

## Models for the transfer of radionuclides in the food chain

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**Abstract.** Airborne particulates containing radionuclides like  $^{238}\text{U}$  decay with several intermediate products to  $^{226}\text{Ra}$ ,  $^{222}\text{Rn}$ , and thence to  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  which are ubiquitous in soils and consequently available for uptake for animals and plants. After decay of  $^{222}\text{Rn}$  or its daughters in air,  $^{210}\text{Po}$  deposits on plants. Grazing animals may take up appreciable amounts of  $^{210}\text{Po}$  from the environment. A dynamic multi-compartmental model is proposed to describe mathematically the radionuclides transfer to the food chain and predict the activity concentration within each sub-compartment. The dynamic model is defined by a system of linear differential equations with constant coefficients describing the mass balances in the different compartments, taking into account the fluxes in and out and the radionuclides decay inside each compartment. The system of differential equations was written in the standard matrix form used in the space state approach and solved numerically using Matlab software. The model output represents the time variation for the considered radionuclide in the *pasture-cow-milk* exposure route and also its distribution within the cow. The aim of the model is to evaluate whether this exposure pathway is relevant in radiological terms; it also allows verifying if more detailed investigations are needed in order to evaluate possible exposure consequences in a specific site. A simulation was done for  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in a hypothetical case study for different site conditions considering as initial state a contaminated pasture that is consumed by a cow. The simulation results fit within the range of values given in the literature, although many assumptions had to be done due to the limited available data; conservative parameters had to be used in some cases.

### 1. Introduction

Radionuclides may enter into the environment as a result of accidental circumstances and adversely affect public health and safety. The radionuclides released into the environment can give rise to human exposure through the dispersion of the radionuclides by wind or water carrying the radioactive and other toxic materials to the atmosphere, aquatic systems or to soil sub-compartments.

One of the principal ways that the public can be exposed to radionuclides hazard is by the inhalation of airborne particulates containing  $^{230}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , as well as uranium, or through the food chain contamination. The exposure may result from direct inhalation of contaminated air or ingestion of contaminated water, or from a less direct pathway - the ingestion of contaminated food products. The most serious contamination problems arises from radionuclides deposition on animal feed crops or on food crops directly consumed by man but the immediate and usually the most significant incorporation of the radionuclides into cow's milk will occur through the ingestion of contaminated pasture by animals. Humans will be subsequently exposed by the consumption of animal products contaminated (dairy or meat).

Mathematical models of environmental and biological behaviour of radionuclides are often formulated as a system of ordinary differential equations with constant coefficients. A dynamic compartmental model is proposed to describe mathematically the radionuclides transfer to the food chain and predict the activity concentration within each sub-compartment following an initial radionuclide deposition. The model output represents the time variation for the considered radionuclide in the *pasture-cow-milk* exposure route and also its distribution within the cow sub-compartments giving the time-dependent concentration of the radionuclides in each compartment or sub-compartment.

## 2. Methods and results

### 2.1.1. Description of the model

The conceptual model is defined as a series of compartments describing the radionuclides distribution in the compartments, the retention and elimination from the system. The conception of the model is based in first-order transfer rates from one compartment to the next using simple mass balance and rate equations. The transfer rates between the compartments represent the fraction of activity in a compartment that is transferred to another per unit of time. The dynamic model is expressed as a system of differential equations, in which radioactive decay and transfer among components are represented as linear processes.

The generic model may be presented as two different approaches: considering the organism as a global compartment or alternatively considering the radionuclide distribution within the cow. The first approach describes the radionuclide transport through the food chain considering as an initial state a contaminated pasture that is consumed by a cow. The starting concentration in pasture and soil compartment can be specified for each radionuclide. For the second approach a more refined model is described taking into account the spread of radionuclides within the cow by including the sub-compartments related to the organs involved in the distribution. Each sub-compartment represents internal organs (tissues) of the organism and the biokinetic transfer material between compartments is represented by first order processes. The transfer coefficients for the sub-compartments within the cow are combined with the respective biological half-lives.

