Overview on the dynamic models for Tritium transfer to animal products

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I. Introduction

Since 2003 IAEA has coordinated the EMRAS (Environmental Modelling for Radiation Safety) programme with a dedicated working group for ^{3}H and ^{14}C. The tritium working group aim is to decrease the uncertainty of the models assessments, focusing on the organically bound tritium (OBT) formation, and its transfer in the environment (humans being the end point). The final report in EMRAS WG2 (IAEA 2009) includes a single case for the models tests for farm animals (IAEA 2008). It was concluded that more tests and models inter-comparisons are needed in order to define the best operational model. The practitioners must also be aware of the user's influence on the model performance. This document is intended to be a step forward for the developing of a simple operational model that is based on the parameters values for animal metabolism and it satisfies the requirements of robustness needed today in radiological assessments.

The tritium contribution to ingestion dose highly depends on dietary habits and can have a large range (5-95 %). In current European diet, it is about 20 %, but for infant can be up to 50 %. For routine release, transfer coefficients and concentration ratio can be used with low uncertainty (IAEA 2010), but for accidental release the experimental data base is very limited (Table 1) and in many cases, there are not dynamic data. The products of interest are milk (cow, sheep), meat (beef, sheep, pork, and broiler), as well as egg. ^{3}H can be ingested by animals as either (or typically both) HTO (food and drinking water) and organic matter, including OBT. Inhalation and skin absorption are also possible routes of HTO intake. Exchangeable organic tritium and HTO rapidly equilibrate with body water. Organically bound tritium form in food is metabolised by animals and partially converted to HTO. Body HTO is also partially metabolised to OBT. If only tritiated water is given to the animal, only a small fraction is metabolized as OBT and the rest (99 %) goes in the water cycle of the animal. The half time for the water turnover is well known for domestic animals ($\approx$ 3.5 days for cow; $\approx$4 days for pig and $\approx$2.5 days for sheep). When a cow ingested HTO, a second component of a longer half time of about 60 days can be observed in the body water, as well as in the milk water. This is due to the catabolism of OBT, but has a low contribution (less than 2 %) to the integrated activity of the body water (Van den Hoek and Tenhave, 1983). In the milk constituents, after the cow was fed with HTO, two components with longer half times are observed: 33 days in casein and more than 200 days in fat. After OBT feeding the milk constituents show in addition a very fast component (half time of 1.5 days) beneath a medium and also a long component (Van den Hoek et al., 1985).

Table 1. The available experimental data

Food item	**Exp. availability**
cow milk after HTO intake	good exp.
cow milk after OBT intake	1 exp.
goat milk after OBT intake	good exp.
goat milk after HTO intake	no exp
Sheep milk after HTO intake	no exp
Sheep milk after OBT intake	no exp
broiler meat after HTO intake	no exp
broiler meat after OBT intake	no exp
egg after HTO intake	Russian exp.
egg after OBT intake	no exp
beef meat after HTO intake	2 exp.
beef meat after OBT intake	no exp.
veal after OBT intake	poor exp.
pig after OBT intake	poor exp.
piglets after OBT or HTO intake	medium exp.
Sheep after OBT intake	partial exp.

In order to understand the experimental data concerning the OBT transfer in milk or meat, it is useful to briefly discuss the fate of the organic food components. The relatively long molecules of carbohydrates, proteins and fats will undergo digestion, which is essentially a process of hydrolytic cleavage, involving the uptake of water. Upon absorption, the resulting smaller molecules (amino acids, monosaccharide and fatty acids) will enter the general pool of metabolic precursors where they can be used for any of the following processes (Van den Hoek 1986):

- *Formation of energy.* This is a metabolic oxidation involving the conversion of OBT to HTO. In case of OBT feeding, about half of the tritium is transferred to milk water (HTO);
- S*ynthesis of functional body constituents* (enzymes, hormones, structural elements, secretion (milk)). This involves conversion from one form of OBT to another form;
- *Synthesis of body reserves, particularly fats.* This again converts one form of OBT in another.

Daily animal feed intake has a large variability due to breed, diet quality, production level, and environment. The average values and ranges are given elsewhere (IAEA 2010), but there is not given an explanation on choosing a specific value. It must distinguish at least between the high efficient industrial farming and subsistence farming in unfavourable environment. A sheep of a similar mass and growth rate can consume in mountain rangeland two times more food than in a stable (Freer 2002). A small sized cow with a milk production of 5 L d^{-1} consumes about 8 kg dry matter (dm) of grass per day, but a large sized cow with a milk production of 40 L d^{-1} needs up to 25 kg dm per day. A high concentrated diet reduces the feed intake comparing with the roughages. Consequently, a variability of up to a factor of 3 rises only from feed intake.

II. Classic approach

Animal intake of tritium in bounded form includes both exchangeable and non-exchangeable OBT and the partition before digestion can be assessed using feed composition (see Annex). Digestion processes can change this partition and the effect is larger for ruminants. The bound hydrogen in the organic matter of plants that is digested to carbohydrates, proteins, and lipids by the animal is more likely to be synthesized into the organic matter of the animal than is the tritium atom that enters the body as water (Peterson 2004). The likelihood of transfers from diets to animals in decreasing order of occurrence is (the names of the transfer factors are given in parentheses):

- hydrogen in water to hydrogen in water (F_{HH});
- hydrogen bound in organic matter to hydrogen bound in organic matter (F_{OO});
- hydrogen bound in organic matter to unbound hydrogen in water (F_{OH});
- unbound hydrogen in water to bound hydrogen in organic matter (F_{HO})

The classical approach for the other radionuclides considers the convolution integral expression for the concentration in animal produce at time T (Müller and Pröhl, 1993):

$$C_{m,k} = \sum_{i=H,O} TF_{m,i,k} \sum_{J=1}^{J} \left\{ a_{m,i.k,J} \int_0^{\sigma} I_{m,i}(t) \lambda_{b,m,i,k,j} \bullet \exp\left\lfloor -\left(\lambda_{b,m,i,k,j} + \lambda_r\right)\left(T-t\right)\right\rfloor dt \right\} \qquad (1)$$

where $C_{m,k}(T)$ is the activity concentration (Bq kg^{-1}) in animal product, m at time T, $TF_{m,I,k}$ is the transfer factor (d kg^{-1}) for animal product, m, J is the number of biological transfer rates, $a_{m,I,k,j}$ is the fraction of biological transfer rate, j, $\lambda_{b,m,I,k,j}$ is the biological transfer rate j (d^{-1}) for animal product, m.

