = 0 = \left[\frac{dM_{veg}}{dt} \right]^{gro} + \left[\frac{dM_{veg}}{dt} \right]^{ali} + \left[\frac{dM_{veg}}{dt} \right]^{dep} \quad (5)$$

where $\left[\frac{dM_{veg}}{dt} \right]^{gro}$ is the foliar biomass growth rate, assumed to be proportional to the foliar biomass itself at each time step (exponential growth assumption, cf. Le Dizès, 2004).

For the other plant components (i.e. large scale farming, leafy vegetables, fruit and root vegetables), we make the assumption that plant biomass (i.e. foliar system or organ) varies through time according to a logistic growth model (cf. Le Dizès 2004).

3.2.4 Principle of ^{14}C mass conservation

The TOCATTA model simulates transfer of atmospheric ^{14}C through plants in response to accidental releases by expressing conservation of mass activity in each plant component according to the following differential equation:

$$\frac{d\{M_{veg} [^{14}\text{C}]_{veg}\}}{dt} = \underbrace{TM_{air,veg}^{npp}}_{\text{AllComponents}} - \underbrace{TM_{veg,ani}^{gra} - TM_{veg,soil}^{dep}}_{\text{Pr airie}} \quad (6)$$

where $TM_{i,j}^p$ are the activity transfer rates from the component i to the component j via the effect of the processes p defined above, $[^{14}\text{C}]_{veg}$ is the mass activity of plant (Bq.kg^{-1}), and M_{veg} plant biomass (kg.m^{-2}), assumed constant through time for prairie or driven by a logistic growth model for other plant components (cf. section 3.2.3).

According to the parametrizations defined above, equation (6) result in the following first order differential equation, used for any plant components:

$$M_{veg} \frac{d[^{14}\text{C}]_{veg}}{dt} = \left[\frac{dM_{veg}}{dt} \right]^{gro} \cdot \left(f_{veg}^c \cdot \frac{pm.[^{14}\text{C}]_{air}}{[^{12}\text{C}_{CO2}]_{air}} - [^{14}\text{C}]_{veg} \right) \quad (7)$$

The analytical solution obtained when temporal derivative of this differential equation is equal to zero (i.e. when mass activity does not vary through time any more) leads to the following equation:

$$[^{14}\text{C}]_{veg}(t \rightarrow \infty) = f_{veg}^c \cdot \frac{pm.[^{14}\text{C}]_{air}}{[^{12}\text{C}_{CO2}]_{air}} \quad (8)$$

where $[^{14}\text{C}]_{veg}(t \rightarrow \infty)$ is the « steady-state » activity, reached under equilibrium conditions, when input fluxes are exactly equal to output fluxes. It is worth noting that this activity corresponds to the one simulated in existing models for chronic releases (cf. Eq. 1).

4. CONCLUSION AND PERSPECTIVES

The TOCATTA model has been developed from bibliographical knowledge on C cycling and common hypothesis to existing ^{14}C transfer models. With the aim of integrating ^{14}C transfer modelling in the SYMBIOSE platform, the model structure is based on its conceptual and mathematical frameworks. In this context, by clarifying important biological and environmental processes of mass transfer between biotic and non-biotic components, the model attempts to overcome some of the shortcomings of the more empirical existing models devoted to assess impacts of chronic contaminations. The TOCATTA model does simulate ^{14}C transfer in responses to accidental releases, by expressing conservation of both isotopic equilibrium and mass activity. It was shown that the analytical solutions obtained when temporal derivatives of the differential equation system are equal to zero correspond to the “steady-state” activities assessed by the classical ^{14}C transfer models for chronic contaminations.

In the near future, the TOCATTA model will have to be calibrated, compared to other several models, and, as far as possible, validated on real activity and/or dose measurements. Sensibility and uncertainty analysis will also have to be carried out.

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