Raport stiintific

octombrie – decembrie 2011

Abordare interdisciplinara a modelarii dinamice a transferului tritiului in plante agricole PN-II-ID-PCE-2011-3-0396

In propunerea de proiect, ne-am agajat pentru 2011, sa facem analiza experimentelor dedicate producerii tritiului legat organic in plante agricole, pe baza analizei detaliate a rezultatelor experimentale. Contributia noastra este parte a unui document tehnic (draft) al Agentiei Internationale de Energie Atomica (AIEA), proiectul EMRAS II (Environmental Modelling for RAdiation Safety), grupul de lucru WG 7 „Tritium Accident", coordonat de directorul de proiect. In prezentul raport includem succint toate contributiile IFIN-HH la acest raport.

Tritiul intra direct in ciclul vietii in multiple forme fizico-chimice. Tritiul emis si scapat din instalatiile nucleare este de regula, sub forma de apa tritiata (HTO), dar o parte din HTO este convertita in tritiu legat organic (OBT) prin intermediul proceselor metabolice din plante si animale. OBT are un timp de rezidenta mai mare in organisme si de aceea, determina un timp de expunere mai mare, care trebuie luat in considerare in studiile de estimare radiologica. In scopul radioprotectiei si securitatii nucleare, este important sa apreciem corect urmarile radiologice al unei emisii accidentale de tritiu, folosind modele robuste, cu incertitudine mica. AIEA coordoneaza proiectul EMRAS (2003-2011). In prima faza, EMRAS I (2003-2007) Romania, prin IFIN-HH, a participat la grupul de lucru "Modelling of Tritium and Carbon-14 transfer to biota and man" (Modelarea transferului tritiului si ^{14}C in flora, fauna si oameni), deoarece acesti radionuclizi sunt de importanta nationala pentru reactorii CANDU, care stau la baza energeticii nucleare romane. In acest grup de lucru, una din activitati s-a concentrat pe modelarea unui accident ipotetic cu emisie de tritiu. Rezultatele diferitilor participanti au demonstrat un grad inaceptabil de incertitudine. Ca urmare, in programul EMRAS II (2009-2011) s-a decis formarea unui grup de lucru dedicat subiectului: WG 7 " Tritium accident". Cu sprijin intern din IFIN-HH si prin accept international, seful de grup a fost ales din IFIN-HH, ca recunoastere a profesionalismului si experientei internationale. Complexitatea problemei si limitele de buget nu au permis dezvoltarea unui model robust apt a fi aplicat in caz operational, dar multe din elementele acestui model au fost acoperite, iar preazentul proiect are ca scop completarea cercetarii pana la nivelul in care aplicatia operationala sa poate fi facuta cu implicarea directa a utilitatii nucleare si cu un raport cost/calitate optim.

Fiecare grup de lucru din EMRAS II are o pagina web dedicata, iar activitatea IFIN-HH in WG7 poate fi monitorata la http://www-ns.iaea.org/projects/emras/emras2/working-groups/working-group-seven.asp?s=8.

Confor politicii stintifice a AIEA, grupurile de lucru trebuie sa finalizeze activitatea prin documente tehnice (TECDOC), care vin sa sprijine activitatea din domeniu a tarilor participante.. Pentru grupul WG 7, la care IFIN-HH este responsabil, redactarea documentatie tehnice este incredintata IFIN-HH pentru draftul care trebuie finalizat anul acesta. In forma finala, publicata de AIEA, unele elemente de redactare vor fi schimbate si aprobarile interne AIEA, cat si publicarea pot cere un interval de timp de 1-3 ani. In acest moment, draftul documetatie tehnice cuprinde 15 capitole, dintre care partea romana este implicata in 9 dintre ele. Continutul acestui draft, aflat in faza finala, este dat mai jos iar autorii din IFIN-HH sunt subliniati. Remarcam ca tematicile abordate de IFIN-HH sunt complex,e iar acest raport de faza nu le va expune explicit. Ne rezuman la o prezentare succinta a doua contributii recente si de impact general, care nu sunt cuprinse in publicatile noastre.

