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Radioecological vulnerability assessment and guideline levels for terrestrial foodchain

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1. Introduction

The release of radionuclides to the environment from nuclear facilities during normal operation or during an unplanned situation leads potentially to radiation exposure of the population and biota. Radionuclides may enter the terrestrial environment via atmospheric releases or releases to aquatic systems not only from nuclear installations but any kind of applications of radioisotopes might be a potential source even the terrorism might bring some threat. Radioactivity released to and dispersed in the atmosphere enters the terrestrial environment due to dry and wet deposition on soil and vegetation causing dose either for human being and biota. Internal exposures to human occur from the use of contaminated plants as food or as feed for domestic animals. Direct contamination of foliage also occurs as well as uptake of radionuclides from soil and deposition of resuspended contaminated soil particles.

The objective of any environmental survey that involves modelling and monitoring might be classified as:

- description and characterisation of the state of the environment,
- prediction of the condition or state of the environment as a result of an activity,
- detection of change and design and assessment of monitoring.

Model building is a kind of creative activity bringing together theoretical considerations, experiment and observation. Visualisation of the model structure and identification of the processes, variables, and parameters utilises current scientific knowledge. Variable or a factor means one of the attributes of interest, parameter means a quantity which might alter in a definite range. The choice of model is determined by available information, knowledge and applicability to the particular context of interest, all may involve combining available environmental information of different origin and this also introduces further challenges in that the information may have been collected under very different protocols and may have widely varying properties. Experimental data regardless they are the results of the laboratory experiment or field studies are used as aid for model design and selection, in parameter estimation and model evaluation.

A good model can be described in many ways, it should be simple, realistic, efficient, valid, reliable, accurate, precise and reproducible. Demonstration that a model is good might be difficult, but always based on the output of a model evaluation, which means studying the criteria being realised. Calibration and validation are two important procedures that are part of the evaluation process. In calibration, we attempt to find parameter estimates that provide acceptable agreement between model predictions and an independent data set. However, there continues to be some debate about model validity, the terminology used and its significance in environmental modelling. One further criterion of particular interest and one often used in environmental radioactivity modelling is the reliability of the model, considering the uncertainty associated with the model predictions (uncertainty about the model quantities, about the model form, about the model completeness), main factors contributing to the uncertainty and acceptability of this uncertainty for the decision making.

In principle two types of terrestrial models can be distinguished, equilibrium models and dynamic models. Equilibrium models are applied for evaluation of radiological consequences of continuous or quasi-continuous releases. They are applied for example to assess exposures during the normal operation of nuclear facilities or the result of weapons' fallout. Under these circumstances, after a certain time the activity concentrations in the environmental media are on average more or less constant. They are appropriate to estimate long-term averages for activities in soil, plants, animal products and activity intakes and dose contribution to human.

Especially after the reactor accident in Chernobyl, radioecology focused on the transfer of radionuclides in the environment subsequent to single or short-term depositions. In this case, the activity concentrations in environmental media can vary drastically with time and equilibrium or quasi-equilibrium in environmental compartments is not achieved. For simulating the behaviour of radionuclides in the environment, the radioactivity-concentrations in the environmental compartments have to be formulated as time-dependent functions, dynamic models.

Figure 1 might help understanding the subject of radioecology, explaining the essential need of continuous developing of an interdisciplinal, complex science. During the development a good model it is essential to emphasize the equal importance of the observations (measurements), theoretical considerations and the mathematical possibilities of description.

The knowledge that was gained during the last fifty years was put to good use, in developing a methodology for radioecological risk assessment and management, which was later transposed to the problems of chemical pollution.

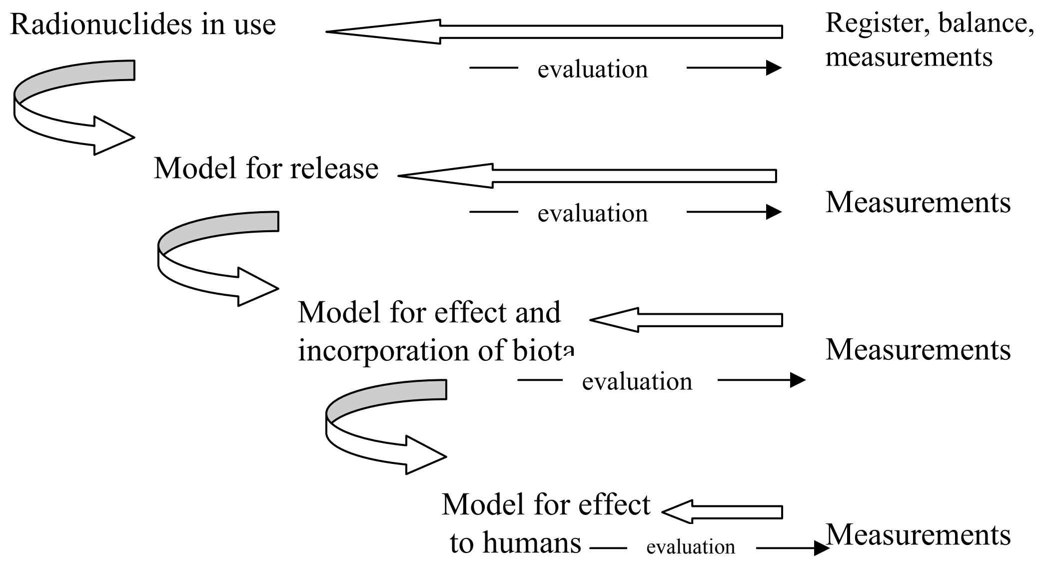

Figure 1. The method of consideration of the effect of radionuclides from getting into environment through foodchain to incorporating of human being, the interactions between measurements and mathematical descriptions

2. Goals

The general aim of the radioecology to provide the scientific and technical knowledge needed to achieve the dual objective:

1. limiting exposure of humans and other biota to radioactivity under all circumstances;
2. post accident management.

