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TOCATTA: a dynamic transfer model of ^{14}C from the atmosphere to soil-plant systems

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ABSTRACT

Many nuclear facilities release ^{14}C into the environment, mostly as $^{14}CO_2$, which mixes readily with stable CO_2. This complete isotopic mixing (equilibrium) is often used as the basis for dose assessment models. In this paper, a dynamic compartment model (TOCATTA) has been investigated to describe ^{14}C transfer in agricultural systems exposed to atmospheric ^{14}C releases from nuclear facilities under normal operating or accidental conditions. The TOCATTA model belongs to the larger framework of the SYMBIOSE modelling and simulation platform that aims to assess the fate and transport of a wide range of radionuclides in various environmental systems. In this context, the conceptual and mathematical models of TOCATTA have been designed to be relatively simple, minimizing the number of compartments and input parameters required, appropriate to its use in an operational mode. This paper describes in detail ^{14}C transfer in agricultural plants exposed to time-varying concentrations of atmospheric ^{14}C, with a consideration also of the transfer pathways of ^{14}C in soil. The model was tested against *in situ* data for ^{14}C activity concentration measured over two years on a grass field plot located 2 km downwind of the AREVA NC La Hague nuclear reprocessing plant. The first results showed that the model roughly reproduced the observed month-to-month variability in grass ^{14}C activity, but under-estimated (by about 33%) most of the observed peaks in the ^{14}C activity concentration of grass. This tends to prove that it is not suitable to simulate intra-monthly variability, and *a fortiori*, the response of vegetation to accidental releases that may occur during the day. The need to increase the temporal resolution of the model has been identified in order to simulate the impact of intermittent ^{14}C releases occurring either the day or night, such as those recorded by the AREVA NC plant.

1. Introduction

The radioactive carbon (^{14}C) released into the environment from nuclear facilities by a routine operation or accident will increase the background of ^{14}C in their vicinity. Mainly released as $^{14}CO_2$ into the atmosphere (Le Dizès et al., 2009), the ^{14}C can easily be incorporated into food webs via photosynthesis by primary producing organisms, such as grass. Grasslands may be pastured by cows, which produce milk and other animal products. The ^{14}C can then be transferred to humans through ingestion of the contaminated plant and/or animal products. Studies of human exposure to ^{14}C have shown that internal radiation following ingestion is the most important exposure pathway (UNSCEAR, 2000). Therefore, a mathematical model to predict the radioactivity of ^{14}C in agricultural plants is a prerequisite to assess ingestion dose to humans (Keum et al., 2008).

In the last two decades, concern about the quantity of atmospheric $^{14}CO_2$ released from nuclear facilities and its subsequent incorporation into the food chain have prompted a number of dynamic modelling approaches to predict the transfer of ^{14}C to plants, soil and animals (Andoh and Amano, 2003; Aquilonius and Hallberg, 2005; Keum et al., 2008; Koarashi et al., 2008; Pearce, 1992; Smith et al., 1994; Takahashi et al., 2008; Tani et al., 2011). All these models can be applied reasonably well to predict the concentration of ^{14}C in plants. A sophisticated time-dependent model can describe actual situations for climate conditions and key ecosystem processes such as photosynthesis, respiration, decomposition, and nutrient cycling (Keum et al., 2008; Peng et al., 2002). However, such a model requires many kinds of input data that are often not easily available. A simplified model with less input parameters is thus often used for a quick and straightforward environmental assessment.

In this paper, a dynamic compartment model (TOCATTA) is presented to estimate the ^{14}C behaviour in agricultural soil and

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plants exposed to spray irrigation with contaminated water and/or airborne $^{14}CO_2$ releases from nuclear facilities under normal operating or accidental conditions. The model has been implemented in the SYMBIOSE modelling and simulation platform that aims to assess the fate and transport of a wide range of radionuclides in various environmental systems, and their impact on humans (Gonze et al., 2011). This platform, co-funded by the Institute of Radioprotection and Nuclear Safety (IRSN) and Electricité de France (EDF), is to be used as a reference tool for assessing doses resulting from radioactive releases of nuclear facilities under accidental, decommissioning or normal operating conditions, as well as from waste disposal facilities. Environmental models in SYMBIOSE address atmospheric, terrestrial, freshwater and marine systems, as well as major transfer processes at their interfaces. Hundreds of compartments and interactions are accounted for in the platform, most of which are modelled using a dynamic and physically based approach. The TOCATTA model computes daily activity concentrations of ^{14}C in various types of agricultural soil, as well as plant and animal products. The subsequent calculation of ^{14}C transfer to man and assessment of doses are performed within the SYMBIOSE platform, based on a typical terrestrial food-chain scenario. The model has been successfully used in a wide scale case study with SYMBIOSE to perform landscape-level dose assessments of a French nuclear power plant (Mourlon et al., 2011).

