

Research and development of environmental tritium modelling, an update

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Sacramento, CA

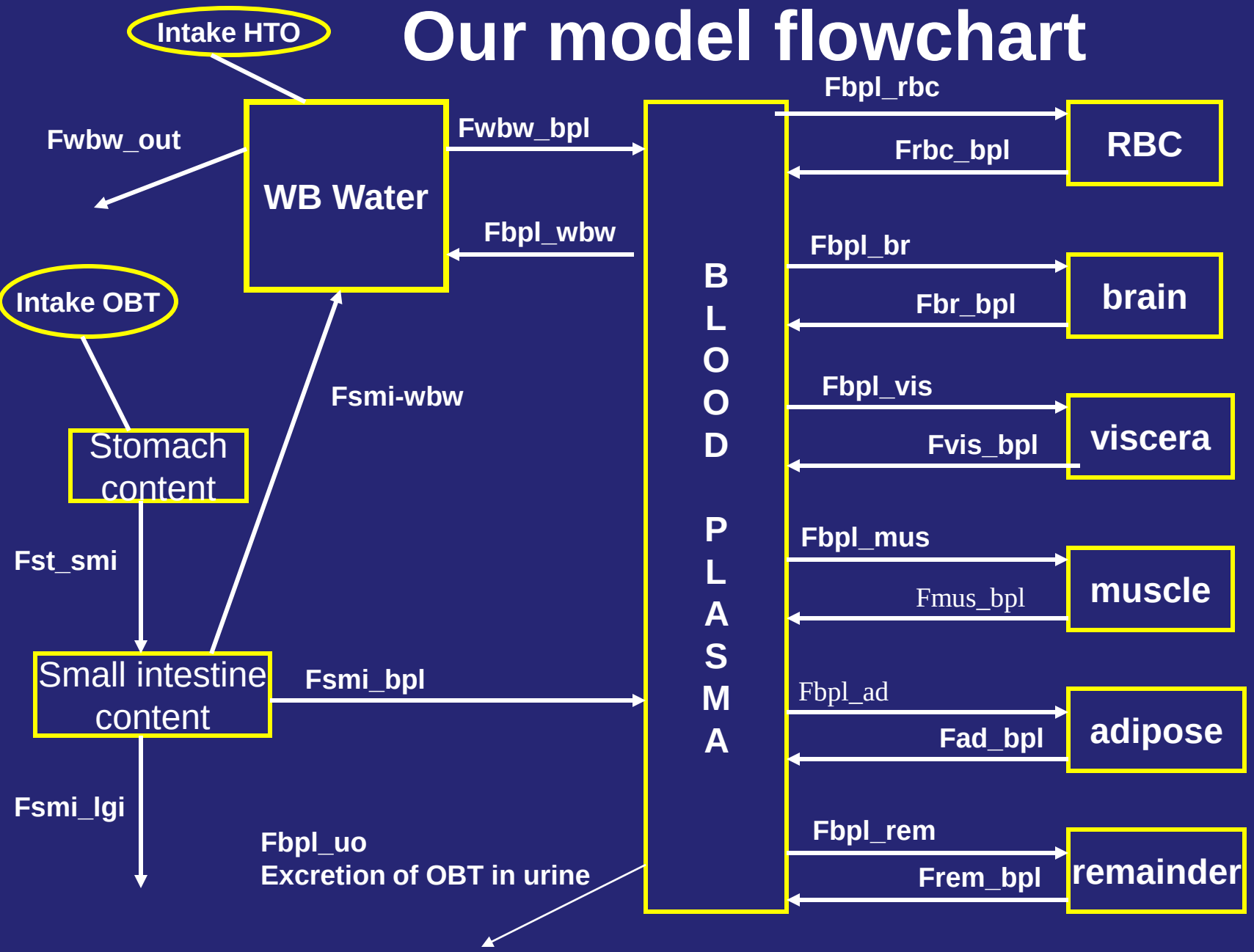
THE INCREASED INTEREST FOR TRITIUM

- Greenpeace actions in Canada, UK, Romania, Japan
- Groundwater tritium near nuclear reactors (USA, Canada)
- The need to preserve public trust in nuclear energy AFTER FUKUSHIMA
- EU Scientific Seminar “Emerging Issues on Tritium and Low Energy Beta Emitters” (Luxemburg, 13 November 2007)
- Canadian Nuclear Safety Commission: Tritium Studies Project (2007-2010) → *Environmental Fate of Tritium in Soil and Vegetation*
- Autorite de Surete Nucleaire (France) → Livre Blanc TRITIUM (2008-2010)
- International Atomic Energy Agency- EMRAS program, Phase I and II - Environmental Modelling for Radiation Safety
- EMRAS I – WG 2 - Modelling of Tritium and Carbon-14 transfer to biota and man working group - Final report on web
- EMRAS II WG 7 – "Tritium" Accidents - TECDOC in preparation

Tritium Human Dosimetry a Metabolic Approach

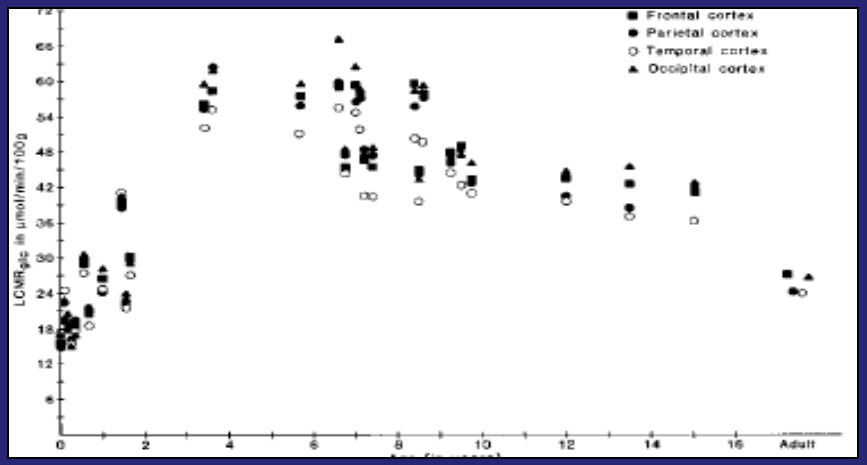
- Actual (ICRP) tritium dosimetry is contested (CERRIE, Greenpeace, IEER);
- AGIR report recommends an RBE=2 (HTO) and leaves the problem of retention open;
- Parameter uncertainty of ICRP model was analyzed before (Harrison 2002);
- A NEW MODEL APPROACH WAS PUBLISHED including the age, gender and race effect;
- Variability, as well as RBE uncertainty;
- RBE for OBT (Chen J., 2006. Radiation quality of tritium. *Radiat. Prot. Dosim.* **122:**546–8)
- Approach based on energy metabolism and probabilistic assessment;
- Galeriu D., Melintescu A., 2010. Retention of tritium in reference persons: a metabolic model. Derivation of parameters and application of the model to the general public and to workers
  J. Radiol. Prot. **30**:445–468

Our model flowchart



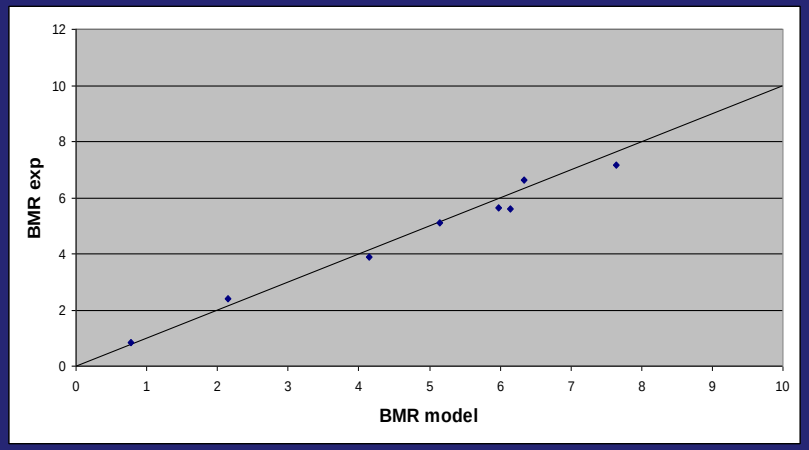
ROLE OF BRAIN

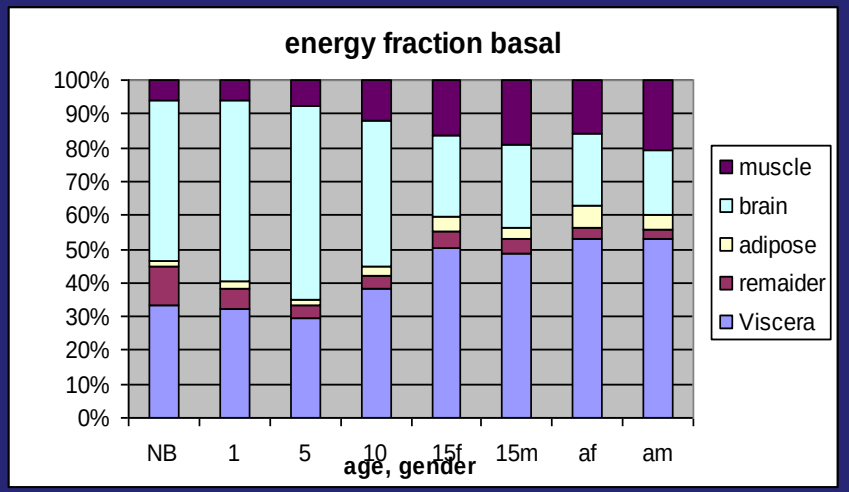
Glucose utilization (metabolic rate) for cortex region at various human ages



- **Various brain regions show a similar pattern;**
- **ADULT AND 1 YEAR OLD CHILD USE A SIMILAR ENERGY FOR BRAIN FUNCTIONING (per kg);**
- **Training for survival uses more energy to establish the base of knowledge and prompt reactions to environment challenge;**
- **At small scale, similar for many mammals**

