IRSN
INSTITUT DE RADIOPROTECTION ET DE SÛRETÉ NUCLÉAIRE
Faire avancer la sûreté nucléaire

Environmental behaviour of tritium released by nuclear facilities in marine and terrestrial ecosystems: *State-of-the-art and examples*

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

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Task force tritium

METI, Tokyo
March 13, 2014

Outlines

- ***Context***
- ***Objectives***
- ***Transfer of tritium in marine and terrestrial ecosystems***
  - Importance of the knowledge of speciation (HT, HTO, OBT)
  - Marine ecosystem
  - Terrestrial ecosystem
  - Interface between marine and terrestrial ecosystems
- ***Conclusions***

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Context

Why is it important to study the transfer of tritium in the environment?

- *Except for rare gases, the amounts (Bq) of tritium from controlled release are higher than for other radionuclides (10-100 times)*
- *Increase of tritium releases in France*
  - Tritium is released by Nuclear Power Plants (NPP), Nuclear Reprocessing Plants (NRP, e.g. AREVA NC La Hague), research centers and defense facilities;
  - In the future, the releases will increase with the commissioning of new built nuclear facilities like the Laser Megajoule (LMJ), the European Pressurized Reactor (EPR) and the International Thermonuclear Experimental Reactor (ITER).
- *Uncertainties on transfers in marine and terrestrial ecosystems*
  - existing uncertainties on transfer knowledge in marine and terrestrial ecosystems, e.g. transfer kinetics of tritiated water (HTO), tritiated hydrogen (HT) and Organically Bound Tritium (OBT) in the different compartments of ecosystems.
- *Anticipate concerns from the public*

Main document in France, white paper on tritium (2010), coordinated by ASN (French Nuclear Safety Authority): see lecture on tritium “the French situation” by J. L. Lachaume (ASN, Deputy Director General) at METI.


asn AUTORITÉ DE SÛRETÉ NUCLÉAIRE

Tritium

Carte des rejets tritium

Accueil > English version

Position et plan d'action de l'ASN

Les groupes de réflexions

Synthèses et recommandations

État des connaissances

Inventaire des sources

Origine du tritium

Métrologie du tritium

Impact sur l'environnement

Impact sur l'homme

Téléchargements et liens

Carte des rejets tritium

Livre Blanc du Tritium

English version

Context

Tritium is a hydrogen isotope that is a low-energy beta emitter (mean energy 5.7 keV). The most commonly-found form in the biosphere is tritiated water and the dominant exposure route is ingestion. The radiotoxicity of tritium is low. The overall impact of tritium releases in France is also low, with an annual effective dose of roughly one µSv or less for the reference groups.

Radioactive releases in the environment around civilian nuclear facilities have significantly decreased over the last few decades, with the exception of tritium. Discharges of this element are forecast to increase due to expected changes in the fuel management methods used by the NPP, and also due to new tritium-emitting facilities, including new power plants that are to be built, and the ITER project.

1. ASN's position and action plan
2. What is Tritium ?
3. Tritium working groups: objectives and approach
4. Summary of work and recommendations
   1. “Tritium Impact” working group
   2. “Tritium: Defence in Depth” working group

In late 2007, papers published in the UK (RIFE 11 report, study by the HPA's Advisory Group on Ionising Radiation (AGIR)) raised questions as to the behaviour of tritium in the environment, in particular focusing on potential accumulation in organisms of organically bound tritium (OBT) from tritium released into the environment and on methods for assessing the biological impact of tritium in humans.

Given this context, ASN wanted to get a clear analysis of the existing studies into the issue and in early 2008 decided to establish two broad working groups, chaired by Dr Patrick Smeesters of the Belgian Federal Agency for Nuclear Control (AFCN) and Mr Roland Masse of the *Académie des technologies*. The groups were formed of experts (from the CEA, CNRS, GSIEN, Institut Curie, IRSN, universities and European Commission "Article 31" experts), representatives of operators (ANDRA, AREVA, CEA, EDF and ITER), associations (ANCCLI, ACRO and CLI) and safety authorities (ASN, DSND). Their findings and recommendations were submitted in early April 2010.

The ASN is pleased with the high quality of the work and the large bibliography provided, which have led to the recommendations issued at the end of each group's summary.

The studies highlight the small impact that tritium releases have in France. However, they do also show the need to carry out further study and research in order to supplement current data and knowledge on the behaviour of tritium in the environment.

The ASN has drawn up its action plan on the basis of the recommendations made by the two working groups. It also hopes that research bodies take into account the requests made by the working groups, as described in the summary of work and recommendations.

Written by a group of experts with various backgrounds (governmental experts, operators, experts from non-governmental laboratories): The white paper encompasses the state of the art and recommendations from IRSN dedicated to the environmental impact of tritium

http://www.asn.fr/sites/tritium/plus/english-version.html

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Objectives

- ***Increase knowledge on the quantification (e.g. fluxes) of tritium transfers in marine and terrestrial ecosystems with a focus on speciation and kinetic of OBT build up:***

  - Estimation of tritium turnover in aquatic biota: e.g. marine seaweed, marine invertebrates such as shellfish, fish ;

  - Quantification of tritium in grassland ecosystem: kinetic of OBT biosynthesis throughout the human food chain; dry and wet deposition;

- ***Improve tritium transfer models between the source of release and the different components of the ecosystems (seawater: MARS, atmosphere: TOCATTA);***

- ***… to have a more realistic human dose assessment.***

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Importance of the knowledge of the speciation

Tritium is present into biota mainly in two forms:

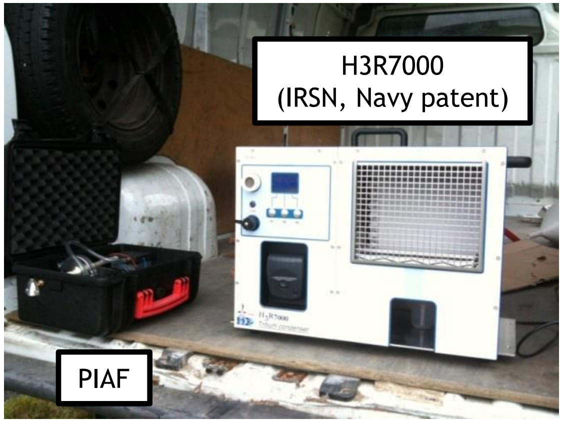
- TFWT (Tissue Free Water Tritium): tritium is incorporated with water exchanges;
  - Quick exchange with the environment (~hour);
- OBT (Organically Bound Tritium): tritium is incorporated with biochemical processes;
  - Relatively slow exchange with the environment (~month).

Tritium speciation of atmospheric or aquatic releases and bioavailability:

- HTO (tritiated water): bioavailable for species with high exchange velocity (few hours) with TFWT;
- Organic molecules: bioavailability depends on the molecule;
- HT (tritiated hydrogen in the atmospheric releases): not bioavailable for species but can be oxidized by biogeochemical reactions and become bioavailable.

Example of speciation in AREVA NC La Hague NRP atmospheric plume in the environment: how can we study the speciation?

Sampling	Treatment	Measurement	Physico-chemical forms
Sampling particles on filter (PIAF) →	Dilution in cocktail →	Liquid scintillation →	Taero
Sampling water vapor (H3R7000) →	Mixing with cocktail →	Liquid scintillation →	HTO
		Liquid scintillation →	HT
Air sampling (pressurized bottle) →	HT and total tritium extraction and mixing with cocktail →	Liquid scintillation →	Torg



Example of speciation in AREVA NC La Hague NRP atmospheric plume (in 2013 ~ 6.0 10^{13} Bq $year^{-1}$)

Number	Sampling date	Aerosol (Bq m^{-3})	HTO (Bq m^{-3})	HT (Bq m^{-3})	Torg (Bq m^{-3})
1	2013/02/22 10:20	<0.02	0.46 ± 0,01	1.2 ± 0,3	<0.4
2	2013/02/22 11:50	<0.02	0.37 ± 0,01	2.8 ± 0,5	<0.5
3	2013/06/13 10:15	<0.02	0.46 ± 0,02	2.0 ± 0,4	<0.4
4	2013/06/13 12:30	<0.02	0.48 ± 0,02	2.5 ± 0,5	<0.6
5	2013/06/27 14:27	<0.02	0.72 ± 0,02	4.1 ± 0,6	<0.9
6	2013/12/10 11:50	<0.02	0.40 ± 0,01	2.1 ± 0,4	<0.6
Mean		<0.02	0.50 (17%)	2.5 (83%)	<0.6