Consequently, it is necessary to have four transfer factors and, for the dynamic case, minimum four biological loss rates. This cannot be accomplished using the experimental data, with the exception of tritium in cow milk after a HTO intake. In this case, there are six data sets in order to infer both the transfer coefficients and biological transfer rates (Mullen et al, 1977; Potter et al, 1972; Van den Hoek and Tenhave, 1983). The data can be analyzed as a contribution of two terms, and the partition factors were normalized to 1. In Table 2 it is seen that the slow turnover of total tritium in cow milk (after HTO intake) has a low contribution to the total transfer, but it involves mostly the conversion of the OBT in the body to the HTO in body-water, as well as OBT in milk. The fast transfer rate (λ_1) corresponds to the body water halftime, but its range is lower than the range given in literature for water (Thorne et al., 2001). In a metabolic model (Galeriu et al, 2001) the transfer coefficient is correlated with the water turnover rate and the body water content. Using the recommended values, an average biological transfer rate of 0.22 d^{-1} can be used. For other animals, the values of the fast transfer rates given by the water turnover rate, were recently revised (Thorne et al., 2001) and can be used as default. It must take care for the seasonal variation of drink water and the influence of diet and production.

Table 2. Transfer coefficients and biological transfer rate for tritium in cow milk after HTO intake

Exp.	$\mathbf{F}^{\#}$	$\lambda_1^{\$}$	$\lambda_2^{\&}$	a_{2n}^{*}
Mullen et al., 1977	0.0128	0.217	0.005	0.007
Mullen et al., 1977	0.0167	0.207	0.0046	0.007
Mullen et al., 1977	0.0221	0.218	0.004	0.003
Mullen et al., 1977	0.02242	0.244	0.006	0.006
Potter et al., 1972	0.01	0.239	0.008	0.001
Van den Hoek and Tenhave, 1983	0.016	0.206	0.024	0.009
Mean	0.0167	0.2212	0.0086	0.0055
sd $^{+}$	0.005	0.016	0.0077	0.0029

transfer factor; $ fast transfer rate; & slow transfer rate; * a_{2n} long term contribution to milk productin; + standard deviation

For cow milk after OBT intake, it is a single report (Van den Hoek et al, 1985) emphasizing the influence of milk production and diet on the transfer, but not on the biological half-times. The transfer coefficient of total tritium varies by 30 % with a higher value for a cow with lower milk production and without concentrate in the diet (as it was expected in the metabolic model (Galeriu et al., 2001)). Analyzing the data in the frame of eq. (1), the biological transfer rates, λ_i, and coefficients, a_i, are obtained and given in Table 3.

Table 3. Biological transfer rates and associated coefficients milk for OBT and HTO after OBT intake

OBT	**fast rate**	**medium rate**	**long rate**
λ_i	6.67E-01	1.25E-01	1.14E-02
a_i	1.10E-01	2.65E-01	6.24E-01
HTO	**fast rate**	**medium rate**	**long rate**
λ_i	2.00E-01	1.49E-02	NA *
a_i	9.48E-01	5.23E-02	NA *

* not available

The values in Table 3 must be used with caution because there is an inherent variability among lactating animals as it was demonstrated by the data on two minigoats (Van den Hoek et al, 1985). On average, the minigoat transfer rates are close with those for cow, but the variability is up to 40 % for the fastest transfer rate and 20 % for the intermediate one. The values in Table 3 show the importance of various processes in the milk contamination: more than 85 % of the OBT in milk is produced by de novo synthesis (the fastest transfer in Table 3), a moderate fraction is linked with the intermediate transfer and very small fraction can be produced by the recycling of the body reserves (in muscle and adipose tissue). The specific activity of milk fat seems to be lower in the minigoat than in the cow (Van den Hoek et al, 1985) and the partition coefficients for cows in Table 3 are not recommended for the minigoats.

The transfer factors and bio-kinetic halftimes can be deduced from complex but robust models and the distinctions can be made comparing with the past assessments. An example is given in Table 4 for pig meat (Galeriu et al., 2009) where the old values used in RODOS (Galeriu et al., 2000) were changed with the results using MAGENTC model (Galeriu et al., 2009).

Table IV. Transfer factors and bio kinetic halftimes for pig meat (OBT)

	Unit	**FDMH**	**MAGENTC**
TF	d kg^{-1}	0.2	0.346
A_1	-	1	0.314
A_2	-	0	0.686
$T_{1/2\ 1}$	d	10	23.6
$T_{1/2\ 2}$	d	NA	219.7

III. OBT bio kinetic rate and basic models

In the absence of a complete experimental data set for bio kinetic transfer rates and transfer factors, covering all farm animals, it is the constraint to use the available models and partial date sets. The hybrid models, in which some of the compartments are counted by the specific activity approach and the others by the transfer factors, are also used. The process oriented models are more complex and used for the accidental releases, but they can be run until the equilibrium conditions are reached, too (and this is not cost effective). Models used for the dynamics of tritium and carbon in farm animals have different levels of complexity, having a single organic compartment (Raskob 1994; Higgins et al., 1995; Smith et al., 1995; Watkins et al., 1998; Maul et al., 2005; Tamponnet 2005; Sheppard et al., 2006a, 2006b) or five-six compartments (see below) and an additional compartment for tritium - the whole body water compartment. The models are based on a specific experimental data set (Takeda et al., 1995; Crout et al., 1998) or developed for the general use (Watkins et al., 1998; Thorne 2003; Galeriu et al, 2005).

The review starts with simple models and continues with complex models. The simplest models use a single organic compartment for the whole body or animal product.

There are many pools of organic carbon or OBT in any developed organism, but using the flux conservation it can define a single half time for the whole body or a specific tissue (organ). The biological half-times have been assessed in the past by a simple balance of body content of the stable carbon and carbon intake (Jones and Jackson 1986) and used also for organic tritium (Raskob, 1994). This approach, of common halftimes for ^{14}C and ^{3}H in organic forms, is supported by the basic science and modelling results (Crout et al, 1998; Galeriu et al, 2001). The use of the simple carbon balance in order to deduce the biological half-times is only an approximation. Traditionally, the whole body Carbon Loss Rate (CLR) was assessed considering the intake of digestible Carbon and the body Carbon content (Jones and Jackson, 1986):

$$CRL = f_d * I_f * \frac{C_{C,f}}{M * C_{C,b}} \tag{2}$$

where: CLR is the whole body carbon loss rate, f_d is the digestible fraction in food intake, I_f is the daily food intake (kg dm d^{-1}), $C_{C,f}$ is the carbon fraction in food (kg C kg^{-1} dm), $C_{C,b}$ is the carbon fraction in body (kg C kg^{-1} fm), M is the fresh body mass

The first estimate have been used in the past (Jones and Jackson, 1986) (see the last column in Table 5) but in the present report it is considered more actual and specific information (for a list of references see Galeriu et al. (2001)) and the growing animals are also included. In Table 5, the carbon loss rate due to maintenance needs only (T_{Cmaint}) and the total growth dilution and maintenance (T_C) are also given. Comparing the update values (Melintescu and Galeriu, 2010) with the old ones (Jones and Jackson, 1986), some differences are observed.

