IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Faire avancer la sûreté nucléaire

Validation d’un modèle d’évaluation des transferts de carbone 14 et de tritium dans l’environnement

Pôle **R**adio**P**rotection - **ENV**ironnement
SErvice de recherche et d’expertise sur les **RIS**ques environnementaux

S. Le Dizès-Maurel, M. Simon-Cornu : IRSN Cadarache
D. Maro, D. Hébert, M. Rozet, L. Solier, D. Boust, : IRSN Cherbourg

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Pourquoi faire un « focus » sur ^{14}C et ^{3}H ?

✓ ^{14}C et ^{3}H sont les principaux radionucléides relâchés dans l'atmosphère par:

- Les réacteurs nucléaires
- L'usine de retraitement des déchets usés AREVA NC La Hague

✓ Dans le futur, ^{3}H sera rejeté significativement dans le cadre d'autres installations nucléaires:

- EPR (European Pressurized Reactor)
- ITER (International Thermonuclear Experimental Reactor)
- LMJ (Laser Megajoule)
- …

✓ ^{14}C et ^{3}H peuvent contribuer significativement à la dose à l'homme (par ingestion), malgré faibles coefficients de dose

✓ Incertitudes rémanentes :

- Sur les cinétiques de transferts du ^{14}C et ^{3}H dans les compartiments biotiques (plantes, animaux,…)

Etude VATO: VAlidation du modèle TOCATTA

Axes d’études

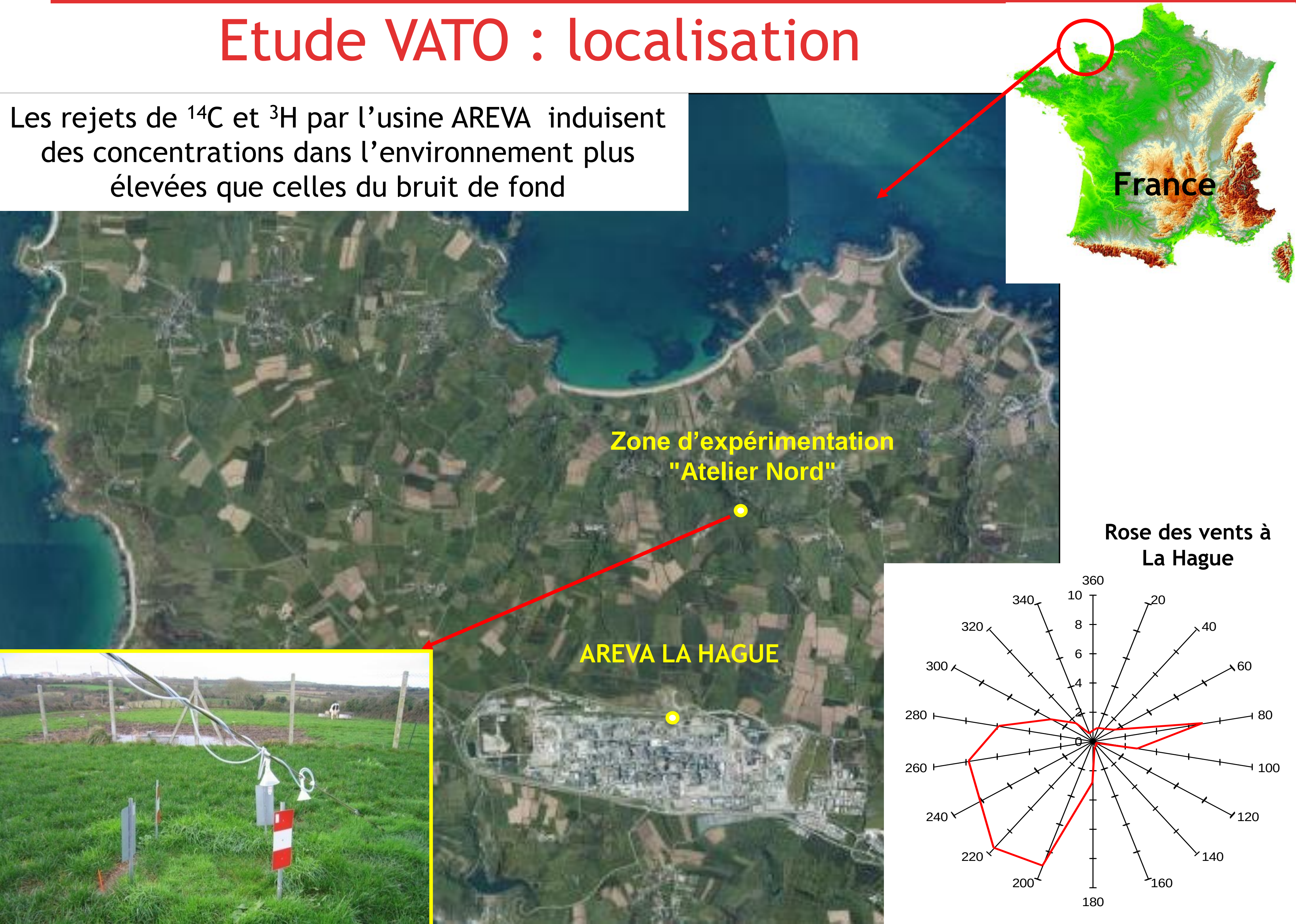
- Modélisation des transferts de ^{14}C et ^{3}H dans un écosystème terrestre: le modèle TOCATTA (module de SYMBIOSE)
- Campagne expérimentale

Objectifs

- Estimer les flux de ^{14}C et de ^{3}H dans un écosystème prairial (air, eau de pluie, herbe, matière organique et eau du sol), en relation avec :
  - l’évolution de la concentration dans l’air (jour/nuit) ;
  - les conditions météorologiques ;
  - les formes chimiques rejetées.
- Estimer les transferts de ^{14}C et de ^{3}H dans le lait de vache en fonction de son régime alimentaire

… pour progresser dans la compréhension des processus et disposer de données bien documentées pour « valider » le modèle TOCATTA