For the cases where the biokinetic of the progeny are different from those of the parent ( $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ), the resulting conceptual model is different as it will also originate different doses from those obtained in the case where the biokinetic of the progeny are assumed to be the same as those of the parent.

In the first approach the generic processes involved in the radionuclide transfer through the *pasture-cow-milk* exposure route, considering ingestion as the only intake are: i) pasture deposition; ii) deposition on the soil; iii) retention of radionuclides by pasture over a certain period of time; iv) root uptake; v) consumption of contaminated pasture by the cow and vi) the secretion of radionuclides into the milk [1]. A scheme of the conceptual model is given in FIG. 1.

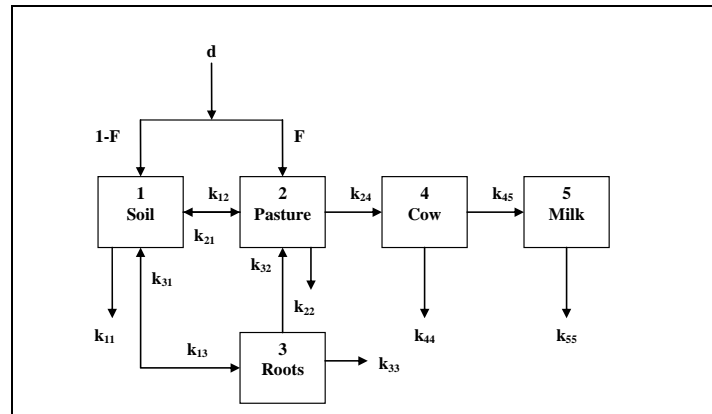


FIG. 1: Conceptual scheme of the pasture-cow-milk exposure route [1].

The total deposition is represented by  $d$  ( $\text{Bq}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ),  $F$  is the interception factor (dimensionless) defined as the fraction of the activity deposited on the ground (soil and pasture) which is intercepted by vegetation during the time of deposition,  $k_{ii}$  are the losses from compartment  $i$  ( $\text{d}^{-1}$ ) and  $k_{ij}$  are the kinetic transfer from compartment  $i$  to compartment  $j$  ( $\text{d}^{-1}$ ) [1].

The losses from the compartment 1 are due to radioactive decay and environmental processes related to migration in soil ( $k_{11}$ ,  $\text{d}^{-1}$ ). For all other compartments the losses considered ( $k_{ii}$ ,  $\text{d}^{-1}$ ) are only due to radioactive decay. The transfer between the compartments results from the resuspension of radioactive

particles from soil and subsequent deposition onto pasture surface ( $k_{12}$ ,  $d^{-1}$ ), root uptake ( $k_{13}$ ,  $d^{-1}$ ), radionuclides weathering from pasture surface ( $k_{21}$ ,  $d^{-1}$ ), ingestion of contaminated pasture ( $k_{24}$ ,  $d^{-1}$ ), translocation from pasture roots to the interior of the grass or to other edible parts ( $k_{32}$ ,  $d^{-1}$ ), transfer from pasture roots to soil ( $k_{31}$ ,  $d^{-1}$ ), considering that root uptake is a reversible process [2], and the segregation into cow's milk ( $k_{45}$ ,  $d^{-1}$ ).

The resulting equations system was written in the standard matrix form used in the space state approach and solved numerically using Matlab. The continuous time models are composed by the state vector and by the input and output vectors. The outputs of the system are quantities that can be measured or observed and the output vector is a linear combination of the state and of the input, often called the observation vector that is represented by the observation equation.

This simple model was already applied to  $^{226}\text{Ra}$  and it is being published elsewhere [1]. For the purpose of this work the simple model was applied to  $^{210}\text{Pb}$ . Although  $^{210}\text{Pb}$  is also excreted through cow's milk, levels in milk are variable and usually low because ruminants absorb less than 3 % of the ingested amount [3]. Lead-210 is included in the bone-volume-seeking radionuclides group among Sr, Ba, and Ra and its behaviour inside the organism is closely related to these radionuclides. The complete model could also be applied to  $^{210}\text{Pb}$  but  $^{226}\text{Ra}$  was chosen for a different example.

