

The logo for IRSN, featuring the letters 'IRSN' in a bold, sans-serif font. The 'I' and 'R' are red, the 'S' is blue, and the 'N' is red with a blue vertical bar on its right side.

INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Faire avancer la sûreté nucléaire

TOCATTÀ status & planned tritium experiments

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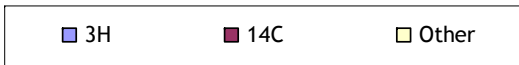
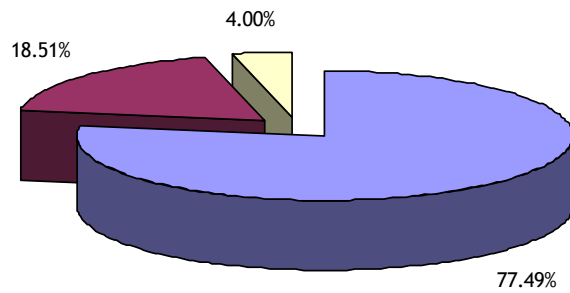
Why studying ^{14}C and ^3H ?

(1)

- Carbon 14 and tritium are substantially released in the atmosphere around:
 - Nuclear Power Plants (NPP);
 - Nuclear Recycling Plants (NRP).

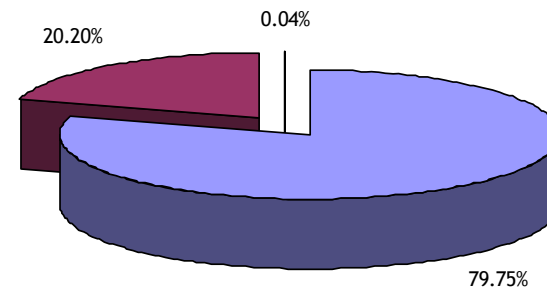
Atmospheric releases

NPP



NRP

$^{85}\text{Kr} = 99.97\%$



Why studying ^{14}C and ^3H ?

(2)

- Still significant uncertainties on ^{14}C and ^3H :
 - sampling and measurement;
 - evolution of the chemical form in the atmosphere, vegetation, soil and groundwater;
 - quantification of dry and wet deposition;
 - quantification of Organically Bound Tritium (OBT).

The VATO project : VALidation of the TOCATTa model

- To estimate ^{14}C and ^3H fluxes in a grassland ecosystem (*raygrass sp.*, soil and groundwater in both saturated and unsaturated zones), in relation with:
 - evolution of air concentrations (day versus night);
 - weather conditions;
 - land use (grazing, maize silage and hay).
- To study ^{14}C and ^3H transfers to cows and cowmilk as a function of