- 83 % of T released by the NRP is in the form of HT

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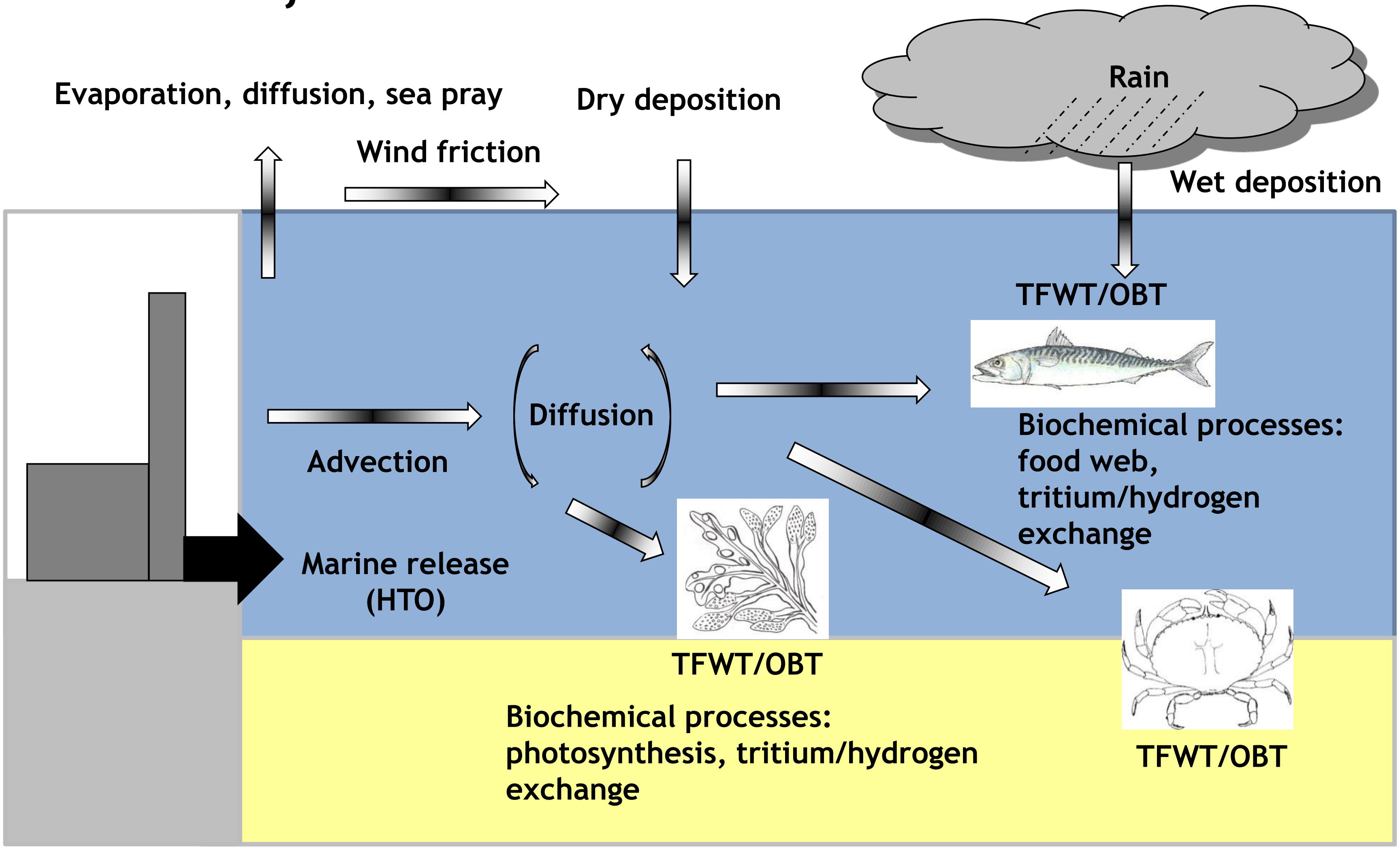
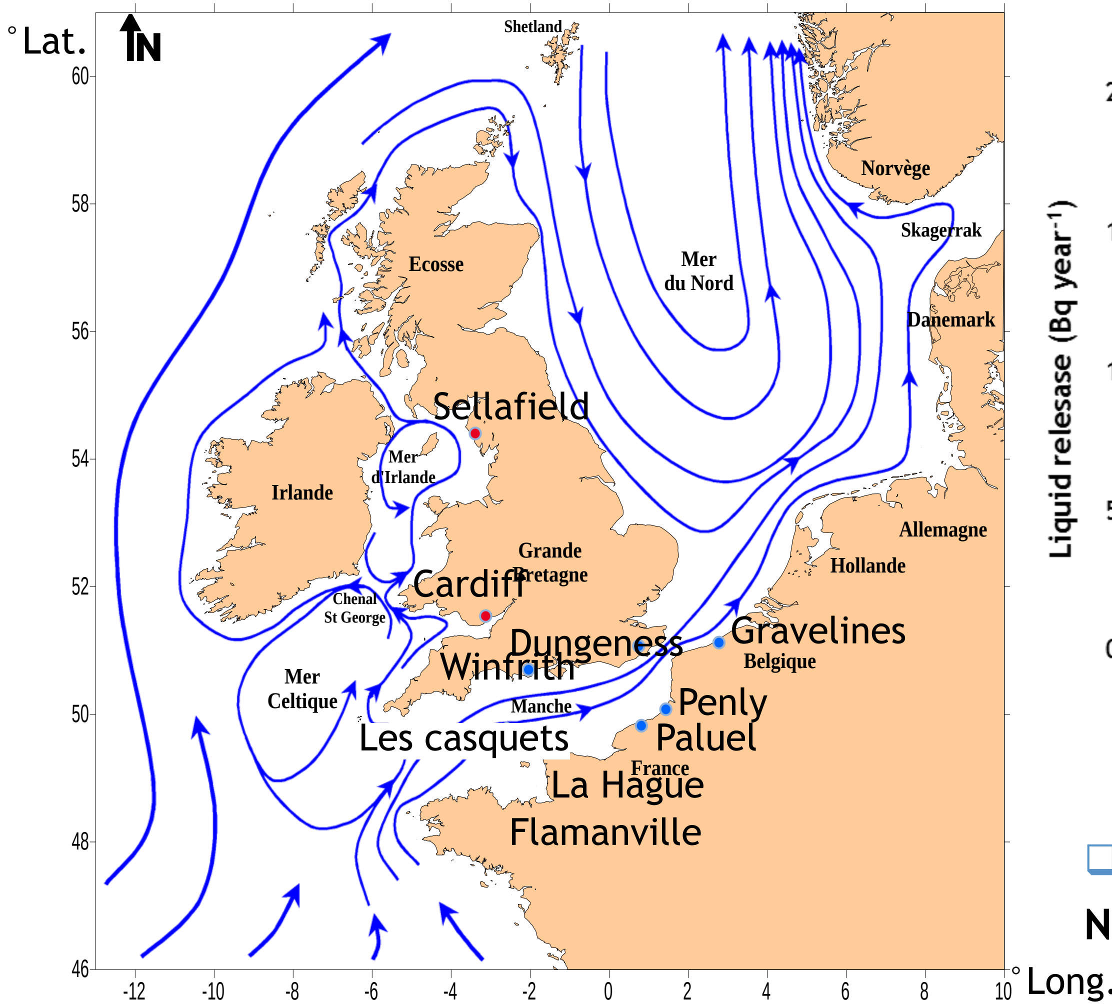
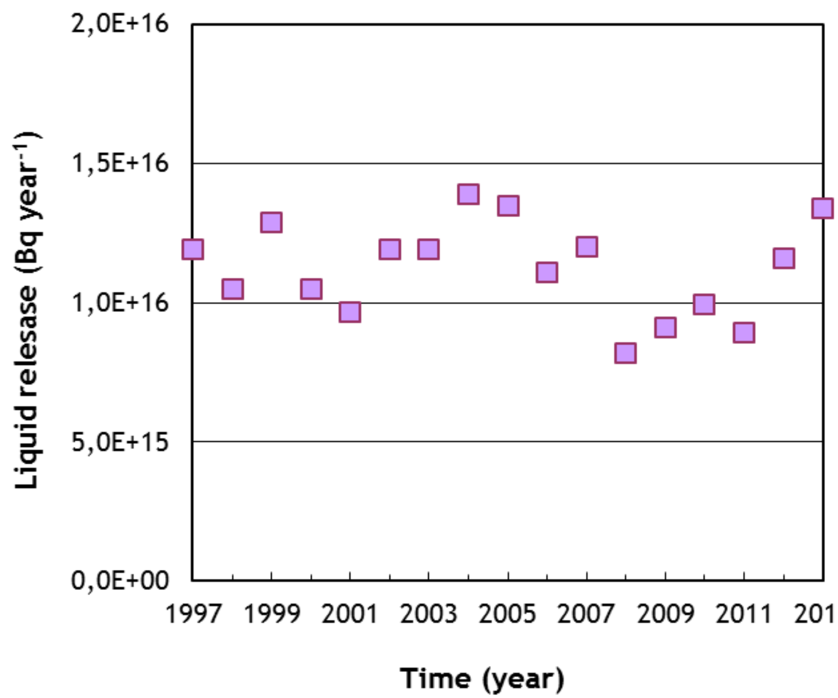
Behaviour of tritium in the marine environment



Liquid releases in the English Channel by AREVA NC: HTO

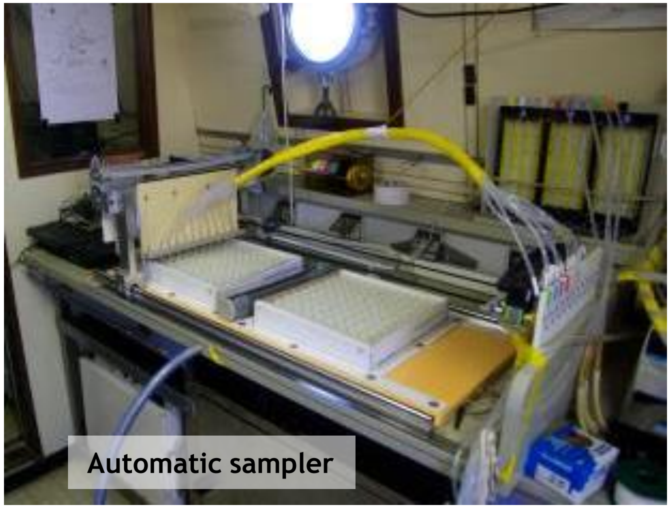


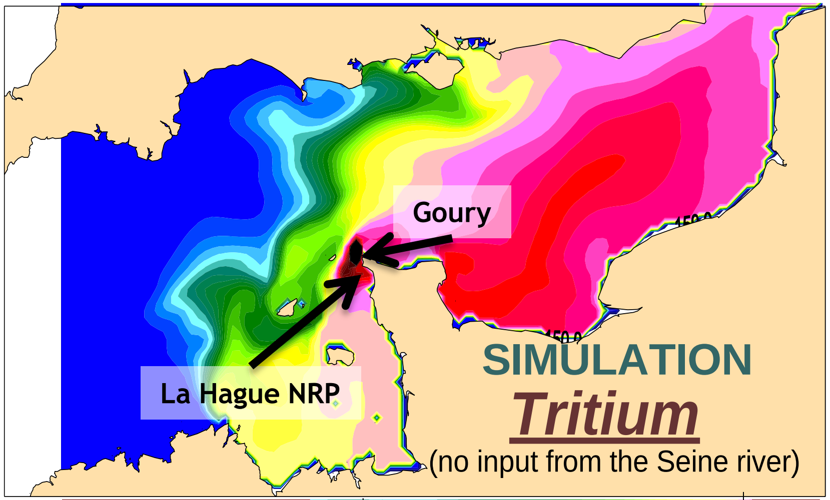


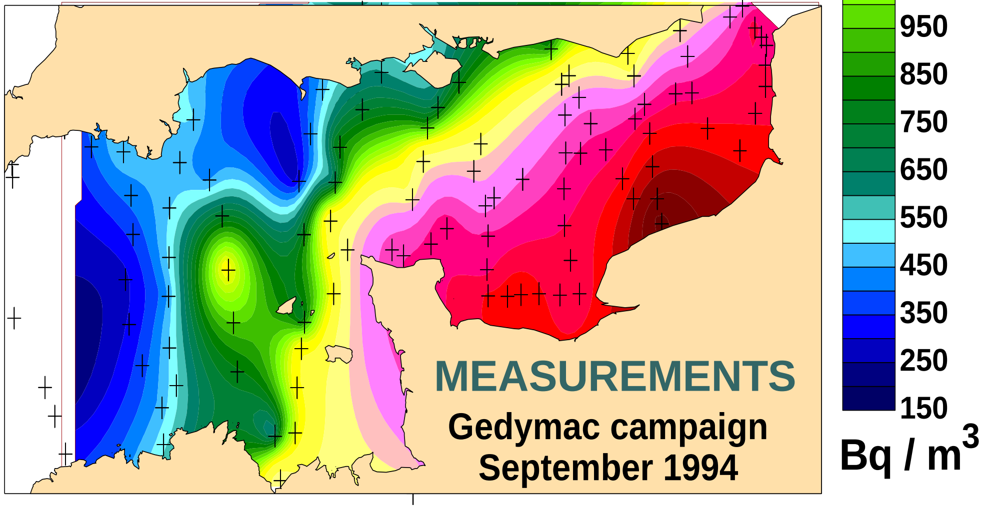
- One PWR NPP: 1/100 of La Hague NRP release