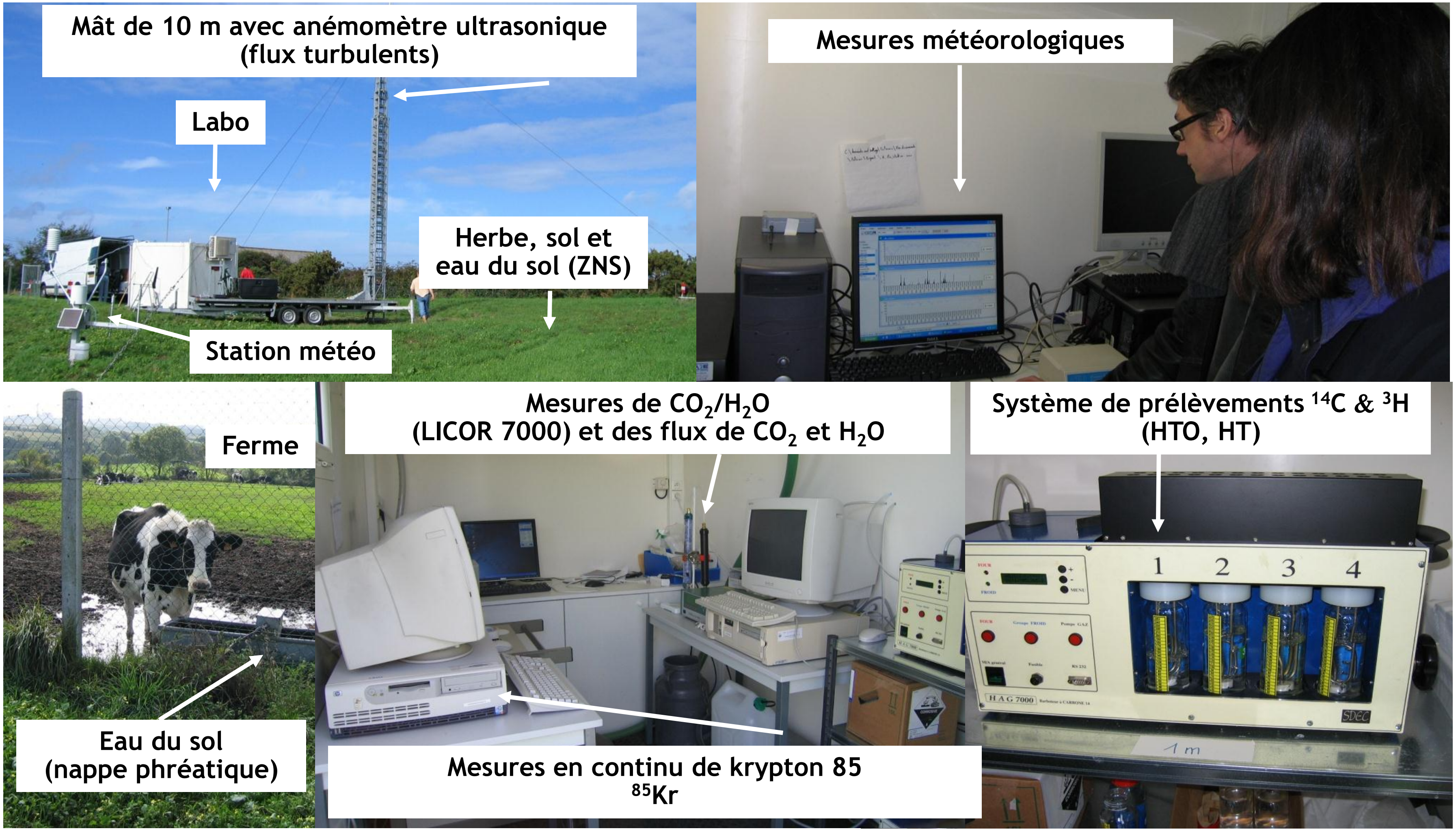
Etude VATO : localisation



Les rejets de ^{14}C et ^{3}H par l'usine AREVA induisent des concentrations dans l'environnement plus élevées que celles du bruit de fond

Rose des vents à La Hague

Etude VATO : laboratoire *in situ*



Mât de 10 m avec anémomètre ultrasonique (flux turbulents)

Labo

Herbe, sol et eau du sol (ZNS)

Station météo

Mesures météorologiques

Ferme

Mesures de CO_2/H_2O (LICOR 7000) et des flux de CO_2 et H_2O

Système de prélèvements ^{14}C & ^{3}H (HTO, HT)

Eau du sol (nappe phréatique)

Mesures en continu de krypton 85 ^{85}Kr

Modélisation des transferts de ^{14}C & ^{3}H

Caractéristiques & hypothèses du modèle TOCATTA

- Milieu d'exposition : écosystèmes terrestres agricoles (sol, plante & animal)
- Types de rejets : atmospherique et/ou liquide (irrigation par aspersion)
- Fonctionnement installation : normal et/ou accidentel
- Formes physico-chimiques des rejets : $^{14}CO_2$, HTO, (HT)
- Echelles de temps:
  - Pas de temps: jour
  - Durée: > 1 an(s)
- Modèle basé sur données empiriques + connaissances mécanistes sur cycle des éléments stables (C,H)
- Modèle dynamique
  - ✓ Basé sur des **courbes de croissance** de la biomasse des plantes, prédéfinies ou issues de données expérimentales
  - ✓ Basé sur hypothèse d'**équilibre isotopique** entre le flux d'assimilation photosynthétique par la végétation et l'air environnant, à chaque pas de temps

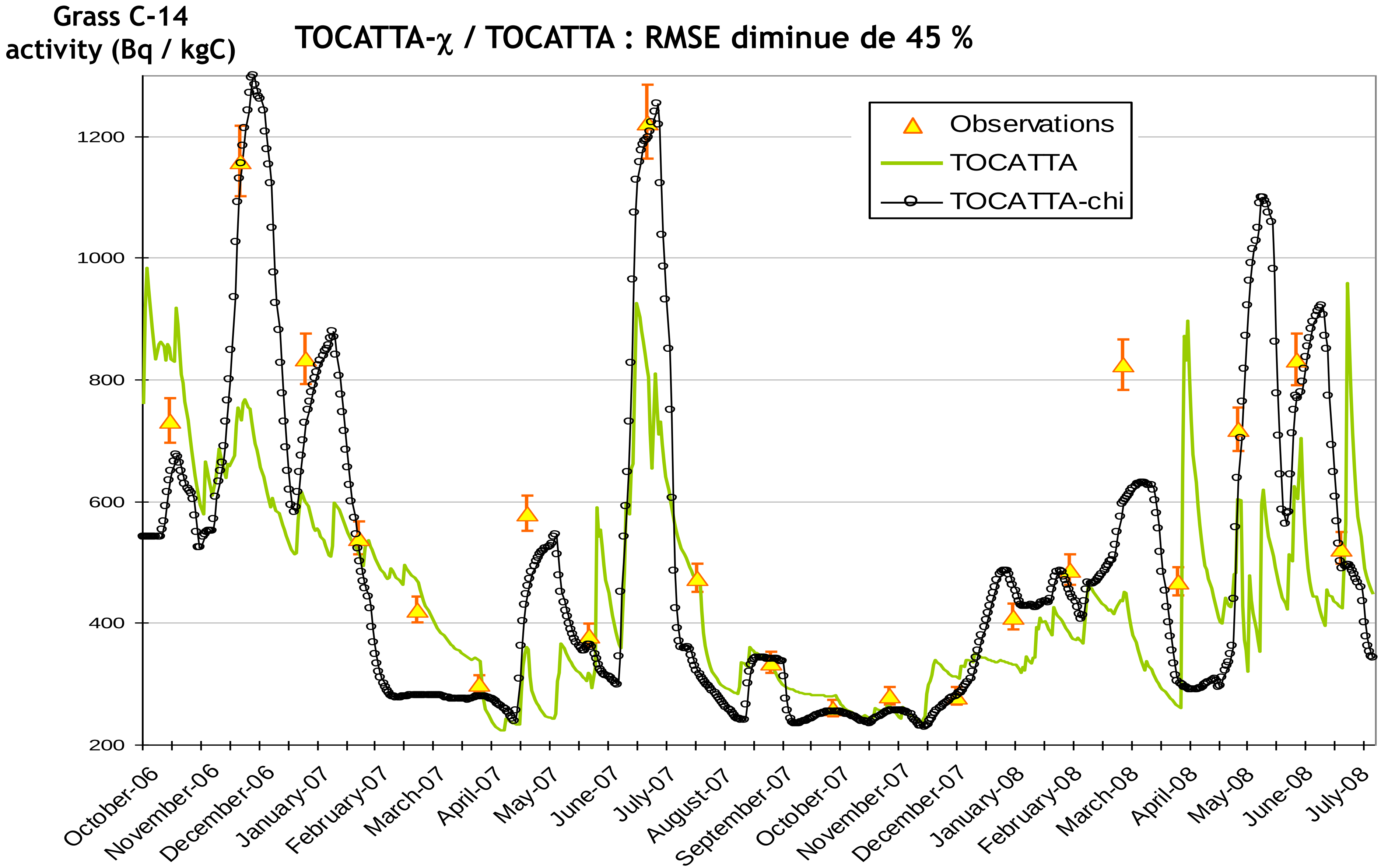
Comparaison modèle / mesures

TOCATTA:

✓Concentrations simulées plus faibles (jusqu'à 40 %) que les mesures

✓Variabilité entre les mois sous-estimée

TOCATTA-χ / TOCATTA : RMSE diminue de 45 %



Conclusions

- **Cas de rejets constants:**
  - pas de difficultés particulières à modéliser les transferts de ^{14}C et ^{3}H dans l'environnement terrestre (cf. « Specific Activity models »)

- **Cas de rejets chroniques (fct normal):**
  - Adéquation du modèle TOCATTA si variabilité infra-journalières des rejets est négligeable

- **Cas de rejets intermittents:**
  - Incertitudes du modèle TOCATTA-^{14}C dans l'application à un écosystème prairial (VATO) proche de l'usine AREVA La Hague (rejets intermittents)

→ Nécessité d'améliorer le modèle en termes de **cinétiques** de transfert du C (et ^{14}C) pour l'adapter à des situations de rejets et de météo variables