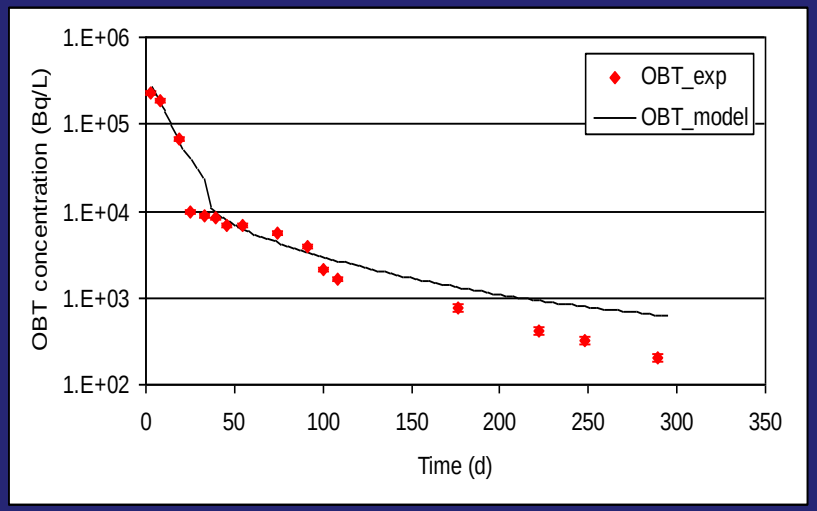
Reconstruction of basal metabolic rate

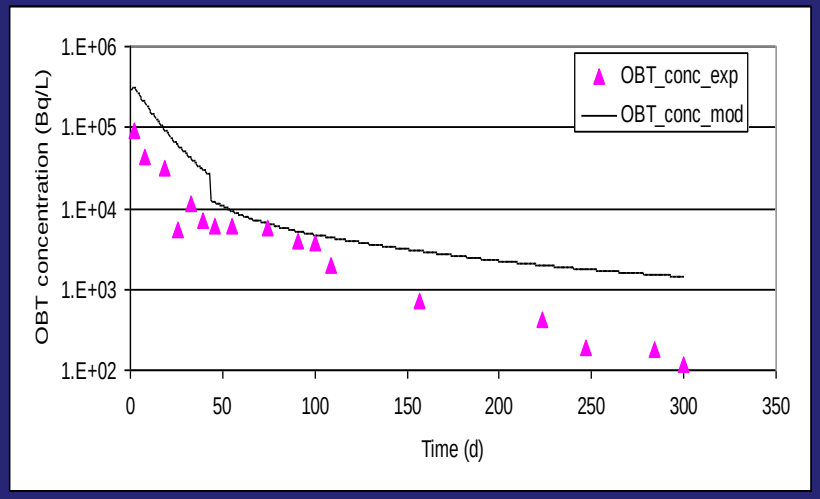




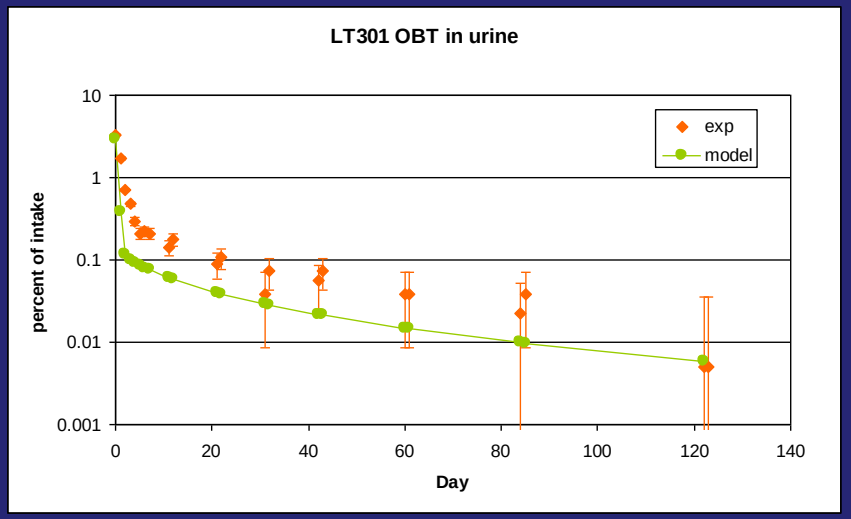
TESTS with human experimental data

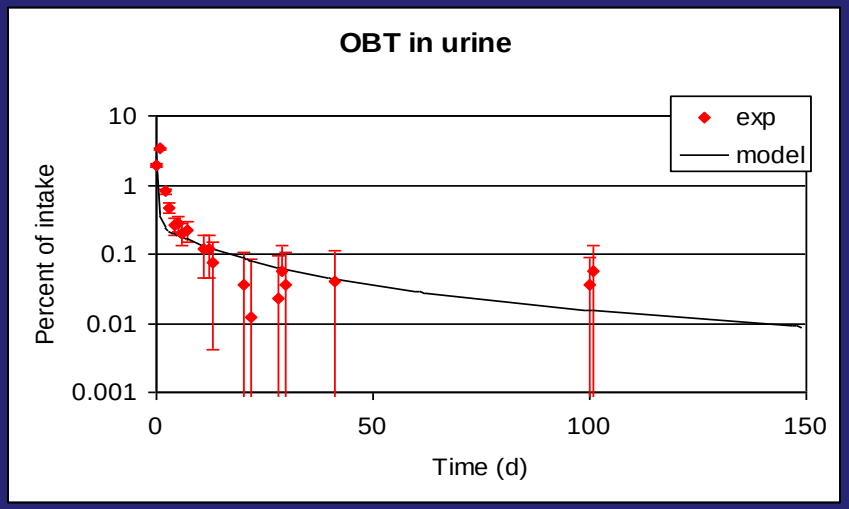
HTO intake





OBT intake





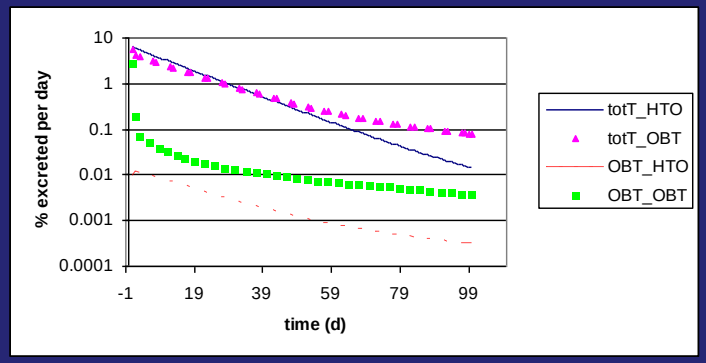
NO other model was tested with OBT experimental data on humans

Predicted doses for a single OBT intake (10^{-11} Sv Bq^{-1}) using different RBE values

age	ICRP	H (uniform, RBE=1)			E (non-uniform, RBE=1)			E (non-uniform, RBE>1)		
		5%	50%	95%	5%	50%	95%	5%	50%	95%
3 months	12	26.4	28.9	31.4	19.4	21.4	23.5	61.6	69.5	77.4
1 year	12	16.6	18.1	19.7	12.4	13.8	15.2	38.7	43.8	48.6
5 years	7.3	9.5	10.5	11.5	7.7	8.6	9.7	23.7	26.8	29.7
10 years	5.7	8	8.7	9.5	5.7	6.2	7.0	17.3	19.5	21.7
15years	4.2	6.4	7	7.6	5.8	5.4	6.2	14.5	17.1	19.2
adult	4.2	6.7	7.2	7.7	4.4	4.9	5.2	13.9	15.6	17.4

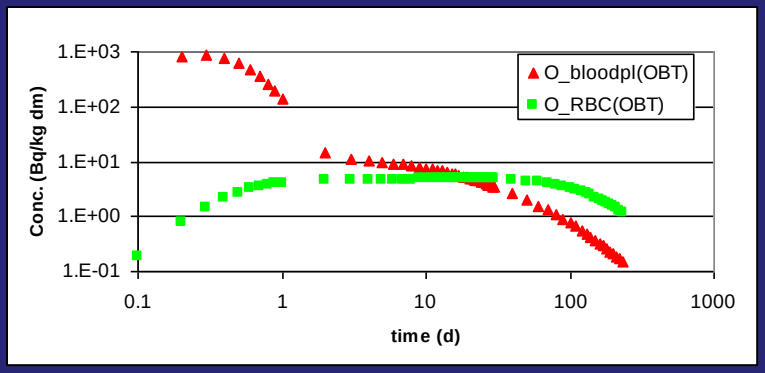
H - uniform distribution, no w_T; E – non uniform distribution to be compared with ICRP
Moderate increase, but infant a factor 2

Total T and OBT in urine after HTO or OBT intake



Use bioassay

Dynamics of OBT in blood plasma and red blood cells OBT after an OBT intake of 1000 Bq



SATISFIES THE RECENT REQUIREMENTS

Transfer of tritium in the environment after accidental releases from nuclear facilities

Report of Working Group 7
Tritium Accidents
of EMRAS II Topical Heading

Approaches for Assessing Emergency Situations
Environmental Modelling for
Radiation Safety (EMRAS II) Programme

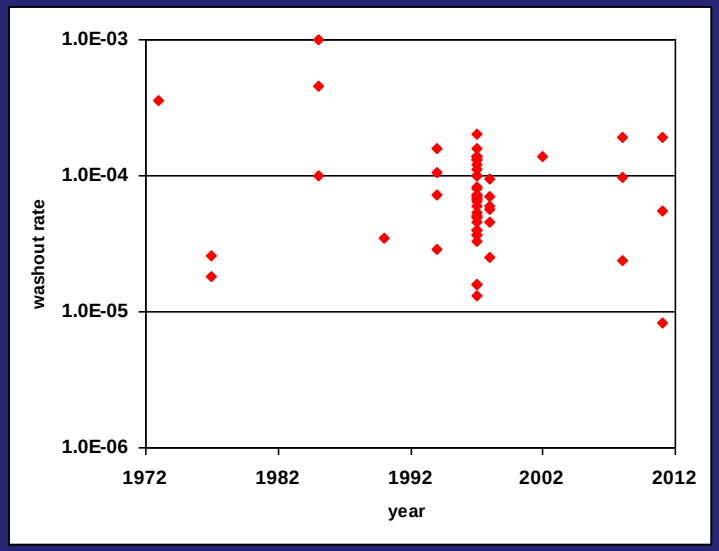
- Developing a standard conceptual dynamic model for tritium dose assessment for acute releases to the atmosphere and water bodies, starting with the external input for tritium dynamics in atmosphere or water from the source to receptor.
- Agreement on common sub-models for the specific transfers or 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.
- Defining the framework for an operational model (requirements for meteorological data, atmospheric transport, site specific data).
- Achieve the capability to assimilate real measured data in the models.