- Annual releases are quite stable ~ 1.2 10^{16} Bq $year^{-1}$

Model validation of hydrodynamic dispersion (numerous in-situ measurements vs. modeling)

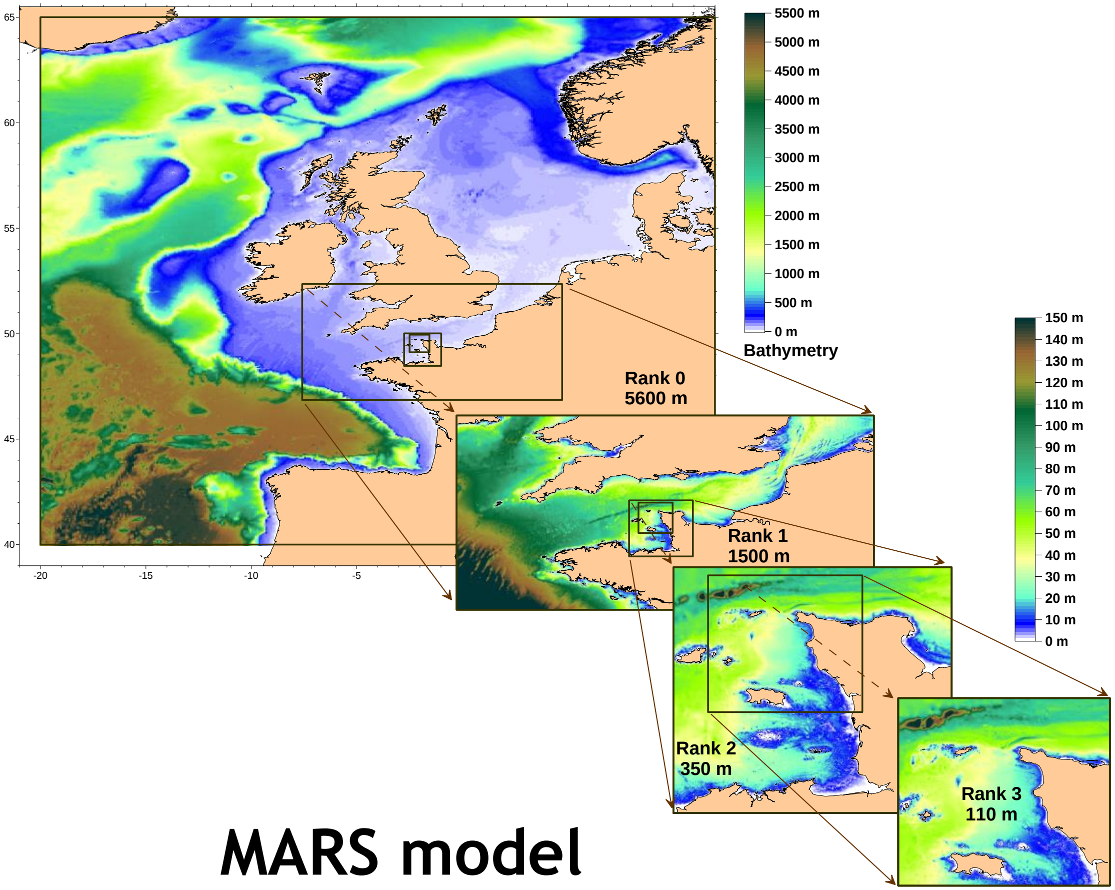




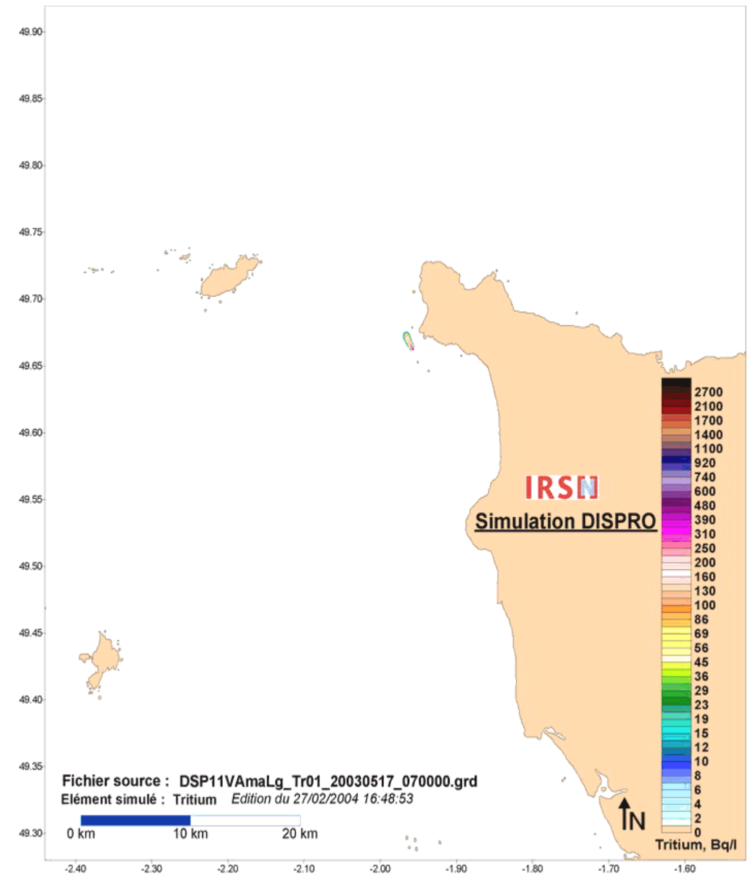


- **Average dilution coefficient close to the outfall of La Hague (Goury) - NRP is well known ~ 0,76 Bq m^{-3} per TBq $year^{-1}$**

Concentration in water: hydrodynamic modeling



MARS model frames and mesh sizes



Real discharges and meteorology, one frame every 15 min

- After model validation, the uncertainties for individual measurements are around 50% (the model is highly reliable)

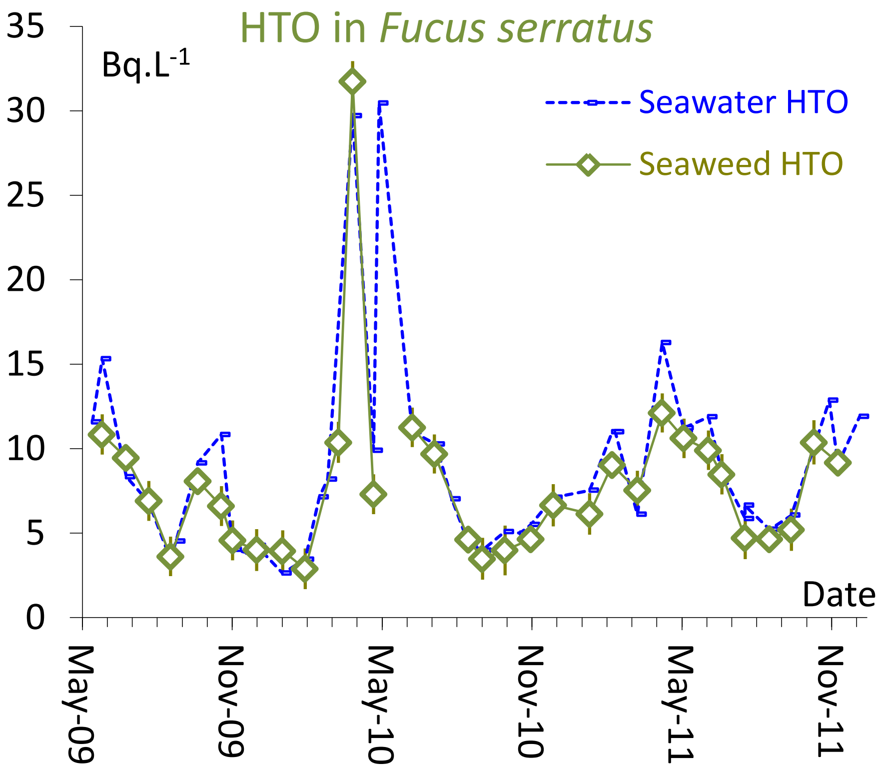
Marine ecosystem: transfer to biota

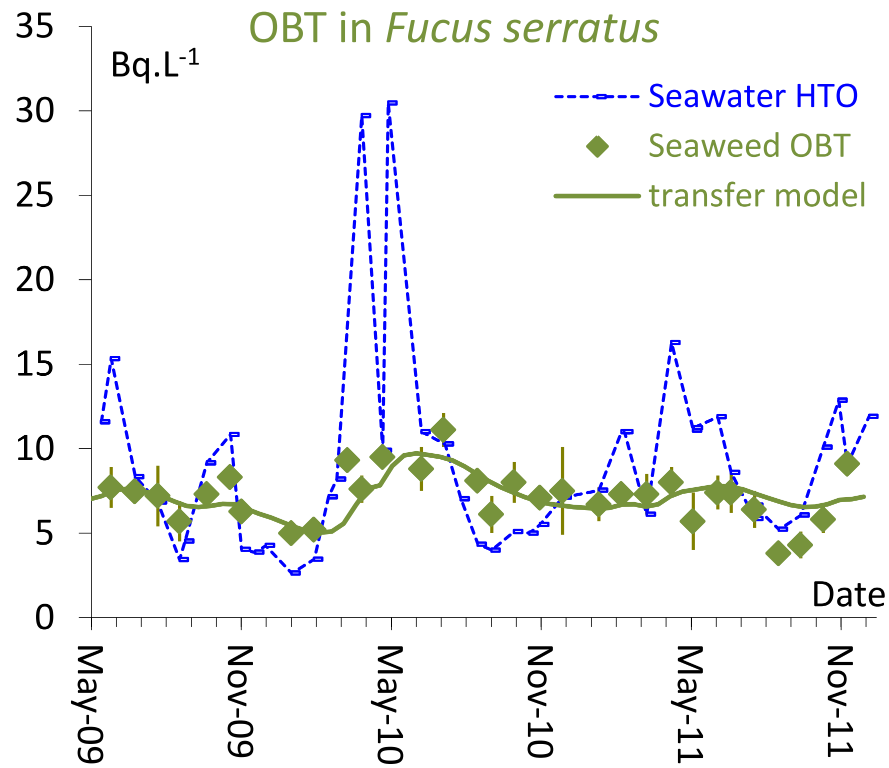
Concentration in marine biota near the point of release