I would like to deal with both of the area mentioned above:

1. limiting exposure of humans to radioactivity regarding the ingestion pathway, that is the regular consumption of foodstuffs under normal circumstances and
2. dealing with the same pathway in the prolonged emergency period (longer than one year), when the economical conditions are not limiting factors, just the protection considerations could be taken into account.

I deal with the monitoring of radioactivity in foodstuffs and agricultural environment for more than fifteen years, so I would like to involve into this work the experience gained during the everyday work and draw conclusions for the sake of the improvement of the quality of the monitoring. I intend to evaluate countrywide monitoring data from radioecological point of view, which we have not done in the annual reports.

I would like to deal with problems of so called agricultural radioecology connected with radionuclide turnover in the agrosphere, ionising radiation effects on agricultural plants and animals, transfer processes of radioisotopes regarding foodchain, vulnerable levels and areas of agroecosystems, and laying down the basis of the legislation that makes possible to obtain non-contaminated agricultural products meeting the general radiological protection standards.

Above all the primary goal of agricultural production is providing safe and enjoyable food for the whole population of the World. My goal is to provide the scientific base of the implementation a regulation which gives the right to just one authority dealing with the foodstuffs for the sake of the effectiveness of the necessary measures, the immediate withdrawal of the foodstuffs or feedstuffs in case of non-compliance. Legislation in force in European Union is a follow up type now and deals with only the periods after emergency or contains indirect dose limit for the population. I would like to provide derived limits for radioactive isotope content for foodstuff and feedstuff for the direct and quick applicability under normal circumstances and prolonged emergency period with the introduction of the upper limit of the background as a base and tolerance and acceptance levels.

3. Procedures and Methods

3.1. Radioecology

Radioecology, the science of the relationship between radioactivity and living things in the environment. This discipline is particularly concerned to encourage the co-operation and the exchange of the information between scientists dealing with many kind of sciences, like Meteorology, Biogeochemistry, Geochemistry, Geophysics, Geography, Hydrology, Physics, Mathematics, Chemistry, Biology, Biochemistry, Cytology, Genetics, Cellular biology and so on. Nowadays following branches of Radioecology might be identified: theoretical and general radioecology, agricultural radioecology, animal radioecology, forest radioecology, marine and freshwater radioecology and finally radioecology of the main natural ecosystems (medows, steppes, tundra, swamp…) The radioecology part of methods is compiled book of Kanyár et al (Kanyár et al, 1996), the book edited by Scott (Scott, 2003) and the book edited by Stricht and Kirchman (Stricht and Kirchman, 2001).

In principle two types of terrestrial models can be distinguished, equilibrium models and dynamic models. Main advantage of the dynamic models is that they take into account the time-dependence. Equilibrium models are applied for evaluation of radiological consequences of continuous or quasi-continuous releases. They are based on two fundamental assumptions: emission rates of the radionuclides are constant in time and duration of releases is long compared to the times needed for radionuclide transfer along the environmental pathway considered. They are applied for example to assess exposures associated the normal operation of nuclear facilities or that of weapons' fallout. Under these circumstances, after a certain time the activity concentrations in the environmental media are on average more or less constant. They are appropriate to estimate long-term averages for activities in soil, plants, animal products and activity intakes and doses to human. The basic approach used by equilibrium models is likely to give conservative exposure estimates. This type of radioecological model is therefore generally used to establish the compliance of routine releases from nuclear facilities with exposure limits.

Most of the models that available to simulate radionuclide distribution and transport in the environment are deterministic models. This means that appropriate point estimates are selected for all model parameters and a single set of results is obtained. I followed this approach in case of evaluation of the results of Pu survey and I would like to follow also during the evaluation of monitoring results regarding ^{137}Cs and ^{90}Sr.

There are three main descriptive and predictive methods for radionuclide transfer in the environment: application of concentration rations, kinetic methods and allometric approach. All of them have advantages and drawbacks. One significant limitation of either the concentration ratio or transfer coefficient approach is that neither can be extrapolated to other systems because they are largely based on empirical observations. On the other side the mentioned fact might be advantageous as well, deriving site specific data for the predictions, taking into account bioavailability and stable isotope or analogue intake.

Bioconcentration factors are integral parts of radioecological risk assessments. They are used to predict transfer from a typically abiotic component of the environment to a biotic one. Usually they are empirically obtained through the field or laboratory studies or extrapolated from the behaviour of chemical analogues. The problem with this aspect is that data exist for several radionuclides in selected environments and transfer processes, data are site specific, and values can range over many orders of magnitude for a given radionuclide. Sometimes useful information gets lost when concentration ratios are compiled, for example:

- equilibrium conditions are supposed, not always totally realised,
- effect of competing element is not always negligible,
- sometimes only a part of the species, sometimes the whole are taken into account,
- some important detail of sample preparation for measurement is not always described (washed, peeled, skinned…).