Following the second approach, the conceptual model describing the radionuclide transfer within the cow was adapted from the International Commission on Radiological Protection (ICRP) biokinetic models [4]. In case of  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$ , the biokinetic model uses the structure of the alkaline earth model: it describes the kinetic in bones, which is the main site of deposition and retention, and also considers retention in blood and other soft tissues, as well as routes of excretion. For the given example,  $^{226}\text{Ra}$  distribution within the cow is modelled by including three additional sub-compartments: the gastrointestinal system (GIT), the plasma and the bones [5]. Two compartments are considered for bones with different retention time. The scheme for the conceptual model describing the radionuclide transfer is represented in FIG. 2.

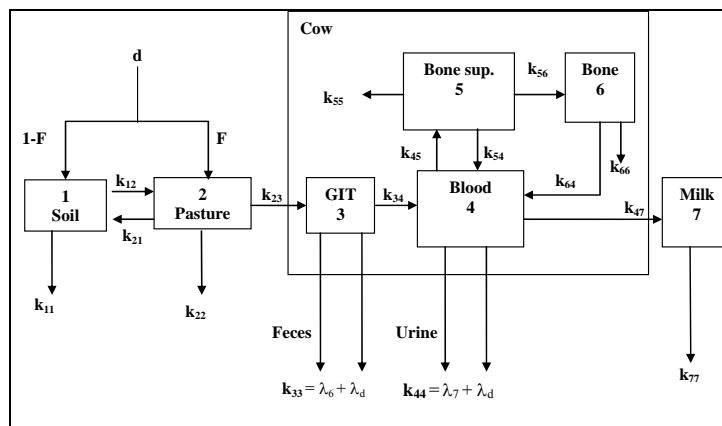


FIG. 2: Conceptual model for  $^{226}\text{Ra}$  kinetics in the cow [5].

The system of linear equations for this conceptual model is obtained as previous by defining a transient mass balance equation for each compartment only now the conceptual model is defined by seven compartments which will result in a system with seven differential equations.

Polonium deposition from the air is rather more expressive than uptake from the soil. Following entry into the transfer compartment (GIT),  $^{210}\text{Po}$  is taken to be distributed to the liver, kidneys, red bone marrow, spleen and all other tissues. For the activity lost to excretion the greater part is assumed to be lost to faeces. Polonium is also secreted in the milk of lactating animals being the concentration in milk a function of time after intake [3]. Polonium-210 long-term toxicity is about five times that of radium.

To model  $^{210}\text{Po}$  transfer processes described in the ICRP biokinetic model, a conceptual model with fourteen compartments was defined. The scheme for the conceptual model is represented in FIG. 3.