The updated values (Galeriu et al., 2003) are higher for pig, sheep and cow, lower for hen and similar for rat. These results show the influence of inputs in eq. (2). For growing animals (veal, lamb, broiler), the updated approach (Melintescu and Galeriu, 2010) considered the maintenance and growth needs in food intake taking into account the animal energy needs, but disregarding the different energy efficiency (McDonald et al., 1995).

Table 5. Updated carbon half times

Animal	**mass (kg)**	**growth (kg d^{-1})**	**Intake rate (kg dm d^{-1})**	**Maintenance intake (%)**	**Body C content (kg C kg^{-1} fw)**	**Digestible fraction (f_d)**	**C food content (kg C kg^{-1} dm)**	**halftime for total C (d)**	**halftime of C due only to maintenance (T_{Cmaint}) (d)**	**Tc (Jones and Jackson, 1986) (d)**
veal	160	0.8	4.85	52	0.173	0.7	0.42	13.45	25.86	
Beef	400	0.8	8.6	55	0.230	0.7	0.42	25.26	45.93	
Cow (no milk	550	0.1	6.7	92	0.254	0.7	0.42	49.15	53.42	36
Lamb	20	0.2	1	40	0.164	0.7	0.415	7.84	19.61	
Sheep	50	0.08	1.22	75	0.273	0.7	0.415	26.71	35.61	27
Pig	100	0.8	2.8	30	0.339	0.9	0.47	19.82	66.08	45
Pig (fast growth)	100	1.25	3.8	35	0.200	0.9	0.47	8.61	24.59	
Hen	2.5	0.007	0.12	78	0.251	0.83	0.47	9.28	11.90	18
broiler	1.7	0.03	0.11	56	0.241	0.83	0.47	6.61	11.81	
rat	0.45	0.0008	0.025	94	0.220	0.9	0.45	6.78	7.22	7

There is also a metabolic approach in defining the whole body loss rate of organic matter, starting with the Metabolic Theory in Ecology (West et al., 2001). West et al. (2001) released a metabolic model for ontogenetic growth with the central assumption that the basal metabolic rate depends on the body mass at power 0.75. There is much criticism for a single and universal exponent, but some results of West et al. (2001) can be used for aim of the present study. The mass equation proposed by West et al. (2001) depends on mass at birth, m_0, the mass at full maturity, M, and a specie dependent parameter, a. The mass equation in West et al. (2001) can be re-written in order to emphasize the gain and loss rate:

$$\frac{dm}{dt} = (a * m^{-1/4} - \frac{a}{M^{1/4}}) * m \tag{3}$$

The loss rate can be defined now as:

$$\lambda_{loss} = 0.1778 * a * M^{-0.25} \tag{4}$$

$$\lambda_{gain} = \lambda_{loss} * (\frac{m}{M})^{-0.25}$$

with masses in kg.

Then, it follows:

$$\frac{dm}{dt} = (\lambda_{gain} - \lambda_{loss}) * m \tag{5}$$

$$\frac{d\mu}{dt} = (\lambda_{gain} - \lambda_{loss)}) * \mu$$

where the maturity (μ = m/M) was used

The Relative Growth Rate RGR is simply:

$$RGR = (\frac{1}{m})\frac{dm}{dt} = \lambda_{gain} - \lambda_{loss} \tag{6}$$

From eqs. (3), (4), (5), (6) it is seen that the gain rate depends on the maturity degree and that the loss rate depends on the specie and mature mass.

An estimate of the loss rates of some animals is given in Table 6.

Table 6. Masses and loss rates for some animals (West et al., 2001)

Animal	a	m_o (kg)	M (kg)	loss rate (d^{-1})	halftime (d)
cow	0.28	33.3	442	0.010858	63.8262
pig	0.31	0.9	320	0.013032	53.17742
hen	0.47	0.043	2.1	0.069418	9.98294

IV. Derivation of the dynamic equation for a single organic compartment

It is used a generic equation for the animal growth (McDonald et al., 1995) and it is combined with the balance of radioactivity.

It is considered the well known energy balance for an animal of mass M:

$$RE = k_g * (MEI - MEm) \tag{7}$$

where: RE is the retained energy in the body (MJ d^{-1}), k_g is the growth efficiency, MEI is metabolisable energy intake (MJ d^{-1}), MEm is the maintenance energy need (MJ d^{-1})

The retained energy, RE is split in energy deposited in protein or lipids and corresponds with an increase in empty body weight, EBW. EBW is the live body weight, LBW, minus the gastrointestinal content:

$$LBW = g(M) * EBW \tag{8}$$

where g(M) is an a-dimensional function

The empty body gain, EBG, is given by an increase of body protein, ash, lipid and water:

$$EBG = \frac{dEBW}{dt} = RE * f(EBW) \tag{9}$$

The relation between EBG and RE needs a function, f, depending on EBW. The function transforms the gain in energy into the gain in mass and has as units kg d^{-1}/(MJ d^{-1}) = kg MJ^{-1}.

So, it follows:

$$\frac{dM}{dt} = EBW * \frac{dg}{dt} + \frac{dEBW}{dt} \tag{10}$$

The first term in eq. (10), because g is quite constant is ignored and eq. (10) is:

$$\frac{dM}{dt} = g * EBG = (g * f) * RE = (g * f) * k_g * (MEI - MEm) \tag{11}$$

Considering the level of nutrition, L=MEI/MEm and the parameterize MEm=aM^b, it is obtained the following eq.:

$$\frac{1}{M}\frac{dM}{dt} = k_g * f * g * aM^{b-1} * (L-1) = \lambda_{gain} - \lambda_{loss} \tag{12}$$

For a specie, parameter "b" is constant, but parameter "a" depends on genotype and the previous nutrition (the influence of level of nutrition on visceral mass and finally, on maintenance energy).
The following equations are defined now:

$$\lambda_{loss} = (k_g * f * g) * a * M^{b-1} = (k_g * f * g) * \frac{MEm}{M} \tag{13}$$

$$\lambda_{gain} = L * \lambda_{loss}$$

$$RGR = \lambda_{loss} * (L-1) \tag{14}$$

Knowing the metabolisable energy density, MD, in animal food and the dry matter ingestion rate, I_d, it follows that $MEm = MD * I_{d,m}$ and $MEI = MD * I_d = L * MEm$

At full maturity, M_{mat}, there is not growth and the intake rate $I_{d,m}$ compensates the loss rate $I_{d,m} = \lambda_{loss} * M_{mat}$.