→ Nécessité d'intégrer la photosynthèse et les dynamiques de croissance des plantes, à pas de temps **plus fin**, en fonction de données agro-météorologiques locales

→ Développement du modèle **TOCATTA-χ horaire** pour la **prairie**, implémenté dans SYMBIOSE v2.1 (choix optionnel)

  - Prise en compte de la variabilité jour/nuit des rejets de ^{14}C

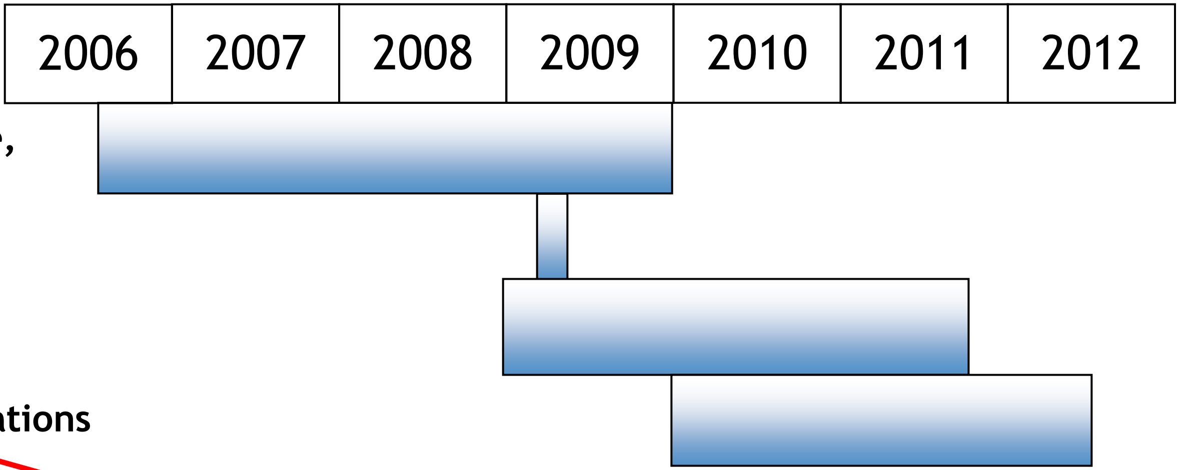
Perspectives

- Nécessité de poursuivre la validation du modèle TOCATTA-χ sur des jeux de données de ^{14}C indépendants obtenus sur d'autres écosystèmes agricoles

- Démarrage d'une **campagne expérimentale** à l'IRSN (2013-2017) pour l'étude des transferts du tritium sur le même écosystème prairial à proximité de l'usine AREVA NC La Hague

Agenda

VATO-^{14}C



Mesures dans échantillons d'herbe, air et sol

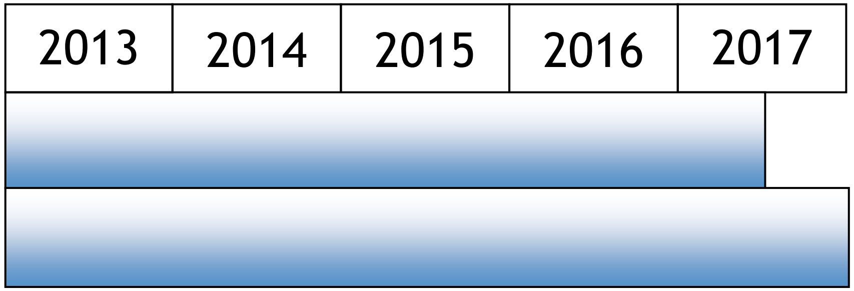
Mesures dans lait de vache

Comparaison modèle-mesures

Amélioration du modèle et publications

1. Le Dizès S., Maro D., Hebert D., Gonze M.-A , Aulagnier C. TOCATTA: a dynamic transfer model of ^{14}C from the atmosphere to soil-plant systems, (2012) J. Environ. Radioact. 105: 48-59.
2. Aulagnier C., Le Dizès S., Maro D., Hebert D., Lardy R., Martin R., Gonze M.-A. Modelling the transfer of ^{14}C from the atmosphere to grass : A case study in a grass field near AREVA-NC La Hague (2012) J. Environ. Radioact. 112, 52-59.
3. Aulagnier C., Le Dizès S., Maro D., Hebert D., Lardy R., Martin R. The TOCATTA-χ model for assessing ^{14}C transfers to grass: an evaluation for atmospheric operational releases from nuclear facilities (2013) J. Environ. Radioact. 120, 81-93

VATO-^{3}H



Mesures dans échantillons d'air, eau de pluie, herbe, sol et ZNS, nappe

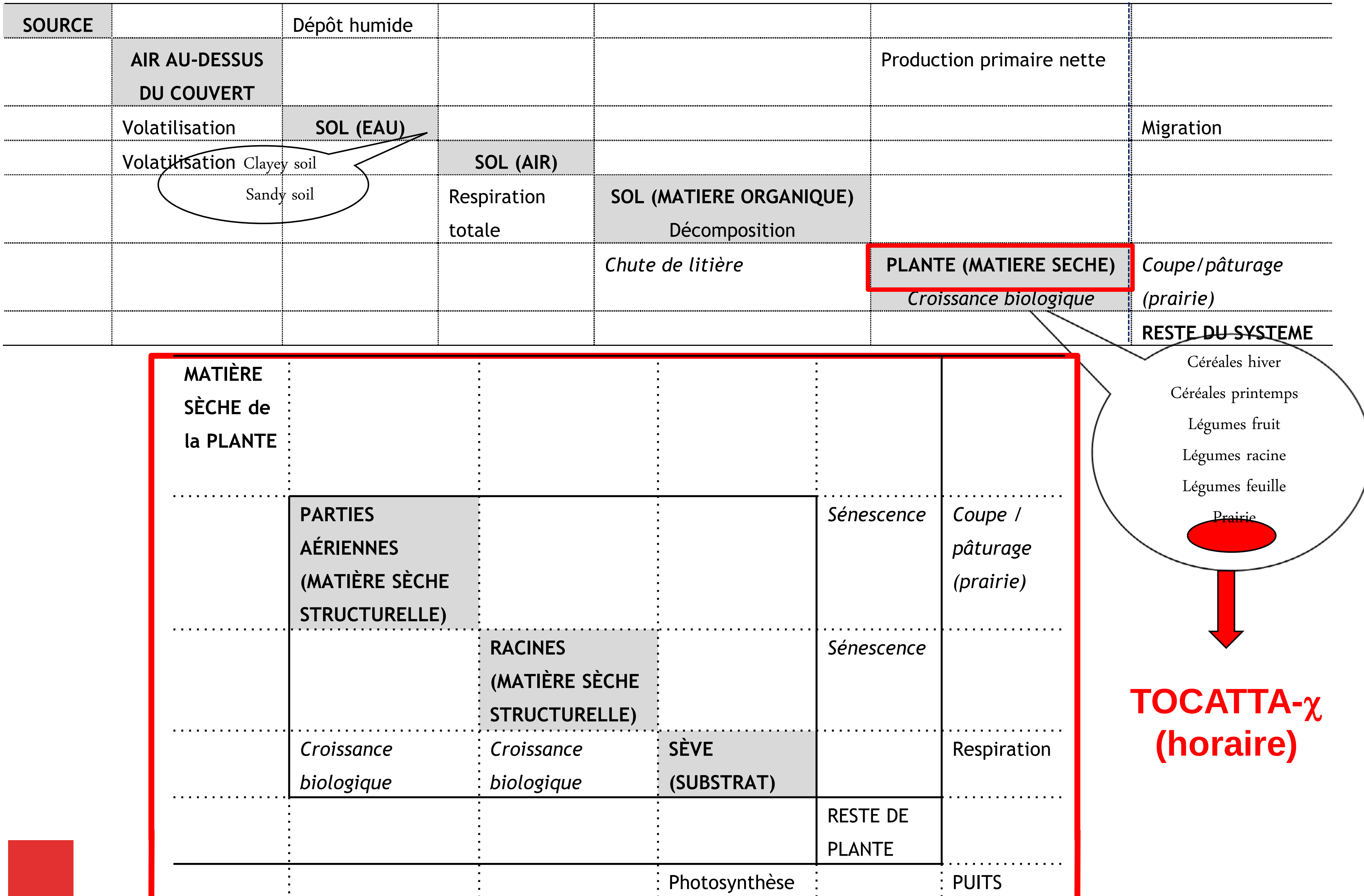
Comparaison modèle-mesures, interprétation, amélioration du modèle, publications…