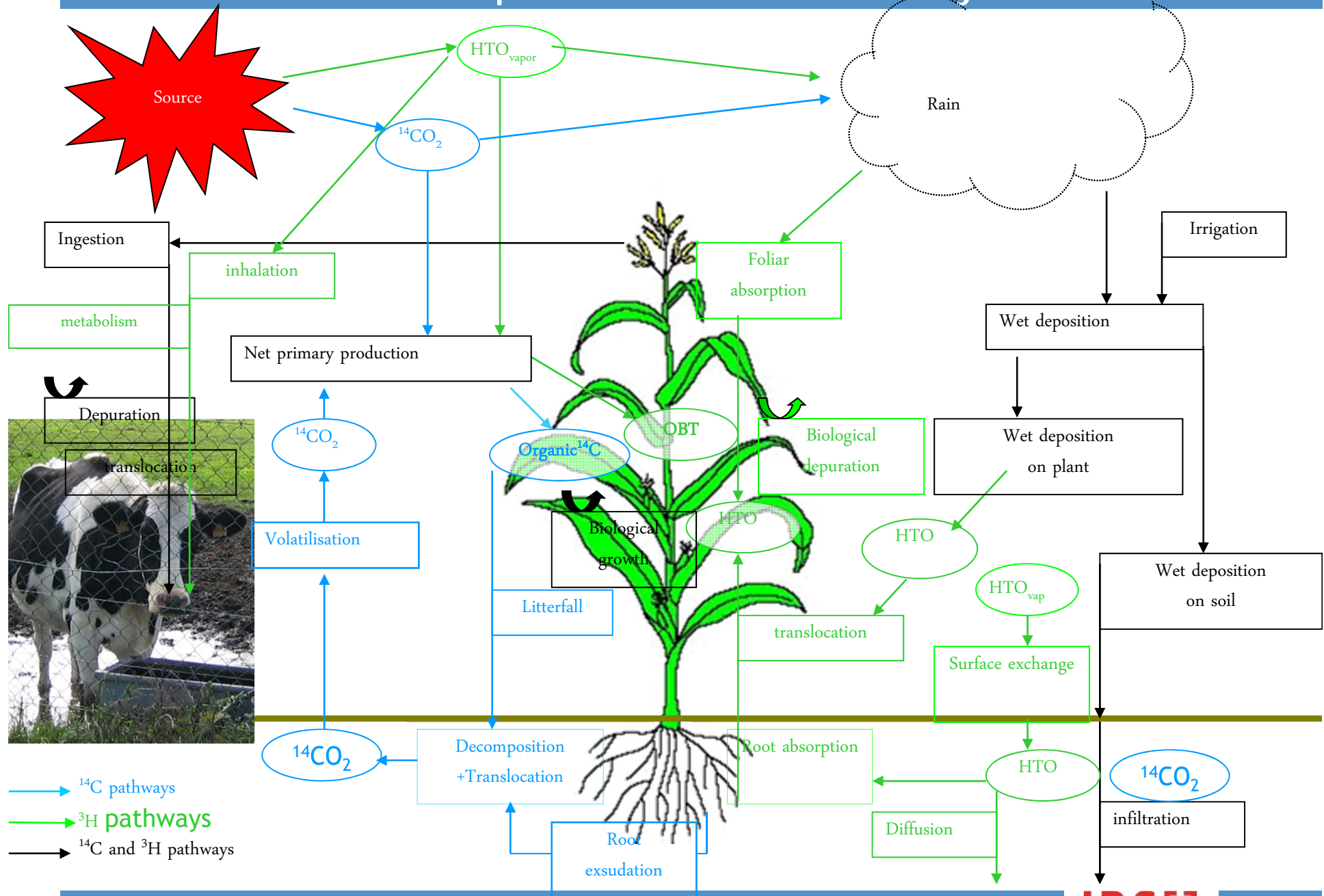
➔ In order to validate the TOCATTa model.

^{14}C & ^3H transfer modelling in TOCATTa (1)

➤ Main characteristics

- Types of releases: atmospheric and / or liquid (spray irrigation)
- Main environmental media : agricultural systems (soil, plant and animals)
- Multiple source term kinetics : normal and accidental modes
- Physico-chemical forms of releases : $^{14}\text{CO}_2$, HTO
- Temporal scales:
 - Daily time step
 - Duration of simulation: ≥ 1 year(s)

^{14}C and ^3H : transfer processes in a rural ecosystem in TOCATA



^{14}C & ^3H transfer modelling in TOCATTA (2)

➤ Other characteristics

A dynamic model

- Based on plant biomass growth whose curves are either predefined or derived from experimental data
- Based on the assumption of isotopic equilibrium between the quantity of newly created plant biomass and the surrounding air, at each time step (i.e. 1 day)

Integrated in SYMBIOSE

- Simple model, flexible, limited number of input parameters and compartments to be used in an operational mode
- Model parameterized for various types of agricultural plants, broken down into three groups : annual crops, vegetable crops and pasture grass. Two categories of soils considered by default: sandy soil and clayey soil.
- Conceptual (interaction matrix) and mathematical models (mass balance)
- Dose man calculations through ingestion of contaminated foodstuffs

The soil-plant system: conceptual model (³H)

SOURCE*						
	CANOPY ATMOSPHERE	Surface exchange Wet Input• (1-Captation)	Net primary production	Foliar absorption Wet Input •Translocation		
		SOIL WATER		Root uptake •Translocation		Diffusion Migration
			PLANT ORGANIC DRY MATERIAL		Rad. decay	Litterfall Grazing
				PLANT WATER MATERIAL	Biol. decay Rad. decay	Grazing
				Litterfall Root exudation	REST OF PLANT	Harvest
						SINK

*Tritiated water (HTO) presents in the atmosphere (air or water droplets) or in irrigation water.