Generally, the loss rate can be assessed knowing the animal composition and the metabolic needs for maintenance. For a lactating animal (constant mass) the metabolisable energy for milk production must be used for assessing the gain rate.

For the balance of radioactivity (^{14}C or OBT) in the whole body, M, there are used the derivation for ^{14}C (Sheppard 2006a), the approach for biota (Beresford et al., 2004) and the fact that the loss rate for radioactive organic matter is the same as for matter (Makarieva et al., 2004), while the gain rate is:

$$\lambda_{gain} = (\frac{I_d}{M}) * D * (\frac{C_f}{C_o}) \tag{15}$$

where C_f and C_o are the radionuclide concentrations in food and animal, respectively (Bq kg^{-1} dm)

Then, the radionuclide concentration in animal is:

$$\frac{dC_o}{dt} = (I_d * \frac{D}{M}) - (\lambda_{loss} + RGR) * C_o \tag{16}$$

It is concluded that the loss rate is driven by the maintenance metabolism (intake rate for maintenance only), while the gain rate depends on the current intake (level of nutrition). In the same time, the factor D is not the digestibility, but the metabolisability of food, q (the ratio between metabolisable and gross energy in animal food).

For tritium, a whole body water compartment is added in all models. For the simplest model, there are two compartments (organic and water) and for the complex models, the organic body compartment is split in many components.

V. Analysis of simple models

There are two simple models: STAR-H3 (Smith et al., 1995; Watkins et al., 1998) and OURSON (Ciffroy et al., 2006), the last one being applied for the pig scenario (IAEA 2008). Their flowcharts are given in Figure 1.The models differ in their intake routes and transfers between HTO and OBT and OBT and HTO, respectively. In STAR model, the input of OBT is only in the fast (body water) compartment, while in OURSON model is only in the slow (Body OBT) compartment. STAR considers the metabolisation of OBT from body HTO, but this is ignored in OURSON.

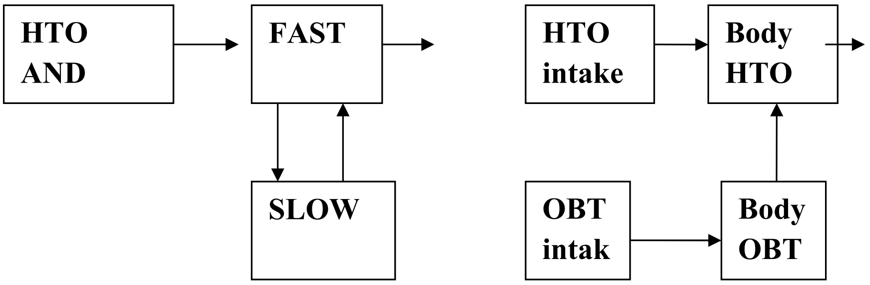

Figure 1. Flowcharts for STAR (left) and OURSON (right) simple models

The simplest model STAR-H3 (Smith et al., 1995; Watkins et al., 1998) was developed in 1995-1998 and has a single organic compartment (slow turnover). The model is implemented in a software platform (AMBER) having a restriction on the intake (only pasture). The fast and slow turnover compartments are considered in the STAR models for both ^{14}C and ^{3}H. In the case of tritium, the fast compartment can be identified with body water and the slow with organically bound tritium in the animal body. In the case of ^{14}C the fast compartment can be linked with the fast component of animal respiration. The model considers 1 kg of animal meat and the intake of water or feed is normalized to animal carcass. Only pasture is considered and pasture equivalent feeding rate is assessed for non-ruminant animals. All intakes come to fast compartment only and all excretion leaves the fast compartment. STAR is intended to be simple and conservative covering animal products needed in UK radiological assessment but the degree of conservatism is not documented. A week point for STAR application in case of tritium is its inability to distinguish the separate intakes of HTO or OBT, as well as its inability to distinguish between milk and egg. OBT in milk and egg being produced mostly in *de novo* synthesis, STAR considers only the slow turnover and the dynamics in these products cannot be predicted. Using only pasture equivalent as animal feed, STAR cannot be applied in cases of dried diets (winter hay, concentrates and grains) used in many countries. For all animals, the slow turnover rate is 0.03 d^{-1} (half-time of 23 d). Documentation of STAR is not clear, but a direct interaction with the key developer was done in the past and the full description and parameters were accomplished and reported in pig scenario (IAEA 2008).

The OURSON model (Ciffroy et al., 2006; IAEA 2008) considers that all OBT in the diet enters the organic compartment, and the dynamic equation is derived for the specific activity (SA) including the growth dilution. Input OBT is corrected for the difference in SA from food and whole body. The transfer rate to Body HTO is given by digestible intake per body dry weight. HTO concentration in urine is considered to be the same as Body HTO, while OBT concentration in urine urea is taken to be equal with body OBT concentration. OBT in faeces correspond to OBT in the non digestible fraction of food. It was assumed that OBT specific activity was identical in the digestible and non digestible fractions. It was considered that whole body OBT was representative for the muscle compartment. Concentrations in other organs were derived from concentration in muscle using a correction factor based on fats and proteins content of each organ, fats and proteins turn-over rate, H content of fats, proteins and carbohydrates. In the original model and its subsequent applications there were unclear explanations of OBT halftime (a range only). The equations in OURSON for growing animal (no lactation) are given below:

$$\frac{dC_{urine}^{HTO}(t)}{dt} = -\lambda_w C_{urine}^{HTO}(t) + \frac{1}{H_2O_{pig}}\left(HTO_{diet} + k_{ing}.OBT_{pig}(t)\right)$$

(17)

$$\frac{dA_{meat}^{OBT}(t)}{dt} = -k_{ing} A_{meat}^{OBT}(t) + k_{ing}.\frac{H_{food}}{H_{meat}}.A_{food}^{OBT}(t) \qquad (18)$$

where: C_{urine}^{HTO} is the HTO concentration in urine (and animal body water H_2O) (Bq L^{-1}), $\lambda_w = \frac{waterconsumption(L)}{bodywater(L)}$, HTO_{diet} is the HTO activity in diet (drinking water +food) (Bq d^{-1}), k_{ing} is the OBT turn-over rate (day^{-1}), given by $k_{ing} = \frac{I \cdot D}{W}$, $OBT_{pig}(t)$ is the total OBT in pig (Bq), I is the food intake (kg dm day $^{-1}$), D is the digestibility (unitless), Wis the animal dry weight (kg), A_{meat}^{OBT} is the OBT specific activity in meat (Bq g^{-1} H), A_{food}^{OBT} is the

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OBT specific activity in food (Bq g^{-1} H), H_{food} is the average food OBH (g kg^{-1} dm), H_{meat} is the average meat OBH (g kg^{-1} dm)

In OURSON model there is not a metabolic transfer from Body HTO to body OBT. The transfer from body HTO to body OBT is ignored and all OBT intakes enter only to the OBT body compartment. This contradicts experimental evidence, at least at equilibrium.