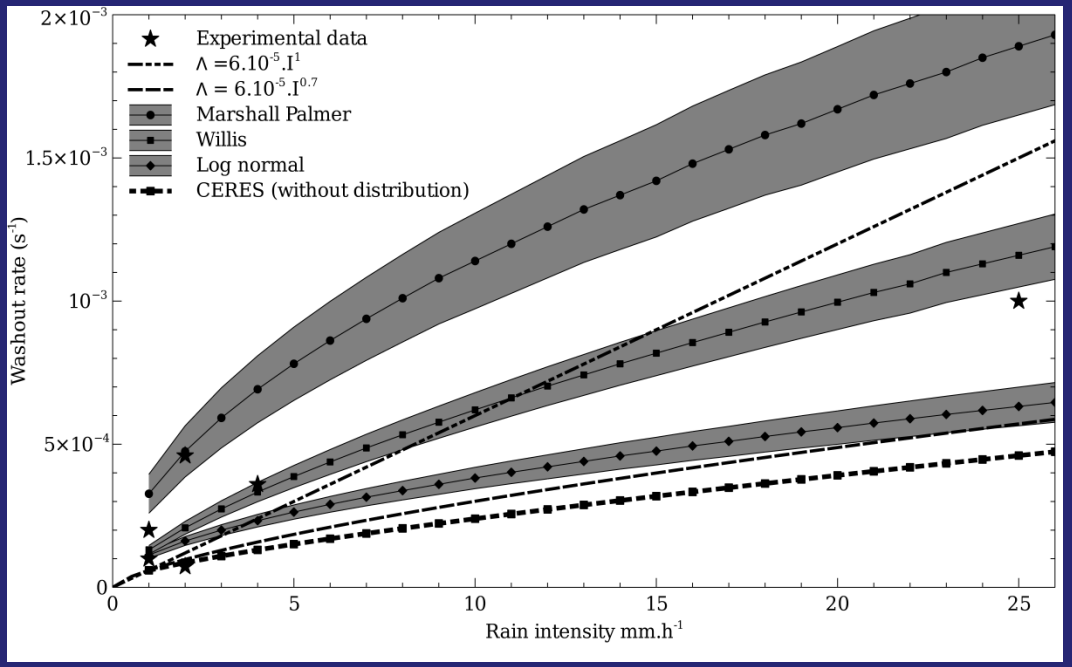
WG leader D. Galeriu, IAEA TECDOC in preparation

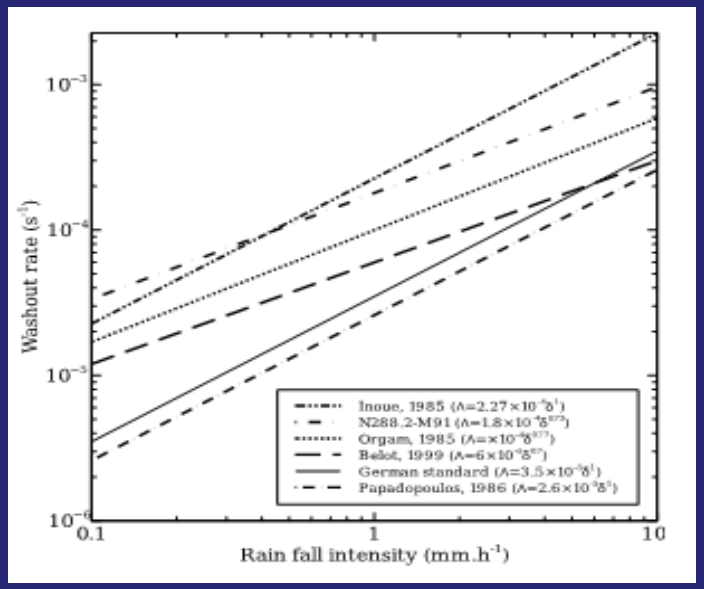
CONTENTS

1. Introduction (P. Cortes, D. Galeriu, 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, D. Galeriu, A. Melintescu)
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)

WG-7 TRITIUM ATMOSPHERIC WASHOUT





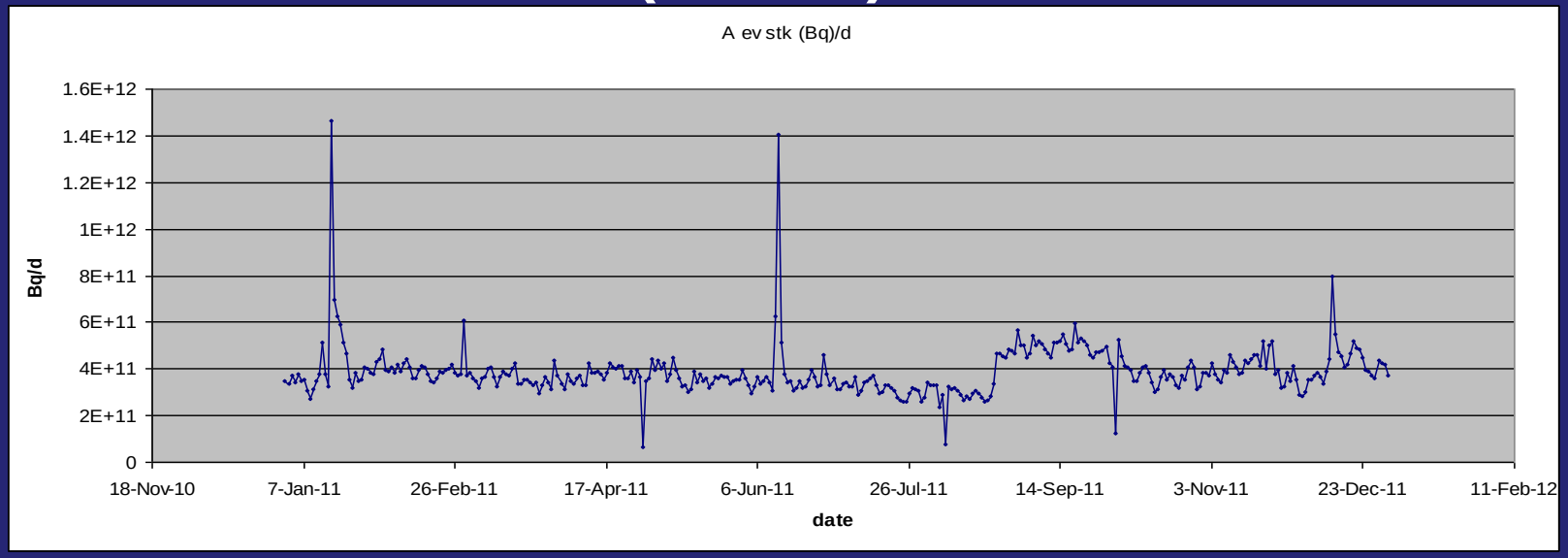


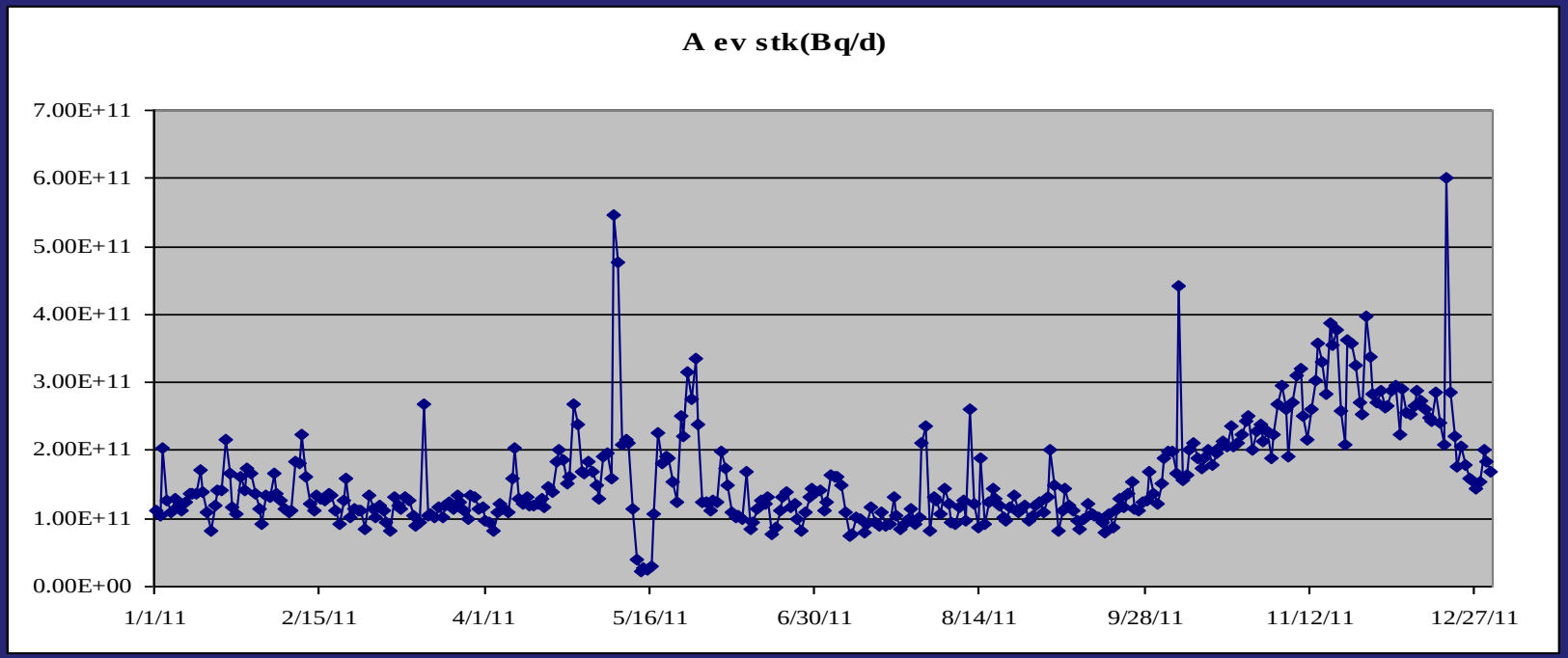
Average and standard deviation washout rate for the Marshall-Palmer, Willis and Log normal distributions computed using Kessler, Seinfield, Andronache, Loosmore and Chamberlain drop velocity equations