Nature	Species	Location	Sample date	$Bq.Kg^{-1}$ fresh ± 10%	TFWT, $Bq.l^{-1}$ ± 10%	OBT, $Bq.l^{-1}$ ± 10%	Ratio OBT/HTO
Mollusc	earshell	Omonville La Rogue	21/07/1997	14.7	14.9	10.6	0.7
Crustacean	edible crab	Omonville La Rogue	21/07/1997	11.7	11.7	11.6	1.0
Mollusc	periwinkle	St Germain des Vaux	21/07/1997	12.5	12.7	9.3	0.7
Mollusc	periwinkle	Herquemoulin	21/07/1997	19.0	19.3	13.1	0.7
Crustacean	edible crab	Jobourg	21/07/1997	12.7	12.5	16.0	1.3
Fish	mackerel	Omonville La Rogue	21/07/1997	11.8	11.8	11.8	1.0
Mollusc	earshell	Herqueville	21/07/1997	15.6	15.9	10.8	0.7
Seaweed	*Fucus serratus*	Carteret	16/03/2006	4.0	3.9	5.0	1.3
Seaweed	*Fucus vesiculosus*	Dielette	15/03/2006	11.0	11.0	11.2	1.0
Seaweed	*Fucus serratus*	Dielette	15/03/2006	9.7	9.5	12.5	1.3
Seaweed	*Fucus vesiculosus*	Goury	14/03/2006	10.8	10.7	12.4	1.1
Seaweed	*Fucus serratus*	Goury	14/03/2006	11.4	11.3	12.3	1.2
Seaweed	*Fucus serratus*	Sciotot	15/03/2006	8.0	8.1	6.6	0.8
Crustacean	lobster	Carteret	23/05/2006	10.7	10.8	8.3	0.8
Crustacean	lobster	Flamanville	26/04/2006	10.1	10.1	9.1	0.9
Mollusc	limpet	Carteret	30/03/2006	4.0	4.0	4.2	1.1
Mollusc	limpet	Dielette	28/03/2006	17.2	17.4	14.2	0.8
Mollusc	whelk	Flamanville	26/04/2006	5.3	5.1	9.5	1.9
Mollusc	limpet	Goury	27/03/2006	16.0	16.1	13.1	0.8
Fish	plaice	Carteret	19/06/2006	5.9	5.8	8.6	1.5
Fish	sea wrasse	Carteret	19/06/2006	7.0	6.9	9.7	1.4
Fish	sole	Flamanville	26/04/2006	13.5	13.5	13.8	1.0
Fish	sea wrasse	Flamanville	26/04/2006	14.0	14.0	13.3	1.0
			Mean	**11.1**	**11.2**	**10.7**	**1.0**

- **Around 10 Bq L^{-1} (TFWT, OBT) for 1.2 10^{16} Bq $year^{-1}$**

Concentration in seaweed as function of water concentration



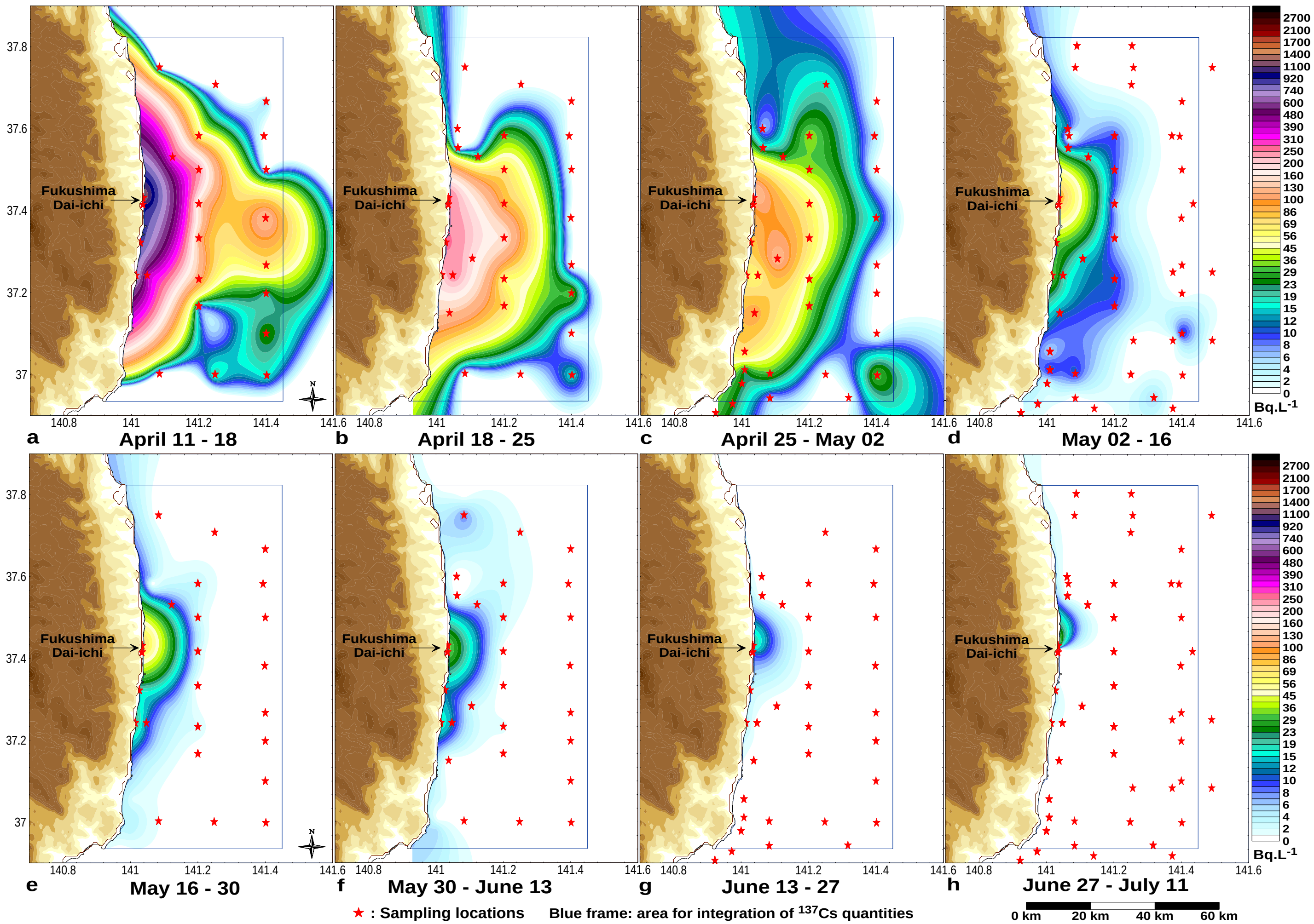


- **Steady state between seawater and seaweed TFWT is rapidly achieved while transfer between HTO and OBT is much slower: need to take this into account when modeling**

Transfer parameters	OBT/HTO(steady state), expecting 1	$t_{1/2}$bio (day) half-life
Fucus serratus	0.78 [0.76-0.82]	128 [99-175]

Fiévet B., Pommier J., Voiseux C., Bailly du Bois P., Laguionie P., Cossonnet C., Solier L. (2013) Transfer of tritium released into the marine environment by French nuclear facilities bordering the English Channel. Environmental Science & Technology 47(12): 6693-6703.

Estimation of ^{137}Cs quantities from measurements



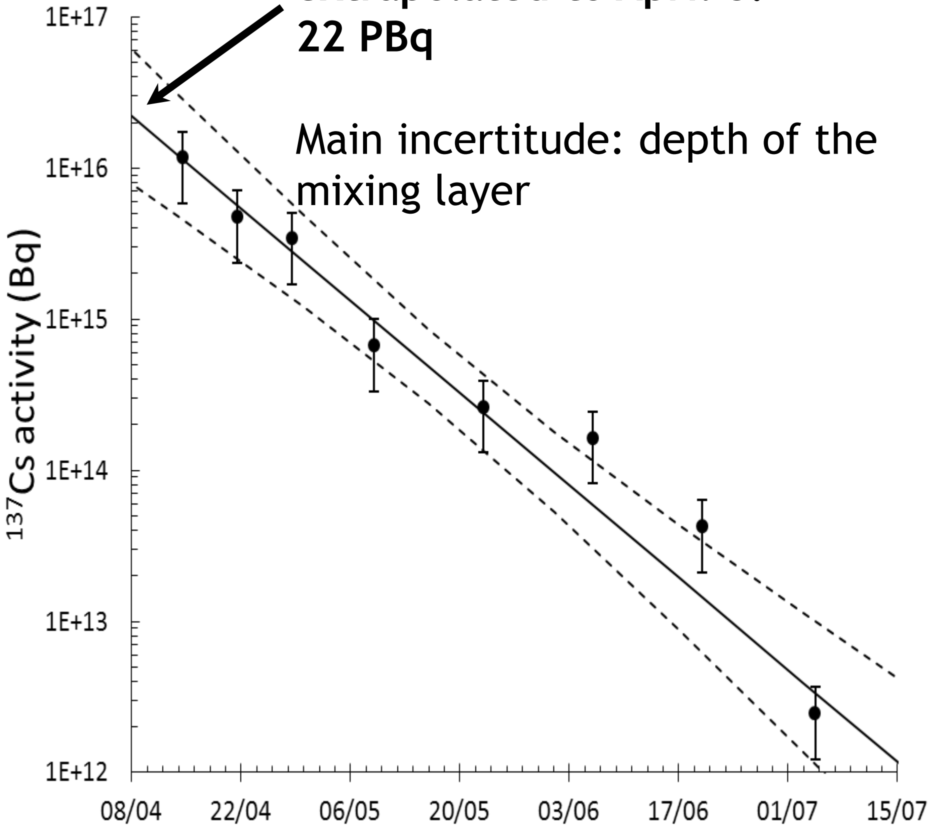
Interpolated ^{137}Cs concentrations from April 11 to July 11