Consequently the intake of organic tritium must be distributed between Body OBT and Body HTO, and not as in STAR or OURSON. This explains why STAR over predicts the OBT in urine and OURSON under predicts it. STAR considering all the organic intakes in the fast compartment, it is normal to under predict the OBT in animal organs. The under-prediction of HTO in meat observed for OURSON is explained again by the intake route of OBT, only in Body OBT, with a subsequent slow transfer to Body HTO.

In the TOCCATA model (Le Dizes 2004) a simple model is also used (Tamponnet 2005) and the maintenance loss is defined but the quantitative relations are not given and the OBT halftime is described in a improper and unpractical approach.

For tritium in cow (milk and meat) a very used model is included in UFOTRI (Raskob, 1994), a standard code for fusion reactor design and licensing. This model is discussed in more details, because it has peculiar accomplishments. Despite its simplicity, it potentially can be generalized for other animals. A single animal organic compartment is used in UFOTRI, but milk HTO and milk OBT compartments are added explicitly. All HTO intakes come to animal body water (HTO) compartment but OBT intake is distributed between body HTO, body OBT and milk OBT. This last direct transfer is not a physical transfer, but it helps to model the *de novo* synthesis. The cow in UFOTRI has a mass of 500 kg and gives 15 L d^{-1} of milk. The model transfer rates are obtained using the mass balance of free and bound hydrogen and few assumptions (e.g. OBT loss rate). The model was tested with published (Van den Hoek and Tenhave, 1983; Kirchmann et al., 1973) or unpublished experimental (van Hess M, personal communication, 2000) data as below:

1. A cow of 461 kg giving 12 L d^{-1} of milk was fed for 30 days with HTO. Milk was monitored for total T and OBT. At the end of the feeding the cow was sacrificed and the organs were measured for OBT. At day 30, the time integral ratio (the ratio between predicted and observed results), Total tritium P/O denoted as T = 0.85, milk OBT=0.64. At day 30, muscle P/O=0.78

2. A cow of 550 kg giving 22 L d^{-1} of milk was fed for 25 days with HTO. Total T and OBT in milk were measured for 70 days. UFOTRI P/O for total T is 0.6 and for OBT is 0.68.

3. Single HTO intake: Total T in milk integral at day 70 is very close to data; OBT in milk, P/O=0.8.

4. A cow of 566 kg giving 9.2 L d^{-1} of milk received OBT in hay for 28 days, followed by milk monitoring (data in the intake phase were not reported). At day 28, UFOTRI under-predicts OBT in milk with a factor of 2 (P/O=0.5) and total T with 70 % from the observed data (P/O=0.7). The integral between day 28 and day 100 is also under-predicted with the same factor (a factor of 2 for OBT).

The UFOTRI model seems to slightly underestimate the OBT in meat and milk with a factor less than 2, remarkably for a simple model. The approach taken in UFOTRI can be expanded for sheep and goat.

VI. Complex models

Complex models with many organic compartments were also published; for sheep (Crout et al., 1998) treated ^{14}C and ^{3}H case using similar transfer rates, but the parameters are derived from sheep experiments (Crout et al., 1998) and cannot be generalized to other animals. The occurrence of fast and slow turnovers of OBT can be included using two organic compartments as in MCT (IAEA 2008) and PRISM (Thorne 2003; Maul et al., 2005). The flowcharts of MCT and PRISM models are given in Figure 2.

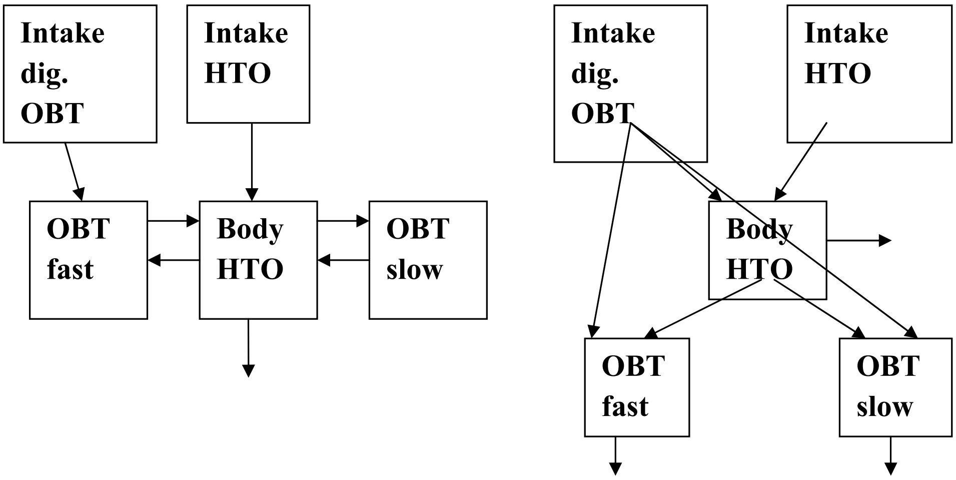

Figure 2. Flowcharts of MCT (left) and PRISM (right) models

In these models, two OBT compartments and one FWT compartment are assumed. Both models have a fast and slow OBT compartment, but MCT transfers the catabolic OBT to the Body water, while PRISM transfers it directly out, and it seems that it is a model simplification. The MCT model was initially developed for humans, including Japanese humans (Saito 1992). Assuming that the pigs can mimic the humans (or vice versa), the MCT model was used with minimal changes for pigs in Pig scenario (IAEA 2008), since the hydrogen metabolism in the pig is expected to be similar to that of humans. PRISM in its initial form does have not urine and faeces, but an extension of the model was released (Walke and Thorne, 2007). PRISM uses a simplified GI tract and OBT intake is partitioned between body water and the two organic compartments. A fraction of 0.79 (range 0.61-0.94) is converted to HTO and distributed to body water. The rest enters both organic compartments with two times more in the fast organic compartment (fast OBT). The fast organic compartment contents two times more hydrogen than the slow one (close with MCT for humans) as average, but the range is very large (between 1/9 and 9). As in STAR, milk *de novo* synthesis cannot be modelled. Transfer routes are very different, as well as many transfer rates, but both models give predictions relatively close with the observations for the pig scenario (IAEA 2008). MCT does not consider the fraction of input organic tritium directly absorbed in the body OBT, and this explains the under-prediction in urine. MCT was only applied for pig and PRISM parameters are given as ranges, with little information on selected animals. PRISM was not tested with experimental data (except the sheep) and is commercial software.