On site tritium studies at Cernavoda NPP - 2011
Application of ISCPRIME

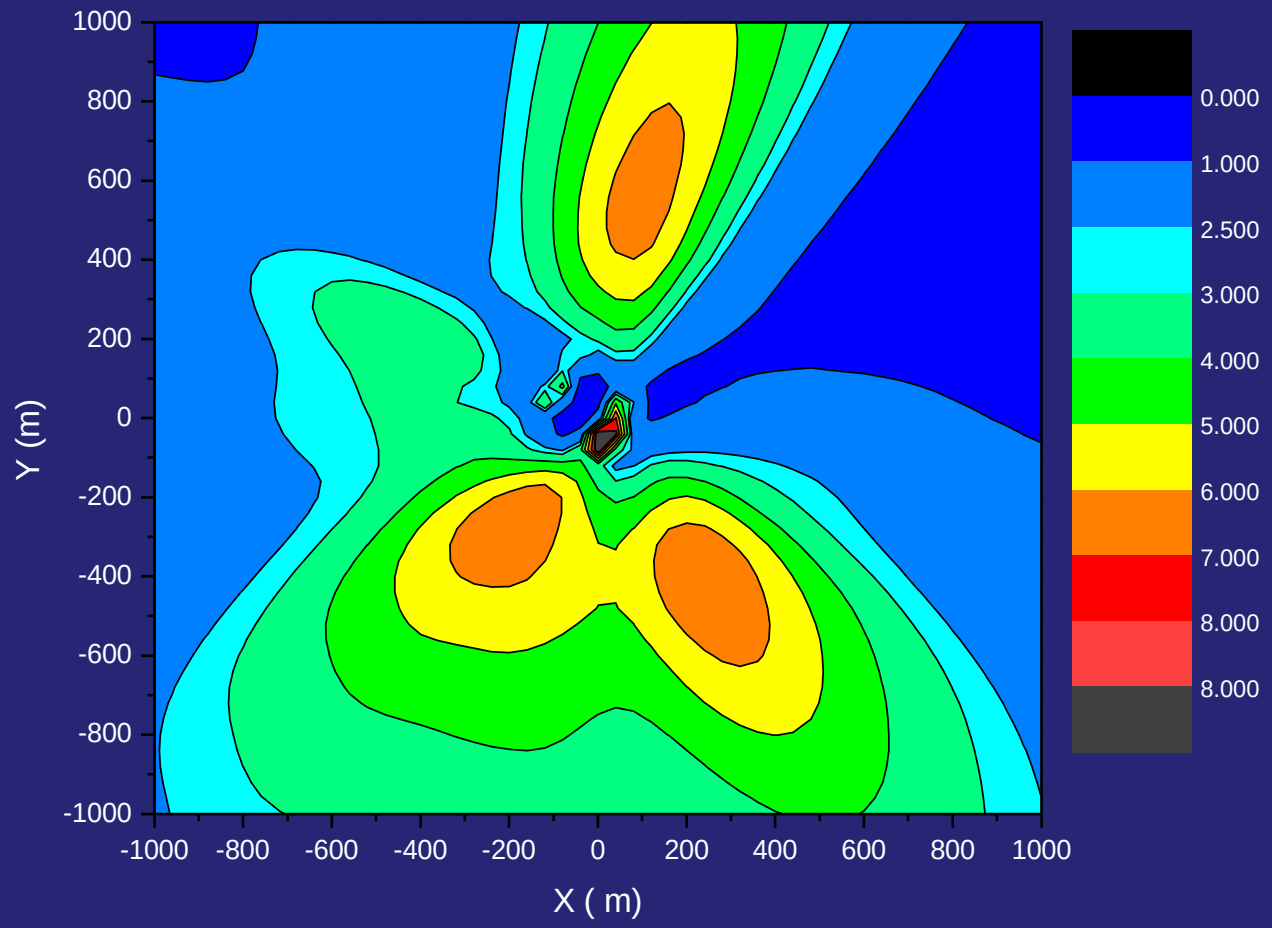


Daily Tritium atmospheric emission U1 (top) and U2 (bottom)

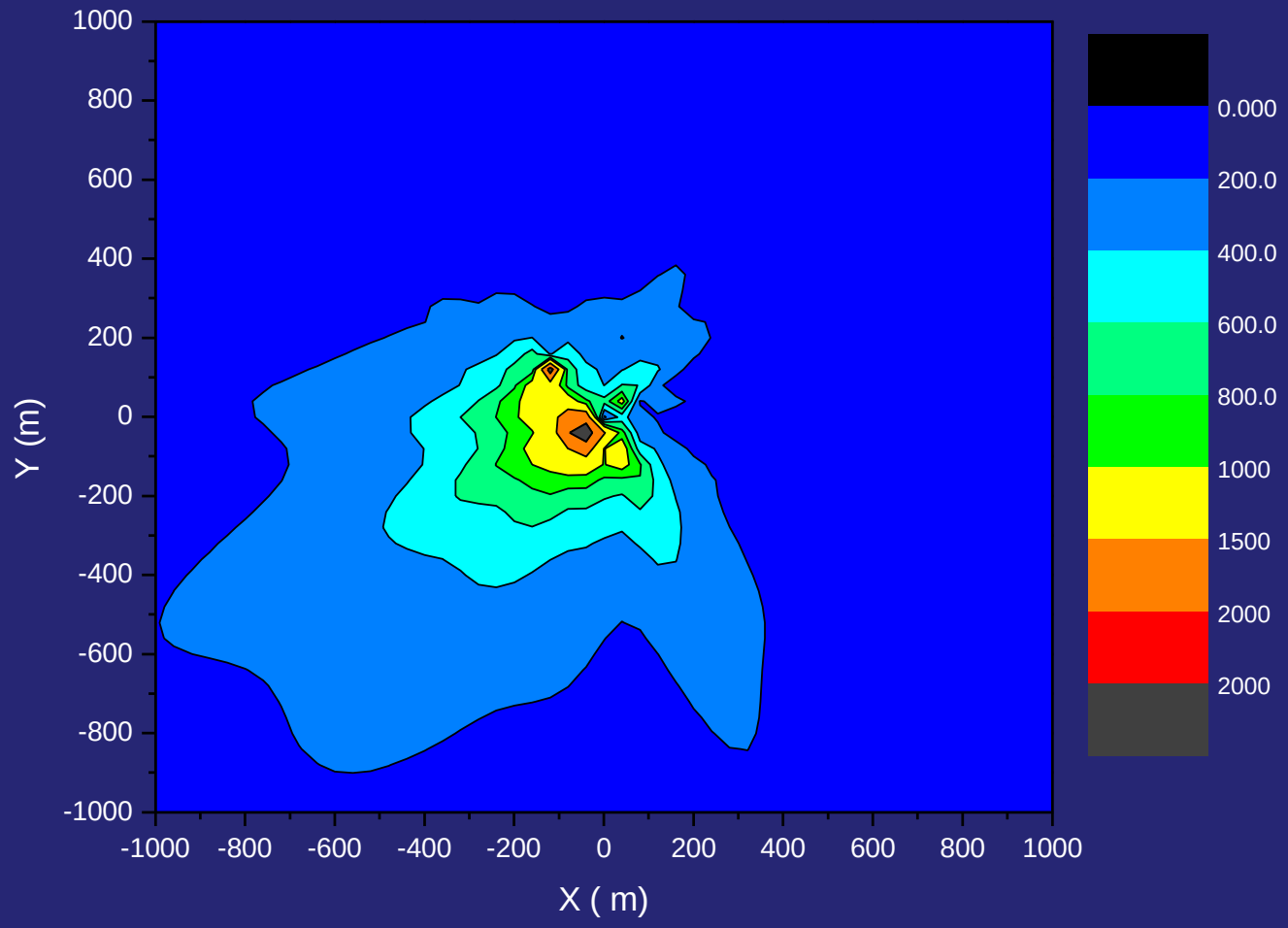




Average tritium concentration in air (Bq/m^3) – 2011 using ISC-PRIME and local meteorology



Average tritium in precipitation (Bq/L) – 2011, using ISC-PRIME and local meteorology



Measurements and Model - air concentration (up), precipitation concentration (down)

station	ADI-02	ADI-04	ADI-05	ADI-06	ADI-08	ADI-09	ADI-10	ADI-11	ADI-12	ADI-13
distance km	11	10	1.6	1.2	2.5	9	12	0.62	0.34	0.62
bearing	80	135	168	220	315	15	215	50	83	315
Measured 2011average	0.24	0.21	0.48	0.25	0.29	0.13	0.06	0.49	3.67	2.04
Model Average	0.09	0.25	2.05	3.05	0.45	0.55	0.21	0.95	0.97	2.35
P/O	0.37	1.18	4.23	12.27	1.57	4.24	3.53	1.92	0.26	1.15

station	ADI-12	SSS-18	ADI-13	ADI-05	ADI-08
distance	0.34	0.43	0.62	1.6	2.5
bearing	75	210	315	170	315
Measured 2011 average	207	172	268	131	25
Model average	84.05	570.31	112.29	195.34	24.80
P/O	0.40	3.32	0.42	1.49	0.98

Follow-up

Monthly values have larger ranges for predicted/observed ratios, explained by:

- PRIME simplicity
- Site complexity
- Quality and number of meteorological instrumentation
- Influence of small hills and Danube valley
- Modelling of tritium washout near buildings
- Need of local rain drop size distribution

Acknowledgements: radiation protection staff at CNE
Peter Eckhoff, U.S. Environmental Protection Agency

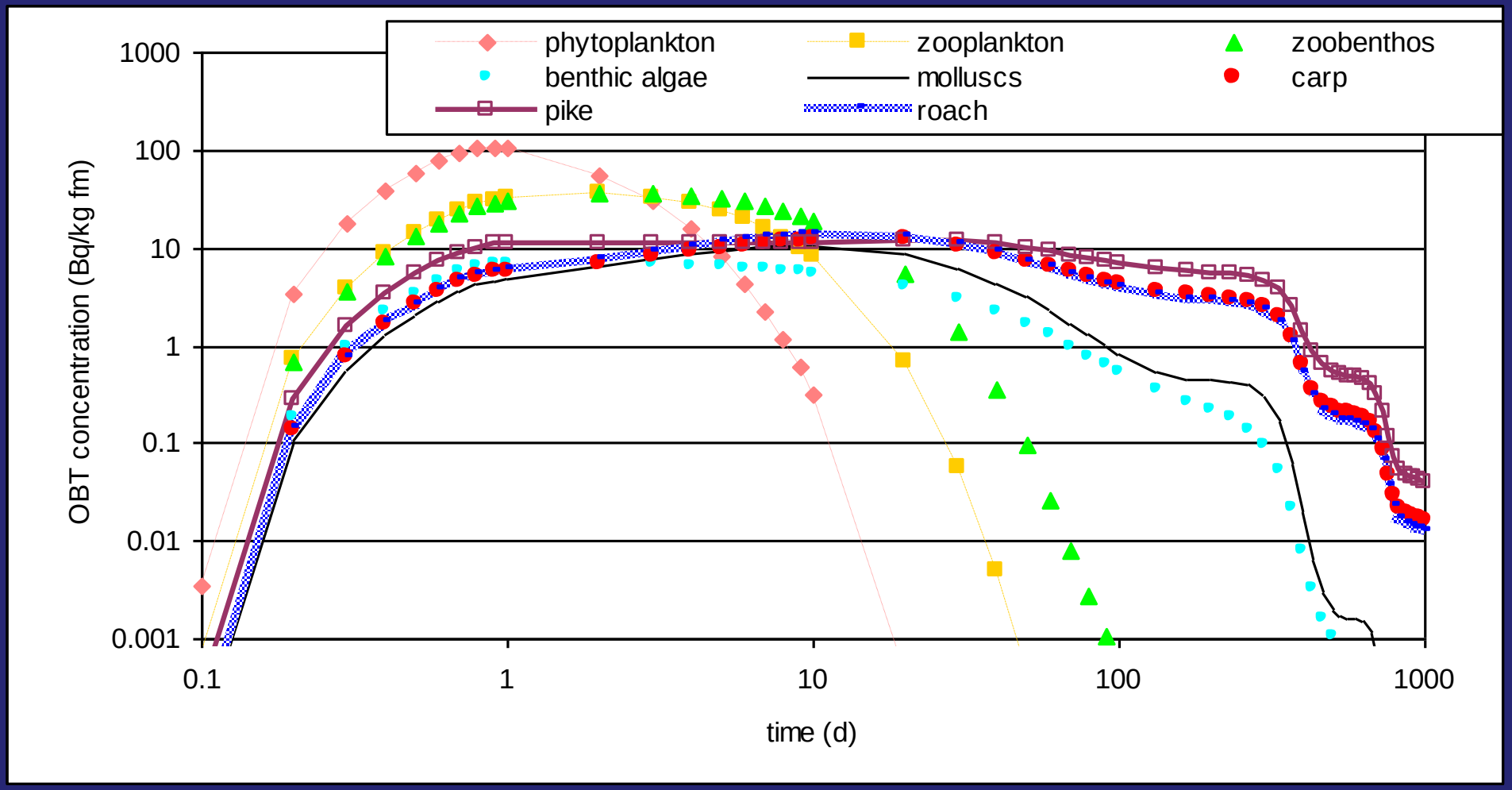
Updated AQUATRIT – AQUAtic TRITium Model

