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# Intercomparison of model predictions of <sup>14</sup>C concentrations in agricultural plants following acute exposures to airborne <sup>14</sup>C

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### 1 1. Introduction

2 Carbon-14 (<sup>14</sup>C) is a naturally occurring beta-emitting radionuclide with a maximum energy of 156 KeV and a half-life of 5730 years. The main natural source of <sup>14</sup>C on Earth is the action of cosmic rays on atmospheric nitrogen and it is present 3 in the atmosphere at a specific activity of around  $242 \pm 6$  Bq kgC<sup>-1</sup> (Roussel-Debet et al., 2006). <sup>14</sup>C is also formed as the 4 5 by-product of nuclear power generation and nuclear fuel reprocessing plants and is produced in nuclear reactors by the neutron capture of nitrogen, carbon or oxygen present as components of fuel, coolant, structural materials and cladding 6 7 metals. The radiological assessment of <sup>14</sup>C is important because it has a long half-life, prolonged persistence, and high mobility in the environment (Aulagnier et al., 2012). Under normal operational releases from nuclear facilities, <sup>14</sup>C is 8 9 assumed to behave similar to its stable analogue, <sup>12</sup>C (IRSN 2012).

<sup>14</sup>C follows the carbon cycle and its transfers between two compartments of the environment are commonly evaluated 10 under the assumption that the isotopic ratio between <sup>14</sup>C and stable carbon between the organism and the surrounding 11 environment is maintained (IRSN 2012). The carbon cycle in the soil-plant terrestrial environment is summarized in 12 Figure 1. The behaviour of atmospheric carbon dioxide (<sup>14</sup>CO<sub>2</sub>) is that it can be incorporated into food webs via 13 photosynthesis by primary producer organisms. Indeed, <sup>14</sup>C is incorporated into plants mainly by photosynthesis. CO<sub>2</sub> 14 emissions from soil organic residue mineralization and root respiration tend to increase the concentration of  $CO_2$  in the air 15 above the plant canopy. The daily flux of CO<sub>2</sub> emitted by the soil seems to contribute around 10% to the total carbon 16 17 assimilated by the leaves during photosynthesis (IRSN, 2012).

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Figure 1. Carbon cycle in the soil-plant-animal system (IRSN, 2012).

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Environmental modelling of the transfer of <sup>14</sup>C in the terrestrial soil-plant environment is complex because its transfer 20 responds rapidly to changes in weather and plant conditions. In order to improve the assessment of the impact of <sup>14</sup>C doses 21 from routine or accidental releases, it is necessary to improve the understanding and modelling of the behaviour of this 22 radionuclide along the food chain. The EMRAS program of the International Atomic Energy Agency (IAEA) 23 24 (http://www-ns.iaea.org/projects/emras/), which ran from 2003 to 2007 (EMRAS I) and 2009 to 2011 (EMRAS II) had a 25 focus on the improvement of environmental transfer models and the development of reference approaches to estimate the radiological impact on humans as well as flora and fauna (see Barry et al., 1999, to illustrate the work carried out for <sup>3</sup>H). 26 This program collected data to test and compare models. It also supported the development of an international consensus 27 on radioecological modelling approaches and parameter values. In this context, a certain number of model-model and 28 model-measurement intercomparison exercises in the field of radioecological modelling have already been carried out at 29 the international level. The EMRAS program, in particular, included a 'Tritium and <sup>14</sup>C Working Group' (TCWG), which 30 aimed to test the ability of <sup>14</sup>C (and tritium) transfer models in the environment to reproduce observations (IAEA, 2014). 31

32 These observations were organized in the form of nine scenarios to facilitate the model-measure intercomparison exercise 33 (Barescut et al., 2009; Yankovich et al., 2008).

In parallel, the TOCATTA- $\gamma$  model was developed at IRSN to simulate and evaluate the transfer of tritium and <sup>14</sup>C for a 34 grassland ecosystem by comparing it with measurements obtained during various in situ experiments (Aulagnier et al., 35 2012; Aulagnier et al., 2013; Le Dizès et al. 2017). Following on from the work on the grassland ecosystem, a 36 complementary project was initiated by IRSN to better quantify the transfer of <sup>14</sup>C to leafy vegetables, fruits and roots. In 37 this context, the present study illustrates the extensive application of the TOCATTA- $\gamma$  model to real case scenarios, and 38 its comparison with other existing international datasets and models for routine releases of <sup>14</sup>C to the environment. The 39 40 article is organized as follows: in the following section, the theoretical aspects of the real-case scenario, called the "potato scenario", are briefly described, along with a conceptual overview of the TOCATTA- $\gamma$  model including its 41 parameterization to this dedicated scenario and the development of modules. The third section deals with a test and an 42 43 evaluation of the results of the TOCATTA- $\gamma$  model by comparing them to existing models and experimental 44 measurements. The main results of the model and their implications for the future are discussed in the last section. 45