- ^{137}Cs quantities were estimated on the basis of individual measurements in a 50 x 100 km (100-1000 measurements for each period) area around the plant (issues: depth, mixing layer, atmospheric fallout, rain water washout, …)

Estimation of the rate of seawater renewal

Exponential decay back extrapolated to April 8: 22 PBq

Main incertitude: depth of the mixing layer



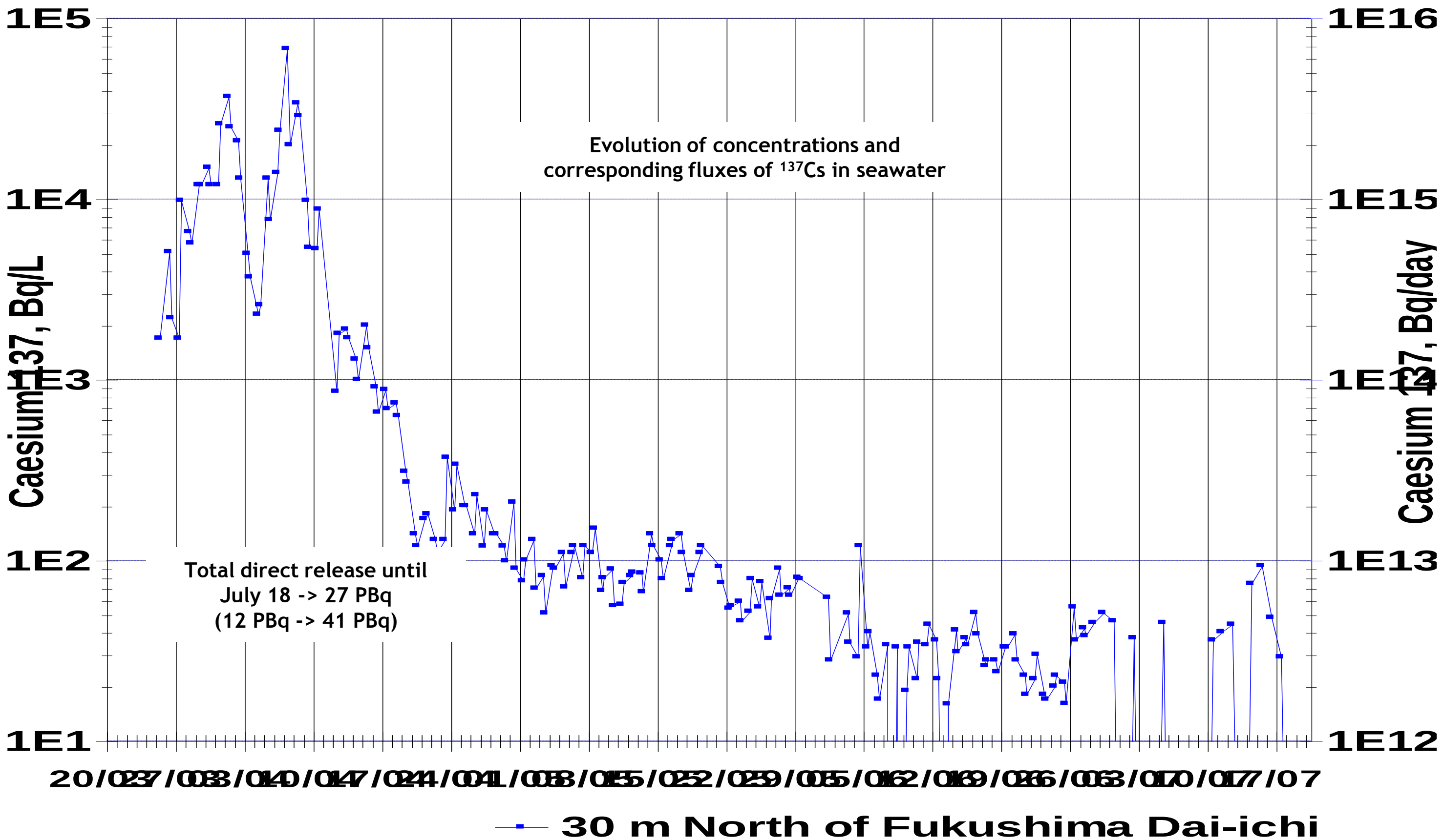
Evolution of ^{137}Cs quantities measured in seawater

Constant dilution by clean water through marine currents due to convergence of Kuroshio and Oyashio currents:

- Seasonal changes in the ocean circulation ?
- Return of contaminated water back in the area ?

- **Environmental half-time exponential decay $t_{1/2}$ = 6.9 days**
- **This source-term could be used in numerical dispersion models**

Flux estimation of ^{137}Cs from direct releases and the dilution coefficient



Assumptions:

- Measurements close to the plant are representative of the released flux;
- Amount of 22 PBq corresponds to the quantity of ^{137}Cs released from March 26 to April 8; (average concentration: 15 716 Bq.L^{-1}, number of values = 28, duration = 13.2 days);
- Right Y axis in figure shows this conversion.

- **Fluxes of ^{137}Cs could be deduced from concentrations by applying the factor: 1.06 10^{11} Bq released per Bq L^{-1} measured**

- **Conversely, concentrations could be estimated from fluxes**

Bailly du Bois P., Laguionie P., Boust D., Korsakissok I., Didier D., Fiévet B. 2011. Estimation of marine source-term following Fukushima Dai-ichi accident. Journal of Environmental Radioactivity. DOI:10.1016/j.jenvrad.2011.11.015

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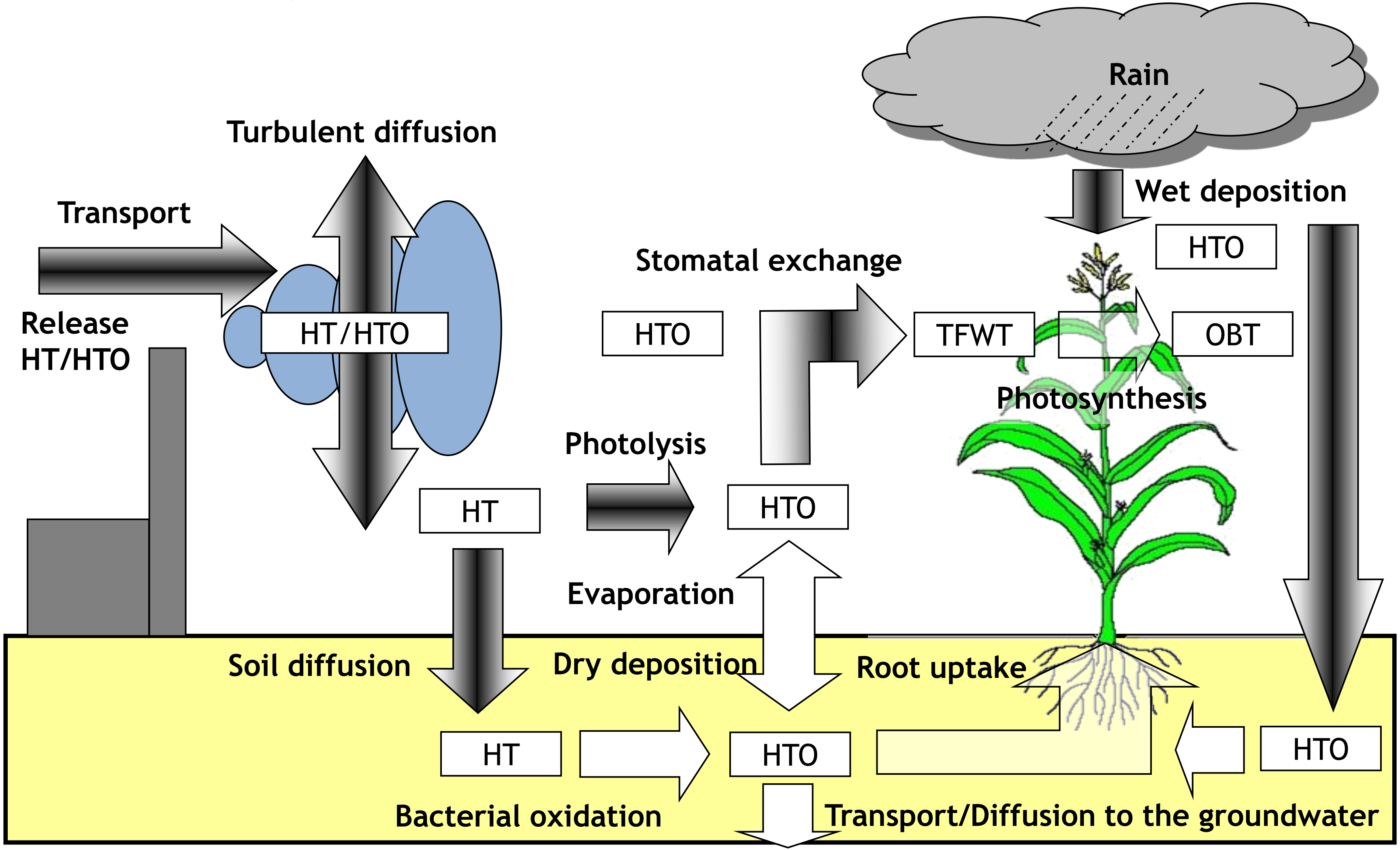
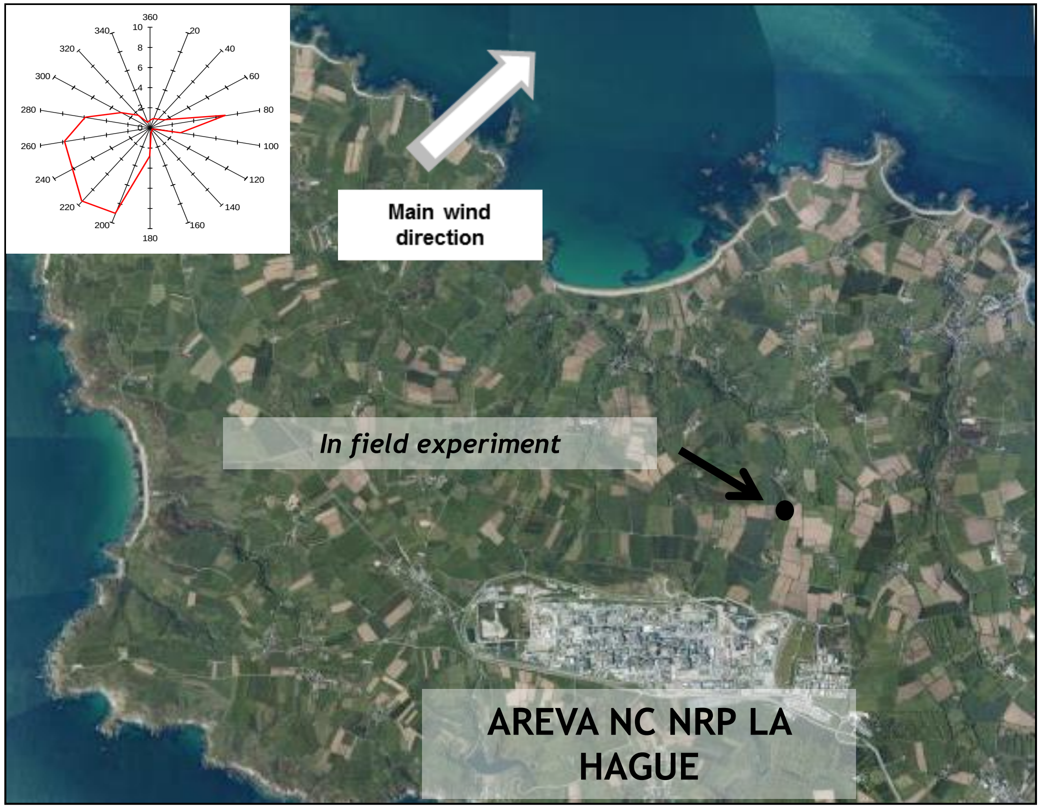
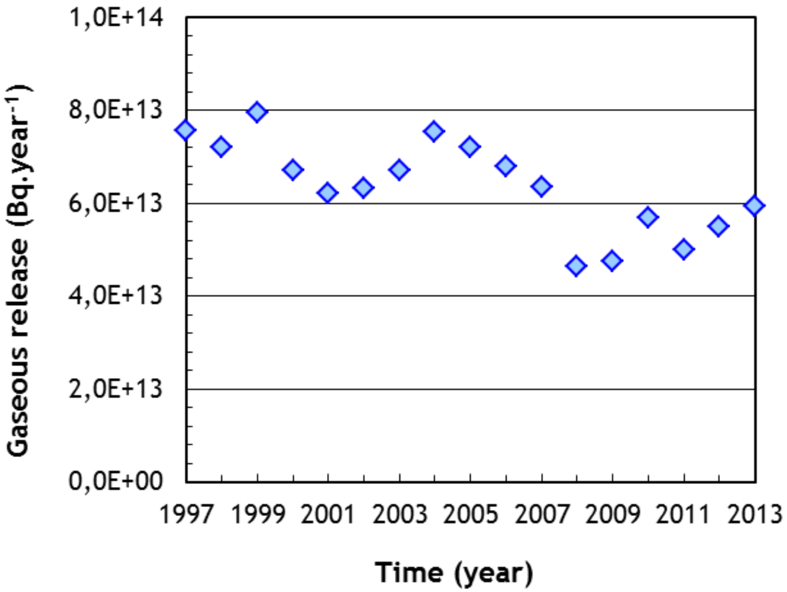
Behaviour of tritium in the terrestrial environment