Mathematical model

- First order differential equations
- Conservation of mass within each compartment

Stock or concentration of stable or radioactive C within compartment i:

$$\frac{dA_i}{dt} = \frac{d\{\chi_i [C]_i\}}{dt} = \underbrace{\sum_{k=1}^m TC_{j,k}^p}_{\text{input}} - \underbrace{\sum_{k=1}^m TC_{k,l}^{p'}}_{\text{output}}$$

Stock (mol.m⁻²)

Concentration (mol.kg⁻¹)

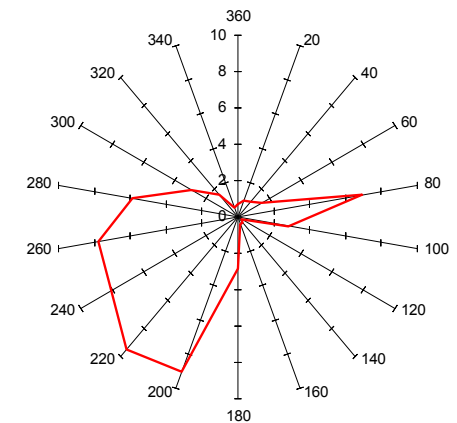
Density (kg.m⁻²)

Mass transfer fluxes (mol.m⁻².d⁻¹)

(with A_i=0 à t=0)

VATO project: *in situ* lab

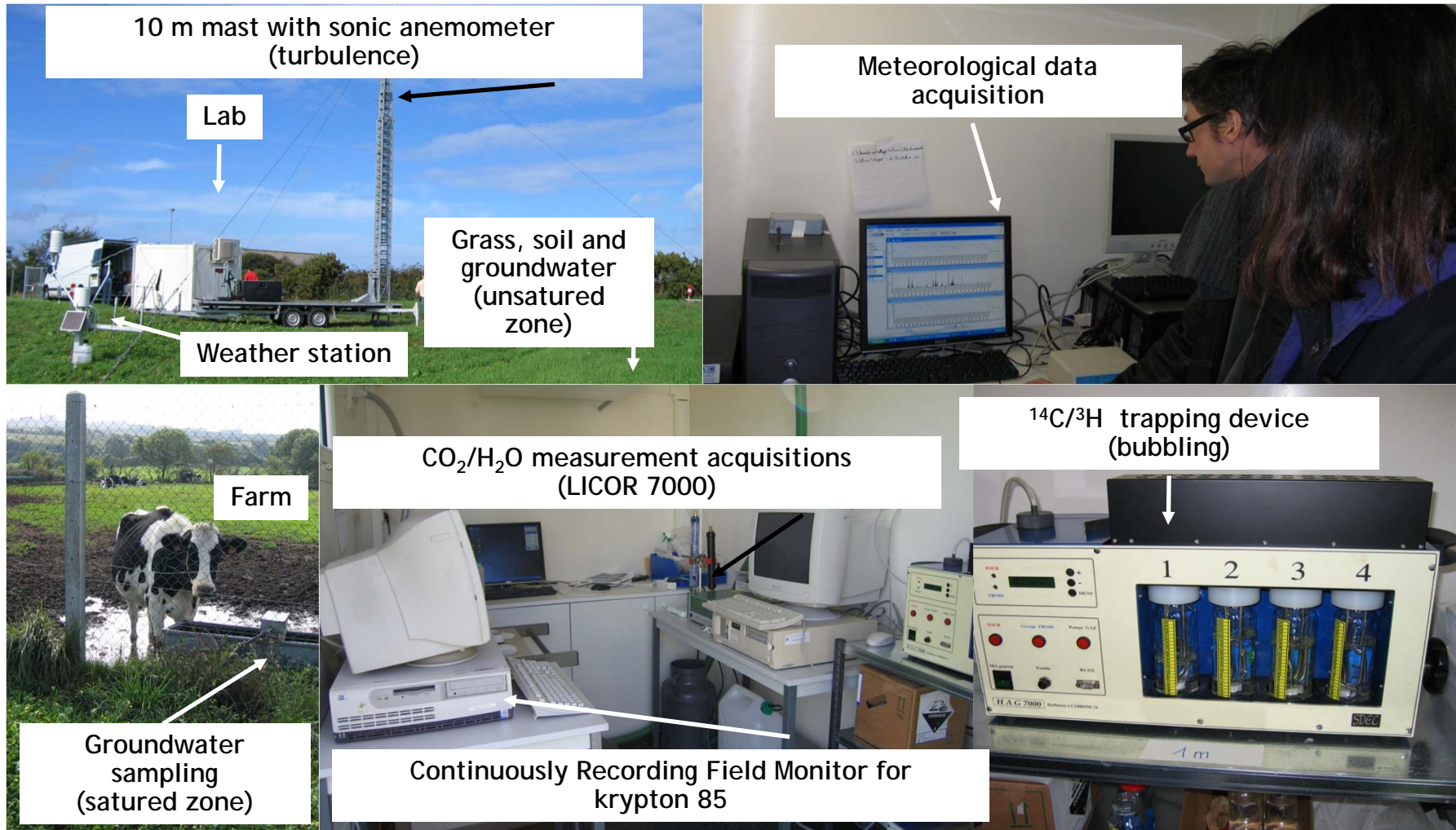
Releases of ^{14}C and ^3H by the reprocessing plant induce greater concentrations than background levels