#### 46 2. Material and methods

#### 47 2.1 Potato Scenario description and participants

The scenario involves an experiment conducted at Imperial College, London (IAEA, 2008). About 100 pots were prepared 48 with 2-3 potato plants in each pot. The plants were fumigated in a wind tunnel with <sup>14</sup>CO<sub>2</sub> (under practical atmospheric 49 boundary layer conditions) and held at six different stages (P1-P6) of their growth cycle for 10-hours. The wind tunnel is 50 51 capable of holding thirty pots. The major meteorological details for each stage experiment and the concentration of air  $^{14}$ C 52 activity in the wind tunnel during fumigation are described in Tables 2-3(a, b) respectively. Randomly selected plants were collected after fumigation to assess the <sup>14</sup>C activity concentration and the remaining plants were transferred to the 53 field for further sampling before harvest (IAEA, 2008). Concentrations of <sup>14</sup>C activity in potato tissues mostly decreased 54 after exposure in the P1 and P2 experiments due to translocation to newly forming tissues, particularly edible tuber 55 tissues. Tuber development began after P3 exposure and the edible tubers had the largest concentrations of all plant tissues 56 in studies of P3 to P6 experiments until harvest. The measured activity concentrations were considered for the inter-57 comparison exercises of the <sup>14</sup>C model within the EMRAS program. 58

59 Four participants from various countries submitted their model results for the potato scenario (Table 1) (IAEA, 2008), with the TOCATTA  $(-\gamma)$  model being added externally for the inter-comparison exercise. Specifically, modelers were 60 61 asked to estimate the concentration of  ${}^{14}C$  in leaves and tubers for the P1, P3 and P6 experiments. The TOCATTA (- $\gamma$ ) model used the results of the participating models to test and validate the estimates for this scenario. The models have 62 adopted different approaches to simulate the transfer of <sup>14</sup>C from the atmosphere to plants (IAEA, 2008) depending on 63 their scope and objectives. Table 4 provides a brief description of the different modelling approaches and assumptions for 64 their application to the potato scenario. 65

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# Table 1 Participants in the Potato Scenario (IAEA, 2008). TOCATTA (-χ) is added at the end of the list for the current intercomparison exercise.

Participant	Affiliation	Model name
F. Siclet	Electricite de France, France	EDF
P. Kennedy	Food Standard Agency, UK	FSA
A.Melintescu and	National Institute For Physics and Nuclear	WOFOST
D. Galeriu	Engineering, Romania	SCOTTISH
S. Uchida et al.	National Institute of Radiological Sciences,	UTTY
	University of Kyoto and Y first Inc. Japan	UTTY revised
Séverine Le Dizès	Institute for Radioprotection and Nuclear Safety, France	TOCATTA(-χ)*

(\*) TOCATTA (- $\chi$ ) model is added to the list externally for the current intercomparison exercise. It was not identified as a participant in the TCWG framework.

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Table 2 Temperature (°C) and Photosynthetically Active Radiation (PAR, W/m<sup>2</sup>) during fumigation in each experiment (IAEA, 2008).

Experiment	Temperature range (°C)	Range in PAR (W/m <sup>2</sup> )
P1	23-27	70-150
P2	21-26	50-160
P3	20-23	40-160
P4	19-24	30-130
P5	19-13	30-130
P6	17-20	30-130

Table 3a Fumigation schedule of potato plants for the experiments (IAEA, 2008).

Experiment	Fumigation date	Time of fumigation (days after
		sowing)
P1	Aug 25, 1995	21
P2	Sep 7, 1995	33
P3	Sep21, 1995	47
P4	Oct 5, 1995	61
P5	Oct 18, 1995	74
P6	Nov 2, 1995	89

8	7	

<b>P1</b>		P2		<b>P3</b>		P4		P5		<b>P6</b>	
Time	Air	Time	Air	Time	Air	Time	Air	Time	Air	Time	Air Conc.
(min)	Conc.	(min)	Conc.	(min)	Conc.	(min)	Conc.	(min)	Conc.	(min)	$(Bq/m^3)$
	$(Bq/m^3)$		$(Bq/m^3)$		$(Bq/m^3)$		$(Bq/m^3)$		$(Bq/m^3)$		
32	65121	32	47090	31	68339	31	55009	30	57453	30	30450
99	43715	99	29804	100	42376	98	34387	97	36612	96	21067
166	21521	166	16279	167	24373	165	18999	163	19576	162	12966
233	12095	233	8297	236	11749	230	10269	236	9906	228	7152
300	6577	301	4405	303	6361	294	5774	304	5028	295	4086
368	3667	369	2490	371	2983	360.5	3359	370	2858	361	2461
435	2325	438	1393	438	1827	430.5	1686	436	1646	426	1452
501	1460	505	801	504	839	496.5	985	501	954	492	900
569	701	570	565	570	694	567	651	568.5	607	566	507

Table 3b Concentration of  ${}^{14}C$  (Bq/m<sup>3</sup>) in the air above the potatoes in the wind tunnel (IAEA, 2008).