Merci pour votre attention…

Système Sol-Plante (modèle conceptuel)

Carbone-14:

TOCATTA

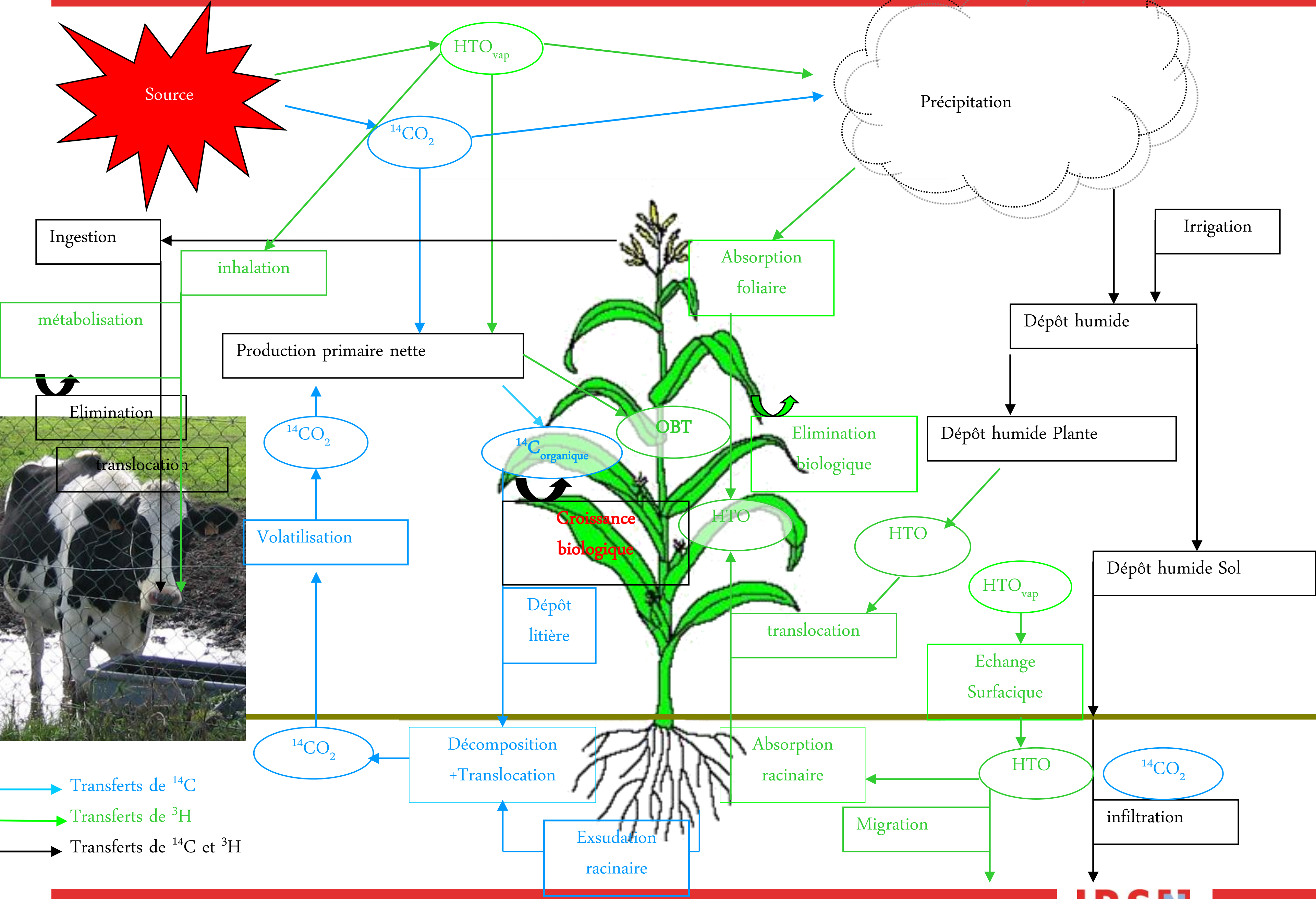


SOURCE		Dépôt humide				
	AIR AU-DESSUS DU COUVERT				Production primaire nette	
	Volatilisation	**SOL (EAU)**				Migration
	Volatilisation		**SOL (AIR)**			
			Respiration totale	**SOL (MATIERE ORGANIQUE)** Décomposition		
			Chute de litière	**PLANTE (MATIERE SECHE)** *Croissance biologique*		*Coupe/pâturage (prairie)*
						RESTE DU SYSTEME

MATIÈRE SÈCHE de la PLANTE					
	PARTIES AÉRIENNES (MATIÈRE SÈCHE STRUCTURELLE)			*Sénescence*	*Coupe / pâturage (prairie)*
		RACINES (MATIÈRE SÈCHE STRUCTURELLE)		*Sénescence*	
	Croissance biologique	*Croissance biologique*	**SÈVE (SUBSTRAT)**		Respiration
				RESTE DE PLANTE	
			Photosynthèse		PUITS

TOCATTA-χ (horaire)

Processus de transfert dans le système sol-plante-animal



Source

HTO$_{vap}$

$^{14}CO_2$

Précipitation

Ingestion

inhalation

métabolisation

Elimination

translocation

Production primaire nette

Absorption foliaire

Irrigation

Dépôt humide

Dépôt humide Plante

OBT

$^{14}C_{organique}$

Elimination biologique

$^{14}CO_2$

Volatilisation

Croissance biologique

HTO

HTO

Dépôt humide Sol

HTO$_{vap}$

Dépôt litière

translocation

Echange Surfacique

$^{14}CO_2$

Décomposition +Translocation

Absorption racinaire

HTO

$^{14}CO_2$

Exsudation racinaire

Migration

infiltration

Transferts de ^{14}C

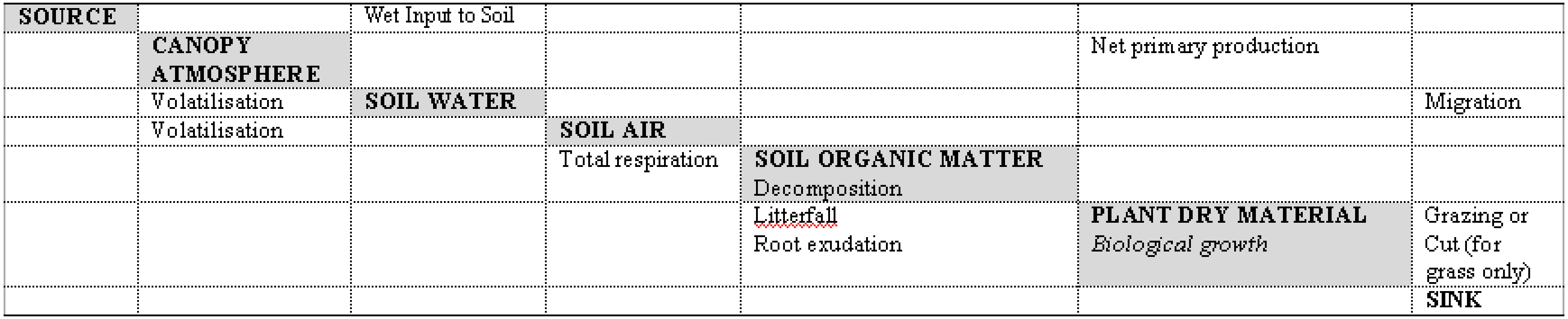
Transferts de ^{3}H

Transferts de ^{14}C et ^{3}H

The TOCATTA soil-plant model: first version

- Le Dizès S., Maro D., Hebert D., Gonze M.-A , Aulagnier C., 2012.« TOCATTA: a dynamic transfer model of ^{14}C from the atmosphere to soil-plant systems », J. Environ. Radioact., 105: 48-59.