- Dynamic model for predicting ^{3}H transfer in aquatic food chain;
- More coherent assessments for the aquatic food chain, including the benthic flora and fauna;
- Explicit application for the Danube ecosystem;
- Model extension to the specific case of dissolved organic tritium (DOT)
- Model tests with data for large trout (S.B. Kim, AECL)
- Full details given in:

A. Melintescu, D. Galeriu, 2011. Dynamic model for tritium transfer in an aquatic foodchain. *Radiat. Environ. Biophys.* **50**:459–473

A. Melintescu, D. Galeriu, S.B. Kim, 2011. Tritium dynamics in large fish – a model test. *Radioprotection*, **46**(6):S431-S436

The dynamics of OBT concentration in different aquatic organisms, considering a tritium release in Danube of 3.7 PBq on August 1 and a river flow of 6000 m^3s^{-1}



DOT- The Cardiff case

- Cardiff case – it should be noted that the tritated waste from GE Healthcare (former Amersham) includes not only the HTO and the by-product, but also the high bio available tritiated organic molecules (*i.e.* hydrocarbons, amino acids, proteins, nucleotides, fatty acids, lipids, and purine / pyrimidines).
- For the model application - the input data: the annual average of total tritium and organic tritium releases from GE Healthcare, tritium concentration in sea water and the monitoring data for mussel and flounder have been taken from literature
- Using the available input data - the model successfully predicts the trend for tritium concentration in mussels and flounders

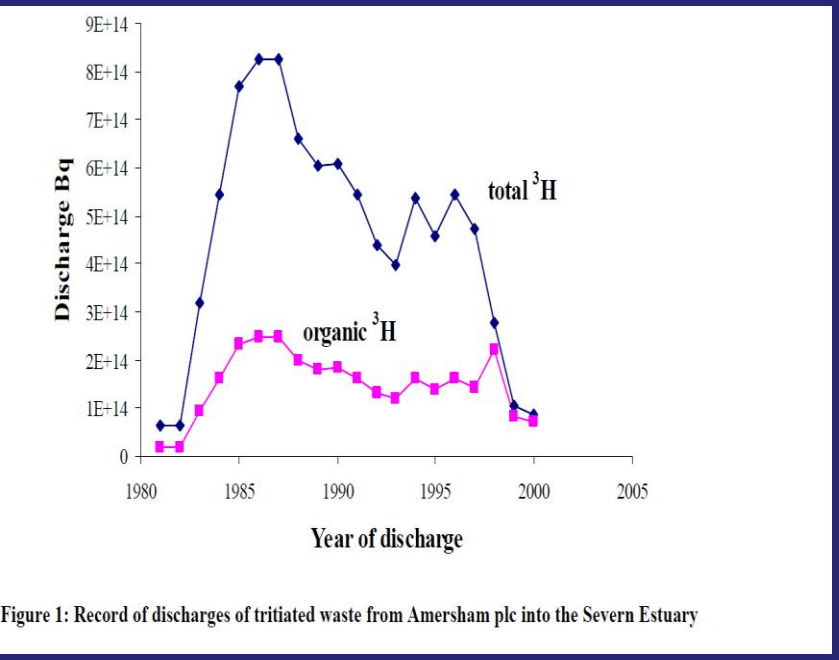
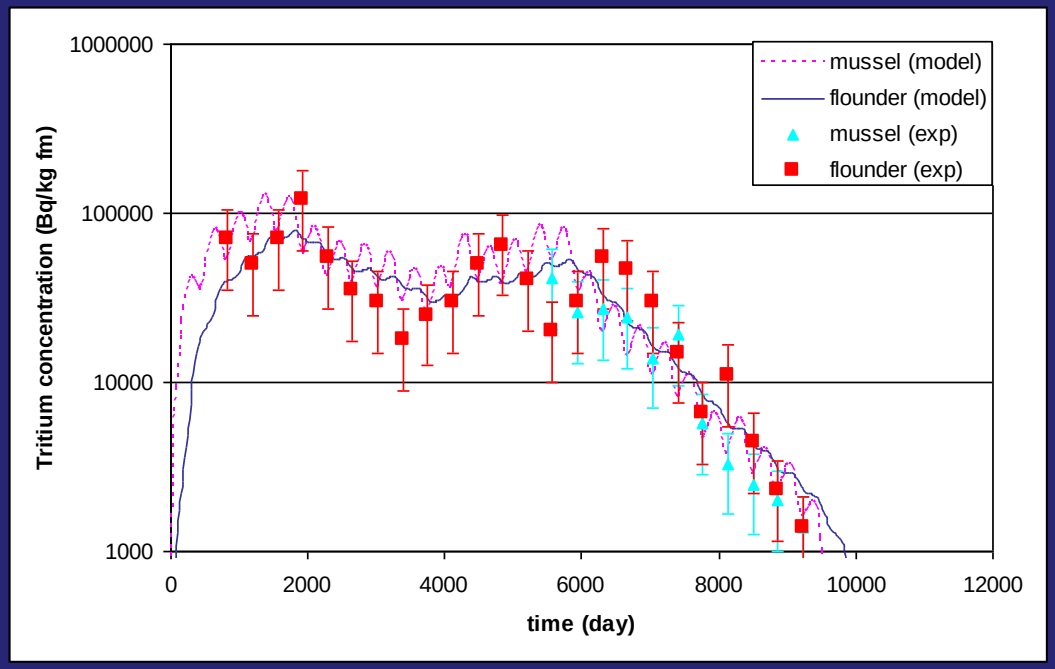
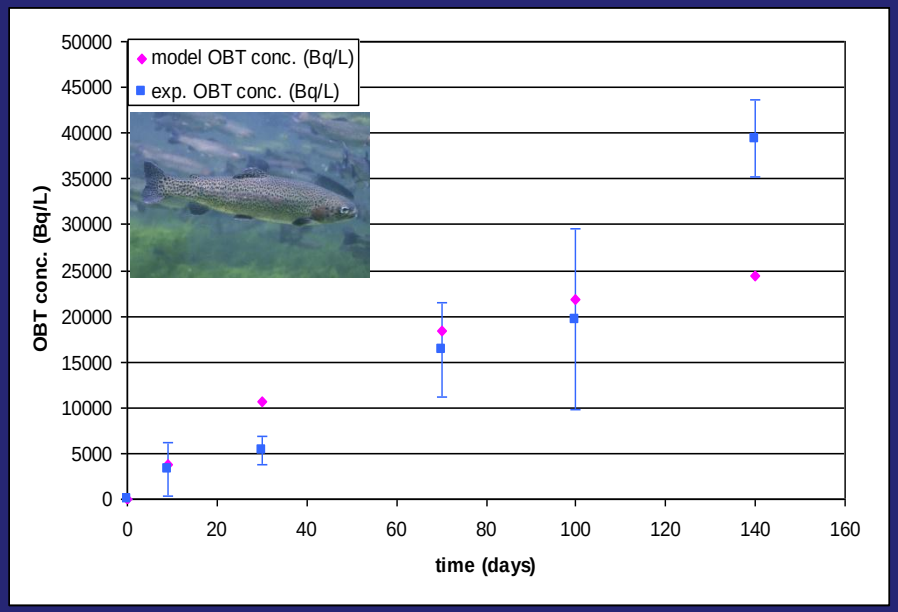


Figure 1: Record of discharges of tritiated waste from Amersham plc into the Severn Estuary

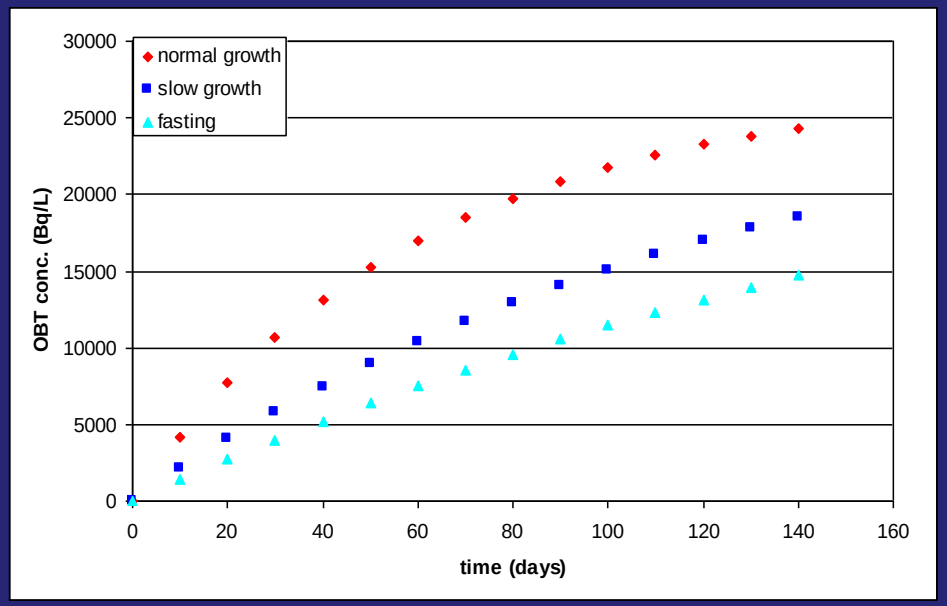


Large trout (S.B. Kim, AECL)

Comparison between model results and exp. data for OBT concentration in fish in the case of OBT uptake



Dependence of OBT concentration dynamics on various feeding regimes for the same final mass



- **Feeding regime - important factor;**

- In the experimental conditions - individual variability in food intake and growth;

Further improvements in the experimental methodology must take into account the fish tagging.