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Model/Model description & assumptions	Type of model	Photosynthesis	Principles and other simplifying assumptions
<b>EDF</b> (IAEA, 2008)	A dynamic model developed to evaluate radionuclide concentrations in the aquatic and terrestrial environment resulting from liquid discharges. Specifically for potatoes two phases are considered: - Vegetative stage (growth of shoots) - Filling stage (tuber growth)	<ul> <li>The incorporation of <sup>14</sup>C in plants is mainly from photosynthetic carbon assimilation.</li> <li>The net photosynthetic carbon assimilation rate is a function of leaf biomass and corresponds to the plant's total growth rate.</li> <li>The photosynthetic daily net rate is taken as 0.0495gC /g leaf of dry matter.</li> </ul>	<ul> <li>Translocation fraction to tubers during vegetative stage and filling stage: 0.10 and 0.50.</li> <li>Fractional carbon content per unit of dry biomass in leaves and tuber is 45%.</li> <li>The air concentration of 0.19gC/m<sup>3</sup> CO<sub>2</sub> is taken as a constant parameter value during fumigation.</li> </ul>

<b>FSA</b> (IAEA, 2008)	<ul> <li>A dynamic, numerical, compartment model.</li> <li>The biological compartments include internal leaf, internal stems, roots, internal fruit, plant water and energy storage.</li> <li>Environmental compartments include soil water, soil organic material and sink.</li> </ul>	<ul> <li>The rate of carbon uptake during daylight hours is controlled by photosynthesis.</li> <li>The assimilation factor for plants is uniformly distributed in the range of 0.4-0.6.</li> <li>The fraction of CO<sub>2</sub> recycled during photosynthesis is log- normally distributed in the range of 0.04-0.3.</li> </ul>	<ul> <li>The biomass fraction in the energy store is log-uniformly distributed in the range of 0.001-0.02.</li> <li>The dry to fresh weight ratio is triangularly distributed in the range of 0.05-0.3 and best estimated as 0.1.</li> </ul>
WOFOST (IAEA, 2008; Melintescu et al., 2013)	<ul> <li>A complex model and used the default potato parameters and parameters specific to a SCOTTISH cultivar.</li> <li>Plant growth is primarily influenced by its genotype and climate condition.</li> </ul>	The model uses the photosynthesis sub model that mainly depends on photosynthetically active radiation (PAR), leaf area index (LAI) and maximum leaf photosynthesis rate.	<ul> <li>The model is based on the following principles:</li> <li>The specific activity of <sup>14</sup>C transferred to the plant in a given time interval is the same as the average specific activity of the source over that interval.</li> <li>It was assumed in the case of potato experiments that under normal conditions, more than 90% of plant carbon comes from the atmosphere.</li> <li>In biochemical reactions occurring in the plant, the discrimination factor between <sup>14</sup>C and <sup>12</sup>C is closed to 1 (0.96 ± 0.02, Sheppard et al., 2005). As a result, modelling of <sup>14</sup>C transfer is the same as modelling stable carbon transfer.</li> </ul>
SCOTTISH (IAEA, 2008; Melintescu et al., 2013)	<ul> <li>A simple model considers dry matter production only.</li> <li>The model can be used together with the partition fractions and growth dilution to predict <sup>14</sup>C concentrations in plants.</li> </ul>		<ul> <li>Principles and the processes implemented in SCOTTISH and WOFOST models are same.</li> <li>Each of the processes can be modelled simply or at a process level.</li> </ul>

UTTY	A dynamic compartment	- The model assumes	The model considered the mean air
(IAEA, 2008)	model. It is made up of five	that the [Plant_inorg]	concentration that was obtained by
	compartments:	and [StemLeaf_org]	dividing the integrated air concentration by
	- One inorganic	compartments transfer	the exposure time of 600 min.
	compartment (whole	the photosynthates to	
	plant)	the tuber.	
	- Two organic	Where, [Plant_inorg]	
	compartments (stem-leaf-	compartment includes	
	root and tuber)	non-fixed (inorganic)	
	- Two environmental	carbon or organic	
	compartments (air and	carbon, which is easily	
	soil)	exchangeable with air.	
		[StemLeaf_org]	
		compartment includes	
		the stem, leaf, and root	
		with fixed	
		photosynthates.	
UTTY revised	The model is same as the		The UTTY revised model improved the
(IAEA, 2008)	UTTY model.		performance of the UTTY model by
			decreasing the external air concentration
			without changing the conceptual model.
ΤΟCΑΤΤΑ - χ	A dynamic compartment	Stomatal resistance and	- The water content of the soil at the
	model, with three	leaf photosynthesis rate	saturation point is 0.4 m <sup>3</sup> .m <sup>-3</sup>
	compartments :	are calculated on an	- The atmospheric CO <sub>2</sub> concentration is
	- Shoot structural dry	hourly basis based on	398 ppm.
	matter	Huntingford model and	
	- Root structural dry matter	Farquhar model,	
	- Sap (substrate)	respectively.	