One of the drawbacks is at the same time the main advantage of this approach like above mentioned, once a certain kind of transfer is specified for a given radionuclide, site and process, the validity of the value is not questionable and could be used as a reliable parameter for the prediction. The generalisation is the main problem, if methods were developed which provide technically proven values for a wide range of species and environmental conditions they would represent a considerable enhancement in radioecological risk assessment.

During the possible contamination of plants the following processes are usually taken into account:

- In many assessment models the <u>dry deposition</u> of radionuclides from the atmosphere to soil and vegetation is quantified by the deposition velocity, which depends on a variety of factors, such as the particle size, the characteristics of the interface surface and air, the meteorological conditions and chemical form of the deposit. Sedimentation is quantified by the Stokes-formula:

$$v_s = \frac{mg}{6\pi\, r\eta}, \qquad \text{Eq 3.1}$$

m	mass of the particle, kg
g	gravitational acceleration, 9.81 m/s^2
r	radius of the particle, m
η	viscosity, at 20^{o}C 1.815E-5 Ns/m^2

Velocity of the deposition is:

$$v_g = \frac{Q_d}{c}, \qquad \text{Eq 3.2}$$

Q_d	deposited material, kg/sm^2
c	activity concentration in air near surface, Bq/kg

The real velocity will be the following:

$$v_d = \frac{v_s}{1 - \exp\left(-\frac{v_s}{v_g}\right)} \qquad \text{Eq 3.3}$$

For particles the minimum deposition velocity was observed for diameters 0.1-1 μm, namely 0.001 m/s, below this size deposition of particles are more effective due to diffusion. Above 1 μm deposition increases with increasing particle size, because of gravitational settling and impaction (coagulation and attachment to larger particles) becomes more important, these particles are lost from the near-surface atmosphere due to sedimentation. The particle size spectrum is influenced by the source, for example routine releases are mainly filtered and cut-off point of the filter device determines the maximum particle size. Unplanned releases might contain the whole particle size spectrum, however with increasing distance from the release point the size distribution of the released particles approaches to the natural aerosols which are in the range of 0.1-1 μm. The deposition is more effective for well-developed plant canopies, since the area of interface between vegetation and atmosphere is increased. The deposition processes might be simulated in analogy to Ohm`s law, this differentiates among the aerodynamic (flux of the substance considered from the atmosphere to laminar layer of the surface), the surface layer (transport within the laminar layer: Brownian movement, impaction on the surface, sedimentation) and the transfer resistance (interaction of the deposit and the interface: chemical process, assimilation, uptake).

- Radioactivity in air is washed out by rain and snow. The vegetation retains some fraction of radionuclides deposited with precipitation, and the rest falls through the canopy to the ground, the fraction that is initially intercepted is a very important quantity in all radioecological models, due to the fact that direct deposition may cause relatively high activity concentrations in feed and food.

Quantification of the process is the following:

$$\frac{dQ'}{dt} = -\Lambda Q' \qquad \text{or} \qquad Q' = Q_0 e^{-\Lambda t} \qquad \text{Eq 3.4}$$

Q' remaining activity in cloud, Bq

Λ washout, 1/s

t duration of raining, s

- Soil is the main reservoir for long-lived radionuclides deposited on terrestrial ecosystems. The behaviour of radionuclides in soil controls their migration in the soil, the possible transport to the groundwater and the long-term radionuclide concentration in vegetation. Uptake of radionuclides takes place via the soil solution, as for all minerals. Complex physico-chemical interactions with soil matrix, where ion exchange is the dominant mechanism determine the radionuclide concentration in soil solution. While ion-exchange has a competitive mechanism, the concentration and composition of major elements, particularly those which are regarded competitive are of primary importance in determining the distribution of radionuclides among soil, soil solution and plant roots. For example the amount and nature of clay minerals present in soil are key factors in determining their radioecological sensitivity with regard to concentrations of radiocaesium. Sorption and desorption of radionuclides and their possible uptake by plants are the result of the complex interaction of soil type, pH, redox potential, sorption capacity, clay content, organic matter and land management practice if relevant. Although these factors are known they are difficult either to quantify or to integrate in a universal model applicable to a wide range of soil conditions. The decrease of transfer factors in the order sand > loam > clay is observed in general, especially for cations. There are some common simplifying agreement in the field of calculation of concentration rations: generally the root zone is regarded the upper 20 cm layer of the soil, the total concentration is taken into account without speciation or chemical form, the aggregated transfer factors are the total deposited amount of activity to a unit area, sometimes root uptake is taken into account radionuclides having more than one year of half life.

The flux from soil to plant depends on the water transport and the concentration gradient in the root zone, the simplified form is the Michaelis-Menten equation:

$$F = \left(\frac{F_{\max}}{k_m + c_a} \right) c_a , \qquad \text{Eq 3.5}$$

F	flux trough the surface of the root, Bq/cm^2,
c_a	concentration in root – soil layer, Bq/kg
F_{max}	maximum of the flux, Bq/cm^2
k_m	Michaelis rate constant, Bq/kg .

When $c_a \ll k_m$, then: $F=\alpha c_a$, where α so called root sorption factor, cm/s. Determination of the parameters for the flux is rather difficult, but with contamination experiences is possible (Oncsik et al 2002).