Review of dynamic models for dose assessment of non-human biota – analysis of questionnaire

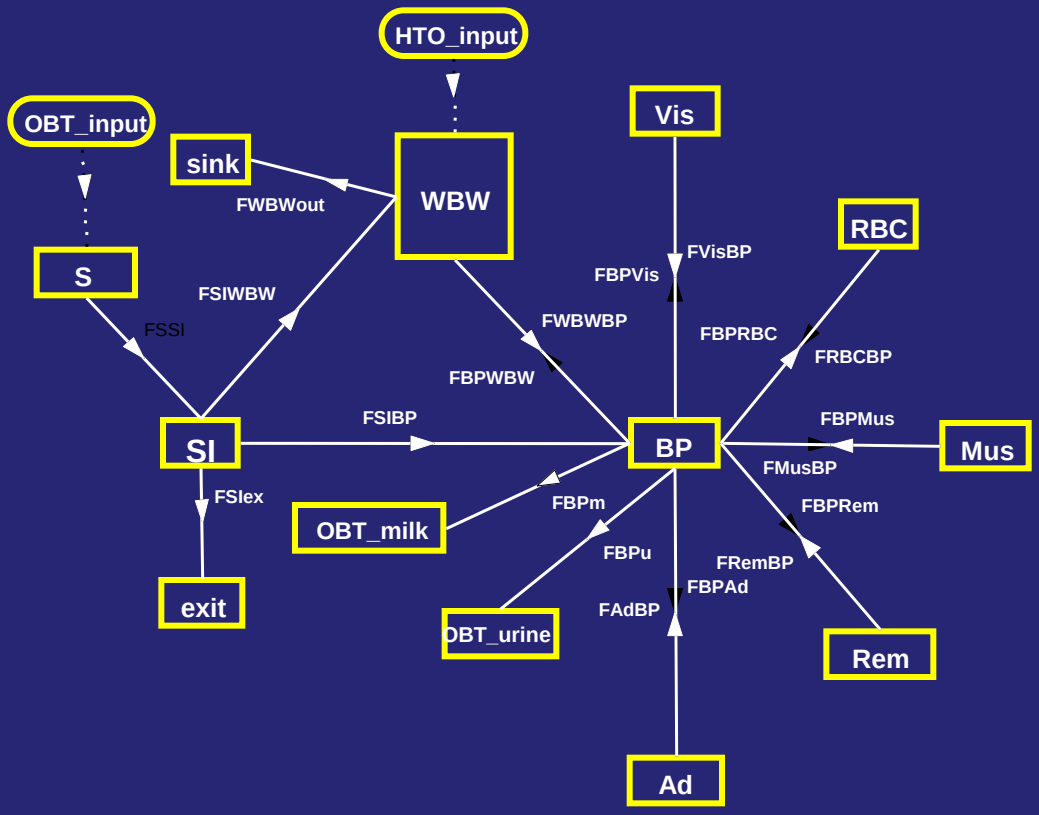
- Interest in recent years regarding dynamic models for protection of non-human biota
- Current assessment models mainly CF-based
- Potential case for dynamic-based models for non-equilibrium scenarios
  - Pulsed discharges
  - Decommissioning situations
  - Accidents
- Modelling group agreed to look at the current state of the art on dynamic modelling:
  - Look at what models are around
  - Assess the need and demand for the dynamic models
  - Release chapter for the final Tecdoc

Identification of fields of interest for the IAEA EMRAS II follow-up programme: Assess impact to humans and biota in an integrated approach

OUR VISION: Complex dynamic model for H-3 and C-14 transfer in mammals using Energy Metabolism (variant of Metabolic Theory in Ecology)

A. Melintescu, D. Galeriu, 2010. *Radiat. Environ. Biophys.* 49:657–672

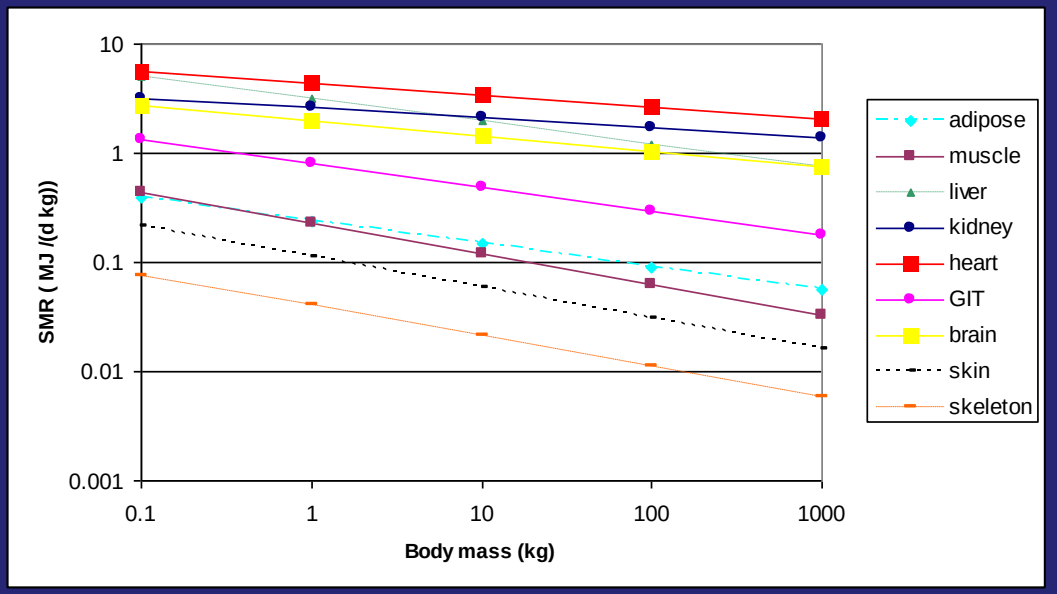
D. Galeriu, A. Melintescu, N.A. Beresford, H. Takeda, N.M.J. Crout, 2009. *Radiat. Environ. Biophys.* 48:29–45

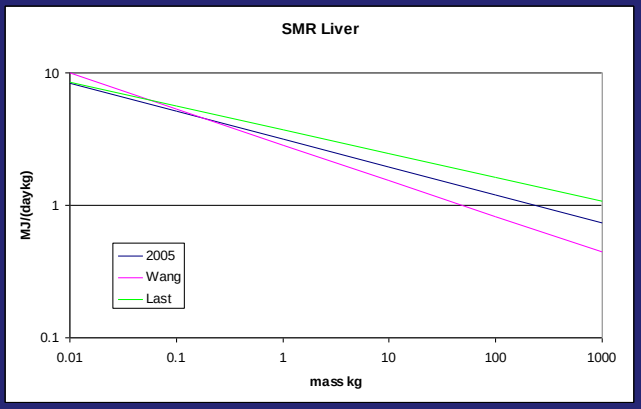


Inputs

Body and organ mass (case specific); organ composition (generic)
Specific organ Metabolic Rates (animal specific?, use generic) - key input
Basal Metabolic Rate (BMR) and Field Metabolic Rate (FMR)
Diet and digestion style; Food and water intake

Known for HUMANS
Good knowledge for
FARM ANIMALS &RAT
SMR for wild by allometry





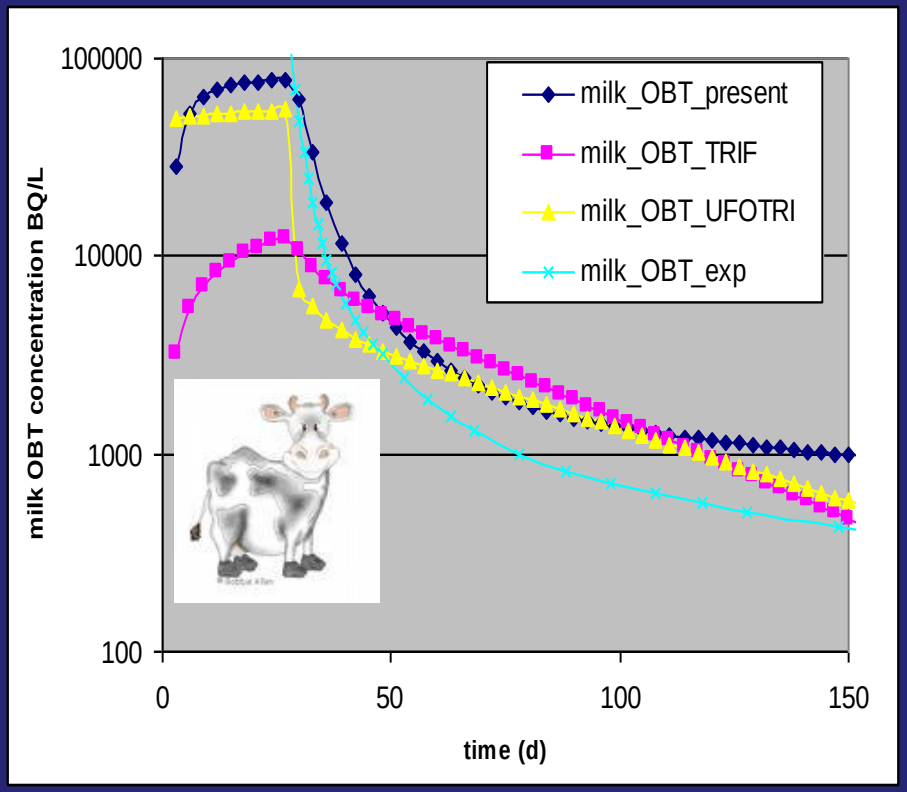
Preliminary allometric relationships for organs specific metabolic rates ,2005

SMR liver, various allometry

Model tests with cow data (no calibration)