### 90 2.2 TOCATTA-χ model

The TOCATTA- $\chi$  model is a dynamic compartment model developed at the Institute of Radioprotection and Nuclear 91 Safety (IRSN) to simulate the transfer of <sup>14</sup>C and Tritium in grassland ecosystems from nuclear facilities operating under 92 normal or accidental conditions (Aulagnier et al., 2013; Le Dizès et al., 2017). The conceptual framework used by the 93 model is summarized in the form of an interaction matrix (see Limer et al., 2015; Table 2). The model operates on an 94 hourly basis and takes into account sub-daily processes for potential application to operational and accidental releases. 95 The model takes into account the main ecophysiological processes (e.g., mainly photosynthesis, biological growth, 96 respiration, etc.), radioecological processes (e.g., radioactive decay) and meteorological data (e.g., mainly the temperature 97 of the air, air humidity, photosynthetically active radiation (PAR)). The model requires input data of <sup>14</sup>C and HTO activity 98 concentrations in air and some growth parameters, i.e., initial and harvested biomass densities. It also requires an initial 99 value for the <sup>14</sup>C activity at the beginning of the simulation (Aulagnier et al., 2012). The initial carbon content is directly 100 proportional to a function of the shoot biomass density and root densities (Le Dizès et al., 2012; Aulagnier et al., 2013). In 101 addition, stomatal resistance and leaf photosynthesis rate are calculated on an hourly basis from the process-based 102

- modeling approaches of Huntingford (Huntingford et al., 2015) and Farquhar (Farquhar et al., 1982), respectively. More detailed information on the TOCATTA- $\chi$  model is given by (Le Dizès et al., 2017) and in the Supplementary Material B.
- 105
- 106 2.2.1 Model parameterization

107 The TOCATTA- $\chi$  model was parameterized for application to the scenario (see Table 5). Experiments P1, P3 and P6 108 were selected for inter-comparison exercises on three of the six experiments, because these experiments cover all the 109 important stages of growth of a potato plant, i.e., the development and translocation to different plant organs such as 110 leaves, stem and tubers. As a result, the following assumptions were made for the simulation:

- V<sub>cmax</sub> and V<sub>omax</sub> are the kinetic parameters, used by the Farquhar photosynthesis model, they define the maximum rate of carboxylation and photosynthesis oxygenation. These parameters are used to estimate the rate of photosynthesis (A, (µmol m<sup>-2</sup> s<sup>-1</sup>)) when Rubisco concentration is limited (Harley et al., 1985; Medlyn et al., 2002; Farquhar et al., 1980), as described by the following equation :
- 115

$$A = V_{C \max} \left[ \frac{C - \Gamma_*}{C + K_c (1 + \frac{O}{K_o})} \right] - R_d$$
(1)

116

Where, C (Pa) and O (kPa) are the intercellular partial pressures of CO<sub>2</sub> and O<sub>2</sub>, respectively.  $R_d$  (µmol m<sup>-2</sup> s<sup>-1</sup>) is residual respiration.  $V_{cmax}$  (µmol m<sup>-2</sup> s<sup>-1</sup>) and  $V_{omax}$  (µmol m<sup>-2</sup> s<sup>-1</sup>) are the maximum rate of carboxylation and photosynthesis oxygenation, respectively.  $K_c$  (µmol mol<sup>-1</sup>) and  $K_o$  (mmol mol<sup>-1</sup>) are the Michaelis constants for CO<sub>2</sub> and O<sub>2</sub>, respectively.  $\Gamma_*$  (µmol mol<sup>-1</sup>) is CO<sub>2</sub> compensation point in the absence of residual respiration and expressed as (Farquhar et al., 1980) follows:

$$\Gamma_* = 0.5 \left[ \frac{V_{o \max} K_c}{V_{c \max} K_o} \right] . 0$$
<sup>(2)</sup>

- To estimate the rate of electron transport (J), the curvature of the light reaction curve (θ) is assumed to be 0.90 (Bernacchi, 2003; Medlyn et al., 2002; eq. 4).
- The extinction coefficient quantifies the reduced amount of light impinging on the plant. The coefficient relies mainly on the canopy arrangement and is used in the photosynthesis model to measure the light use efficiency. The coefficient value for potatoes is presumed to be 0.48 (Van Heemst, 1988).
- For each experiment, time series of varying shoot and root growth rate densities have been reconstructed to assess the plant biological growth by using the available data on biomass dynamics and the dry weight fraction from the description of the scenario (cf. Supplementary Material A).
- 130 131

