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

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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**
Outlines

Context

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Conclusions
**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**
**Context**

**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.

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
Context

Objectives

Transfer of tritium in marine and terrestrial ecosystems

- Importance of the knowledge of speciation (HT, HTO, OBT)
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Conclusions
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.
Outlines

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**Transfer of tritium in marine and terrestrial ecosystems**

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- **Conclusions**
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.
Importance of knowledge of the speciation

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

**Sampling**
- Sampling particles on filter (PIAF)
- Sampling water vapor (H3R7000)
- Air sampling (pressurized bottle)

**Treatment**
- Dilution in cocktail
- Mixing with cocktail
- HT and total tritium extraction and mixing with cocktail

**Measurement**
- Liquid scintillation

**Physico-chemical forms**
- Taero
- HTO
- HT
- Torg

**Equipment Images**
- PIAF
- H3R7000 (IRSN, Navy patent)
- Bottle B20
- In Lab extraction
### Importance of the knowledge of the speciation

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

<table>
<thead>
<tr>
<th>Number</th>
<th>Sampling date</th>
<th>Aerosol (Bq m$^{-3}$)</th>
<th>HTO (Bq m$^{-3}$)</th>
<th>HT (Bq m$^{-3}$)</th>
<th>Torg (Bq m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2013/02/22 10:20</td>
<td>&lt;0.02</td>
<td>0.46 $\pm$ 0.01</td>
<td>1.2 $\pm$ 0.3</td>
<td>&lt;0.4</td>
</tr>
<tr>
<td>2</td>
<td>2013/02/22 11:50</td>
<td>&lt;0.02</td>
<td>0.37 $\pm$ 0.01</td>
<td>2.8 $\pm$ 0.5</td>
<td>&lt;0.5</td>
</tr>
<tr>
<td>3</td>
<td>2013/06/13 10:15</td>
<td>&lt;0.02</td>
<td>0.46 $\pm$ 0.02</td>
<td>2.0 $\pm$ 0.4</td>
<td>&lt;0.4</td>
</tr>
<tr>
<td>4</td>
<td>2013/06/13 12:30</td>
<td>&lt;0.02</td>
<td>0.48 $\pm$ 0.02</td>
<td>2.5 $\pm$ 0.5</td>
<td>&lt;0.6</td>
</tr>
<tr>
<td>5</td>
<td>2013/06/27 14:27</td>
<td>&lt;0.02</td>
<td>0.72 $\pm$ 0.02</td>
<td>4.1 $\pm$ 0.6</td>
<td>&lt;0.9</td>
</tr>
<tr>
<td>6</td>
<td>2013/12/10 11:50</td>
<td>&lt;0.02</td>
<td>0.40 $\pm$ 0.01</td>
<td>2.1 $\pm$ 0.4</td>
<td>&lt;0.6</td>
</tr>
<tr>
<td>Mean</td>
<td></td>
<td>&lt;0.02</td>
<td>0.50 (17%)</td>
<td>2.5 (83%)</td>
<td>&lt;0.6</td>
</tr>
</tbody>
</table>

- 83 % of T released by the NRP is in the form of HT
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Behaviour of tritium in the marine environment

Marine ecosystem: schematic transfer pathways

Evaporation, diffusion, sea spray
Dry deposition
Wind friction
Rain
Wet deposition
TFWT/OBT
Biochemical processes:
food web, tritium/hydrogen exchange

Marine release (HTO)
Advection

TFWT/OBT
Biochemical processes:
photosynthesis, tritium/hydrogen exchange
Liquid releases in the English Channel by AREVA NC: HTO

- Annual releases are quite stable ~ $1.2 \times 10^{16}$ Bq year$^{-1}$
- One PWR NPP: 1/100 of La Hague NRP release
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\(^{-1}\)
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

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

**Mean**

- **11.1**
- **11.2**
- **10.7**
- **1.0**

- Around 10 Bq L⁻¹ (TFWT, OBT) for 1.2 \(10^{16}\) Bq year⁻¹
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.

<table>
<thead>
<tr>
<th>Transfer parameters</th>
<th>OBT/HTO (steady state), expecting 1</th>
<th>( t_{1/2} \text{bio (day)} ) half-life</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fucus serratus</td>
<td>0.78 [0.76-0.82]</td>
<td>128 [99-175]</td>
</tr>
</tbody>
</table>
Estimation of $^{137}$Cs quantities from measurements

- $^{137}$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, ...)

Interpolated $^{137}$Cs concentrations from April 11 to July 11
Marine ecosystem: dispersion around Fukushima NPP

**Estimation of the rate of seawater renewal**

Exponential decay back extrapolated to April 8: 22 PBq

Main incertitude: depth of the mixing layer

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?

Evolution of $^{137}$Cs quantities measured in seawater:

- 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

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

Conversely, concentrations could be estimated from fluxes.

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: $15.716 \text{ Bq.L}^{-1}$, number of values = 28, duration = 13.2 days);
- Right Y axis in figure shows this conversion.

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Terrestrial ecosystem: schematic transfer pathways

**Behaviour of tritium in the terrestrial environment**

- **Turbulent diffusion**
- **Transport**
- **Release HT/HTO**
- **Soil diffusion**
- **Dry deposition**
- **Evaporation**
- **Bacterial oxidation**
- **Root uptake**
- **Transport/Diffusion to the groundwater**
- **Stomatal exchange**
- **Photolysis**
- **Wet deposition**
- **Rain**
- **Wet deposition**
  - HTO
  - OBT
  - TFWT

- **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

- Automatic sampler
- Captive balloon

Strong discrepancy between models and measurements

Atmospheric Transfer Coefficient (ACT) $\sim 3 \times 10^{-6}$ s m$^{-3}$

Concentration in the atmosphere: 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)

- 10 m mast with sonic anemometer (turbulence & wind)
- Lab
- Grass and soil (unsaturated zone)
- Weather station
- Lysimeters
- CO₂/H₂O measurement acquisitions (LICOR 7000)
- Continuously Recording Field Monitor for ⁸⁵Kr
- ¹⁴C/³H trapping device (bubbling)
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

- 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)
  - Photosynthesis

- SOIL WATER (HTO)
  - Root uptake (HTO)

- PLANT WATER (TFWT)
  - Net formation (OBT)

- PLANT DRY MATTER (OBT)
  - Shoot structural dry matter
  - Root structural dry matter
  - Ageing
  - Cutting (grass)

- REST OF PLANT
  - Biological growth
  - Substrate (sap)
  - Respiration
  - Ageing
  - Rest of plant

- SINK

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

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- **Conclusions**
Interface between marine/terrestrial ecosystems

**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

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Conclusions
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$_2$O. 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 $^3$H/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: ³He 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.**

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.
Example, the gaseous/liquid releases contribution to the dose to the public in 2010: 8.7 $\mu$Sv y$^{-1}$ for the farmer and 4.7 $\mu$Sv y$^{-1}$ for the fisherman

- Gaseous releases: $^3$H contribution $\sim$ 1% for 57 TBq y$^{-1}$
- Liquid releases: $^3$H contribution $\sim$ 1% for 10 000 TBq y$^{-1}$
**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 June 10-13, 2014