Tabla de materii TECDOC draft "Tritium Accident", elaborata sub conducerea IFIN-HH:

7. OVERVIEW EXPERIMENTS ON TRITIUM TRANSFER FROM AIR TO PLANTS AND THE SUBSEQUENT CONVERSION TO OBT (D. Galeriu, A. Melintescu, S. Strack, S.B. Kim, M. Andoh-Atarashi)
8. REVIEW ON SOIL-PLANT TRITIUM TRANSFER (V. Korolevych)
9. TRITIUM TRANSFER IN WHEAT EXPERIMENTS AND MODELS TESTS (D. Galeriu)
10. TRITIUM TRANSFER IN FARM ANIMALS (D. Galeriu, A. Melintescu)
11. BRIEFING OF COMPLEX MODEL (H. Nagai, M. Ota)
12. TRITIUM IN AQUATIC FOODCHAIN (A. Melintescu, F. Siclet, D. Galeriu).
13. QUALITY ASSURANCE OF DATA (S.B. Kim)
14. QUALITY ASSURANCE OF MODELS (J. Duran, D Galeriu).
15. STATUS AND PERSPECTIVES OF ACCIDENTAL TRITIUM MODELLING (D. Galeriu)

In capitolul dedicat depunerii umede a tritiului, contributia originala a IFIN-HH consta in analiza influentei cladirilor de pe amplasamentul nuclear asupra dispersiei tritiului si a depunerii umede, fenomen ce impune o tratare complexa a dispesiei poluantului si o analiza profunda a factorilor ce influenteaza depunerea umeda a tritiului.. Analiza rezultatelor experimentale privind rata de depunere (washout rate) evidentiaza un domeniu larg de valori - 2 ordine de marime conform Figurii 1. In Figura 2, comparam valorile masurate cu cele teoretice, in care distributia diametrului picaturilor de ploaie este parametrizata empiric si demonstram importanta acestui parametru care trebuie masurat in conditii locale. Se evidentiaza in acelasi timp ca modelul francez curent subestimeaza datele cu un factor 2-3.

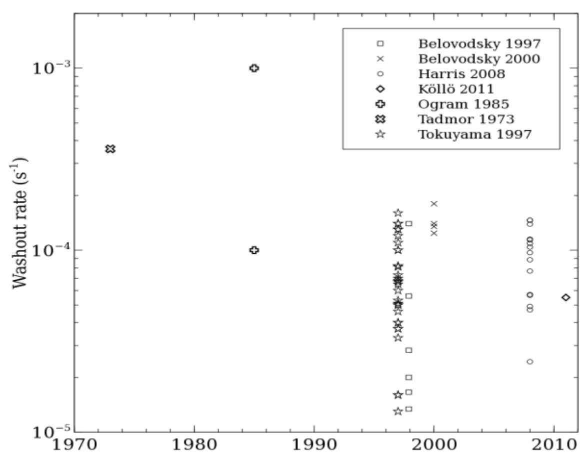


Figura 1. valori experimentale pentru rata de depunere

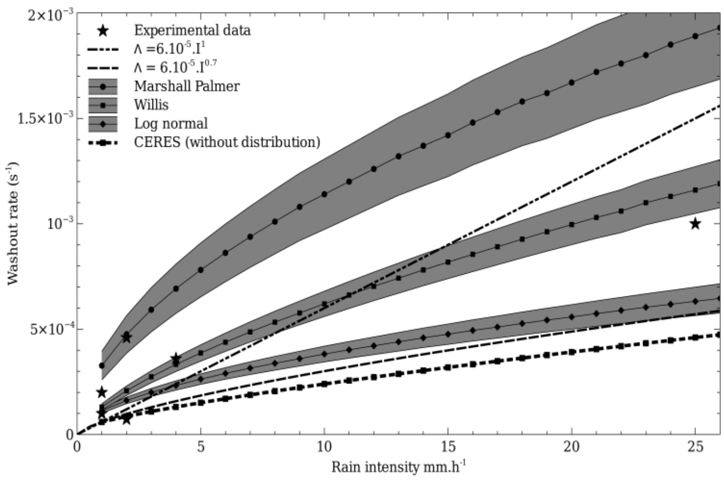


Figura 2. Valori teoretice si experimentale pentru rata de depunere, modelul francez subestimeaza