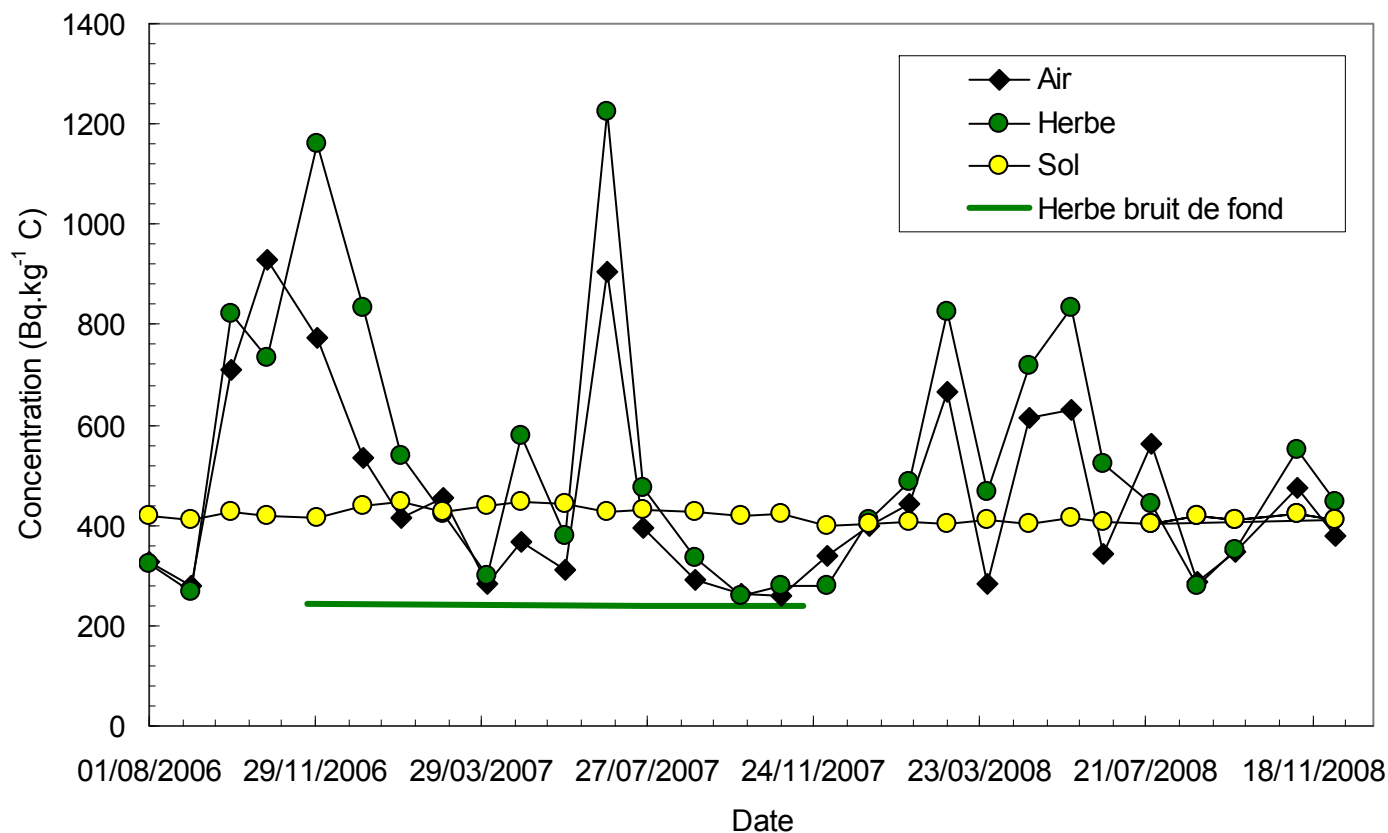
Wind rose at La Hague

"Atelier Nord": a well located experimental site, considering the most frequent wind directions