- The soil-plant system : conceptual model



SOURCE		Wet Input to Soil				
	CANOPY ATMOSPHERE				Net primary production	
	Volatilisation	**SOIL WATER**				Migration
	Volatilisation		**SOIL AIR**			
			Total respiration	**SOIL ORGANIC MATTER** Decomposition		
				Litterfall Root exudation	**PLANT DRY MATERIAL** *Biological growth*	Grazing or Cut (for grass only)
						SINK

Module Plante (modèle conceptuel)

Tritium

SOURCE (HTO)	Dispersion gazeuse	Apport humide (HTO, via précipitation et irrigation par aspersion) ●Interception par le sol	Apport humide (HTO, via précipitation et irrigation par aspersion) ●Interception par la plante			
	AIR AU-DESSUS DU COUVERT (HTO)	Échange de surface (HTO)	Absorption foliaire (TFWT)	Production primaire nette (OBT)		
		EAU DU SOL (HTO) Décroissance radioactive	Absorption racinaire (HTO)			Migration
			EAU DE LA PLANTE (TFWT) ●translocation Décroissance radioactive *Croissance biologique*		Elimination biologique (HTO)	
				MATIERE SÈCHE DE LA PLANTE (OBT) Décroissance radioactive *Croissance biologique*		*Chute de litière* *Coupe/Broutâge (prairie)*
					RESTE DE LA PLANTE	
						RESTE DU SYSTEME

Key features of the ^{14}C soil-plant transfer in the TOCATTA model

Type of contamination	Gaseous atmospheric releases of and/or spray irrigation with contaminated water
Driving variables (inputs)	Daily atmospheric $^{14}CO_2$ concentration, monthly data on climate (e.g. temperature, rainfall height) and, if irrigation, concentration in irrigated water and monthly irrigation depth
Plant growth	Predefined curves of plant growth for all vegetation categories. Possibility of use of empirical dry biomass data for grass
Mass balances	Stable and radioactive C fluxes between air and vegetation fully balanced at a daily time-step
Operation (time-step)	Daily fluxes relatives to stable and radioactive C, daily plant growth
Outputs	concentration or stock in plants and soil, plant growth rate and dry matter quantity, soil $^{(14)}C$ dynamics

- Major **soil-plant transfer processes** considered in TOCATTA following a gaseous release of ^{14}C in atmosphere and/or spray irrigation, modelling approaches, and original source

Process	Approach	Sources
Growth of a plant	Based on predefined plant growth curves for dry biomass evolution: shaped logistic equations (annual crops), linear (vegetables), exponential or based on user-defined data (grass)	-
Net primary production	Based on an isotopic equilibrium hypothesis with air and plant growth characteristics	-
Litterfall	Based on a single exponential function characterised by a first-order rate kinetic formulation. Litterfall rate is assumed constant for grass and related to crop yield and the amount of crop not removed for other plant categories	Sheppard et al. (2006b)
Root exudation	Based on a single exponential function characterised by a first-order kinetic formulation	Jouven et al. (2006a, 2006b)
Soil decomposition & microbial respiration	Based on the five-compartment Rothamsted soil C model formulated as a system of first-order differential equations where decomposition rates are empirical functions of percent clay, moisture content, and temperature	Jenkinson (1990); Coleman and Jenkinson (2005)

Process	Approach	Sources
Interception by vegetation & soil	Estimated as a function of plant dry biomass	Chamberlain (1970)
Wet input to soil	Based on radioecological calculations of interception factors and wet inputs associated with rain and/or spray irrigation	-
Volatilisation	Based on a single exponential function of the stock activity of soil inorganic characterised by a first-order empirical rate constant. The specific activity of the $^{14}CO_2$ volatilized from the soil is then estimated from an empirical canopy dilution factor and the simplified assumption that soil outputs as CO_2 are balanced by inputs of plant C	- Sheppards et al. (2006a; 2006b)
Migration in soil	Based on a single exponential function characterized by a first-order kinetic formulation	-

Plant module in TOCATTA: Mathematical model (1)

- ✓ Computes the daily ^{14}C concentration (mol.kg DW^{-1}) and specific activities (mol.kg^{-1} C) in dry plant material
- ✓ Equation based on the assumption of an **isotopic equilibrium** between the net primary productivity flux assimilated by vegetation and the canopy atmosphere at each time step of the simulation (e.g. 1 day)

$$\frac{\partial}{\partial t}\left\{\chi_p[^{14}C]_{plant}\right\} = TC14^{Npp}$$

kgC.kg DW^{-1}

$$\frac{\partial}{\partial t}[^{14}C]_{plant} = [^{12}C]_{plant} \times \frac{\partial}{\partial t}[^{14}C]^{sp}_{plant} = (\underbrace{TC14^{Npp}}_{\text{NetPrimaryProduction}} - \underbrace{\frac{1}{\chi_p}\frac{\partial \chi_p}{\partial t}[^{14}C]_{plant}}_{\text{Biological growth}})$$

mol.kg DW^{-1}

mol.kg C^{-1}

Plant module in TOCATTA: Mathematical model (2)

Net primary production

$$TC14^{Npp} = \boxed{\lambda_P^{Gro}} \times [^{12}C]_{plant} \times \frac{[^{14}C]_{AirCanopy}}{[^{12}C]_{Air}}$$

Relative growth rate (d^{-1})

$$\lambda_P^{Gro}(t) = \frac{1}{\boxed{\chi_P}} \left[\frac{d\chi_P}{dt} \right]$$