- Background level in North hemisphere < 1 Bq L^{-1} water vapor (10^{-2} Bq m^{-3} air)

Gaseous release in the atmosphere by AREVA NC: HTO/HT

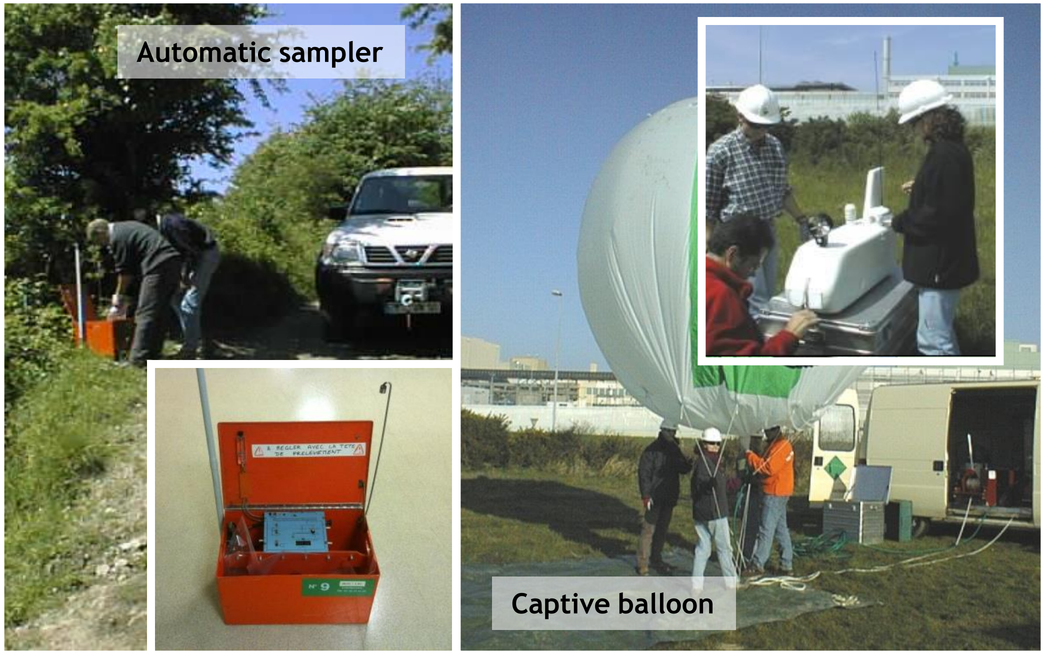


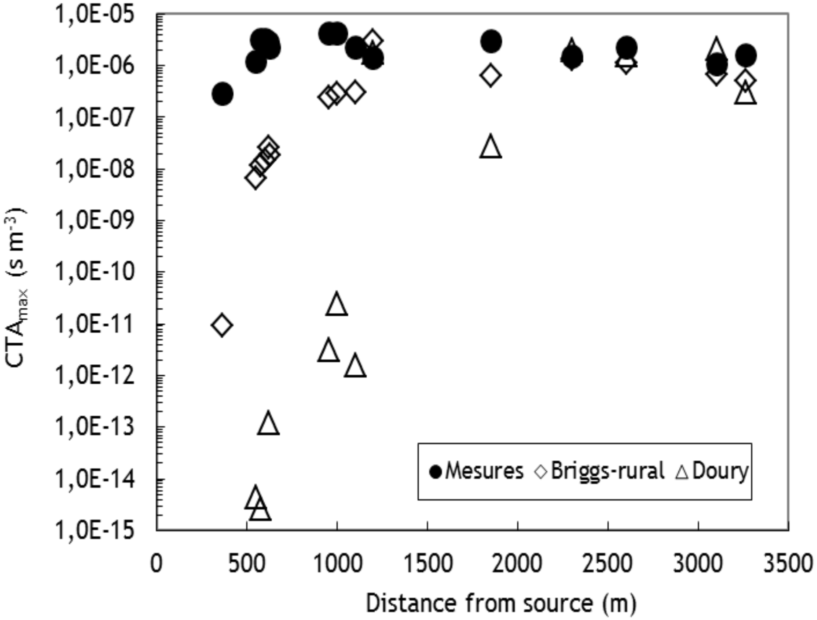


- One PWR NPP: 1/50 of La Hague NRP release

- Gaseous release decrease: in 2013 ~ 6.0 10^{13} Bq $year^{-1}$

Model validation: krypton 85 as a tool to validate atmospheric dispersion modeling