VATO project: monitoring system



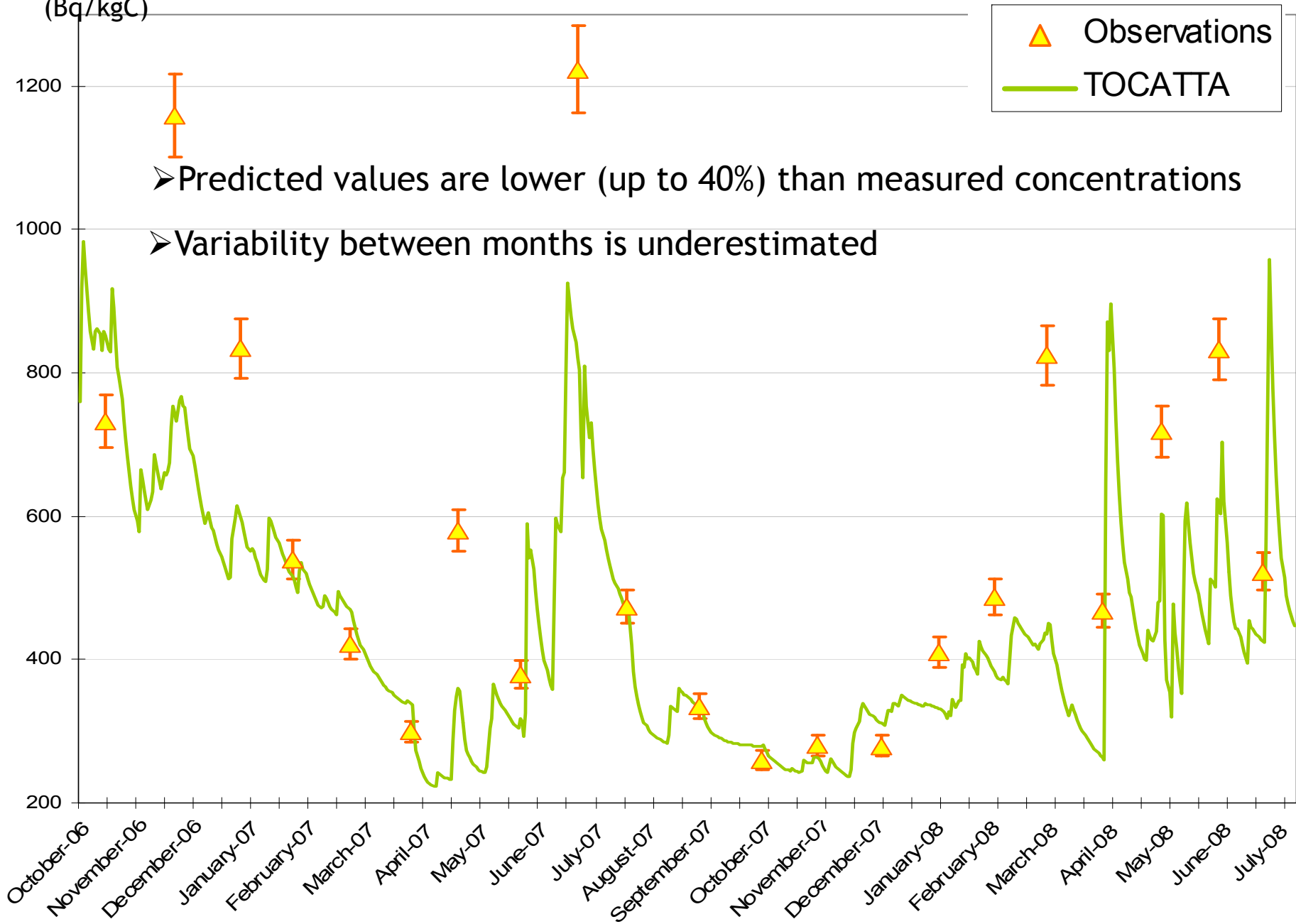
^{14}C concentration measurements in air, grass and soil



- 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.