Table 5 Parameterization of the TOCATTA	-χ model	to the	potato	scenario
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Parameters	Definition and	Parameter values for each experiment & references					
	Related Process or Variable	P1	Р3	P6			
V <sub>cmax</sub> (Farquhar	The maximum rate	31.46 µmol/m <sup>2</sup> /s	26.53 µmol/m <sup>2</sup> /s	29.94 µmol/m²/s			
Model)	of carboxylation in	( Medlyn et al., 2002;	( Medlyn et al., 2002;	( Medlyn et al., 2002;			
	Photosynthesis	Harley et al., 1985)	Harley et al., 1985)	Harley et al., 1985)			
V <sub>omax</sub> (Farquhar	The maximum	0.21*V <sub>cmax</sub>	$0.21*V_{cmax}$	0.21*V <sub>cmax</sub>			
Model)	oxygenation velocity	(Harley et al., 1985)	(Harley et al., 1985)	(Harley et al., 1985)			

Light attenuation in	0.48	0.48	0.48
canopy	(Van Heemst 1988)	(Van Heemst 1988)	(Van Heemst 1988)
Maximum rate of	122.5 μmol/m <sup>2</sup> /s	110.67 μmol/m <sup>2</sup> /s	118.56 µmol/m²/s
electron transport in	(Bernacchi, 2003)	(Bernacchi, 2003)	(Bernacchi, 2003)
photosynthesis			
	0.6 L/kg DW	0.6 L/kg DW	0.6 L/kg DW
	(Peterson and Davis	(Peterson and Davis	(Peterson and Davis
	2002)	2002)	2002)
	76%	76%	76%
	(Van Heemst 1988)	(Van Heemst 1988)	(Van Heemst 1988)
	Light attenuation in canopy Maximum rate of electron transport in photosynthesis	Light attenuation in canopy0.48 (Van Heemst 1988)Maximum rate of electron transport in photosynthesis122.5 μmol/m²/s (Bernacchi, 2003)0.6 L/kg DW (Peterson and Davis 2002)0.6 L/kg DW (Peterson and Davis 2002)	Light attenuation in canopy0.48 (Van Heemst 1988)0.48 (Van Heemst 1988)Maximum rate of electron transport in photosynthesis122.5 μmol/m²/s (Bernacchi, 2003)110.67 μmol/m²/s (Bernacchi, 2003)0.6 L/kg DW (Peterson and Davis 2002)0.6 L/kg DW (Peterson and Davis 2002)0.6 L/kg DW (Peterson and Davis 2002)76% (Van Heemst 1988)76% (Van Heemst 1988)1988)

\*Water equivalent factor is the amount of water generated through the combustion of the dry material in the sample.

Weq (water equivalent factor (L/kg)) = [(percent protein × 0.07) + (percent fat × 0.12) + (percent carbohydrate × 0.062)]/ 100 × (1/fraction of mass of water that is hydrogen)

Where, 0.07 = fraction of hydrogen in proteins, 0.12 = fraction of hydrogen in fats, 0.062 = fraction of hydrogen in carbohydrates, 2/18 = fraction of mass of water that is hydrogen.

132

### 133 2.2.2 Module Development

This section describes the main module that underlies our modelling approach (the modules are the building blocks of the 134 overall model). The evaluation of the concentration of <sup>14</sup>C in edible organs such as potato tubers is not present in the 135 TOCATTA- $\gamma$  model, therefore a module had to be specifically developed. The module is mainly related to the source 136 compartments, to the transfer pathway and to the pre-estimated concentration of  ${}^{14}C$  in the leaves via the TOCATTA- $\gamma$ 137 model. It was assumed that the transfer of photosynthates occurs directly from leaves to tubers and depends mainly on the 138 growth stage of the tubers. A transfer factor equation was taken from the UTTY's potato model under EMRAS, TCWG 139 (IAEA, 2008). The most relevant compartments and the transfer pathway involved for the development of the module are 140 illustrated in Figure 3. The [Stemleaf organic] compartment consists of photosynthates fixed in the stem, leaves, and root. 141 142 Due to dark respiration, the amount of photosynthate in this compartment decreases over time and most of the photosynthates are carried to the [Tuber organic] compartment. Likewise, photosynthates in the [Tuber organic] 143 compartment also decrease over time by dark respiration only. 144



Figure 3. Transfer pathways of <sup>14</sup>C between air, Stemleaf\_organic and Tuber\_organic compartments.

145

146 The relocation of the photosynthates from [Stemleaf organic] compartment to [Tuber\_organic] compartment is thus 147 calculated by a transfer factor equation ( $k_{le}$ ) (IAEA, 2008) as follows:

$$k_{le} = \{ (1 - \gamma) . \alpha_{E_{org}} . (1 + \beta_{E_{res}}) . dW_E / dt ) ) / ((W_P - W_E) . \alpha_{org}) \}$$
(3)

Here,  $\alpha_{\text{org}}$  is the ratio of the weight of organic carbon to the weight of the whole plant without the tuber ( $\alpha_{\text{org}} = 0.37$ ) or to the weight of the tuber ( $\alpha_{\text{E}_{org}} = 0.40$ ),  $\beta_{\text{E}_{res}}$  is the ratio of organic carbon used in respiration to the tuber alone ( $\beta_{\text{E}_{res}} = 0.15$ ) (IAEA, 2008). W<sub>P</sub>, W<sub>E</sub> (g) are the total dry weight of plants and tubers.