Principala contributie pe care IFIN-HH o aduce in domeniul evaluarii riscurilor emisiilor de tritiu consta in analiza exhaustiva a rezultatelor experimentale privind producerea tritiului legat organic (OBT-Organically Bound Tritium) in plante agricole. Vegetatia absoarbe HTO din atmosfera sau sol si in procesul de fotosinteza o converteste in forme organice. Tritiul (cat si hidrogenul) poate fi legat de atomi diversi, dar daca este legat de carbon, legaturile sunt mult mai stabile si persistenta in mediul viu este mai mare. OBT este o denumire generica a multor compusi organici in care tritiul este legat de carbon in forme biogene.. Ca urmare, producerea de OBT in plante agricole este una din temele majore de cercetare. Experimentele de laborator au demonstrat ca OBT se produce atat in timpul zilei, cat si in timpul noptii, iar procesele fizice si biochimice sunt complexe si insuficient intelese. Conform unor estimari [Raskob and Barry, 1997], pentru emisii accidentale de tritiu in atmosfera, contributia OBT la doza de ingestie poate fi de 80 %. Din acest motiv, clarificarea mecanismelor de producere si analiza rezultatelor experimentala este esentiala pentru a asigura robustetea modelului radiologic. La solicitarea IFIN-HH, mai multi cercetatori ne-au pus la dispozitie date experimentale nepublicate, cat si detalii importante, astfel incat documentul elaborat de noi reprezinta prima analiza exhaustiva a rezultatelor experimentale din mai multe tari: Germania, Canada, Japonia, Corea de Sud, Franta si cuprinde experimente pe mai multe plante de cultura, la diferite stadii de dezvolatre, ziua si noaptea. Revizia noastra cuprinde informatii experimentale despre grau (soiuri germane de primavara si iarna), orez (soiuri japoneze si coreene), fasole si soia (soiuri germane, japoneze si coreene), cartof, rosii, salata, varza, ridichi si mandarine. Experimentele prezentate sunt de mare complexitate, desfasurate in anii 1994-2010, si datele au fost analizate sub doua aspecte: a) transferul apei tritiate din atmosfera in frunze in functie de tipul plantei si conditiile de mediu; b) formarea OBT in frunze si transferul in partea comestibila a plantei.

Se pune in evidenta importanta factorilor meteo cu variatie diurna in transferul aer-planta care este mediat de stomate si influentat de lumina, temperatura, umiditatea aerului. Considerand timpul de injumatatire (legat de rata de schimb), datele pentru graul de iarna demonstreaza ca transferul este maxim in timpul zilei (pranz) si mult mai lent seara si noaptea (vezi Tabelul 1).

Tabelul 1. Timpul de injumatatire pentru apa tritiata din planta (min)

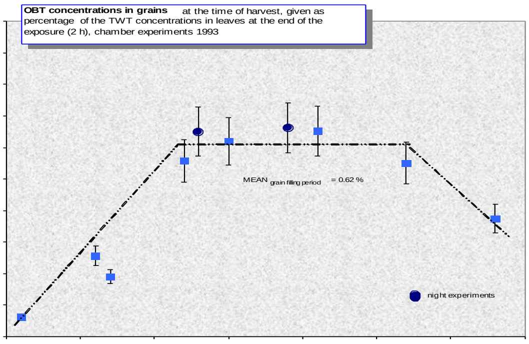
Plant parts	Exposure at dawn (3 exp.)	Exposure at day time (6 exp.)	Exposure at dusk (2 exp.)	Exposure at night (2 exp.)
Leaves	40-60	25-49	230-660	110-170
Stems	45-49	20-26	130-320	60-190
Ears	79-91	50-126	210-330	150-920
Total plant	50-72	27-60	220-340	100-250