^{14}C in grass: model versus measurements

Grass C-14 activity
(Bq/kgC)



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.
- A new model is being built based on the **PASIM*** model.

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

- collaboration between IRSN-INRA
- Post-doctoral fellowship (C. Aulagnier, 01/2011-)
- It will take into account plant physiology and local meteorology.

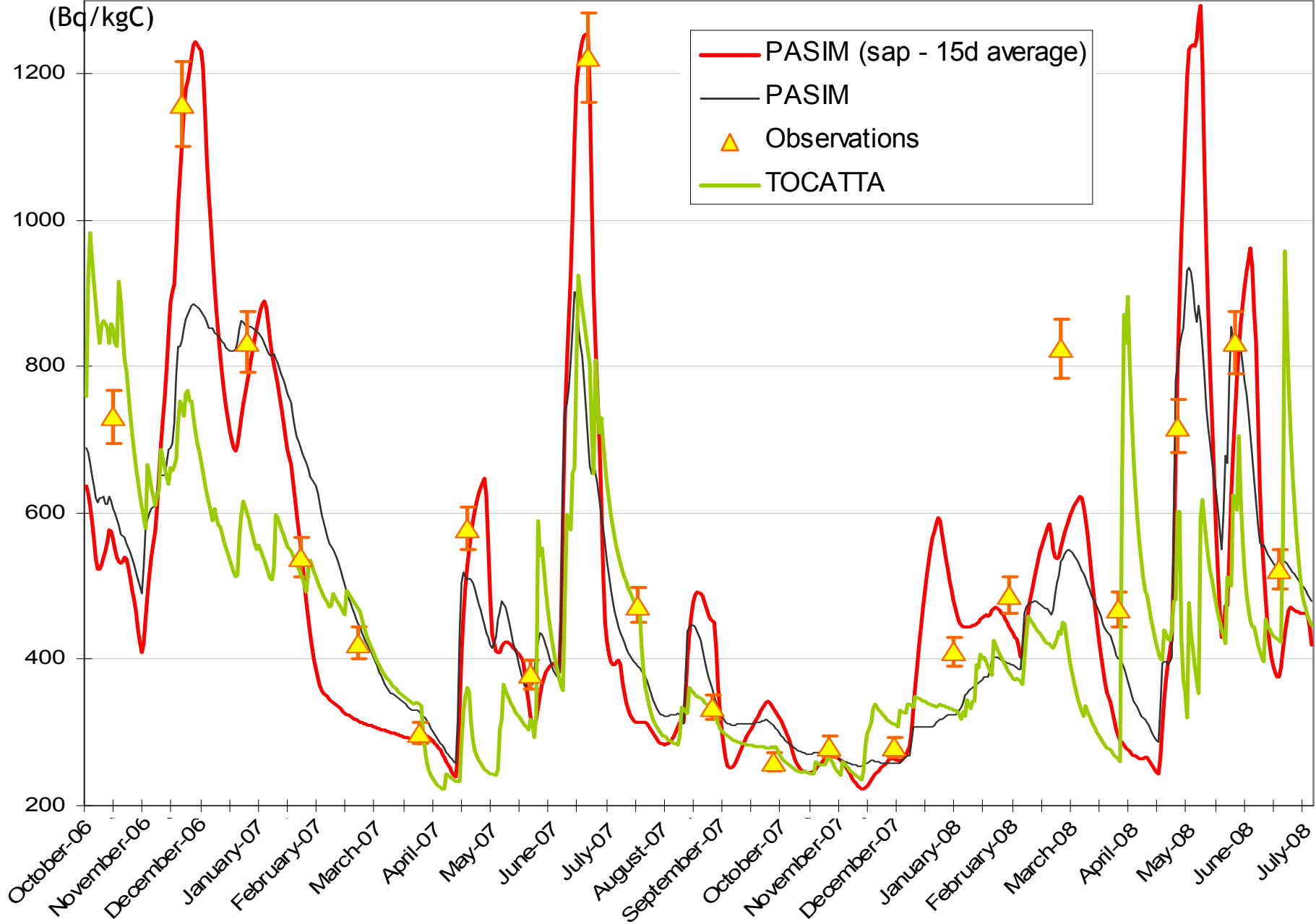
Conclusion (1)

- The new model still needs further adaptations to take into account the acute variations in radionuclide releases and weather fluctuations. An hourly time-step is required:
 - to simulate photosynthesis: carbon 14 cycling;
 - to simulate water exchange: tritium cycling.