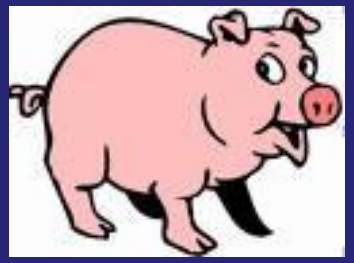
- Several exposures
  - Single HTO intake
  - Continuous HTO intake
  - Continuous OBT intake
- Cow mass, feed and water intake, milk and urine production taken from experiments
- All other model parameters taken from literature – no calibration with tritium data
- **Model gives predictions within a factor 2 of the observed data**

Experimental data and model predictions for OBT in milk after OBT fed for 26 days.
Experimental data were reported only after stop dosing



Tests with growing pigs and veal

Few experiments



1. Pigs of 8 weeks old fed for 28 days with HTO:
   Muscle P/O ~ 1
   Viscera P/O ~1

2. Pigs of 8 weeks old fed for 28 days with milk powder contaminated with OBT:
   Muscle P/O ~ 3
   Viscera P/O ~ 2

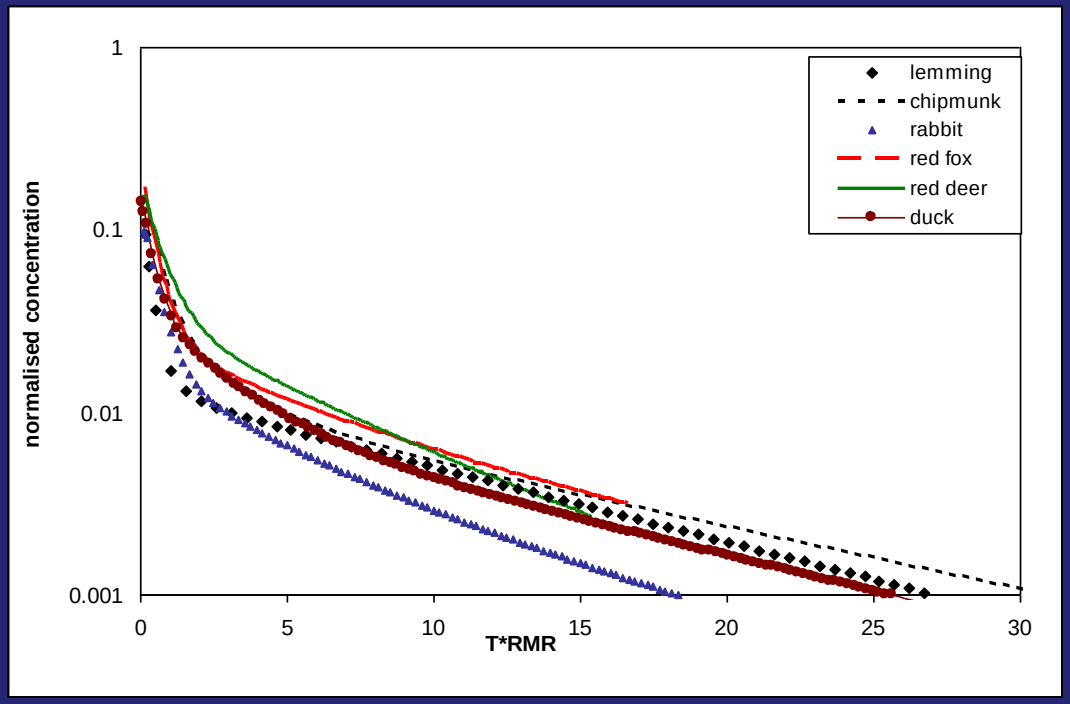
3. Pigs of 8 weeks old fed for 21 days with boiled potatoes contaminated with OBT:
   Muscle P/O ~ 0.2
   Viscera P/O ~ 0.3
   } Not quite sure about these values → Potential explanation: old and insufficiently reported experimental data

4. Two calves of 18 and 40 days old, respectively fed for 28 days with milk powder contaminated with OBT:
   Muscle P/O ~ 1
   Viscera P/O ~ 2.5



Generalized coordinates:
normalized concentration (same per kg whole body)
non dimensional time (use scaling with average metabolic rate)

$$\mathrm{Re}\,MR = 0.073 * BW^{-0.266}$$



On average FMR/RMR~3
For specific case 2-5
Generalized coordinates
are useful to avoid
mass difference
Life span must be considered
Lemming - ReMR=0.1543 d^{-1}.
Lifespan ~2 years
Deer - ReMR 0.0211 d^{-1}
Lifespan 10 years
Reference duck included
Only matures considered
Humans to be added.

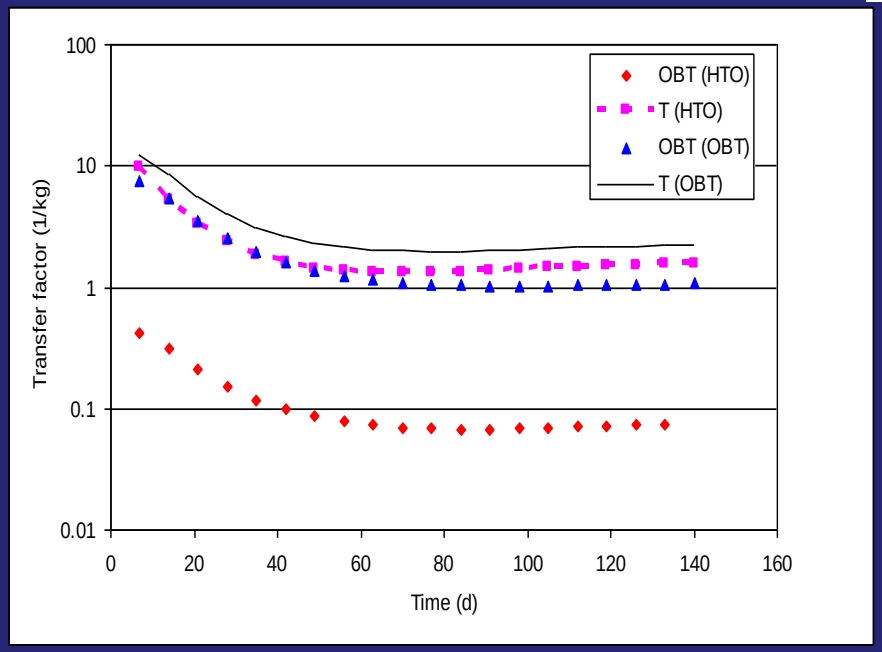


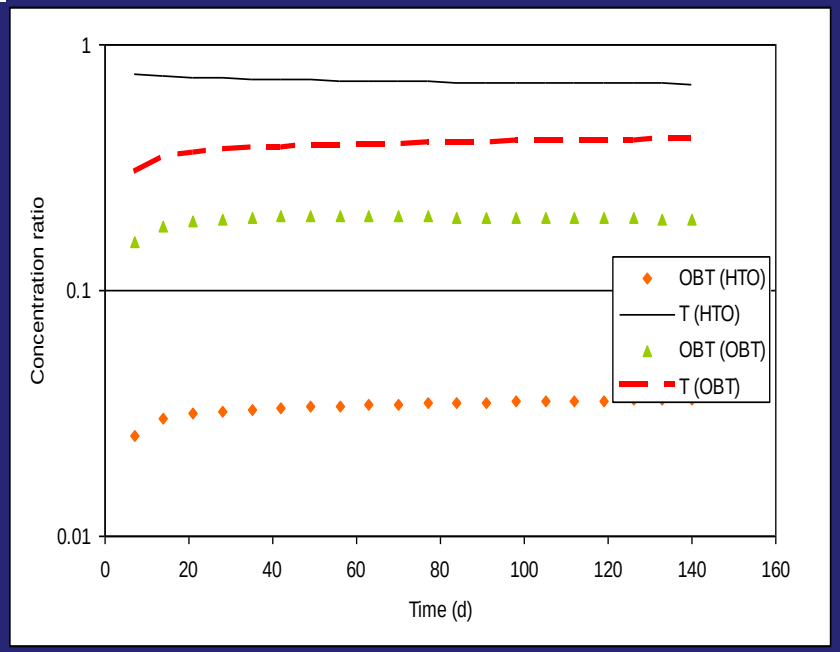
Extension to birds - application to broiler

Transfer factor for tritium in broiler



Concentration ratio for tritium in broiler





In the case of fast growing broiler, at the market weight of about 2 kg (42 days old) the model predicts lower transfer factors (TF) than for the equilibrium case

The predicted concentration ratios (CR) for our fast growing broiler are close to those obtained for “equilibrium” .

In absence of any experimental data or previous modelling assessments, our results give a first view on the transfer of ^{3}H and ^{14}C in birds.

CONCLUSIONS for animal model

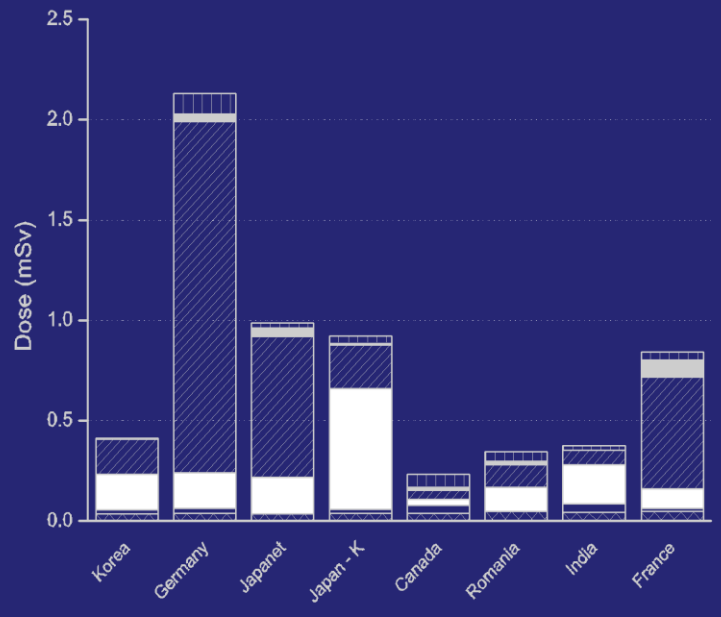
- We developed a process level oriented model for mammals (human included), birds, and aquatic foodchain based on a unified approach – animals energetic;
- Model inputs are taken from Life Science research, concerning animal metabolism, nutrition and growth → **Interdisciplinary Research**;
- The aim was to develop a robust model avoiding calibration, in order to be used directly in practice