Considerand ansamblul datelor experimentale privind transferul HTO din aer in frunze, cateva observatii sunt de mare interes practic:

- in conditii de mediu similare, rezistenta stomatala poate varia cu pana la un ordin de marime intre diferite plante agricole de interes, si ca urmare si rata de transfer;
- in timpul noptii, rezistenta stomatala este semnificativ mai mare decat valoarea presupusa in trecut (4000 s/m) si variaza mult intre plante (minima la varza - 250 s/m). Rezulta ca stomatele nu sunt complect inchise si avem o contributie semnificativa a transportului cuticular;
- noaptea, transferul aer-frunze este mai intens la orez, urmat de grau, fasole si cartof, iar pentru salata este mic;
- pentru a avea un model robust sunt necesare determinari experimentale ale rezistentei stomatale la plantele de interes local;
- dupa perioada de expunere, scaderea concentratiei de HTO in frunze este rapida in timpul zilei si mai lenta noaptea. Dupa 24 de ore, nivelul apei tritiate scade cu doua ordine de marime.

Conversia tritiului in materie organica prin procese biologice in plante poate fi evaluata sintetic prin „Indexul de Translocatie (translocation index TLI)" care este definit ca raportul intre concentratia de

OBT la recolta si cea din apa din frunza (HTO) la sfirsitul expunerii. Precizam ca aici intervine ca unitate de masura concentratia de OBT in apa de combustie si nu per kg de materie uscata. Experimentele de laborator cu grau de primavara au demonstrat dependenta TLI de stadiul dezvoltarii plantei (Figura 3) cu valori maxime si aproape constante in perioda de umplere liniara a bobului. Ulterior s-au efectuat experimente de camp mai elaborate, in perioada de umplere liniara a bobului, dar la diferite ore ale zilei, prin expunere pe o durata de o ora si cu purjare rapida. Precizam ca datele eperimentale complete ne-au fost puse la dispozitie pentru revizia noastra, fiind nepublicate de echipa din Germania. In prezentul raport ne concentram pe analiza acestor date, de importanta pentru Romania si acest proiect. In Figura 4 reproducem valorile TLI pentru aceste experimnente. Se remarca domeniul restrans al valorilor, slaba dependenta de ora experimentului, valori comparabile pentru zi si noapte.