- In order to evaluate the concentration of ^{14}C and ^3H (HTO, OBT) in the different compartments of rural ecosystem, from the atmosphere to the groundwater *via* grassland.

^{14}C in grass: models versus measurements

Grass C-14 activity



Conclusion/perspectives (2)

- Development of a new model (TOCATTA_χ) for the atmosphere-soil-plant system
 - Integrates the key physiological processes of PASIM (photosynthesis, growth) at an hourly time-step
 - Intermediate level of complexity between TOCATTA and PASIM:
 - ✓ Retains the “mechanistic” aspect of PASIM in modelling C cycle,
 - ✓ While being simpler and therefore more operational
 - Accounts for the intra-day variability of ¹⁴C releases
- Replacement of the TOCATTA model by TOCATTA_χ in SYMBIOSE ??

Posters & publications

2 posters: "Proceedings ICRRER 2011, 19-24 June, Hamilton (Canada)":

- Maro et al.: Modelling and validating ^{14}C transfer in terrestrial environments in response to ^{14}C releases
- Le Dizès et al.: Modelling ^{14}C transfer in terrestrial environments in response to chronic and accidental ^{14}C releases

S. Le Dizès, D. Maro, D. Hebert, M.-A. Gonze, C. Aulagnier. TOCATTA: a dynamic transfer model of ^{14}C from the atmosphere to soil-plant systems.

✓ Accepted to the *J. Env. Radioact.* (Sept 6th, 2011)

C. Aulagnier, S. Le Dizès, D. Maro, D. Hebert, R. lardy, R.Martin, M.-A. Gonze. Modelling chronic and accidental releases of ^{14}C to the environment : A case study in a grass field near AREVA-NC La Hague.

✓ Submitted to the *J. Env. Radioact.* (Sept. 1st, 2011)

C. Aulagnier, S. Le Dizès, D. Maro, D. Hebert, R. lardy, R.Martin, M.-A. Gonze. New TOCATTA model developments for accidental release scenarios.

✓ To be submitted (end of year 2011)