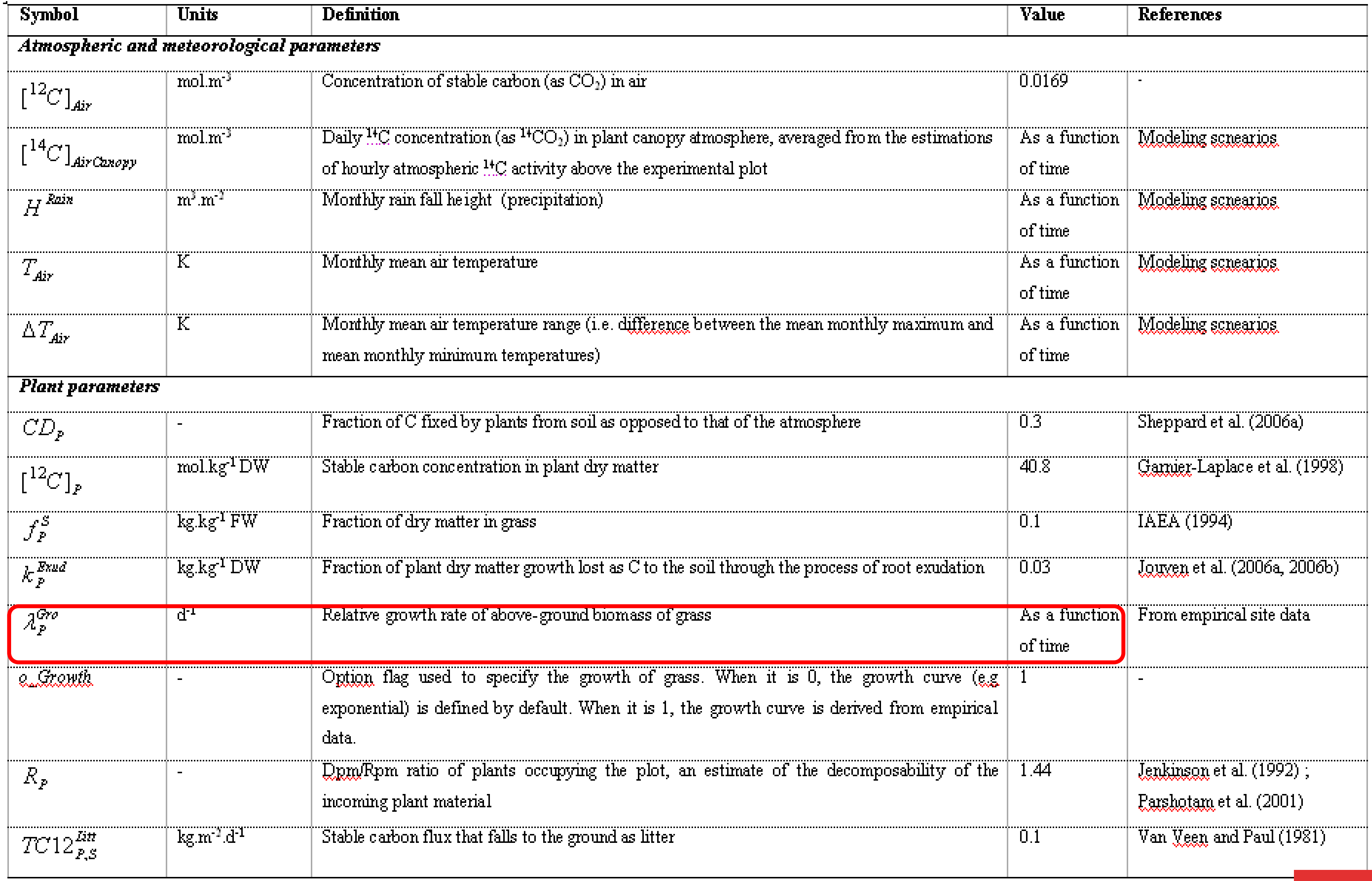
Plant dry density

derived from **time-dependant predefined growth curves** or **experimental data if available**

Biological growth

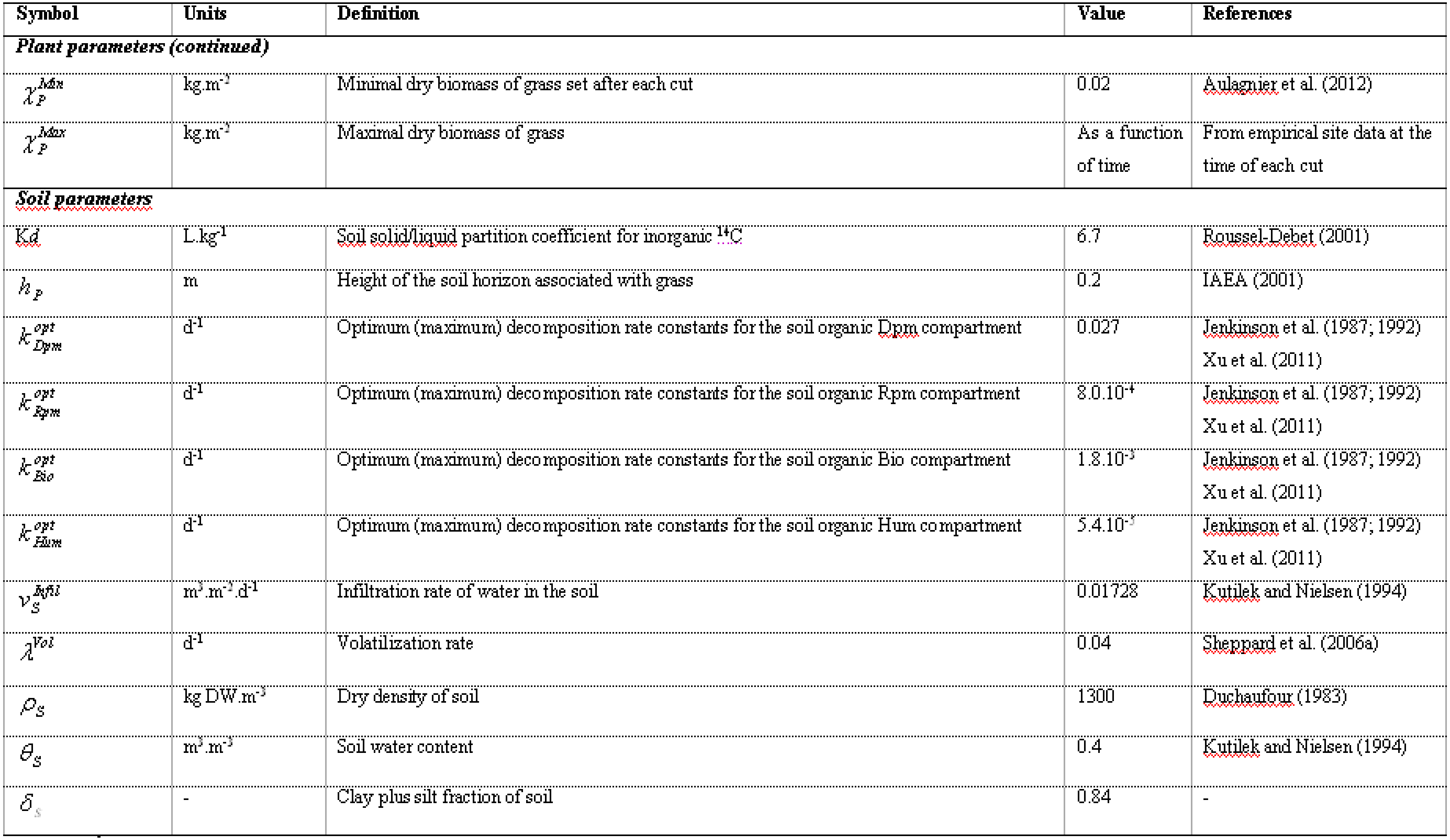
$$TC14^{Growth} = \boxed{\lambda_P^{Gro}} \times [^{14}C]_{plant}$$

Input parameters (1)