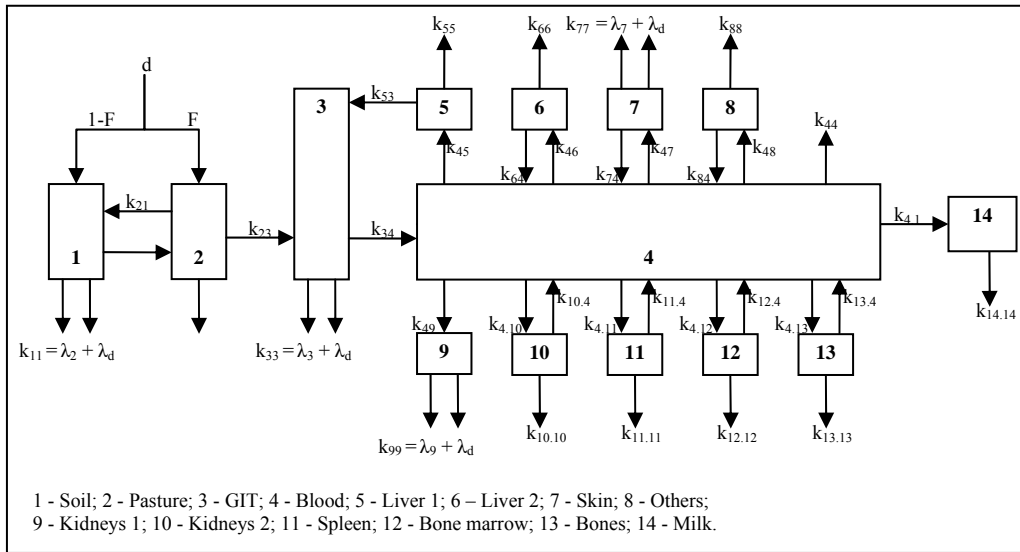


FIG. 3: Conceptual model for  $^{210}\text{Po}$  kinetics in the cow (adapted) [6].

To transcribe mathematically the conceptual model it is necessary to establish a differential equation for each one of the compartments which will result in a system with fourteen differential equations as it is necessary to define the same number of differential equations as the number of compartments.

### 2.1.2. Example of model simulation

In the simpler model the processes involved in radionuclides transfer to pasture are deposition, resuspension and root uptake. Any direct contamination of the pasture surface will dominate the activity concentration in the compartments. Processes such as root uptake and direct soil contamination will only be important when there is no direct deposition onto the pasture. Deposition of contaminants resuspended from the soil surface and subsequently deposited on plant surfaces is considered under resuspension and is relevant in these scenarios.

Model parameters were adopted from several different sources: some parameters were adopted from measurements referring to a particular contaminated site [7] and others were adopted from published data on radionuclide behaviour in animals, such as distribution or retention in different organs and tissues and subsequent excretion routes [4]. The unknown parameters were estimated from available data.

An initial deposition of  $1 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  was considered as an input in a hypothetical case study for different site conditions. Pasture contamination results from the deposition fraction that is intercepted by grass: a default value of 0,42 was adopted for the interception fraction (F) [8]. An initial activity value was assumed for the two first compartments: soil and pasture. For the other two compartments the initial activity was considered to be inexistent.

The weathering and decay constant accounts for removal of deposited material from plant surfaces as a result of weathering processes and radioactive decay. The constant is estimated with the weathering half-life which is the time required for half of the originally deposited material to be lost from the plant. A default value of 14 days was adopted for all radionuclides which is consistent with literature values [2]. The value for the kinetic constant representing this transfer process ( $k_{21}$ ) is  $0,495 \text{ d}^{-1}$ . The radionuclide fraction removed by radioactive decay ( $k_{22}$ ) is  $8,63 \times 10^{-5}$  for  $^{210}\text{Pb}$ ;  $1,17 \times 10^{-6} \text{ d}^{-1}$  for  $^{226}\text{Ra}$  and  $0,05 \text{ d}^{-1}$  for  $^{210}\text{Po}$ .

The rate of contaminant removal from soil is usually equated with the radioactive decay constant and a loss from the soil compartment based on a soil migration half-life. The default value adopted from literature is  $2,5 \times 10^4$  days [8] and the value for the kinetic constant ( $k_{11}$ ) which represents the radionuclide decay losses and the losses due to its migration in soil is  $1,13 \times 10^{-4} \text{ d}^{-1}$  for  $^{210}\text{Pb}$ ;  $2,82 \times 10^{-5} \text{ d}^{-1}$  for  $^{226}\text{Ra}$  and  $0,00503 \text{ d}^{-1}$  for  $^{210}\text{Po}$ . Resuspended radioactive particles from soil with later deposition on grass surface ( $k_{12}$ ) follow the resuspension factor approach and it has a value of  $0,016 \text{ d}^{-1}$  for all the radionuclides in this study.