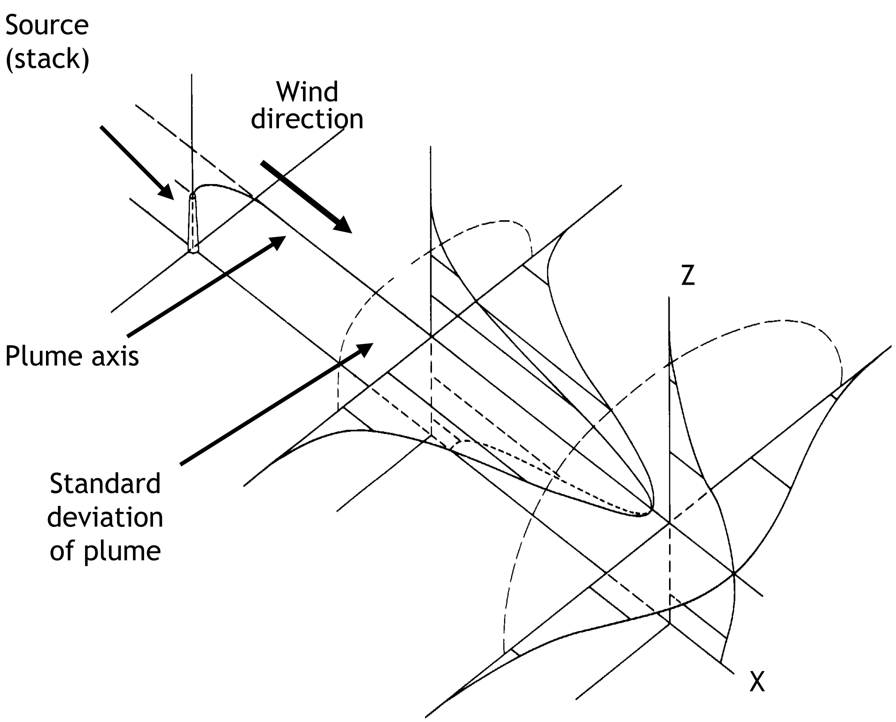


- Strong discrepancy between models and measurements

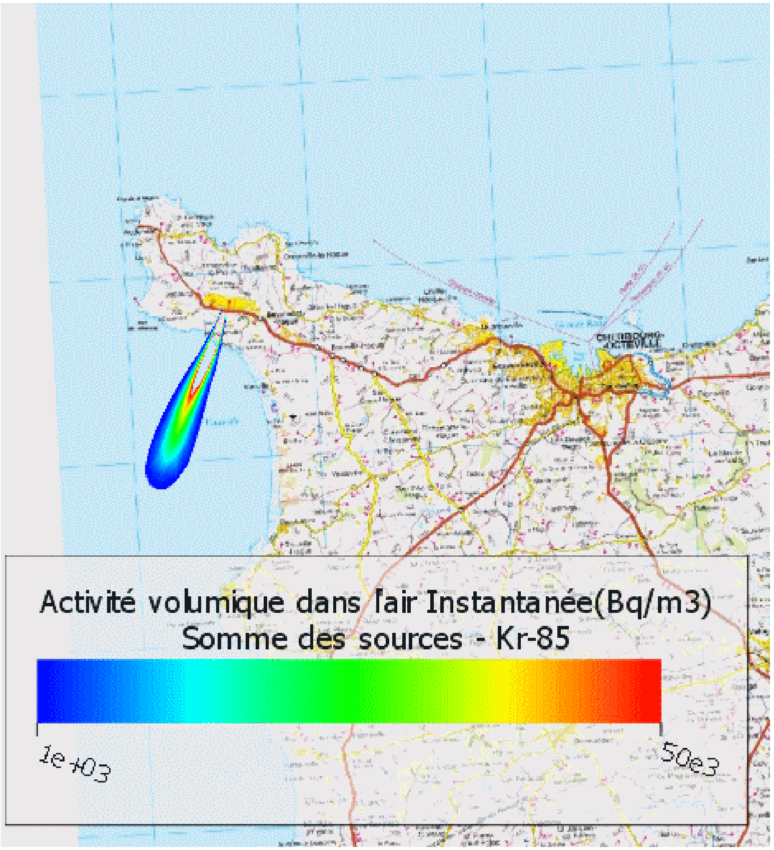
- Atmospheric Transfer Coefficient (ACT) ~ 3 10^{-6} s m^{-3}

Leroy C., Maro D., Hébert D., Solier L., Rozet M., Le Cavelier S. and Connan O. (2010). A study of the atmospheric dispersion of a high release of krypton-85 above a complex coastal terrain, comparison with the predictions of Gaussian models (Briggs, Doury, ADMS4). Journal of Environmental Radioactivity, 101, 11, 937-944.

Concentration in the atmosphere: Gaussian modeling



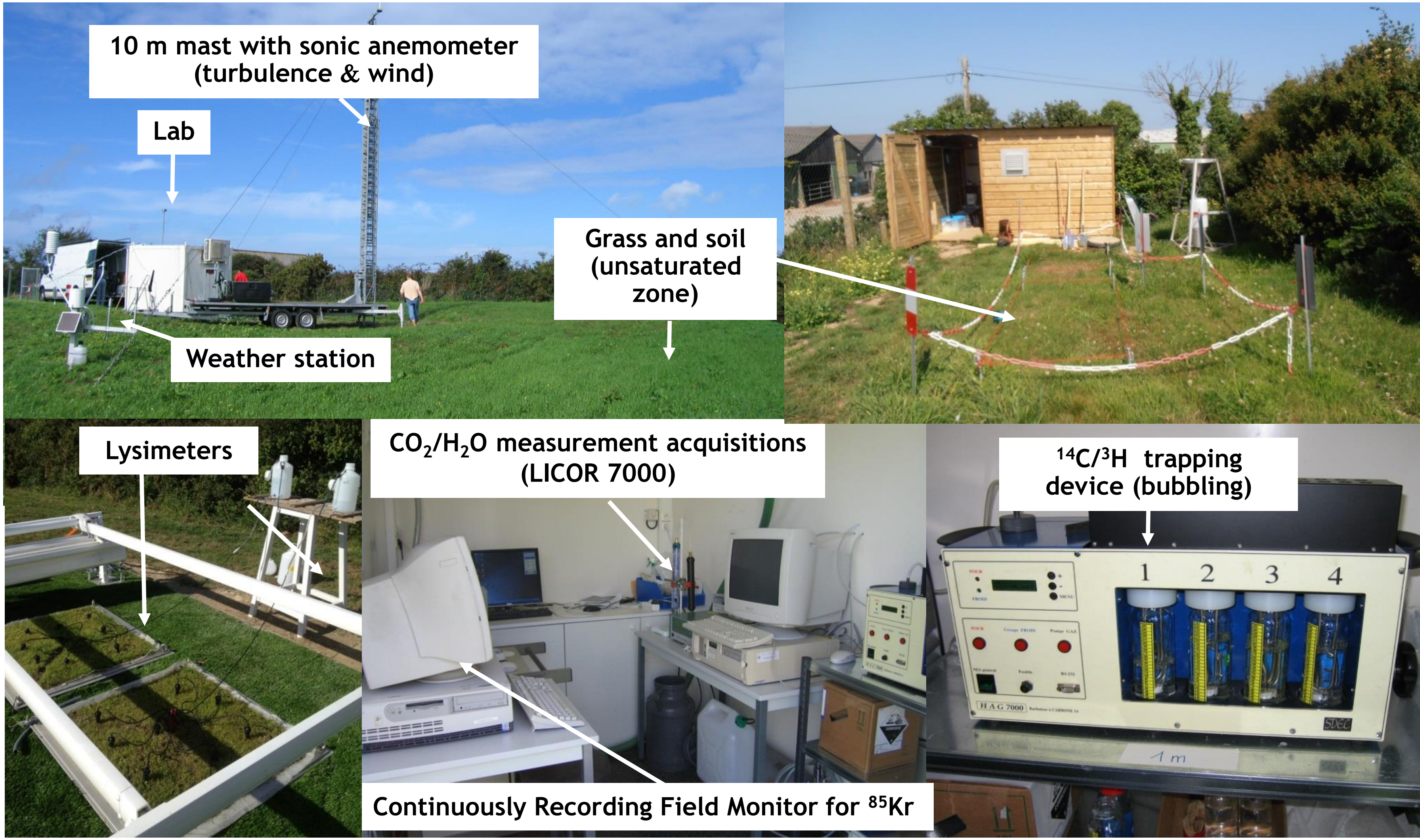
Gaussian modeling



- ❑ **After model validation the uncertainties are below a factor of 3 for in all meteorological conditions**

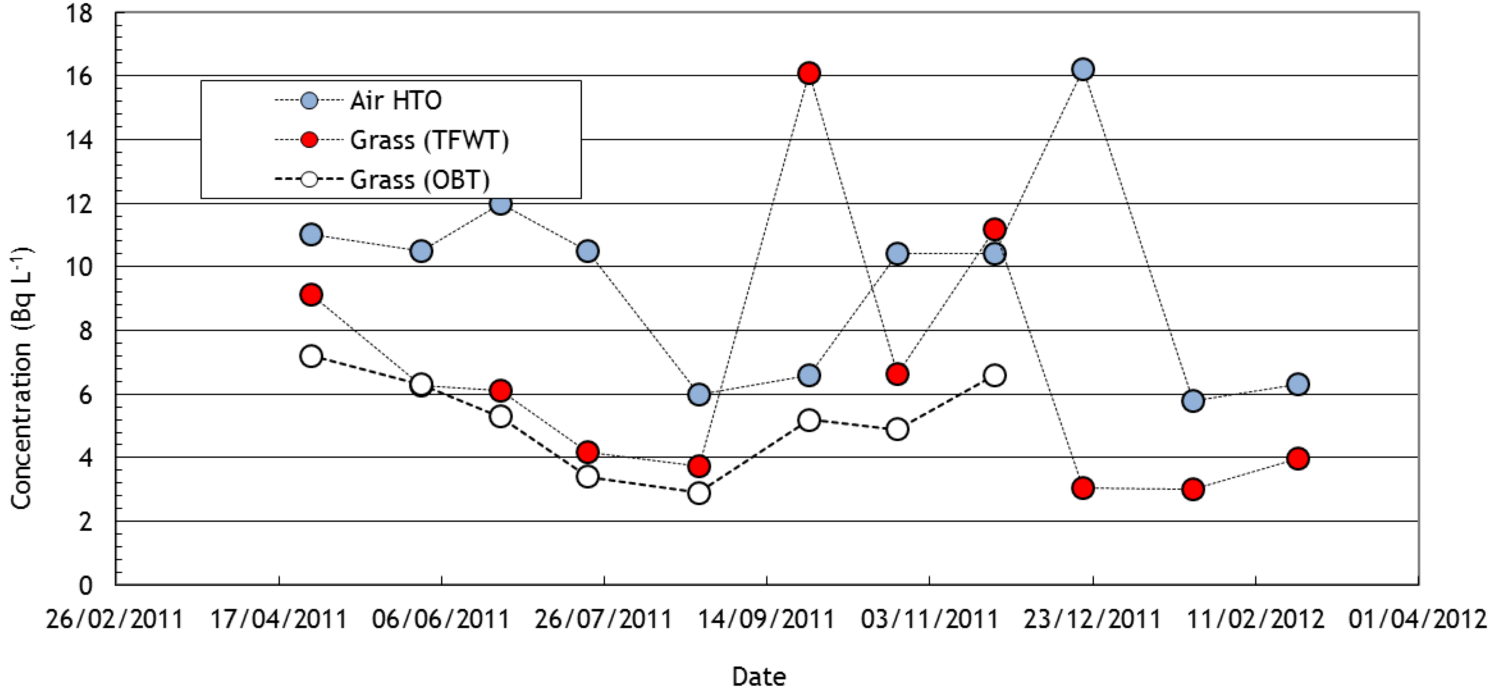
Terrestrial ecosystem: transfer to biota

- ***"In field experiment" technical platform (2 km downwind distance) to study tritium transfer (kinetics of OBT, dry and wet deposition, microbial oxidation of HT in soil)***



Terrestrial ecosystem: concentration to biota

- ***Concentration in grass around the NRP: “In field experiment” technical platform (2 km downwind distance)***



- **around 10 Bq L^{-1} for 6.0 10^{13} Bq $year^{-1}$**

- ***In La Hague area, concentrations in vegetable are in the same order of magnitude***

Modeling of tritium transfer in terrestrial ecosystem (TOCATTA), a part of the SYMBIOSE Platform

SOURCE	Gas dispersion (HTO)	Wet input (HTO, via precipitation) • Interception by soil	Wet input (HTO, via precipitation) • Interception by plant						
	CANOPY ATMOSPHERE	Soil surface exchange (HTO)	Foliar diffusion (TFWT)				Photosyn -thesis		
		SOIL WATER (HTO) Radioactive decay	Root uptake (HTO)						
			PLANT WATER (TFWT) Radioactive decay	Net formation (OBT)					
				PLANT DRY MATTER (OBT) Radioactive decay					
					Shoot structural dry matter			Ageing	Cutting (grass)
						Root structural dry matter		Ageing	
					Biological growth	Biological growth	Substrate (sap)		Respiration
								REST OF PLANT	
									SINK

- TOCATTA is a hourly time-step model, implemented within the SYMBIOSE modeling platform

Le Dizès S., Aulagnier C., Henner P., Simon-Cornu M. (2013) TOCATTA: a dynamic transfer model of 3H from the atmosphere to soil-plant systems. Journal of Environmental Radioactivity, 124: 191-204.