MAGENTC (Mammals GENeric model for the transfer of Tritium and Carbon) was developed as a research model and is more complex. The model is well documented in the literature and the default parameters are given (Melintescu and Galeriu, 2008; Galeriu et al., 2009; Melintescu and Galeriu, 2010). The model considers six organic compartments and a single body water compartment and its flowchart is given in Figure 3.

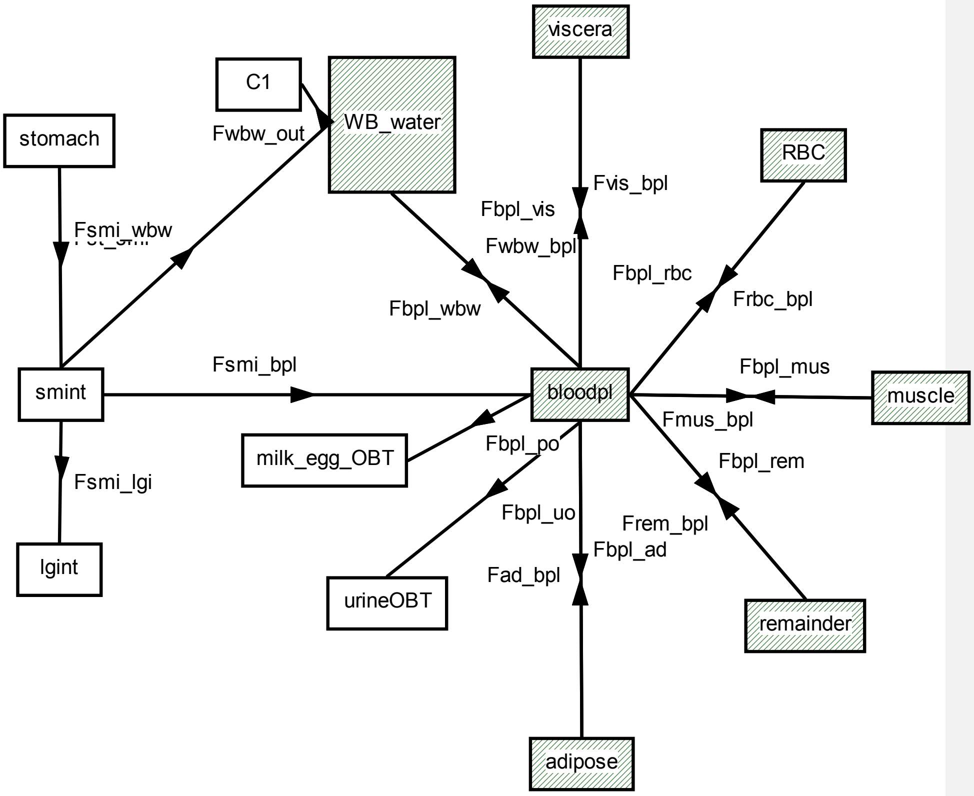

Figure 3. Flowchart of MAGENTC model

For the adult mammals, the model for T&C transfer in body compartment is based on the following assumptions:

- The most important body organic compartments are viscera (including the heart), muscle, adipose tissue, blood (plasma and red blood cells (RBC)) and remainder (including the brain). Their masses and compositions are known.
- Tritium in body water equilibrates fast with water and a single body water compartment is enough, if tritium is of concern.
- Loss rate from the organic compartment is similar for the intakes of HTO, OBT or OBC and can be assessed directly from energy turnover rate (net maintenance).

- Net maintenance is considered as a sum of the basal metabolic needs and activities and the thermal stress is not considered. Basal metabolic rate is a sum over the multiplication between the organs specific basal metabolic rate and the organ mass.
- Specific metabolic rate (SMR) for organs in adult mammals varies marginally, excepting the muscle and comparing with the basal state. Basal SMR shows a mature mass dependence. SMRs are obtained experimentally for few mammals and a zero order approximation is generally used depending on the mature mass.
- There is a metabolic conversion of HTO to OBT and equilibrium value does not vary among mammals for the ration of OBT derived from HTO or OBT intake.
- The energy (heat) and the additional matter lost in transformation of the metabolisable input into the net requirements are considered as a single, fast process.

The net daily energy expenditure of animals is referred to as the net ‘field’ (for active animals) metabolic rate (FMR; MJ d^{-1}), whilst the daily energy expenditure per unit fresh body mass is termed the specific metabolic rate (SMR; MJ kg^{-1} d^{-1}). The energy turnover rate or relative metabolic rate (ReMR; d^{-1}), can be defined as the ratio of SMR and the energy content of the body (determined by the body composition (protein, lipids, and carbohydrates)):

$$\mathrm{Re}MR = \frac{FMR}{EBW * BED} = \frac{SMR}{BED} \tag{19}$$

where EBW is the empty body mass (kg) defined as the live-weight less the mass of the gastro-intestinal contents and BED is the body energy density (MJ kg^{-1} fresh weight (fw)). The BED is estimated from the body tissue composition of lipids and protein, and combustion energy of lipids and protein

Under these hypotheses the model gives reliable predictions with no calibration.

Previously to the model development, a revision of the animal growth and nutrition was done (Galeriu D and Melintescu A, personal communication, 2002), as well as a revision of the experimental data base (Melintescu 2002).

For the growing farm animals the model has an additional growth model, which gives both the growth rates of the organic compartments in the model and changes in body composition with age and management (Melintescu and Galeriu 2010). The growth model depends on the animal and can be adapted to each country research results. For ruminants, a generic model was used (Tedeschi et al. 2008), for pigs a French models was implemented (van Milgen et al., 2005), while for hen and broiler different literature reports (Gous et al., 1999; Lopez et al., 2007) were used.

For a constant OBT concentration in the diet (1 Bq kg^{-1} dm) administrated to three pig genotypes, the results are given in the Figure 4. In Figure 4, it is seen we that the genotype is not particularly important for the continuous intake and the whole body overestimates the muscle concentration by a factor depending on the pig obesity.