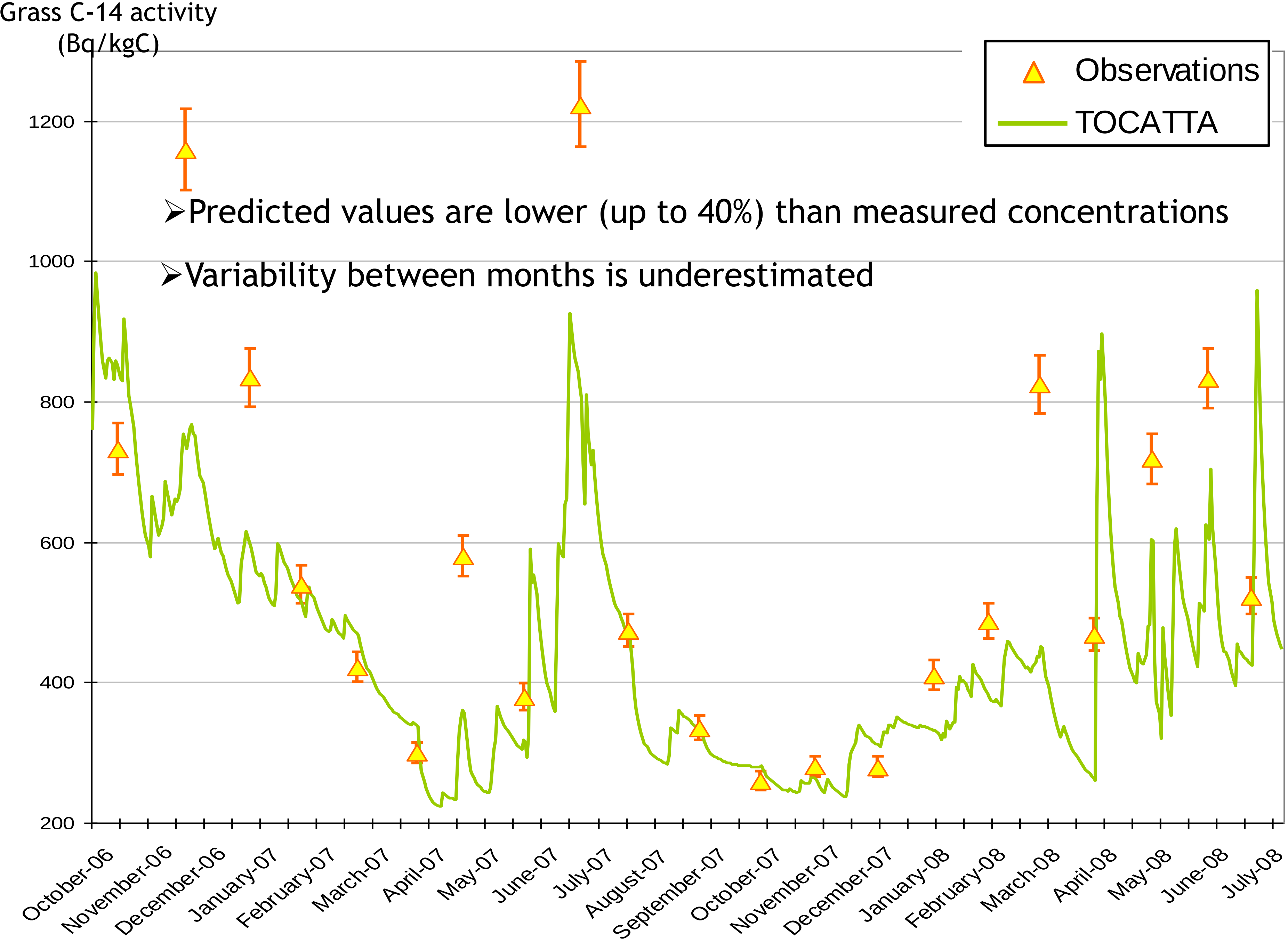
Symbol	Units	Definition	Value	References
Atmospheric and meteorological parameters				
$[^{12}C]_{Air}$	mol.m^{-3}	Concentration of stable carbon (as CO$_2$) in air	0.0169	-
$[^{14}C]_{Air\,Canopy}$	mol.m^{-3}	Daily ^{14}C concentration (as ^{14}CO$_2$) in plant canopy atmosphere, averaged from the estimations of hourly atmospheric ^{14}C activity above the experimental plot	As a function of time	Modeling scnearios
H^{Rain}	m^3.m^{-2}	Monthly rain fall height (precipitation)	As a function of time	Modeling scnearios
T_{Air}	K	Monthly mean air temperature	As a function of time	Modeling scnearios
ΔT_{Air}	K	Monthly mean air temperature range (i.e. difference between the mean monthly maximum and mean monthly minimum temperatures)	As a function of time	Modeling scnearios
Plant parameters				
CD_P	-	Fraction of C fixed by plants from soil as opposed to that of the atmosphere	0.3	Sheppard et al. (2006a)
$[^{12}C]_P$	mol.kg^{-1} DW	Stable carbon concentration in plant dry matter	40.8	Garnier-Laplace et al. (1998)
f_P^S	kg.kg^{-1} FW	Fraction of dry matter in grass	0.1	IAEA (1994)
k_P^{Exud}	kg.kg^{-1} DW	Fraction of plant dry matter growth lost as C to the soil through the process of root exudation	0.03	Jouven et al. (2006a, 2006b)
λ_P^{Gro}	d^{-1}	Relative growth rate of above-ground biomass of grass	As a function of time	From empirical site data
o_Growth	-	Option flag used to specify the growth of grass. When it is 0, the growth curve (e.g exponential) is defined by default. When it is 1, the growth curve is derived from empirical data.	1	-
R_P	-	Dpm/Rpm ratio of plants occupying the plot, an estimate of the decomposability of the incoming plant material	1.44	Jenkinson et al. (1992) ; Parshotam et al. (2001)
$TC12_{P,S}^{Litt}$	kg.m^{-2}.d^{-1}	Stable carbon flux that falls to the ground as litter	0.1	Van Veen and Paul (1981)

Input parameters (2)



Symbol	Units	Definition	Value	References
Plant parameters (continued)				
χ_P^{Min}	$kg.m^{-2}$	Minimal dry biomass of grass set after each cut	0.02	Aulagnier et al. (2012)
χ_P^{Max}	$kg.m^{-2}$	Maximal dry biomass of grass	As a function of time	From empirical site data at the time of each cut
Soil parameters				
Kd	$L.kg^{-1}$	Soil solid/liquid partition coefficient for inorganic ^{14}C	6.7	Roussel-Debet (2001)
h_P	m	Height of the soil horizon associated with grass	0.2	IAEA (2001)
k_{Dpm}^{opt}	d^{-1}	Optimum (maximum) decomposition rate constants for the soil organic Dpm compartment	0.027	Jenkinson et al. (1987; 1992) Xu et al. (2011)
k_{Rpm}^{opt}	d^{-1}	Optimum (maximum) decomposition rate constants for the soil organic Rpm compartment	$8.0.10^{-4}$	Jenkinson et al. (1987; 1992) Xu et al. (2011)
k_{Bio}^{opt}	d^{-1}	Optimum (maximum) decomposition rate constants for the soil organic Bio compartment	$1.8.10^{-3}$	Jenkinson et al. (1987; 1992) Xu et al. (2011)
k_{Hum}^{opt}	d^{-1}	Optimum (maximum) decomposition rate constants for the soil organic Hum compartment	$5.4.10^{-5}$	Jenkinson et al. (1987; 1992) Xu et al. (2011)
v_S^{Infil}	$m^3.m^{-2}.d^{-1}$	Infiltration rate of water in the soil	0.01728	Kutilek and Nielsen (1994)
λ^{Vol}	d^{-1}	Volatilization rate	0.04	Sheppard et al. (2006a)
ρ_S	$kg\ DW.m^{-3}$	Dry density of soil	1300	Duchaufour (1983)
θ_S	$m^3.m^{-3}$	Soil water content	0.4	Kutilek and Nielsen (1994)
δ_S	-	Clay plus silt fraction of soil	0.84	-

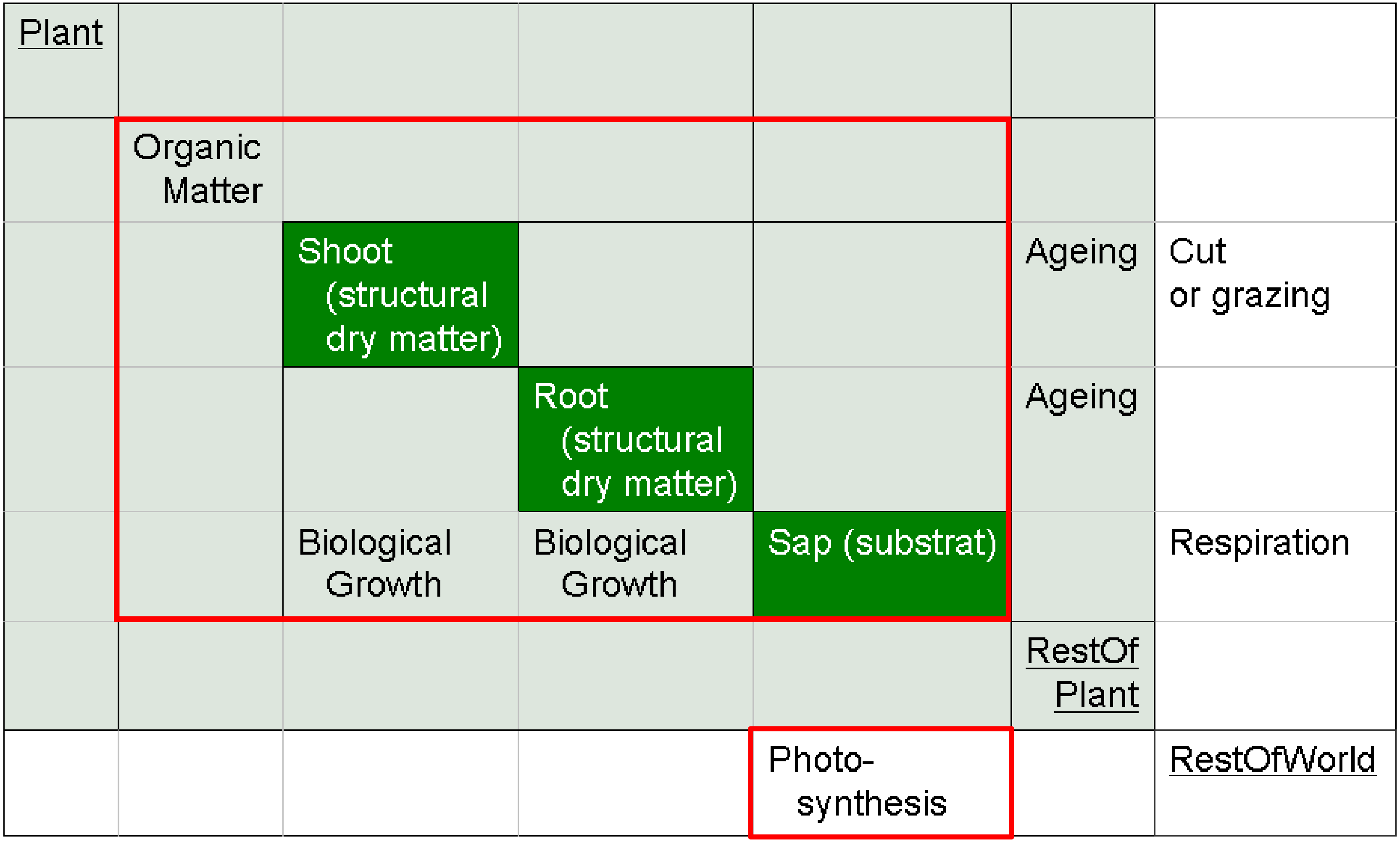
TOCATTA- ^{14}C applied to grass: model versus measurements