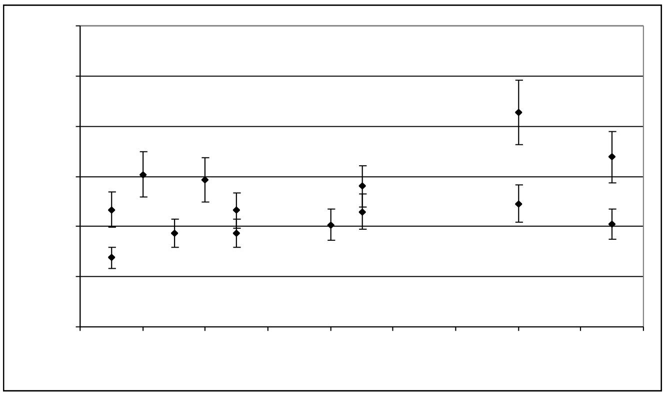


Figura 3. Dependenta TLI de stadiul de dezvoltare

Figura 4. Dependenta diurnala a TLI

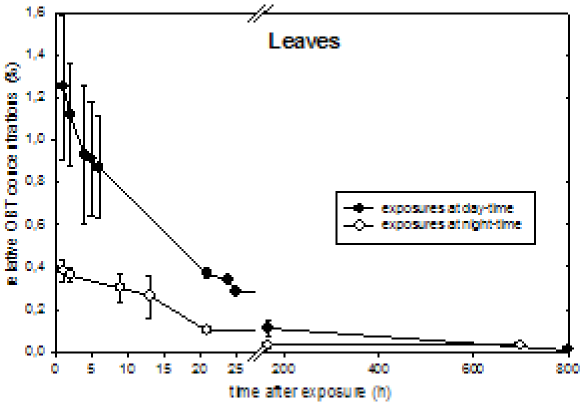


Figura 5. Dinamica concentratiei relative de OBT in frunza de griu, ziua si noaptea

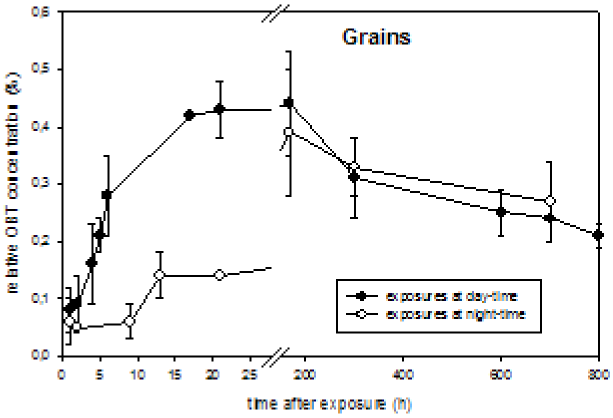


Figura 6. Dinamica concentratiei relative de OBT in bobul de griu, ziua si noaptea

Dinamica concenttratiilor de OBT in frunze si bobul de griu (Figurile 5 si 6) demonstreaza diferente intre zii si noapte. In figura reprezentam concentratiile de OBT normalizate la nivelul concentratiei de HTO in frunza,la sfirsitul expunerii. In timpul nopti avem nivele mai reduse de OBT in frunze si o scadere mai lenta. In bobul de griuOBT creste rapid in tiimpul zilei dar mai lent si cu intirziere in timpul noptii. De remarcat ca la recolta, nivelul de OBT in griu este similar pentru zi si noapte.

Analiza rezultatelor experimentale pentru orez, efectuate in diferite conditii in Japonia si Korea, arata comportamente similare cu graul, dar cu nivele TLI mai mari cu 30 % pentru unele soiuri. Rezultatele experimentale arata ca procesul de transfer frunza-bob ests rapid, o zi, iar nivelul de OBT in intreaga planta este maxim la 1-3 zile, scade apoi datorita respiratiei si dupa 2 saptamani ramane practic constant.

In experimentele cu soia (Japonia si Corea de Sud), nivelul de OBT in boabe variaza in funcrtie de stadiul dezvoltarii plantelor, apar cazuri in timpul noptii, cand valorile TLI sunt putin mai mari ca ziua ,dar comportarea generala este similara cu cerealele, cu diferenta unor valori TLI mai mari. Pentru fasole, putine data au fost obtinute, dar remarcam valoarea TLI sensibil mai mare noaptea decat ziua.

Ca o concluzie generala pentru toate culturile studiate, formarea OBT si transferul in partea comestibila depinde de stadiul dezvoltarii plantei, indicele de translocare TLI variaza intre 0.1 si 1.5 % si s-au

Datele experimentale analizate evidentiaza ca procesul formarii OBT nopatea nu poate fi ignorat in modelarea accidentului de tritiu. Pentru aceeasi emisie accidentala, in timpul noptii, concentratiile de HTO in aer, la receptor, sunt mult mai mari decat ziua, de pana la 40 de ori. Transferul aer-frunze este de numai 3-10 ori mai putin intens noaptea, iar valorile experimentale ale TLI sunt comprabile cu cele in timpul zilei.