This paper describes in detail ^{14}C transfer to plants, with a consideration also of the transfer to soil. In particular, ^{14}C transfer to animals is not described. Processes considered in this paper are assigned to two major sub-models: (1) a plant sub-model that estimates above-ground biomass and the dynamics of ^{14}C specific activity (activity of ^{14}C per mass of C) in various categories of agricultural plants, (2) a soil sub-model that simulates ^{14}C dynamics in soil and litter pools, and exchange processes at the soil- canopy atmosphere interface. The TOCATTA model was tested against ^{14}C measurements performed on a grass field plot located near the AREVA NC La Hague nuclear reprocessing plant. The objectives of this paper are (1) to document the scientific basis, major assumptions, conceptual modelling and mathematical formulations of the ^{14}C transfers in the model, (2) to discuss model test results based on a real-case scenario and identify its strengths and weaknesses, and (3) to discuss the future developments of the model to improve its predictive ability.

2. Model description

2.1. Main assumptions and characteristics

The TOCATTA model is based on a daily time step and is mainly driven by daily atmospheric $^{14}CO_2$ concentration and meteorological data. The only atmospheric emissions taken into account are in the form of $^{14}CO_2$, as this is thought to be the main chemical form of ^{14}C released by nuclear facilities (Fontugne et al., 2004; Koarashi et al., 2005); the remaining organic ^{14}C, mostly emitted as methane (CH_4) plays no role in photosynthesis and is therefore not directly transferred to plants. Although methane released to atmosphere can exchange with the soil atmosphere/soil solution and there be microbially metabolised, to some degree, to carbon dioxide, these processes are not considered in our application. It is also possible to ignore isotopic discrimination of ^{14}C in the biosphere because there are other sources of variability and uncertainty with larger effects than expected from isotopic discrimination (Killough, 1980). Moreover, given the long radioactive half-life of ^{14}C, it is not necessary to consider its radioactive decay since the simulations performed by the model do not exceed the time scales of decades. The wet input flux of airborne $^{14}CO_2$ brought by rain and irrigation to plants is assumed negligible compared with the atmospheric transfer pathway entering plants through photosynthesis (Sheppard et al., 2006a). Consequently, this transfer pathway is not explicitly modelled in TOCATTA, which assumes that the contribution of ^{14}C in precipitation and/or irrigation water is incorporated by wet inputs directly to the soil (cf. §3.2.3), and in this case passes indirectly to the plant via the emission of CO_2 from the soil. It is also assumed that root uptake of ^{14}C by plants is negligible in our atmospheric scenario. Indeed, the root incorporation of carbon into the plant from small organic molecules or soluble carbonates would represent less than 5% of total carbon incorporated (Amiro et al., 1991; Garnier-Laplace and Roussel-Debet, 2001). The model has the following main characteristics:

- A dynamic approach based on time-dependent growth curves for plants is adopted, on a daily basis. Moreover, photosynthesis and foliar respiration are not explicitly considered as separate processes. Instead, in the current version of the model, the net primary productivity is estimated from the simulated growth rate and dry matter quantity, either derived from experimental data or from the predefined curve of plant growth. If the model were to use a time step of less than one day, photosynthesis would have to be handled separately from foliar respiration, as the two exhibit different diurnal patterns.
- The assumption is made that an isotopic equilibrium condition between the plant compartment and canopy atmosphere is reached at each time step of the simulation (e.g. 1 day); each step corresponding to a given period of time during which the ^{14}C concentration in air is assumed to be constant. Consequently, the quantity of newly created plant biomass has the same isotopic $^{14}C/^{12}C$ ratio as the surrounding air (Le Dizès, 2004; Tamponnet, 2004).
- The model is parameterized for various types of agricultural plants, broken down into three groups according to well-defined criteria: annual crops, vegetable crops and pasture grass, comprising a total of fifteen SYMBIOSE categories (Calmon, 2009). Two categories of soils are also considered by default: sandy soil and clayey soil.
- Conceptual modelling deals with describing and analysing the structure (potential stocks of contaminant) and functioning (potential fluxes of contaminant) of the biosphere, by defining compartments and elementary processes of mass transfer in an Interaction Matrix (cf. §2.2). Mathematical modelling is based on a system of differential equations defined for time-varying release conditions, expressing conservation of radionuclide activity for each compartment of the conceptual model. The time rate of change in the activity or concentration in a given compartment i can be generally expressed as:

$$\frac{\mathrm{d}S_i}{\mathrm{d}t} = \frac{d\{\chi_i[C]_i\}}{\mathrm{d}t} = \underbrace{\sum_{j,p} TC^p_{j,i}}_{\text{inputs}} - \underbrace{\sum_{k,q} TC^q_{i,k}}_{\text{outputs}} \quad (1)$$

Where S_i and $[C]_i$ are respectively the stable or radioactive carbon stock (i.e. concentration of ^{12}C or ^{14}C expressed per unit soil surface, e.g. mol m^{-2}) and concentration (i.e. concentration of ^{12}C or ^{14}C expressed per unit of mass, e.g. mol kg^{-1}) of a compartment i, χ i is the compartment areal density (i.e. mass per unit soil surface, e.g. kg m^{-2}), $TC^p_{i,l}$ are the mass transfer fluxes (e.g. mol.m^{-2} d^{-1}) from the compartment i to the compartment l through the processes p. The two terms on the right side of the Eq. (1) is the sum of the inputs and outputs of the processes p and q and other interacting j and k compartments. At the initial time, the concentration (or activity) of all compartments is zero.