- The interception fraction is often assumed to be constant for all elements, all plants, and all precipitation events, however, investigations in recent years have demonstrated that interception of wet deposited radionuclides is actually much more complex. According to the work of Müller and Pröhl (Müller and Pröhl 1993) the differences among plants were of minor importance compared with the variations among the radionuclides, the interception of polyvalent cations being higher as much as a factor of 8 than for anions (iodine or sulphate). The element dependent retention factor describes the ability of the element to be fixed on the leaves and it is mainly controlled by the chemical form, since the plant surface is negatively charged, the absorption of anions is less effective than cations. The research of the last years proved that the intercepted part increasing with the surface of the leaf and decreasing with the amount of rain:

$$f_w = \left(1; \frac{LAI \cdot k \cdot S \cdot \left(1 - e^{-\frac{\ln 2}{3S} R} \right)}{R} \right) \qquad \text{Eq 3.6}$$

f_w	interception fraction,
LAI	leaf area index, m^2/m^2
k	chemical element specific constant,
S	the rain-retaining ability of the surface, mm
R	amount of rain, mm

- Once deposited on the vegetation, radionuclides are lost from plants due to removal by wind and rain, either through leaching or by cuticular abrasion. The increase of biomass during growth does not cause a loss of activity, however, it does lead to a decrease in activity concentration due to growth dilution. The decreasing rate depends on the season, for example for well managed pasture in middle Europe, the mean daily increase in biomass varies from 10 g dry/(day m^2) in May to 2 g dry/(day m^2) in October. Some parts of the growth rates might be described and are equivalent to half times in the order of 10-50 days. These processes occur simultaneously, the exact contribution of the different processes are difficult to quantify, therefore the total loss is expressed by the weathering half time, which is empirically determined by regression of the radionuclide concentration in vegetation with time. The analysis of observations after Chernobyl accident resulted in weathering half times for iodine 8 days, caesium 10 days. In general, longer half times are observed for slowly growing or dormant vegetation.
- <u>Resuspension</u> is the process when the activity once deposited get back to the near surface atmosphere with the aid of any process like wind, traffic, cultivation processes, this means the redistribution of the activity close to the surface. Being a very complex process empirical estimation is used generally by resuspension factor:

$$K = \frac{c_{air}}{c_{surface}}, \qquad \text{Eq 3.7}$$

K resuspension factor, 1/m,

c_{air} activity concentration in air, Bq/m^3,

$c_{surface}$ activity concentration in surface, Bq/m^2.

$$K = A \cdot e^{B \cdot t} + C, \qquad \text{Eq 3.8}$$

K resuspension factor, 1/m,

$A+C$ the original amount of the resuspension, C is valid for long term a (constant), 1/m,

B velocity of decreasing, 1/day.

This process is influenced by many factors, like type of the soil, soil-texture, moisture content of the soil, quality of the plants grown there, density of plants, strength of the wind. The resuspension factor can be described by time dependent expression in Eq 3.7, for A+C original amount was found 4-50E-9 1/m in Europe after Chernobyl accident (Scott, 2003). For the B half life analogue quantity is 10-

800 day with the median of 1 year. This process has big importance in case of actinides. When the resuspension factor reaches 1E-7 1/m, this process became the determining one, in dry conditions it may be 1E-5 1/m.

- Translocation between the parts of a plants or systemic transport of radionuclides has little influence on the long-term fate of radioactivity in the environment, but it is a key issue when estimating the radionuclide content of food, especially in case of foodstuff used partly like cereals, for plants such as leafy vegetables are used as a whole translocation is not so relevant process. There are two transport systems in the plant, the xylem and pholem, first one transports water and minerals from the soil to plant parts, driving force is the transpiration, transport direction is from the bottom to the top of the plant, there is no discrimination among elements to be transported once they have reached the system. The main function of the other system, pholem is to transport carbohydrates from the leaves, transport is possible in it in both upward and downward directions, and transport is element specific: alkali elements, magnesium, sulphur, chlorine and phosphate are transported; alkaline earth metals, lead, plutonium, lanthanides and actinides are not. The amount of translocated activity is highly dependent on the life-stage of development of the examined plant at which the deposition occurs.

Animals can be contaminated by the three different routes, by inhalation, through the skin and by ingestion of contaminated feed. Skin lesions may provide a direct entry into animal's circulation system, but these processes will not considered here. Inhalation is potentially more important, because the lung surfaces are more permeable to a wide range of elements. The most important pathway to animals is the ingestion of contaminated feed, water or soil. The ingestion of feed and processes influencing absorption and retention determine the radionuclide content of animals. The degree of absorption from the gastro-intestinal tract is the most important factor of in determining the radionuclide content of animal tissues. The radionuclide uptake of animals supposing the equilibrium conditions, equations are described by dependence of the daily radionuclide uptake. This kind of description is valid only for adult animals and steady-state condition. Taking into account the time-dependence one or two biological half life is introduced for the absorption and for excretion, the following estimates the radionuclide content in animal tissue, or food of animal origin.

$$C_m(T) = TF_m \sum_{j=1}^{J} \left\{ a_{mj} \cdot \int_0^T A_{a,m}(t) \cdot \lambda_{b,mj} \cdot \exp\left[-\left(\lambda_{b,mj} + \lambda_r\right)(T-t)\right] dt \right\}, \qquad \text{Eq 3.9}$$

C_m	activity concentration in *m* animal tissue, in *T* time, Bq/kg,
TF_m	transfer coefficient of *m* animal tissue, day/kg,
J	number of biological transfer factors,
a_{mj}	the ratio of *j* biological transfer,
$A_{a,m}(t)$	radionuclide uptake of the animal, Bq/day,
$\lambda_{b,mj}$	the biological transfer rate of *j* for *m* animal tissue, 1/day.