Open problems in OBT modelling in crops

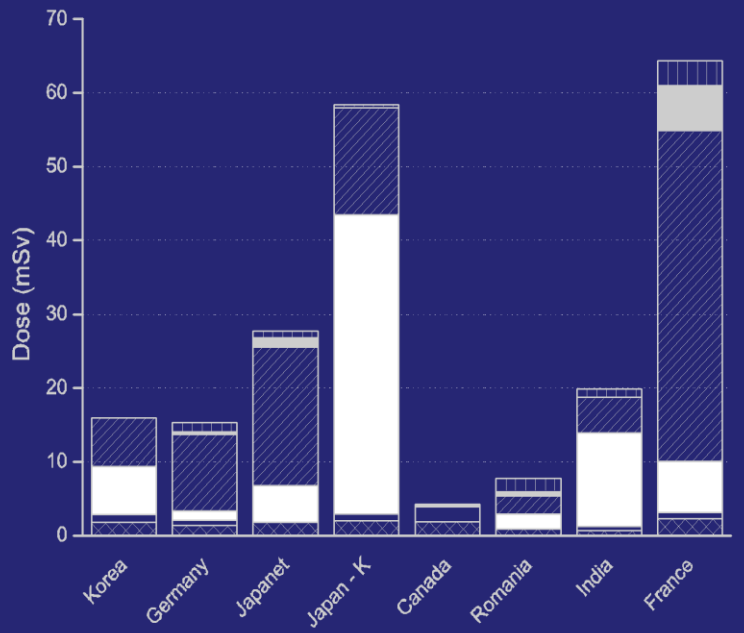
- Conversion of HTO to OBT imply HTO uptake (and release), photosynthesis and respiration and many biochemical reactions
- There is no deposition velocity but exchange velocity
- OBT is produced also in night
- Clear dependence on genotype, crop specie, development stage and environmental factors
- Large uncertainty in modeling

HYPO scenario EMRAS I
1 h HTO emission 10 g
at 1 km from stack

- **Case 1 day** Normalized by 6.10^{9} $Bq.s.m^{-3}$



- **Case 3 night** Normalized by 3.10^{11} $Bq.s.m^{-3}$

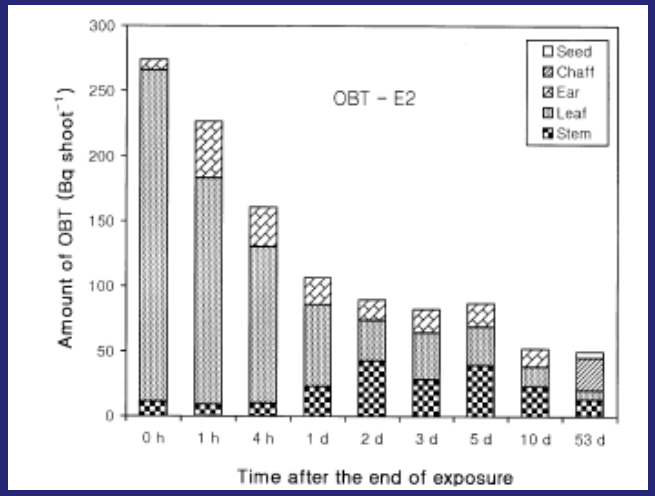


Too large uncertainty. Greenpeace will be happy. Not me

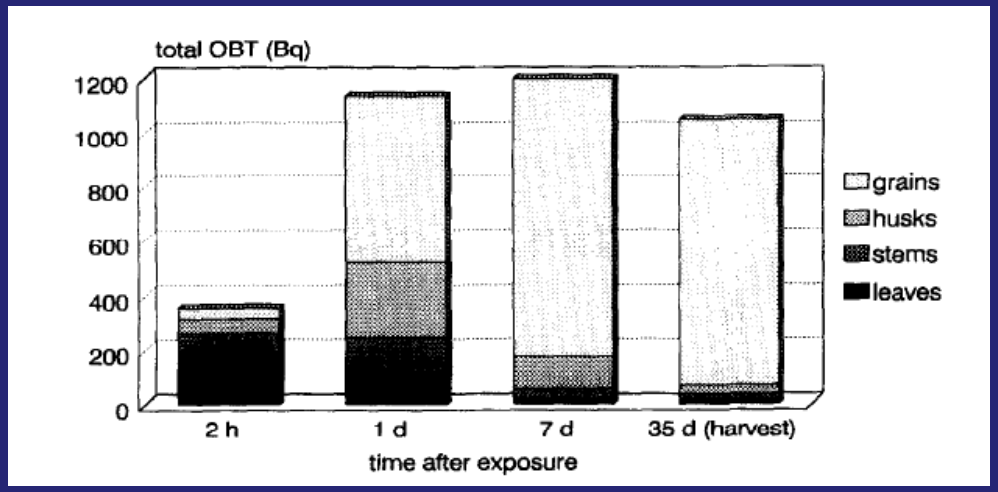
Half-Lives of TWT concentration in wheat within 1 h after the end of exposure to HTO

	TWT half-lives (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

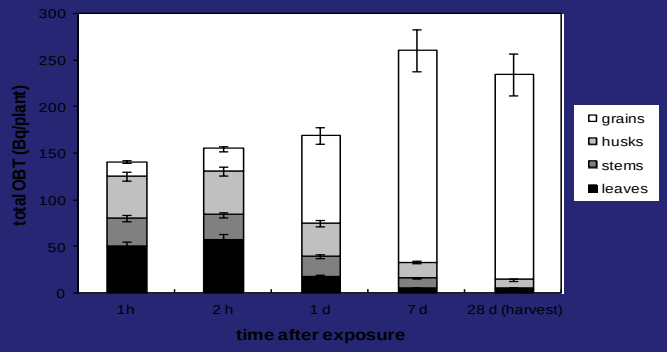
Dynamics of total OBT in wheat and rice



Rice 4 days after flowering



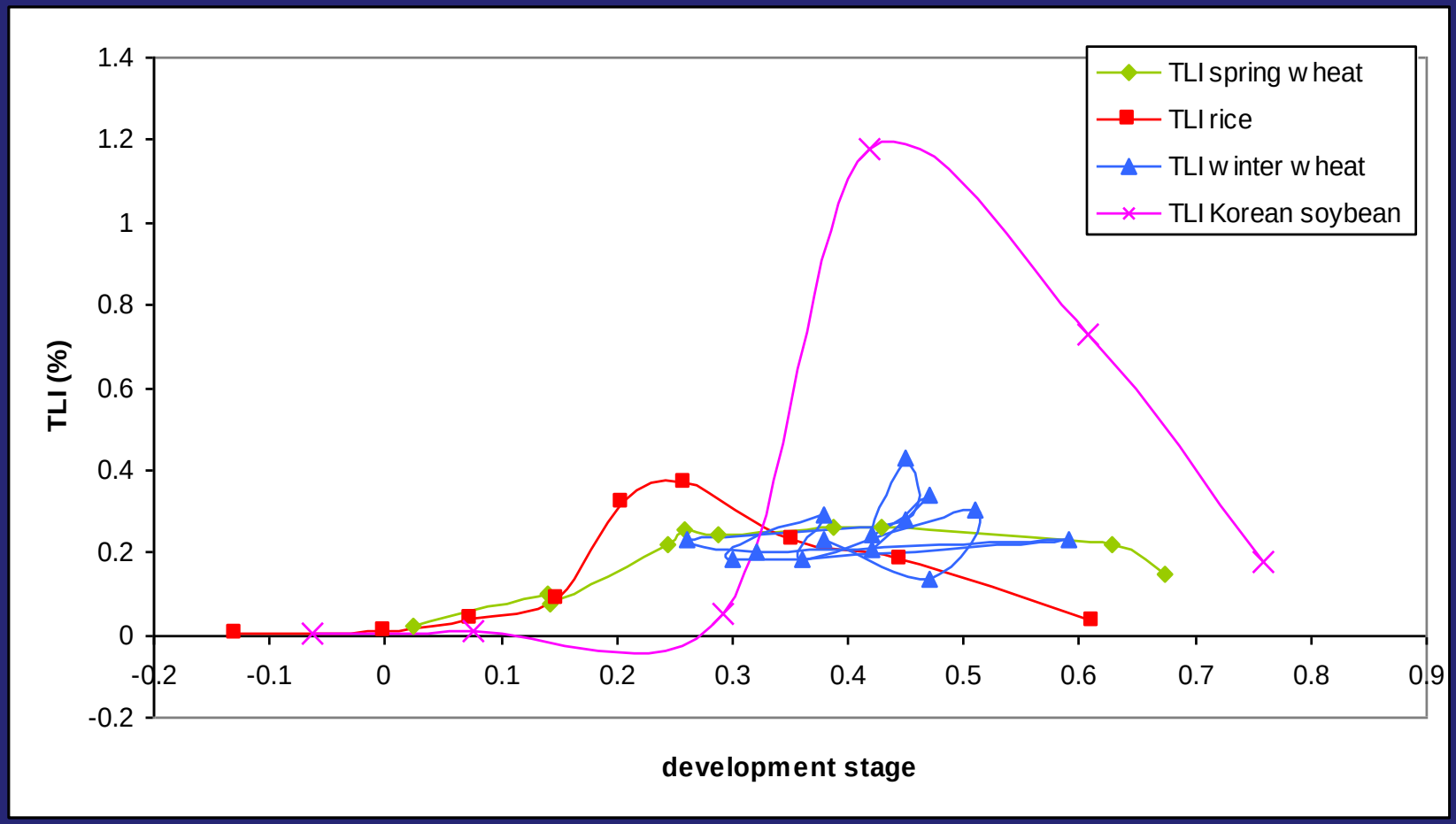
Wheat 20 days after flowering, ½ full light



Wheat 20 days after flowering at dusk

- Clear dependence on development stage;
- Processes are complex and respiration is very important;
- In the period of linear grain filling, formation needs 2-5 days and thereafter, it is a slow decreasing due to respiration

TLI comparison



TLI translocation index = the percentage of OBT concentration in grain (edible plant part) at harvest (Bq mL^{-1} water of combustion) related to the TFWT concentration in leaves (Bq mL^{-1}) at the end of exposure to HTO

CONCLUSIONS

"Tritium is one of the most benign of radioactive materials that I've worked with in my career, and I've worked with many of them. But on the other hand, the perception of tritium as a potential risk in the environment to the public is huge; it is absolutely huge. It is the industry's biggest problem since the Three Mile Island accident in 1979."



Dr. John E. Till, Author of
Risk Analysis for Radionuclides Released to the Environment - Oxford University Press 2008
(but Chernobyl? And Fukushima !!)
CURRENT CHALLENGES:
NIGHT FORMATION OF OBT IN CROPS
HARMONIZATION FOR CONCEPTUAL MODEL
PREGNANT WOMEN AND FOETUS
OPERATIONAL MODEL DESIGN - GENERAL CONCEPT
Budget!
IAEA MODARIA
Acknowledgments - huge list