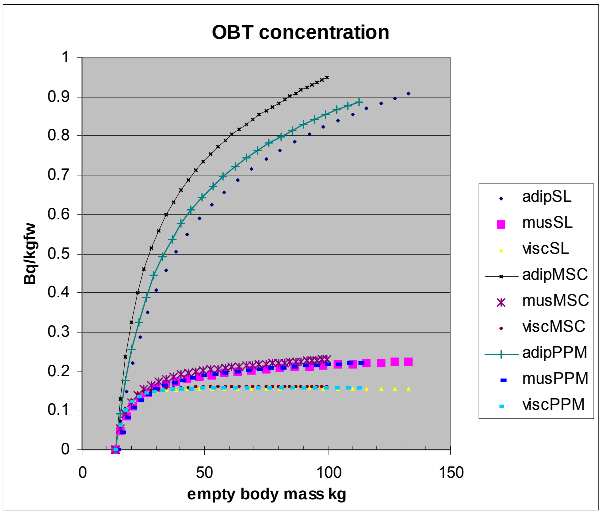

Figure 4. OBT concentration in pig organs (Bq $^{kg-1}$ fw) for different pig genotype (SL$^{\#}$, MSC$^{\$}$, PPM*) for viscera, muscle and adipose tissue
(# - conventional genotype; $- fat genotype; * - lean genotype)

VII. Quality assurance for models

For the operational application, any environmental model must be subject to the quality assurance procedure (EPA 2009) including sensitivity and uncertainty studies, but also test (validation) with the experimental data. For a research model, the recommendation is to fulfil the same way in order to assess the robustness of its predictions. In the present study, the only dynamic models subjected to intensive tests are MAGENTC model based on energy metabolism and the tritium cow-milk model in UFOTRI. Below some examples which emphasize these tests are given

MAGENTC model includes a parameter for the non-exchangeable fraction of organic intake which remains after the digestion. Prior to digestion this value is known from the diet composition. But the digestion can change the value at its half (for ruminants). It is expected a high sensitivity for the predicted concentration in meat. The model considers the homeostatic control for tritium and the sensitivity of OBT concentration in meat is low. A change with a factor of 2 in the non-exchangeable fraction results in change of 10 % in meat concentration.

For OBT in milk after OBT feeding an inter-comparison with the experimental data was done, including the TRIF (Higgins et al., 1995), UFOTRI (Raskob 1994) and MAGENTC (Galeriu et al., 2009) models, as it is shown in Figure 5. It is clear that TRIF model underestimates by a factor more than 5 in the uptake phase, but it overestimates later. UFOTRI and MAGENTC models give reasonably close values comparing with the experimental data.

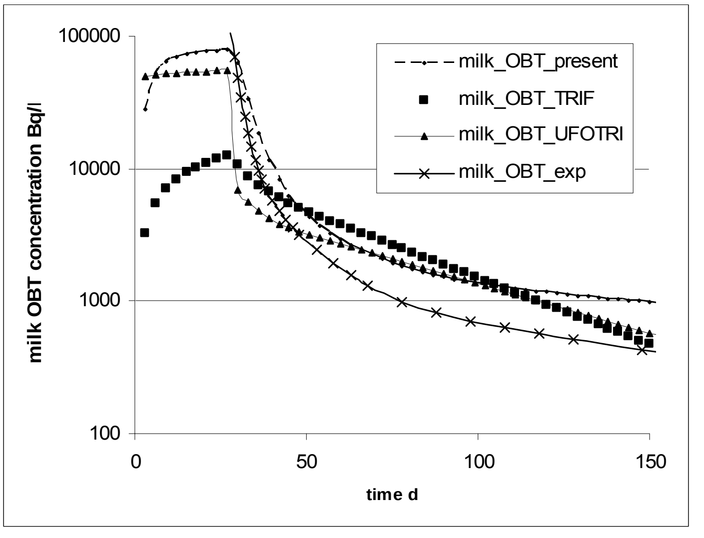

Figure 4. Test of TRIF, UFOTRI and MAGENTC models with the experimental data for OBT in cow milk

The MAGENTC model was intensively tested with all available experimental data on cow milk and the results are given in Table 7 as average and standard deviation of the predicted to observed ratio. There is an under prediction of OBT in milk after a HTO intake and an over prediction after OBT intake. This is explained by the specific of the ruminant digestion, which was not considered in the model. The large part of the intake is in form of carbohydrates which are transformed by rumen digestion in exchangeable forms and the equilibration with hydrogen (tritium) in water is almost complete. The precursors of milk glucogenesis in ruminants have more tritium after an HTO intake and less tritium after an OBT intake than in monogastric animals. This shortcoming of the model is not of major concern in practical application, because the miss-prediction is less than a factor of 2, the same order as the variability of cow productivity and diet in the assessment context.

Table 7. Model performance for dairy cow; NA: not calculated/available

Experiment	**R^2**	**Milk total ^{3}H**	**Milk OBT**	**Urine HTO**
		Mean ± standard deviation P/O (range presented in parenthesis)		
Cow_P	0.97	2.60 ± 1.7 (0.8 -1.9)	1.68 ± 0.8 (0.5 - 2)	2.90 ± 2.34
Cow_C	0.89	0.97 ± 0.08 (0.9 -1.4)	0.73 ± 0.17 (0.65 - 1.7)	0.97 ± 0.06
Cow_H3	0.67	1.02 ± 0.15 (0.9 - 1.5)	0.49 ± 0.12 (0.4 - 0.9)	1.36 ± 0.42
Cow_H	0.88	1.45 ± 0.59 (0.6 - 2.3)	1.86 ± 0.38 (0.55 - 2.12)	NA

In the past there was a single exercise for the models tests at the international level: the pig scenario in EMRAS I (IAEA 2008; Melintescu and Galeriu, 2008). A blind test for a pregnant sow asked the OBT in organs after 84 days of feeding. The predictions for organs HTO and OBT concentrations were supplied by Food Standard Agency (FSA), UK (PRISM model), Japan (MCT model), Electricite de France (EDF), France (OURSON model) and National Institute for Physics and Nuclear Engineering (IFIN-HH), Romania (MAGENTC model, but also STAR and PRISM). The HTO predicted to observed ratio shows that, excepting FSA, all models give good predictions, but EDF underestimates with a factor of 5. Table 8 gives the predicted to observed ratio for OBT in organs. STAR, as expected, underestimates with a factor of 10 due to the model assumptions of OBT intake in the fast compartment (body water). EDF, with all OBT intakes in the organic compartment is close to the observations, but the OBT in muscle is more overestimated. This must be analysed and understood. MCT overestimates with a factor between 2 and 4, while IFIN-HH (in the initial version) overestimated with a factor of 2. The reconstructed PRISM overestimated only by 50 %. It is observed the large range of overestimation in FSA results and this demonstrates the user influence.