151  $\gamma$  is an empirical relationship derived by analyzing the growth curve from the data provided under IAEA potato scenario 152 description (eq. 3). It depends on different stages of tuber growth (IAEA, 2008), as described in the following equation:

$$\gamma = 1 - (dW(t))/dW_{max}) * 0.7$$
 (4)

Here,  $dW(t) = differential increase in tuber weight (g) in hourly time steps, <math>dW_{max} = maximum differential increase in tuber weight (g), dW(t) and dW_{max} have been estimated from logistic growth curves (cf. Supplementary Material C).$ 

155

Fundamentally, the following equation calculating <sup>14</sup>C concentration activity per gram of dry matter in potato tubers (Bq/g dm) is specifically developed for the TOCATTA- $\chi$  model, as a function of the transfer factor equation (see eq. 3) and <sup>14</sup>C concentration in leaves estimated by TOCATTA- $\chi$  model.

$$[C14]_{tuber} = k_{le} * [C14]_{leaves}$$
<sup>(5)</sup>

Here,  $k_{le}$  is a unitless parameter of transfer pathway between the stem and tuber organic compartment, [C14] <sub>leaves</sub> (Bq/g dm) is the <sup>14</sup>C concentration activity in leaves.

161

### 162 **3. Results and Discussion**

163 3.1 Concentration of <sup>14</sup>C in leaves for P1, P3 and P6 experiments

164 The results of the TOCATTA- $\chi$  model are compared to P1 measurements and other existing model results (cf. Figure 4). 165 On one hand, it is observed that the TOCATTA- $\gamma$  model is larger by up to a factor of 2.5 than the measurements and is somewhat underestimated at 21 days and 97 days (Figure 4). This could be explained by the dilution in growth due to the 166 increase in plant and tuber biomass over time. Indeed, some simulated processes (growth and senescence) of the 167 TOCATTA-χ model are very sensitive to the biomass densities which are used to estimate the growth rate of the plant 168 (Aulagnier et al., 2013). On the other hand, the results of the UTTY and EDF models overestimated the measurements by 169 a factor of up to 20 and 6, while the predictions of the WOFOST and SCOTTISH models underestimated the 170 measurements by up to a factor of 5. In addition, the RMSE of TOCATTA- $\gamma$  compared to the other models (SCOTTISH, 171 172 UTTY and EDF) is reduced by a factor ranging from 1 to 16 (cf. Table 6a). Because the TOCATTA- $\chi$  model has the advantage of data disclosure from the scenario description, it helps to better determine the biomass fraction in leaves, 173 resulting in a better prediction of the <sup>14</sup>C concentration in leaves for P1 fumigation as compared to other models (cf. 174 Supplementary Material A). 175

Likewise, the results of the TOCATTA- $\gamma$  model were compared with the measurements and with other models for the P3 176 experiment (cf. Figure 5). The EDF model predictions overestimated the measurements by a factor of 3. The FSA, 177 WOFOST, and SCOTTISH models overestimated the measurements by a factor of 6 to 10. At the same time, the 178 predictions of the TOCATTA- $\chi$  model are within the measurement uncertainty limit and overall the RMSE of the 179 TOCATTA- $\chi$  model is decreased by up to a factor of 8 compared to all other existing models (cf. Table 6a). In the 180 TOCATTA- $\chi$  model, the average of the hourly time steps on a daily basis was used to provide the closest fit of the 181 measurements. In addition, the TOCATTA- $\gamma$  model versus measurements was underestimated by up to a factor of 10 at 182 the age of 47 days (Figure 5) due to the rough calculation of the initial carbon content of the leaves. This could cause a 183 variation in the biological growth of the plant during the simulation. The TOCATTA- $\gamma$  model is sensitive to the flux of 184 photosynthesis. The temperature (18-23°C) was linearly interpolated to determine the carboxylase and oxygenase 185 activities, which were then used by the photosynthesis model to estimate  $CO_2$  uptake in plants. 186



Figure 4. Comparison between model prediction and observations for the concentration of <sup>14</sup>C (Bq/g dm) in leaves following P1 fumigation.