Sometimes more complex compartment models are used to describe the kinetics of radionuclides in animals by a set of differential equations to simulate additionally the recycling of radionuclides between the organs. Such approach requires numerous transfer rates to quantify the activity exchange among the compartments. The absorption, excretion and transfer between tissues are described by transfers between model compartments; the model requires the solution of a set of simultaneous first order differential equations with general form:

$$\frac{dN_i}{dt} = \sum_{\substack{j=1 \\ j \neq i}}^{m} a_{ji} N_j - \sum_{\substack{j=1 \\ j \neq i}}^{m} a_{ij} N_j \qquad \text{Eq 3.10}$$

N_i	activity of compartment i, Bq,
a_{ij}	the rate coefficient for the transfer from compartment i to j, 1/day
t	time, day.

This type of approach is more sophisticated, the structure of the model based on a simplified representation of biological processes, the parameterisation is empirically based on comparisons to observed data. Generally such models are implicitly calibrated to a particular situation and take no account of changing metabolic circumstances.

3.2. Different levels applied in the controlling of the radioactivity

For the explanation and comparison of the different levels IAEA Safety Series 115 (IAEA, 1996), IAEA-TECDOC-953 (IAEA, 1997a) and IAEA-TECDOC-955 (IAEA, 1997b), IAEA-SRS-44 (IAEA, 2005) ICRP 63 (ICRP, 1992), ICRP 64 (ICRP, 1993a), ICRP 65 (ICRP, 1993b), ICRP 75 (ICRP, 1977a), ICRP 77 (ICRP, 1977b), ICRP 81 (ICRP, 1998), ICRP 82 (ICRP, 1999), ICRP 96 (ICRP, 2005), were used. The order of the explanation gives the order of their extent.

First of all it is necessary to distinguish the levels and limits, because the limit is the value of quantity used in certain specified activities or circumstances that must not be exceeded. There are several levels in use in connection with radioactivity. The more general is reference level, which might be an action level, intervention level, investigation level or recording level. Such level may be established for any of the quantities determined in the practice of radiation protection.

The other level for rather general use is the action level, the level of dose rate or activity concentration above which remedial or protective actions should be carried out in chronic or emergency exposure situations. Generic action level is the level of activity concentration in food, milk or water above which remedial or protective actions should be carried out in chronic or emergency exposure situation.

Intervention level (generic intervention level) is the level of avertable dose at which a specific protective or remedial action is taken in an emergency exposure situation or a chronic exposure situation. For the aid of quick decision having of derived levels are essential, the more general is the operational intervention level which is a calculated value for ambient dose rate or for radionuclide concentration measured by instruments or determined by laboratory that correspond to the generic intervention level or generic action level.

Dangerous quantities usually used as D-values, the term cover the dangerous quantity of radioactive material, the source natural or artificial origin, which could, if not under control give rise to exposure sufficient to cause severe deterministic effect.

Exemption, exclusion and clearance level are derived activity concentrations, which are valid for all types of solid material containing radionuclides of artificial or natural origin except foodstuffs and drinking water. Exemption level is the activity concentration of all bulk amounts of solid material, it covers the removal from control of material containing very low levels of radioactivity originating from regulated practices, like nuclear installations, hospitals, research institutes, material from interventions, materials containing radionuclides of natural origin that need to be considered for exclusion. Exclusion levels are the activity concentration values for radionuclides of natural origin, the intention is to exclude from regulation virtually all soils, but to not ores, mineral sands, industrial residues and wastes which are recognized as having significant activity. The values of clearance level are established by Regulatory Authority and expressed in terms of activity concentration and/or total activity, at or below which sources of radiation may be released from regulatory control.

Guideline level is a specified quantity above which appropriate actions should be considered. In some circumstances, actions may need to be considered when the specified quantity is substantially below the guideline level.

Investigation level is the value of quantity such as effective dose, intake or contamination per unit area, weight or volume at or above which an investigation should be conducted.

Recording level is a level of dose, exposure or intake specified by the Regulatory Authority at or above which values of dose, exposure or intake has to be recorded (this term was created originally for the workers to keep the register about their individual dose)

Reporting level defined in 2000/473/Euratom is used for the forming the common European database about the environmental level of radioactivity including foodstuff and drinking water. Uniform reporting levels have been defined on the basis of their significance from an exposure point of view, irrespective of the detection limits applied by the different laboratories, as it is shown in Table 1.

Table 1: Reporting levels in the European Union

Sample type	**Radionuclide category**	**Reporting level**
Air	Gross beta (based on Sr-90)	5 E-03 Bq/m³
	Cs-137	3 E-02 Bq/m³
Surface water	Residual beta (based on Sr-90)	6 E-01 Bq/l
	Cs-137	1 E+00 Bq/l
Drinking water	H-3	1 E+02 Bq/l
	Sr-90	6 E-02 Bq/l
	Cs-137	1 E-01 Bq/l
Milk	Sr-90	2 E-01 Bq/l
	Cs-137	5 E-01 Bq/l
Mixed diet	Sr-90	1 E-01 Bq/d.p*
	Cs-137	2 E-01 Bq/d.p*
* Becquerel per person per day		

There are some useful derived levels, for example notification level related to inform the public by communicé, when discharge from nuclear installation reaches the observable level in the environment according the recommendation of Kanyár et al (B. Kanyár et al, 2008).