Table 8. Predicted to observed ratio (P/O) for organs (day 84 after the start of contamination)

			P/O (OBT)			
Organs	**MCT**	**FSA**	**IFIN_HH (2005)**	**PRISM (DG)**	**STAR**	**EDF**
Heart	2.05	9.89	1.40	1.51	1.29	1.29
Lungs	2.79	4.11	1.90	2.06	0.13	1.30
Liver	1.92	1.04	1.11	1.20	0.08	0.84
Jejunum	3.00	3.23	1.73	1.88	0.12	1.09
Ileum	2.24	13.00	1.53	1.65	0.10	0.96
Colon	3.28	2.23	2.24	2.42	0.15	1.40
Kidney	2.17	8.46	1.48	1.60	0.10	1.17
Muscle	4.44	0.23	1.90	3.65	0.23	3.11
brain	3.91	4.69	na	3.17	0.20	1.65
blood	3.04	969.56	1.27	1.92	0.12	1.22

VIII. Final discussion

An operational radiological model must satisfy some requirements as following:

- To use the available input information (model parameters etc);
- NOT ask the specific calibration experiments;
- To give the predictions with an uncertainty better than a factor of 5 and desirable of about 2;
- To be able to be used for the dose assessment in human food chain and for biota radioprotection;
- It is preferred an over prediction;
- To be as simple as possible (but not simpler);
- To be internationally agreed.

For routine emissions, the transfer coefficients are mostly used, while the concentration ratios seem to be a more robust estimator. The ^{14}C and tritium part of the IAEA report (IAEA 2010) is robust and internationally accepted. Many experimental results are still not published in open literature and the disclosure of the important information remains to be accomplished. The practices and regulatory bodies' conservatism vary among EMRAS participant countries, as well as the transparency. To agree on a common recommendation for an accidental case seems difficult.

For the recommendations of the bio kinetic transfer rates there are progresses. Too few experimental data are available and the new experiments seem less probable due to the financial and ethics constraints. For the mature mammals, the recent advances in animal sciences can be of much help, but for the growing animals there are still uncertainties in the partition of needs between maintenance and growth. This can explain, partially, the divergence in the models assumptions and predictions, shortly described in the present study.

To further progress, experiments and more expert judgment are needed, as well as FULL communication between modellers, in order to guaranty the understanding of the models details, in their applications. To be internationally agreed, the models must be subjected to benchmarks and validation tests. MAGENTC is relatively complex, but it needs only available input data and shows reasonably agreement with the experimentally data, up to now.

It is proposed to use the eq. (16) for the simple dynamic approaches in case of non-lactating farm animals, because the regulatory bodies prefer quite simple equations with a minimum of parameters. In absence of a full experimental data base, it must use both the experimental data and models and it must also appeal to the required conservatism degree in various countries. In an accidental release of a special radionuclide it must first run a screening model to establish the mandatory needs of countermeasures or, at least, the special monitoring programs. This is based on a prediction of the first year ingestion dose and first days feed contamination level. Later it must have the first measurements on the feed contaminations and can improve the radiological assessment. A simple operational model needs more collaboration between EMRAS members and interaction with each country regulatory body. The aim of this study is to determine a relaxed communication between members and not to promote a certain view.

A simple but robust model for tritium transfer must:

- use a proper definition of the loss and gain rate;
- make a distinction between gain in mass and gain in radioactivity;
- consider the splitting of the organic tritium intake in the body water and body organic bound tritium;
- include the formation of OBT from body HTO;
- include the approach taken in UFOTRI for the lactating animals;
- test the model with the experimental data and compare with the complex MAGENTC model.

This activity can be included in the following (?) EMRAS III.

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ANNEX

Table A1. Hydrogen and carbon content of the basic constituents of food (Stoica 1997; McDonald et al., 1995).

Food constituent	Free H	Organically bound H	Total organic H	C
Water	0.11	0	0	0
Carbohydrate	0.02	0.044	0.064	0.44
Protein	0.017	0.051	0.068	0.52
Lipids	0.003	0.117	0.12	0.77

Table A2. Carbon and bound hydrogen content of basic animal food

Food class	C content (kg C kg^{-1} dm)	CV	Organic H content (kg H kg^{-1} dm)	CV	NE organic H content (kg H kg^{-1} dm)
Grasses	0.418	0.03	0.06	0.03	0.043
Hay	0.424	0.012	0.061	0.02	0.043
Silages	0.403	0.09	0.058	0.07	0.041
Roots	0.414	0.05	0.059	0.04	0.041
Concentrate	0.457	0.06	0.066	0.05	0.045
Shrots (ind res)	0.45	0.02	0.063	0.02	0.043

Table A3. Typical hydrogen and carbon contents of animal products (kg H or kg C per kg fw) (Geigy 1981)

Animal product	Free H	Organically bound H	Total organic H	C
		Milk		
Cow	0.096	0.008	0.010	0.067
Sheep	0.090	0.014	0.016	0.107
Goat	0.095	0.009	0.010	0.070
		Meat		
Beef	0.077	0.022	0.025	0.178
Veal	0.077	0.021	0.024	0.173
Mutton	0.074	0.026	0.029	0.203
Lamb	0.077	0.021	0.025	0.176
Goat	0.077	0.021	0.024	0.172
Pork	0.066	0.034	0.038	0.258
Hen	0.077	0.022	0.025	0.178
Chicken	0.080	0.019	0.22	0.155
Egg	0.074	0.018	0.021	0.142

Table A4. Ruminant feed parameters

Feed	Dry matter	Org. matter digestibility	Metabolisable energy density (kJ kg^{-1} fw)	Metabolisable energy density	q	Km	Kl	Kg
hay	0.86	0.592	7160	8326	0.45	0.66	0.577	0.357
concentrate	0.88	0.815	10690	12148	0.64	0.74	0.657	0.528
grain	0.88	0.87	11528	13100	0.715	0.75	0.667	0.564
straw	0.88	0.84	1147	1303	0.302	0.6	0.525	0.241
pasture	0.215	0.72	2181	10144	0.56	0.7	0.617	0.443
upland pasture	0.376	0.51	2200	5851	0.344	0.65	0.5404	0.27432