- Predicted values are lower (up to 40%) than measured concentrations
- Variability between months is underestimated

TOCATTA($-\chi$) model: improved version (grass)

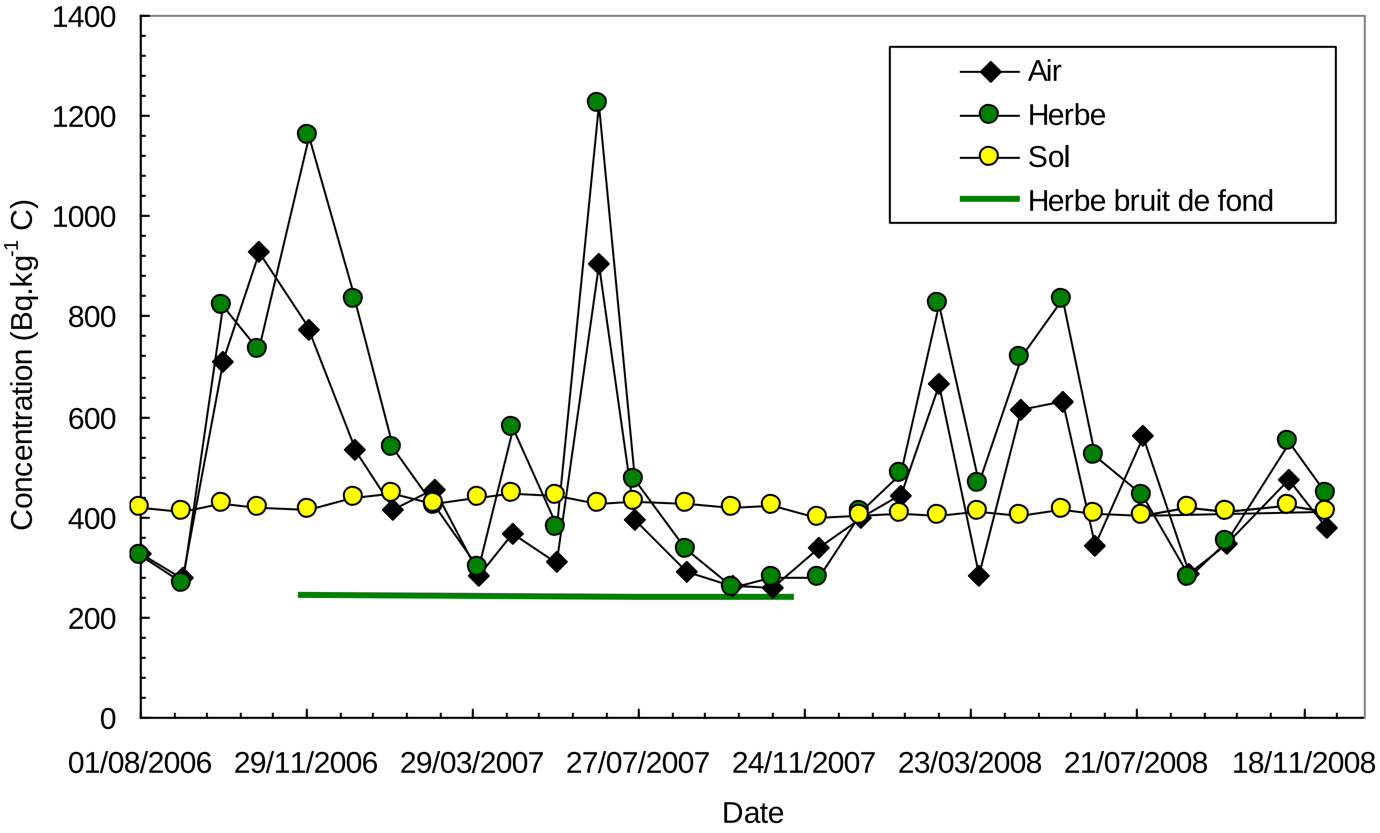
- Aulagnier C., Le Dizès S., Maro D., Hebert D., Lardy R., Martin R., Gonze M.-A , 2012. « Modelling the transfer of ^{14}C from the atmosphere to grass : A case study in a grass field near AREVA-NC La Hague, Journal of Environmental Radioactivity, 112, 52-59.



Plant						
	Organic Matter					
		Shoot (structural dry matter)			Ageing	Cut or grazing
			Root (structural dry matter)		Ageing	
		Biological Growth	Biological Growth	Sap (substrat)		Respiration
					RestOf Plant	
				Photo-synthesis		RestOfWorld

- An hourly time-step model, integrated in SYMBIOSE as a modelling option for grass

Measurements of ^{14}C activity in air, grass and soil



- ➢ Great fluctuations of the signal in air and grass due to the wind direction and the operation of the facility
- ➢ No fluctuation in soil due to a poorly reactive pool of organic matter.

Why this model under-estimation?

- The model is based on a daily isotopic equilibrium between the quantity of newly created plant biomass and the surrounding air
  - In particular, there is no difference whether a release occurs during the day or during the night
  - In other words, the model is better adapted for chronic releases than for accidental releases
- Need to improve the model on the **kinetics** of transfer of C (and 14C) to adapt it to variable releases and weather conditions
- A new model (TOCATTA $\_\chi$) has been developed for grass to simulate intraday ^{14}C transfer in the soil-plant-atmosphere system in the event of accidental releases
  - ✓ Integrates the key physiological processes of the **PASIM model*** (photosynthesis, growth ...) at an **hourly time-step**, according to local agro-meteorological data
  - ✓ Takes into account the intraday variability of ^{14}C releases
  - ✓ Intermediate level of complexity between PASIM (mechanistic) and TOCATTA (simple and operational)

***Grassland ecosystem odel simulating the flow of carbon, nitrogen, water and energy at the soil-plant-atmosphere interface (Riédo et al., 1998; Vuichard, 1997)**

Conclusions and perspectives (1)

- To adapt the model to time varying releases and meteorology, an hourly time-step is required :
  - To estimate ^{14}C air concentration inputs to the model, based on hourly ^{85}Kr data
  - To simulate photosynthesis and plant growth dynamics
- TOCATTA-χ : better correlation with obs. and optimal reproducing of the variability of obs.
- Conclusions restricted to a single case study - grassland ecosystem with cuts of grass each month // grazed meadow with resting T = 1 month.
- Importance of the management of grass on the ^{14}C transfer to vegetation (adjustment of the mean turnover time due to different management modes is required)
- TOCATTA-χ needs to be validated on other independent data sets obtained on terrestrial ecosystems

Conclusions and perspectives (2)

- TOCATTA-χ needs to be validated on other independent data sets obtained on agricultural ecosystems
- Need to develop (or use) a dynamic model of C14 transfer in aquatic (freshwater biota) systems
- …and validate it against data on aquatic systems as much as possible
- An ambitious experimentation is starting at IRSN on tritium transfer (2013-2017) in the same grassland ecosystem of the AREVA NC La hague environment