The decreasing order of derived levels of interest of this work is the following:
intervention > guideline > investigation > recording > reporting/actual level (in normal situation)

The Table 2 contains the comparison of levels and a series of useful values for practice.

Table 2: Comparison of levels for practice

Level with use	Values
Intervention levels	
Radiological emergencies	
Intervention for foodstuffs	10 mSv/year
Sheltering	50 mSv
Evacuation	500 mSv/day or for the duration of evacuation
Distribution of stable iodine	500 mSv thyroid
Relocation	1000 mSv
Radiological attack	
Sheltering	10 mSv in 2 days
Temporary evacuation	50 mSv in 1 week
Distribution of stable iodine	100 mSv thyroid
Relocation	1000 mSv or 100 mSv first year
Long-lived solid radioactive waste disposal	
Not likely to be justifiable	10 mSv/year
Almost always justifiable	100 mSv/year
Prolonged exposure situation	
Not likely to be justifiable	< 10 mSv/year
Almost always justifiable	> 100 mSv/year
Action level	
Naturally occurring radioactive materials	1-10 Bq/g (1-2 mSv/year)
Radon	200-600 in home 3-10 mSv/year 500-1500 Bq/m^3 for workers
Exclusion level	
for consumer goods	
Artificial alpha-emitters	0.01 Bq/g
Artificial beta-/gamma-emitters	0.1 Bq/g
Head of decay chain ^{238}U, ^{232}Th	1.0 Bq/g
Exemption level	
Practices under regulation	0.01 mSv/year
Practices other	0.01 mSv/year
Intervention – radioactive substances in commodities	1 mSv/year
Public exposure	
Radioactive waste disposal	< 0.3 mSv/year and < 1 mSv/year
Long-lived solid radioactive waste disposal	0.3 mSv/year
Prolonged exposure	0.3 mSv/year and < 1 mSv/year

3.3. Current legislation for the radioactive isotope content of foodstuffs

The compliance with requirements for application of dose limits on effective dose shall be determined by comparing the total effective dose with relevant dose limit calculated the following formula (IAEA, 1996):

$$E_T = H_p(d) + \sum_j e(g)_{j,ing} I_{j,ing} + \sum_j e(g)_{j,inh} I_{j,inh} \quad \text{Eq 3.10}$$

E_T	total effective dose, Sv/year,
Hp	dose from exposure to penetrating radiation, Sv/year
$e(g)_{j,inh}$	effective dose per unit intake from inhalation by the group of *g*, Sv
$e(g)_{j,ing}$	effective dose per unit intake from ingestion by the group of *g*, Sv
$I_{j,inh}$	intake via inhalation, Bq/year
$I_{j,inh}$	intake via ingestion, Bq/year

There was a study in Hungary in 1986 about the radioactivity content of foodstuffs and other agricultural products, they provided some derived limits used in controlling the foodstuffs, but the base was the dose of 1 mSv/year and these derived concentrations were joined to the average consumption in Hungary at that time (Fehér et al, 1986).

ICRP regularly reviews the increasing knowledge of the effects of exposure to ionising radiation in order to decide whether new recommendations are needed. The system of protection was summarised in ICRP Publication 60 in 1991 (ICRP, 1991). According new scientific data produced Supporting Guidance 5 (ICRP, 2007a) was published in 2007 and one of the latest publications serve as a basis of my consideration, namely the Publication 103 (ICRP, 2007b). However ICRP is an organisation of the experts without any legal basis their standpoint is impossible to leave out of consideration, sooner or later their commitments will be accepted by some kind of regulation.

The most important document in the field of nuclear and radiological safety is the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources, Safety Series No. 115 (IAEA, 1996) published by International Atomic Energy Agency (furthermore IAEA) in 1996. IAEA take care on the summarization of the worldwide knowledge regularly and publishes many documents in different fields related all nuclear and radiological field, so Safety Series 115 incorporate the opinion of Food and Agriculture Organisation of the United Nations, International Labour Organisation, Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, Pan American Health Organisation and last but not least World Health Organisation.

The Committee on Food Additives and Contaminants belonging to FAO-WHO Codex Alimentarius issues time to time revised versions of guideline levels for radionuclides in foodstuffs, which are recommended to apply mainly in international trade. The latest one was issued in 2006, it will be explained in detail.

Nowadays being one of the member states Hungary is obliged to apply the European Legislation. According to "Council Directive 96/29/Euratom of 13 May 1996 laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionising radiation" the general effective dose limit for members of the public shall be 1 mSv in a year (nationalization of this regulation is 16/2000. (VI. 8.) decree of Health Ministry). Hungary should keep the European regulations, recommendations and directives in evidence. All of the regulations of the European Community regarding the radioisotopes are found on the Euratom website (http://www.euratom.org).

The dose limit serving the basis of the calculation of the guidance level have to be understood as an additional dose to the site specific background level of the ingestion dose determined by the monitoring of foodstuffs. Neither exemption levels (IAEA, 1996) nor "D-values" (IAEA, 2006) are applicable for foodstuffs. D-value term cover the dangerous quantity of radioactive material, the source natural or artificial origin, which could, if not under control give rise to exposure sufficient to cause severe deterministic effect. The intentional ingestion of radioactive material and intentionally exposing oneself to a radioactive source are not involved in the scenarios. During the derivation of exemption or clearance levels different scenarios were taken into account, but all of them belong to the principle of emission, while appearance of radioactivity is clearly an immission situation from the point of view of this work.