Ca urmare, pentru un accident in tipul noptii, implicatiile radiologice sunt comparabile cu accidentul in timpul zilei. Modelaraea robusta a transferului si conversiei tritiului in timpul noptii in materia organica a culturilor agricole este principala problema inca nerezolvata spre care ne vom indrepta atentia in anul 2012.

Contributiile IFIN-HH la documentul tehnic al AIEA sunt exhaustive, cu multe elemente de noutate si merita a fi diseminate in literatura mai repede decat durata publicarii documentului AIEA. Cu acordul AIEA si al colaboratorilor, o versiune mai concisa a reviziei noastre asupta rezultatelor experimentale privind OBT in culturi agricole va fi publicata in curand, ca suport al efortului de modelare.

Director proiect,

Dr. Dan Galeriu

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Scientific report-extended summary

October – December 2011

Interdisciplinary approach for dynamic modelling of tritium transfer in crops PN-II-ID-PCE-2011-3-0396

Some nuclear units have large tritium loads and the subsequent environmental impact. During the normal operation, the public dose is very low and the present monitoring and modelling techniques are sufficient. The technological incidents or unplanned accidents can produce large release of tritiated water (HTO) in a short time. Tritium has a complex environmental behaviour once released into the environment. The peculiarities regarding the tritium behaviour must be taken into account for the understanding of its environmental transport and for the estimation of the radiological consequences of the releases into the environment. Among other international organisation, a leader in promoting the robust tritium modelling was the International Atomic Energy Agency (IAEA) by its Environmental Monitoring for Radiation Safety (EMRAS) programme, which coordinated inter-comparison exercises for the predictions of the doses after a release of 10 g of tritium during an hour. [2] The results of the models participated at that inter-comparison exercise showed an extremely large range of predictions and pointed out that the releases occurred during a rain event can have larger consequences than those occurred during a clear day. The largest consequences are for the releases occurred during the night time, up to the values which impose countermeasures. These large uncertainties are not acceptable for the licensing purposes and the accident preparedness at the nuclear facilities with large tritium loads. Consequently, IAEA decided to coordinate a dedicated working group in order to improve the models and to reduce the uncertainty. Certain key objectives were proposed: (1) Developing a standard conceptual dynamic model for tritium dose assessment for acute releases to the atmosphere and water bodies, starting with the external input for tritium dynamics in atmosphere or water from the source to receptor; (2) Agreement on the common sub-models for the specific transfers or processes, based on an interdisciplinary approach involving the understanding of the processes and key parameters, based on the recent research in Life Sciences. Quality assurance requires a moderate conservatism; (3) Defining the framework for an operational model (requirements for meteorological data, atmospheric transport, site specific data); (4) Achieving the capability to assimilate real measured data in the models.

The needs to identify the main contributors to the uncertainty and the critical periods during the year in relation to the resulting exposures to tritium were emphasised, as well as the identification of the most important and sensitive parameters (*i.e.* hourly, daily and annual variations of the parameters/processes) and the exploration of the practical possibilities for determining those parameters. It was also emphasised the need for getting an idea about the achievable reliability of the tritium modelling for practical, accidental field conditions. Due to the complexity of the problems, the goals were partially achieved and included in a report.

The work group technical document (TECDOC) covers some key sub-topics with contributions from the group members as follows:

1. Introduction (P. Cortes, <u>D. Galeriu</u>, V. Berkovskyy)
2. Key mechanisms for tritium transfer in terrestrial environment (P. Guetat)
3. Interaction matrices and associated processes for terrestrial pathways of tritium transfer (S. Le Dizes-Maurel)
4. Tritium atmospheric washout (L. Patryl, <u>D. Galeriu</u>, <u>A. Melintescu</u>)
5. HT and HTO dry deposition and reemission (M. Ota, H. Nagai)