Available data on root uptake is in the form of a transfer factor defined as the ratio of activity concentration in the edible part of the plant (Bq/kg fresh mass) to that in the soil (Bq/kg dry mass), once equilibrium has been reached. To model this process, a pair of transfer rates between the soil and the roots compartments was used considering that root uptake is a reversible process described with first order kinetic equations. An equilibrium rate was set to one day and the resulting value for  $k_{31}$  is  $0,693 \text{ d}^{-1}$ . The soil-plant transfer factor ( $k_{13}$ ) suggested in literature for pasture is  $0,05 \text{ d}^{-1}$  for  $^{210}\text{Pb}$ ;  $0,08 \text{ d}^{-1}$  for  $^{226}\text{Ra}$  and  $0,023 \text{ d}^{-1}$  for  $^{210}\text{Po}$  [5], [7]. A default value of  $1 \text{ d}^{-1}$  was considered for the radium translocation rate ( $k_{32}$ ) from roots to the edible parts of the grass. For  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  a translocation factor of 0,01 was used based in the assumption that root uptake and translocation of these radionuclides are less than 1 %.

The endpoints are  $^{210}\text{Pb}$  concentrations in soil, pasture, cow (whole body) and milk, in the simpler model. The results of the model calculations for  $^{210}\text{Pb}$  activity can be seen in the FIG. 4.

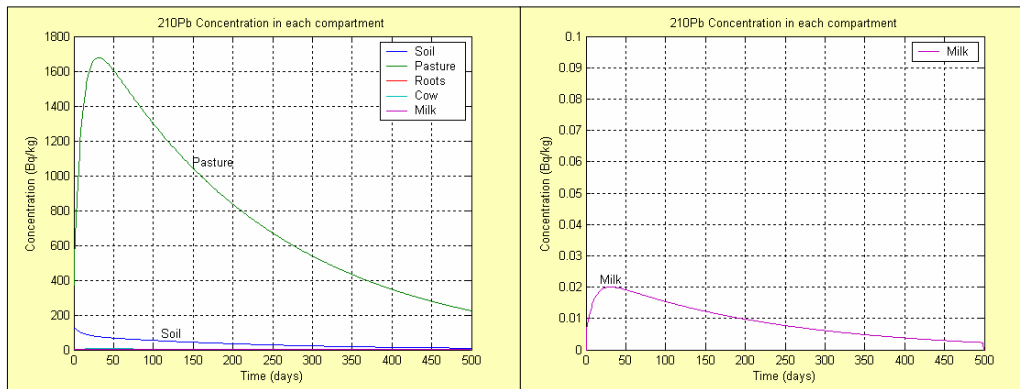


FIG. 4: Time variation of  $^{210}\text{Pb}$  concentration, Bq/kg.

For the complete model root uptake was not included for model simplicity and considering the fact that this transfer pathway is not relevant when the pasture contamination is primary due to deposition. Radium transfer rates within the cow were selected from specialized literature. The  $^{226}\text{Ra}$  and  $^{210}\text{Po}$  concentration may be calculate for all sub-compartments, although we are specially concerned with the concentration in milk. The results can be seen in FIG. 5 and FIG. 6.