All guideline regulations has to cover all agricultural products and processed agricultural products intended for human consumption and the determined guideline levels apply to food in a state of ready for consumption, not to concentrated or dried foods.

European legislations in the field of radionuclide content of foodstuffs are intended to use after an accident. First group of the legislations was introduced governing the situation after Chernobyl accident and deals with only with radiocaesium content of foodstuffs, like 737/90/EEC, 616/2000/EC, 1609/2000/EC, 1635/2006/EC, 2003/274/EC. These regulations define the conditions governing imports of agricultural products and processed agricultural products intended for human consumption originating in third countries which means further narrowing the field of applicability. 2000/473/Euratom recommendation requires the continuous monitoring of the environment and foodstuffs. 2003/274/EC recommendation requires the monitoring of radiocaesium in certain wild food products for example meat of wild game, different berries and mushrooms and it suggest taking also care of carnivorous freshwater fish from lakes. In summary these legislations define the “maximum permitted” levels of radiocaesium as following:

- 370 Bq/kg for milk and milk products and for foodstuffs intended for feeding of infants;
- 600 Bq/kg for all other products concerned;
- for minor foodstuffs the multiplication factor is 10.

There is no time limit given in these regulations, it means that the foodstuff just not exceeding the limit is accounted consumable therefore it may be placed on the market and kept it there and allowed for free circulation in the community.

Second group deals with the maximum permitted levels of radioactive contamination of foodstuffs and feedingstuffs following a nuclear accident or any other case of radiological emergency, 3954/87/Euratom, 2218/89/Euratom, 2219/89/Euratom, 944/89/Euratom, 770/90/Euratom. Foodstuffs or feedingstuffs not in compliance with the maximum permitted levels shall not be placed on the market either originating from inside of EU or in third countries. These regulations give maximum permitted levels for foodstuffs and feedingstuffs for more isotopes as Table 3 shows. Regarding the maximum permitted concentrations of radiocaesium these regulations allow even higher levels than the currently applied regulation for follow up of Chernobyl accident. I would like to draw the attention that according to current regulation there is 3 month time limit for the application of the given maximum permitted levels, but there is possibility for the application of longer period. It means that the foodstuff may be placed on the market and kept it there for a long time and may cause even a chronic exposure. There is no procedure established in the case when foodstuff contaminated with not only one radionuclide.

The mentioned levels apply to food after reconstruction or as prepared for consumption, not to dried or concentrated foods.

New guideline levels for radionuclides in foods were issued by Codex Alimentarius recently, under the name of CAC/GL 5-2006, Table 4. This is intended to use for foodstuffs contaminated following a nuclear or radiological emergency for use in international trade. The guideline levels are based on “an intervention exemption level of 1 mSv in a year”. There was used another important assumption, that the mean fraction of major foodstuffs quantities imported is 0.1. This regulation accepts that the national governments may wish to adopt different values for internal use within their own territory.

Table 3. Maximum permitted levels for foodstuffs and feedingstuffs in Bq/kg following a nuclear accident or any other case of radiological emergency (as it is in 2218/89/Euratom and 770/90/Euratom)

	Foodstuffs				Feedingstuffs
	Baby foods	Dairy produce	Other foodstuffs except minor foodstuffs	Liquid foodstuffs	
Isotopes of strontium, notably Sr-90	75	125	750	125	
Isotopes of iodine, notably I-131	150	500	2000	500	
Alpha emitting isotopes of plutonium and transplutonium elements, notably Pu-239, Am-241	1	20	80	20	
All other radionuclides of half life grater than 10 days, notably Cs-134, Cs-137	400	1000	1250	1000	Pig 1250 Poultry, lamb, calves 2500 Other 5000

Table 4. Guideline levels for radionuclides in foods according to CAC/GL 5-2006

Radionuclides in Foods	Guideline Level (Bq/kg)	
	Infant Foods*	Other Foods
^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am	1	10
^{90}Sr, ^{106}Ru, ^{129}I, ^{131}I, ^{235}U	100	100
^{35}S**, ^{60}Co, ^{89}Sr, ^{103}Ru, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{192}Ir	1000	1000
^{3}H***, ^{14}C, ^{99}Tc	1000	10000

* When intended for use as such
** This represents the value for organically bound sulphur
*** This represents the value for organically bound tritium

Even the groupings of the isotopes are not the same in 2218/89/Euratom and CAC/GL 5-2006, showing the lack of some consensus on international level. Isotopes of strontium especially ^{90}Sr and isotopes of iodine especially ^{131}I are regulated separately in the EU regulation with the ratio of 10 for foodstuffs for adults to foodstuffs for infants, while in CAC/GL 2006 are the same.