Agenda

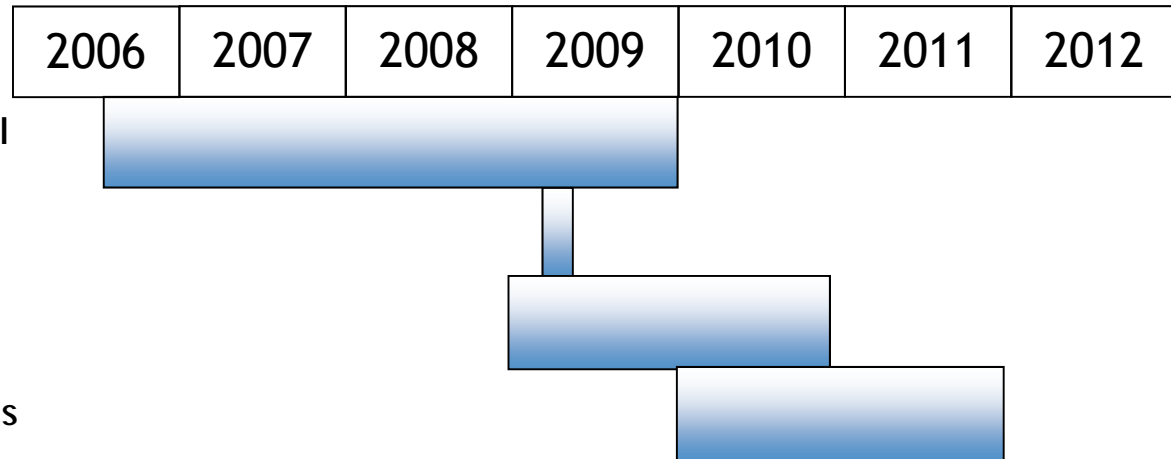
Carbon 14

Measurements in air, grass and soil

Measurements in cow milk

Model-measures comparison

Model adjustment and publications



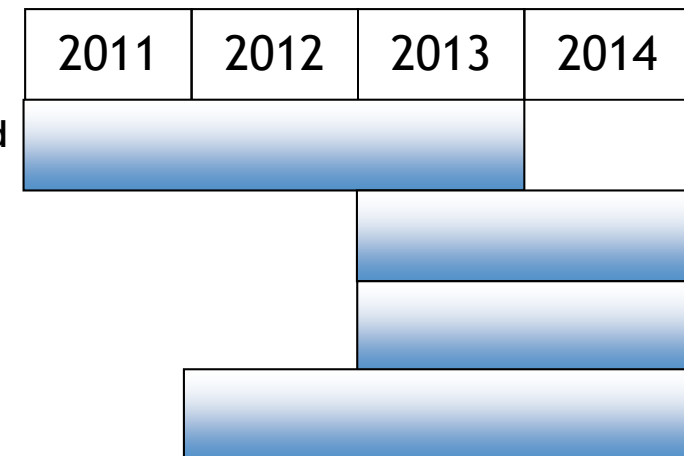
Tritium

Measurements in air, rain water, grass, soil, soil unsaturated zone, and ground water

Measurements in cow milk

Model-measures comparison

Model adjustment and publications



Planned ^3H studies

Probably a collaboration of IRSN with the Canadian Nuclear Safety Commission (CNSC, Ottawa) that will be discussed and validated in November

“Transfer of tritium in a grassland ecosystem”: main characteristics:

- A 4-year project
- Comparison of materials and sampling and measurement techniques in vegetation, soil and groundwater
- Study and comparison of mass balance and fluxes of water and tritium in two grassland ecosystems
- Model analysis and comparison
- Other aspects/uncertainties :
 - atmospheric dispersion of the plume,
 - chemical forms of ^3H release (HTO/HT) in the atmosphere
 - quantification of wet and dry deposition
 - quantification of OBT
 - ...

SYMBIOSE: A Modeling and Simulation Platform for Environmental Radiological Risk Assessments (1/2)

- **Purpose: Risk assessment calculations**
 - fate and transport of radionuclides in ecosystems;
 - dose to man.
- **Flexible approach to deal with a wide range of situations**
 - multiple radioactive releases (atmospheric, fluvial, and/or marine) from nuclear facilities under normal, incidental or accidental conditions;
 - multi-media (atmospheric, terrestrial, freshwater and marine) and interfaces;
 - multiple exposure routes (external, inhalation, ingestion...).

SYMBIOSE: A Modeling and Simulation Platform for Environmental Radiological Risk Assessments (2/2)

➤ Calculations

- reference data bank with generic versus site-specific data;
- dynamical (physically-based) and spatially-distributed modeling;
- including daughters (radioactive decay chains);
- specific models to deal with hydrogen, carbon and chlorine.



TOCATTA model

Mathematical model: the plant module (1)

Example : Transfer of tritium from atmosphere to vegetation (organic pool) :

$$\frac{d}{dt}[OBT]_P = \frac{1}{\chi_P} \cdot \left(\underbrace{TOBT_P^{Npp}}_{\text{NetPrimaryProduction}} - \underbrace{TOBT_P^{Gra}}_{\text{Grazing}} - \underbrace{TOBT_P^{litt}}_{\text{litterfall}} - \underbrace{TOBT_P^{Rad}}_{\text{Radioactive decay}} - \underbrace{\left[\frac{d\chi_P}{dt} \right]}_{\text{Biological growth}} \cdot [OBT]_P \right)$$

Plant dry density

Predefined growth model
or empirical growth data



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

Relative growth rate (d⁻¹)

Mathematical model: the plant module (2)

Relative growth rate (d⁻¹)

$$TOBT_P^{Npp} = \frac{(1 - f_P^S)}{f_P^S} \times \lambda_P^{Gro} \times [H]_P \times \frac{p_{HTO} \times [C - 3H]_{Air} \times DI_P}{[H]_{Air}}$$

Hypothesis:

- Daily time-step (current version)
- **Isotopic equilibrium** between the quantity of newly created plant biomass and the surrounding air, at each time step (i.e. 1 day)
- **Logistic** growth curves (cereals), **exponential** (prairie) or **linear** (leafy vegetables, fruit vegetables, root vegetables), or empirical data, if available
- **Isotopic discrimination** of tritium when entering vegetation