6. HTO uptake in plants and the OBT formation during the day time (A. Melintescu, D. Galeriu)
7. Overview experiments on tritium transfer from air to plants and the subsequent conversion to OBT (D. Galeriu, A. Melintescu, S. Strack, S.B. Kim, M. Andoh-Atarashi)
8. Review on soil-plant tritium transfer (V. Korolevych)
9. Tritium transfer in wheat experiments and models tests (D. Galeriu, L. Patryl, S. Strack, A. Melintescu, M. Ota)
10. Tritium transfer in farm animals (D. Galeriu, A. Melintescu)
11. Briefing of complex model (H. Nagai, M. Ota)
12. Tritium in aquatic foodchain (A. Melintescu, D. Galeriu, F. Siclet, F. Lamego)
13. Quality assurance of data (S.B. Kim)
14. Quality assurance of model (D. Galeriu, J. Duran)
15. Status and perspectives of accidental tritium modelling (D. Galeriu)

An important contribution of IFIN-HH regards the tritium washout parameters which have large uncertainties and the washout process is extensively analysed, together with the current models and key parameters. The washout rate varies between 1.3×10^{-5} – 3.6×10^{-4} s^{-1}, with a mean value of $(9.2\pm5.8) \times 10^{-5}$ s^{-1} for 54 data points. Much of the variation can be attributed to the different tritium release characteristics and meteorological conditions. The washout rate is closely affected by the precipitation characteristics, such as rain intensity, drop size distribution and drop characteristics. At short distances, the washout rate can also be affected by the building wake effects.