189 In addition to P1 and P3, the observations and model results for the P6 experiment are shown in Figure 6. SCOTTISH and FSA underestimated the measurements by up to a factor of 2 and 6. The EDF model overestimated by a factor between 2 190 and 7. TOCATTA- $\gamma$  works slightly less well than the other models compared to the measurements. The RMSE of the 191 TOCATTA-y model exceeds the measurements by a factor between 4 to 12 (cf. Figure 6). The fumigation for experiment 192 P6 was scheduled 89 days after sowing. At this stage, the growth of the plant is complete, including the presence of one or 193 two large tubers. It is assumed that after exposure, the transfer of <sup>14</sup>C to the tubers increases rapidly with the age of the 194 plant. These tubers may contain a higher <sup>14</sup>C concentration than the leaves and contribute to a significant dose rate. The 195 TOCATTA-y model greatly overestimated the experimental results. This could be explained by insufficient 196 197 parametrization of biological growth and maintenance respiration. Indeed, photosynthesis flux and biological growth rate are the key parameters of the TOCATTA- $\gamma$  model. These parameters are mainly influenced by the leaf index area (LAI), 198 199 photosynthetically active radiation (PAR), maximum rate of carboxylation (V<sub>cmax</sub>), local leaf temperature, initial carbon content, moisture content, and biomass densities. It was assumed that the plants are developed under normal physiological 200 conditions - based on the underlying assumption that water and nutrients do not limit growth - but this was not the case in 201 the scenario. In general, plant growth is influenced by climatic conditions. In the scenario, the seed potatoes were planted 202 much later (August 4, 1995) than usual in England, which prevented the seedlings from completing their normal growth, 203 204 resulting in a high partition for the P6 fumigation (IAEA, 2008). In the meantime, no meteorological information is 205 available in the scenario description for the year 1995. Therefore, it was difficult to estimate parameters with precision considered in the formalization of time-dependent ecophysiological processes such as photosynthesis, biological growth, 206 207 and respiration.



Figure 5. Comparison between model prediction and observations for the concentration of <sup>14</sup>C (Bq/g dm) in leaves following P3 fumigation.



Figure 6. Comparison between model prediction and the observations of the concentrations of <sup>14</sup>C (Bq/g dm) in leaves following P6 fumigation.

209 3.2 Concentration of  ${}^{14}$ C in tubers (at harvest)

Concentrations in tubers during harvest are more radiologically significant than those in leaves because the tubers are the 210 edible part of the plant and are more prone to a higher dose rate, resulting in a higher dose assessment in humans (IAEA, 211 212 2008). It can be seen from figure 7 that the EDF model overestimated the measurements by up to a factor of 6. UTTY is 213 substantially overestimated by a factor between 3 and 12. The WOFOST and Scottish models did not predict tuber contamination for fumigation P1, underestimated for fumigation P3, and then overestimated by a factor of 6 for 214 experiment P6. The UTTY revised model made better predictions but still overestimated by up to a factor of 4. All the 215 models predicted that the <sup>14</sup>C concentrations in the tubers were higher than those in the leaves. This could be due to 216 compensatory errors, at least for the models that greatly overestimated the initial contamination of the potato plant (IAEA, 217

2008; Melintescu et al., 2013). For experiment P3, the result of the TOCATTA- $\chi$  model is within the uncertainty limit of the measurements (cf. figure 7) and the RMSE of TOCATTA- $\chi$  is reduced by a factor between 1-5 as compared to the other models (WOFOST, Scottish, EDF, UTTY revised) (cf. Table 6b), while with the UTTY model it is decreased by up to a factor of 34 due to the different modeling approach. TOCATTA-y predictions are overestimated for P1 and P6 experiments by up to a factor of 3 and 11, respectively. This could be owing to insufficient respiration, which is one of the important processes that govern tuber growth and maintenance and has a considerable impact on <sup>14</sup>C allocation in tubers. Nevertheless, it was very difficult to draw a specific conclusion on the expected results of all participating models due to the large uncertainties associated with the experimental measurements and the lack of metrological information available outside the wind tunnel. 



Figure 7. Comparison between model prediction and observations of the concentrations of <sup>14</sup>C (Bq/g dm) in tuber during the final harvest for the experiments P1, P3 and P6, respectively.

Table 6a Comparison of Root Mean Square Errors (RMSE) of the TOCATTA- $\chi$  model with other models for P1, P3 and P6 fumigation of leaves.

Experiment P1		Experiment P3		Experiment P6	
Model	RMSE	Model	RMSE	Model	RMSE
ΤΟϹΑΤΤΑ-χ	376.98	ΤΟϹΑΤΤΑ-χ	132.37	ΤΟϹΑΤΤΑ-χ	218.28
WOFOST	313.10	WOFOST	1092.58	FSA	40.23
UTTY	6325.57	FSA	188.43	SCOTTISH	34.26
SCOTTISH	391.00	SCOTTISH	440.00	EDF	146.59
EDF	2943.25	EDF	388.21		

Experiment P1		Experiment P3		Experiment P6	
Model	RMSE	Model	RMSE	Model	RMSE
ΤΟϹΑΤΤΑ-χ	36.42	ΤΟϹΑΤΤΑ-χ	77.4	ΤΟϹΑΤΤΑ-χ	449.4
WOFOST	Not predicted	WOFOST	86.11	WOFOST	243.16
UTTY	312.86	UTTY	2648.5	UTTY	107.63
SCOTTISH	Not predicted	SCOTTISH	85.8	SCOTTISH	215.70
UTTY revised	43.92	UTTY revised	85.25	UTTY revised	18.4
EDF	4.556	EDF	410.16	EDF	273.45

Table 6b Comparison of Root Mean Square Errors (RMSE) of the TOCATTA-χ model with other models for P1, P3 and P6 fumigation of tubers.