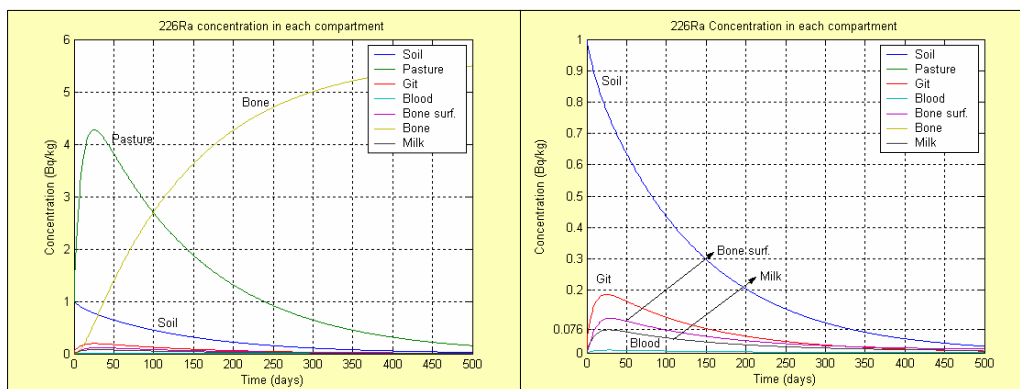


FIG. 5: Time variation of  $^{226}\text{Ra}$  concentration, Bq/kg.

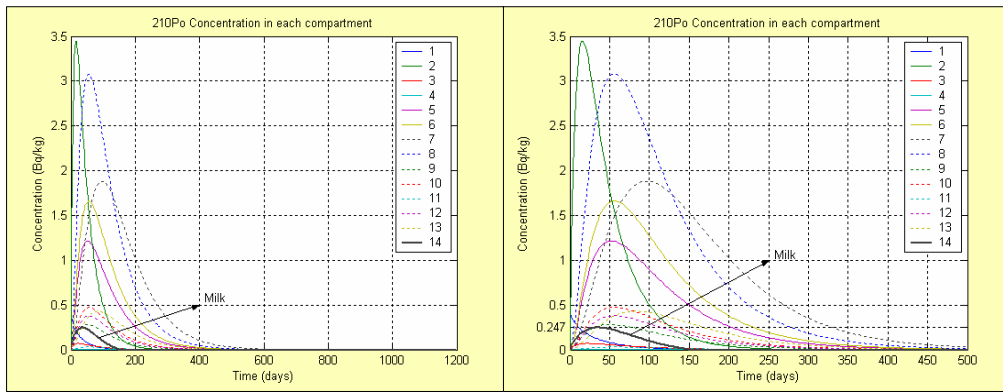


FIG. 6: Time variation of  $^{210}\text{Po}$  concentration, Bq/kg.

### 2.1.3. Results – model performance

The model output represents the time variation for the considered radionuclide in the *pasture-cow-milk* exposure route and also its distribution within the cow. The general tendency is a slow decrease of radionuclides content in each compartment following the maximum value achieved within 30 days. After one year achieving the maximum value,  $^{210}\text{Pb}$  content in milk decreases about 77,5 % (0,0045 Bq/kg);  $^{226}\text{Ra}$  content decreases about 85 % (0,011 Bq/kg) and  $^{210}\text{Po}$  content is null (0 Bq/kg).

Table 1. Comparison between the model simulation results and reference values

Radionuclide	Maximum Concentration (Bq/kg)	Time (d)	Average concentration $\pm$ Std (Bq/kg)	Reference values (Bq/kg) [5], [6]
$^{210}\text{Pb}$	0,020	30	0,009 $\pm$ 0,005	0,007 - 0,023
$^{226}\text{Ra}$	0,076	28	0,029 $\pm$ 0,02	0,008 - 0,036
$^{210}\text{Po}$	0,247	30	0,017 $\pm$ 0,05	0,008 - 0,122

### 3. Conclusions

A multi-compartmental model is presented to predict the activity concentration in the *pasture-cow-milk* exposure route. A more complete model describes the radionuclide distribution, retention and elimination within the cow. The main endpoint is the cow's milk. The aim of the model is to evaluate whether this exposure pathway is relevant in radiological terms; it also allows verifying if more detailed investigations are needed in order to evaluate possible exposure consequences in a specific site for a critical group. Simulation results fit within the range of values given in the literature although many assumptions had to be done due to the limited available data; conservative parameters had to be used in some cases. A more comprehensive site characterisation may improve the model performance but the experimental data will always and only represent a limited set of environmental conditions. Further work is needed for transfer models to animals and for the sensitive parameters related with the distribution in the compartments, retention and elimination from the system.

### ACKNOWLEDGEMENTS

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