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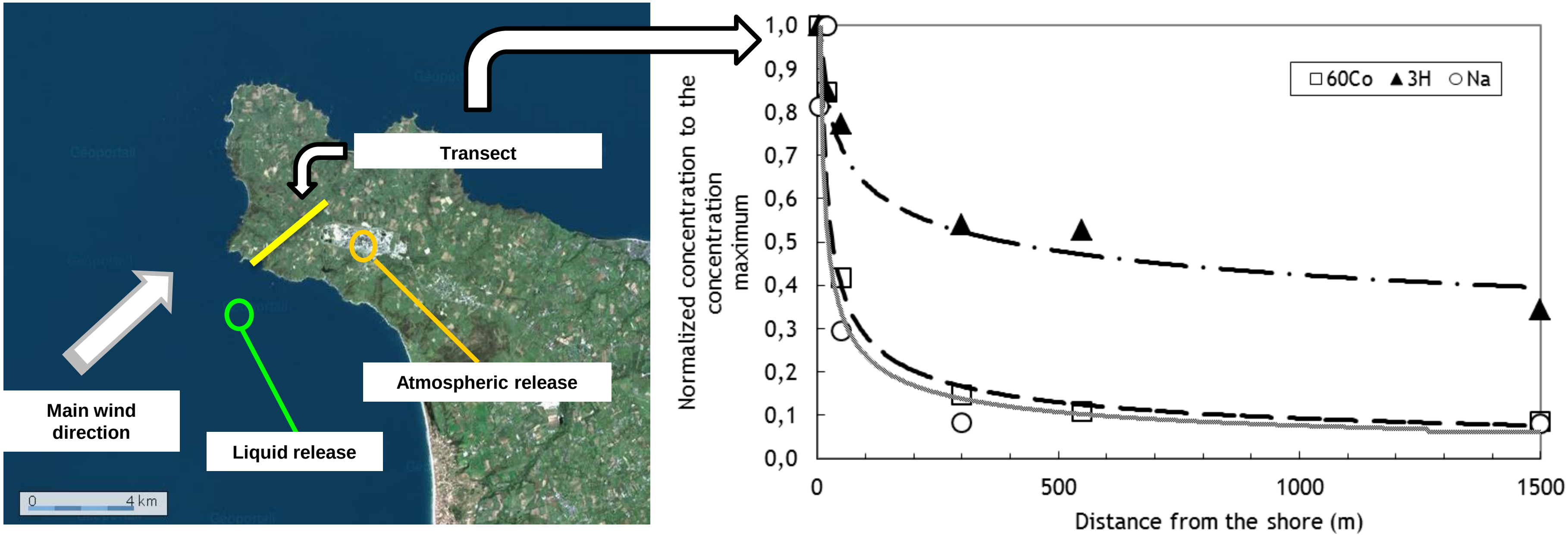
Tritium evaporation and partial pressure equilibrium induce transfer between water and the atmosphere



- Near the shore the atmospheric tritium concentration due to seawater is around 5 Bq L^{-1} of water vapor

Maro D., Tenailleau L., Fontugne M., Germain P., Hébert D. and Rozet M. (2005). Tritium transfer between sea and atmosphere in the English Channel (North Contentin and Bay of Seine). Radioprotection, 40, 1, 589-594.

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Conclusions: evaluation

What do we need to estimate the human dosimetric impact of tritium releases into the environment?

- Speciation of releases (HTO/HT/organic molecule): Tritium released in the environment as HTO behaves like H from H_20. The transfer to the biota, is very quick (hours) for TFWT and slower (months) for OBT;
- Dispersion: validation of dilution coefficients or dilution models are prerequisites to estimate the transfer to biota (e.g. Fukushima, 1.06 10^{11} Bq released per Bq L^{-1} measured);
- Transfer to biota: a constant ratio $^3H/H$ is kept in all compartments of the environment (e.g. water/air and TFWT/OBT);
- Water to atmosphere: this pathway could be taken into account for population close to seawater.

- ***Monitoring the tritium concentrations in the environment:***
  - Water: direct sampling;
  - Air: HTO by cold trap (e.g. H3R7000) and speciation by bubbling device (e.g. MARC 7000);
  - Biota and specifically food web: freeze drying to separate TFWT and OBT (combustion water extraction from dry matter).

- ***Measurement by counting scintillation (DL ~ 1 Bq L^{-1}). For lower levels: ^{3}He ingrown (Mass Spec);***

- ***For example, in the vicinity of La Hague NRP, average tritium concentrations are 10 Bq L^{-1} in the marine and the terrestrial ecosystems.***

Maro D. and Baron Y. (2007). Patent WO2008003853 A2. Method for automatically sampling tritium in the water vapour in air (PREVAIR).
Maro D and Hébert D. (2013). Patent FR 135319. System for the speciation of chemical forms of tritium from air (TRITAIR).

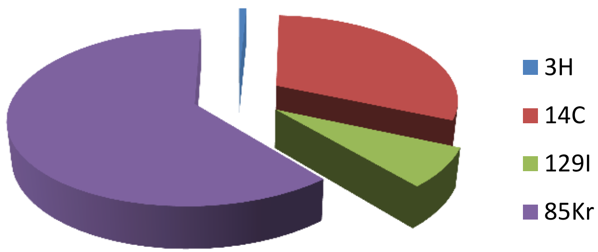
Conclusions: an example of stakeholders involvement in France

- ***This methodology was used by North-Cotentin Radioecology Group:***
  - Indeed, epidemiological studies have shown in 1997 a trend towards an excess number of leukaemia cases in the region of Nord-Cotentin (France) and it was suggested that the risk of leukaemia was associated with some aspects of lifestyle;
  - To respond to public concern, the French Ministries of the Environment and Health decided to commission complementary epidemiological studies and a detailed radioecological analysis;
  - The radioecological study was entrusted to a group of experts with various backgrounds (inspectors, governmental experts, operators, experts from non-governmental laboratories and foreign experts)-the North-Cotentin Radioecology Group;
  - Its principal objective was to estimate the exposure levels to ionizing radiation and associated risk of leukemia for populations in the Nord-Cotentin.

Conclusions: application

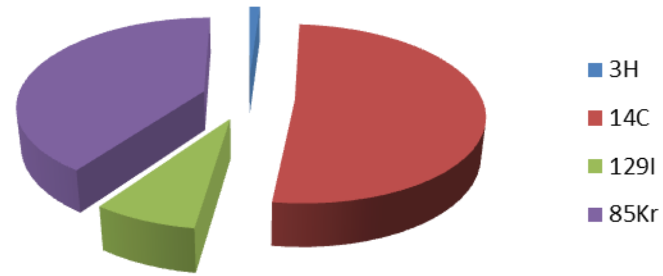
- ***Example, the gaseous/liquid releases contribution to the dose to the public in 2010: 8.7 µSv y^{-1} for the farmer and 4.7 µSv y^{-1} for the fisherman***
  - Gaseous releases: ^{3}H contribution ~ 1% for 57 TBq y^{-1}

Farmer (Digulleville)



^{3}H dose: 6.3 10^{-2} µSv y^{-1}

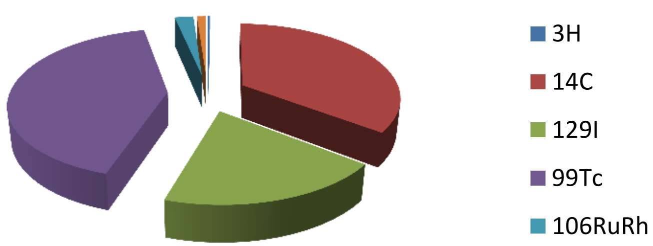
Fisherman (Goury)



^{3}H dose: 2.4 10^{-2} µSv y^{-1}

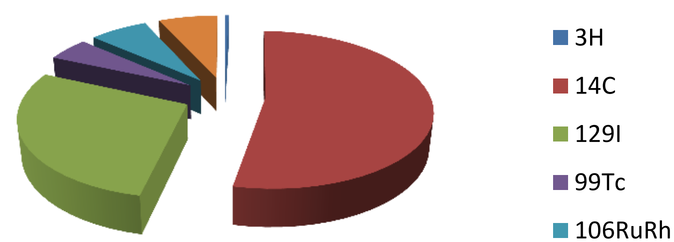
  - Liquid releases: ^{3}H contribution ~ 1% for 10 000 TBq y^{-1}

Farmer (Digulleville)



^{3}H dose: 2.6 10^{-3} µSv y^{-1}

Fisherman (Goury)



^{3}H dose: 9.6 10^{-3} µSv y^{-1}

Conclusions: general

- *In France tritium releases will increase in the future with new built nuclear facilities.*
- *ASN has coordinated a work on tritium (White book, 2010) and recommended to improve knowledge on tritium transfers in the ecosystems: see lecture on tritium “the French situation” by J. L. Lachaume (ASN, Deputy Director General) at METI.*
- *IRSN studies tritium behaviour to have more realistic dose assessment for human and biota.*
- *Uncertainties remain in the marine and terrestrial ecosystems (kinetic of OBT formation, wet deposition…).*
- *IRSN will carry on developing specific programs on these topics (e.g. VATO project).*

Conclusions: IRSN

- ***How can IRSN help for calculating dosimetric impact of controlled release of tritium?***
  - IRSN has developed up-to-date parameterized models of tritium transfers in marine & terrestrial ecosystems where various scenarios can be tested and compared in terms of resulting human dosimetric impact, including sensitivity and uncertainty analyses;
  - IRSN has developed sampling methodologies for environmental monitoring of tritium in various compartments and ecosystems;
  - IRSN has developed low-level tritium metrology adapted to environmental monitoring.


Future IAEA tritium meeting group
MODARIA
will be organized by IRSN in Cherbourg
June10-13, 2014
Last tritium workshop CNSC – IRSN 2011