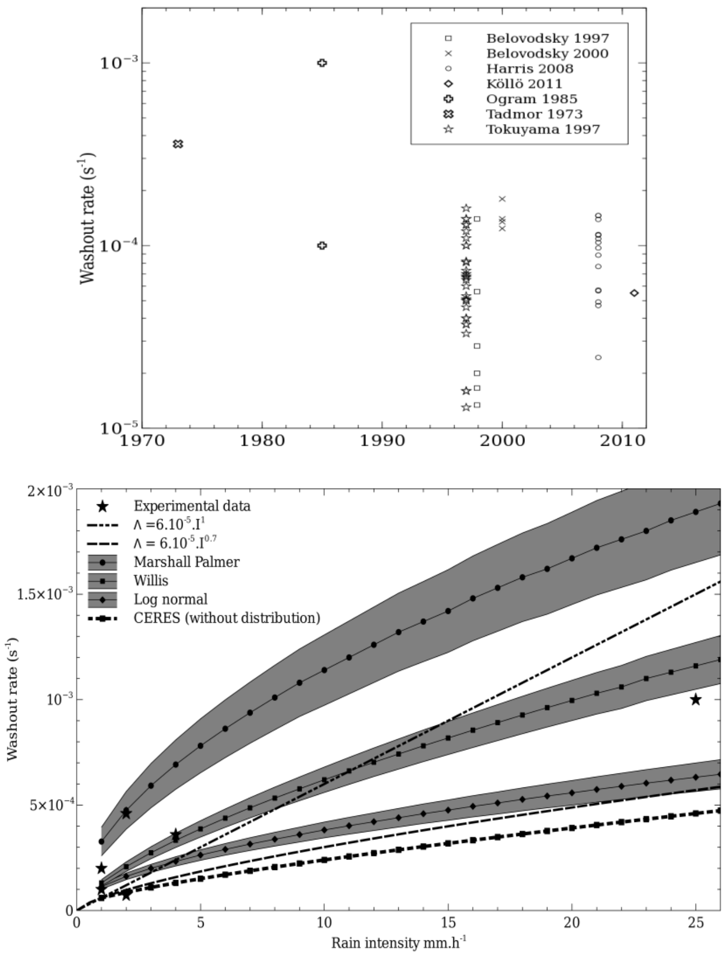

Fig 1. Experimental values for deposition rate

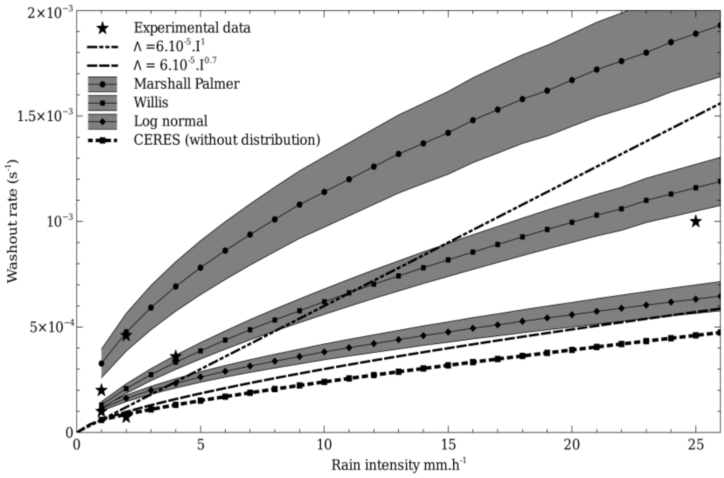

Fig. 2. Theoretical and experimental values for deposition rate**,** the French under-estimates

The HTO uptake by leaves and the subsequent conversion to OBT was considered under the light conditions, because the processes regarding the OBT formation during the night time are more or less understood. An update of the knowledge and modelling approaches was presented for the case when the atmospheric HTO only is the source. The role of the exchange velocity and canopy conductance was emphasized. Different approaches for the stomatal resistance were reviewed. The water and carbon dioxide, CO_2, move by diffusion in opposite directions between stomata and air. The water evaporates from the cell walls, and moves from the stomata to the air, whereas CO_2 moves from the air, via the stomata into the mesophyll, where it is reduced to sugars by the chemical reactions in the Calvin cycle. This behaviour gives a clear dependence of the stomatal resistance on photosynthesis. A simple functional model for OBT production during the day was described. Up to present, any attempt to analyse the available models from the point of view of their transparency, user friendliness and robustness of the predictions was not achieved. Much work is still needed to cover all the situations which may appear in an operational application.

The OBT formation in night condition is well known, but any model is not able to explain its formation. As a preliminary step, a revision of all the experimental data (for day and night) were carried out covering wheat, rice, soy bean, potato, tomato, cabbage, radish and tangerine. The ratio between OBT combustion water at harvest and HTO concentration in leaves at the end of exposure (*i.e* the translocation index (TLI)) was used in order to compare different crops.

In the day light, the sunflower, grape, rice and wheat show higher uptake rates than the komatsuna and cabbage, while the tomato and orange have the lowest uptake rate. In night conditions, even if the stomata are completely closed, the cuticular conductance occurs and varies among the plant species. The HTO uptake during the night time can be relatively high. The orange and tomato have very low loss rates in night, while the rice and wheat have high loss rates. The translocation index depends on the crop, exposure time and environmental conditions. For wheat in the period of the linear grain filling, the translocation index depends weekly on the time of exposure (Fig. 3) and the night values are comparable with those in the day, this is not valid for the other periods of the crop growth.

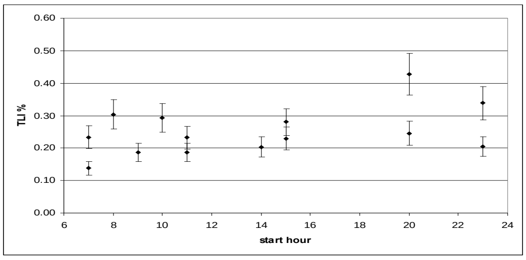

Fig 3. Translocation index for wheat at different times during a day

The environmental tritium models should be modified to reflect the knowledge acquired in the recent experimental work in the areas previously discussed.

Further work along the lines previously suggested will result in better understanding of the tritium behaviour and concentrations in the environment, and an improved ability to estimate doses due to exposure to tritium released to air or water including the climate change.