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### **4. Conclusions and perspectives**

238 The models that participated in this intercomparison exercise are the research tools intended for regulatory assessment or licensing applications. The real case scenario that was chosen in this study concerns the experiment carried out at Imperial 239 College, London, in which potato plants were fumigated with <sup>14</sup>CO<sub>2</sub> for 10 hours within a wind tunnel at six stages (P1 -240 P6) of the crop's growth cycle. The modelers applied various assumptions and parameters relevant to their respective 241 242 modeling approach (Melintescu et al., 2013). The TOCATTA- $\gamma$  model has an edge over other models in terms of data disclosure, allowing it to estimate <sup>14</sup>C concentrations in leaves and tubers better than other models. When compared to 243 other models, the Root Mean Square Error (RMSE) of the TOCATTA- $\gamma$  model is reduced by up to a factor of 8. For most 244 of the experimental data, the TOCATTA- $\gamma$  model and other model predictions for leaves and tubers are overestimated. It 245 246 has been assumed in many models, including TOCATTA- $\gamma$  that plant growth occurs under normal conditions - based on the underlying assumption that water and nutrients do not limit the development of plants - but that was not the case in the 247 current scenario. Indeed, due to late sowing (August 4), the plants could not complete their normal development, which 248 necessitated the complex processes of partition and translocation caused by the onset of weather conditions at the start of 249 the fall shortly after the exposure. Plant genotype is also important in determining  ${}^{14}C$  concentrations since the partitioning 250 of new photosynthate to leaves, stems, and tubers depends on the cultivar, as has been highlighted in several crop growth 251 studies (Melintescu et al., 2013). A high uncertainty was associated with the experimentally measured <sup>14</sup>C activities, 252 which were field variability in leaf properties and growth rate; hence the difficulty of drawing specific conclusions related 253 to the performance of the models. The discrepancy between the predictions of the TOCATTA- $\gamma$  model and the 254 measurements could be further reduced by considering reliable parameter values with respect to ecophysiological 255 processes. In addition, a contribution to the TOCATTA- $\gamma$  model was attempted through the development of a module 256 aimed at evaluating the concentrations of <sup>14</sup>C in edible organs such as potato tubers. It was observed that by using this new 257 module, the TOCATTA- $\chi$  model can predict the concentration of <sup>14</sup>C in tubers for the P1-P3 experiments and on average 258 259 reduces the RMSE by up to a factor of 5 compared to the other existing models. For the P6 experiment, the performance of the module was not good compared to the other models and observations due to inappropriate respiration and 260 translocation. The module is adequate for simple screening applications, but it might be improved in the future by adding 261 a few more compartments or transfer pathways to better capture complex processes for radiological assessments. 262

Several characteristics of the behavior of <sup>14</sup>C in the atmosphere-soil-plant continuum and effective approaches to 263 modelling processes require further consideration, such as night time release of <sup>14</sup>CO<sub>2</sub> by plants, soil emanation, and 264 transfer rate of <sup>14</sup>CO<sub>2</sub> from soil to tubers. To make such progress in the modeling of <sup>14</sup>C transfer processes (including 265 translocation), a good collaboration between experimenters and modelers is necessary. Such collaboration was made 266 possible, thanks to recent studies of the BIOPROTA project, an international collaborative forum (BIOPROTA, 2019), 267 268 which worked in parallel with the IAEA MODARIA II Working Group 6 (IAEA, 2016). These two studies aimed to improve the modelling of the biosphere for long-term safety assessments of radioactive waste facilities (IAEA, 2021). 269 Collaborative activities were organized as part of the BIOPROTA forum on long-term dose assessment for <sup>14</sup>C. The last 270

271 project of this forum focused on model-data and model-model comparisons based on experimental data provided by IRSN

relating to the deposition of <sup>14</sup>C released into the atmosphere towards pastures at the La Hague site in north-western

France (Limer et al., 2015). All of these activities have contributed to better justification and reduced uncertainties in  ${}^{14}C$ radioactive waste disposal assessments.

In summary, the work of the TCWG, illustrated here by the results of the potato scenario as well as these different working groups, helps to establish the degree of confidence that can be placed in the predictions of environmental  $^{14}$ C (and tritium) models, as well as the level of effort that may be required to make such estimates.

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