First of all it is necessary to lay down the fact that there is no limit for the normal condition for consumption of foodstuffs, namely when there is no announced emergency situation in force, no guideline level available even as a suggestion. European legislation as a follow-up measures uses nowadays the 370 Bq/kg for infants and 600 Bq/kg for adults regarding radiocaesium as consumption limit, which was originally designed for shorter time after the Chernobyl accident. Application of these limits means, that just below this value everything is legally placed on the market for free circulation inside the European Union, consequently it is possible that cause exposure for longer time, because there is no other limit in force. Using the consumption rate suggested by World Health Organisation 550 kg/year for adults and 200 kg/year for infants, and ingestion dose conversion factors (committed effective dose per unit intake via ingestion) from IAEA, Safety Series 115 (IAEA, 1996), the results are 4.3 mSv/year for adults and 1.6 mSv/year for infants, supposing the worst case when the contamination of all food consumed is equal with the limit during a year. My opinion is that these values are too high, they has to be considered as additional dose coming from only one source, from food consumption. Furthermore these results are not in line with the Council Directive 96/29/Euratom, they are higher than the suggested general dose limit for public exposure of 1 mSv. An additional prolonged exposure of more than 1 mSv/year from one source, may easily lead to an annual dose greater than 10 mSv if all kind of commodities are considered (ICRP Supporting Guidance 5).

3.4. Current legislation for feed

There are lot of regulations about the preparation and quality of different kind of feedingstuffs, among them several regulations dealing with radionuclides, these measures regulate mainly the import into the European Union, like in case of foodstuffs, but there is only one in force, which contains definite values for radionuclide content of foodstuffs. This one is a Commission Regulation (Euratom) No 770/90 of 29 March 1990 laying down maximum permitted levels of radioactive contamination of feedingstuffs following a nuclear accident or any other case of radiological emergency, the maximum permitted levels intended to apply to feedingstuffs as ready for animal consumption, and are listed in the Table 5.

Table 5. Maximum permitted levels of radioactive contamination (caesium-134 and caesium-137) of feedingstuffs (as it is in the regulation 770/90/Euratom):

animal	Bq/kg
Pigs	1250
poultry, lambs, calves	2500
other	5000

The main statement of this regulation is “Whereas consideration of the relative quantities of individual radionuclides liable to be released in the event of a nuclear accident in conjunction with their half-lives and transfer from feedingstuffs to animal produce leads to the conclusion that maximum permitted levels of radioactive contamination of feedingstuffs are needed only for the caesium isotopes”. The measure was formed almost 30 years ago, it is the time to involve the new scientific findings in the field of legislation of radiological protection.

Table 6 contains the main parameters used for the calculations of radiocaesium concentration in meat for different animals and cow milk applying the maximum permitted levels for radiocaesium according to the current legislation. The results are higher than any regulation in force for foodstuffs, not to mention the later recommended, lower 9 Bq/kg tolerance or 30 Bq/kg acceptance level.

Table 6. Parameter values (IAEA TECDOC in preparation) for the calculations of radiocaesium concentration in meat and milk from current maximum permitted level of feedingstuff (770/90/Euratom)

Animal	Daily dry matter intake, kg dry/day	Transfer coefficient, d/kg or d/l	Concentration in meat (cow milk), Bq/kg
pig	2.4	0.24	1286
poultry	0.07	10	2059
lamb	1.1	0.49	2406
calf	7.2	0.05	1607
other (cow milk)	16.1	0.0079	1136

Just an additional information that there is one regulation in force, which is recommends using $NH_4Fe[Fe(CN)_6]$ compound of 50-500 mg/kg feed concentration in the feed as radiocaesium binding additive in emergency situation when needed (70/524/EEC).

4. Recommended Guideline levels for radionuclide content of food and feedstuffs

My goal is derivation of guideline levels for radionuclide concentration in foodstuffs consumed day by day in normal situation and in case of prolonged emergency situation. My wish is make these proposals part of the legislation, giving the base of rapid intervention, when needed. However International Commission on Radiological Protection (ICRP furthermore) declared that "existing situations include naturally occurring exposures as well as exposures from past events and accidents, and practices conducted outside the Commission's Recommendations", I believe that everybody have right to get safe food in good quality without any contaminants at least causing no higher risk than so called trivial risk. In the event of a nuclear emergency resulting in significant releases of radioactive material to the environment, emergency response organisations will be called upon to provide guidance on the management of radiological contamination, to assess the radiation doses that may be received by members of the public and to introduce countermeasures to reduce or avert doses, according to the unavoidable cost benefit analysis. An understanding of the factors affecting the transfer of radionuclides to, and their behaviour in animals is essential if we are supposed to be able to interpret monitoring results in case of emergency situation, accurately predict activity concentrations in animal-derived food products and develop effective and appropriate countermeasures.

4.1. Determination of background level

Since the additional effective doses serving the basis for the determination of guideline levels, they have to be added to the exposure from the isotopic activity concentrations representing the background level, if relevant. Therefore it is essential to know the background level of the country for both the artificial and natural isotopes. The Chernobyl accident happened more than twenty years ago, so only the long lived artificial radionuclides are in Hungarian environment, but not only from Chernobyl, but also from global fall-out of former nuclear weapon tests. These two sources represent nowadays the artificial background. Determination of background level of artificial isotopes has to be based on the fact that their distribution in the environment is lognormal, so the geometric mean and the associated 1σ upper bound of geometric standard deviation is recommended as of the upper limit of the background for artificial isotopes. Naturally occurring radioisotopes are regarded to follow the normal distribution, mean plus 1σ standard deviation recommended for accepting as a background level. The basis of this have to be a long-term, careful, countrywide monitoring of foodstuffs, preferably without any further contamination determining the activity concentrations, with other words, there is a need of long, source free period.

At the end of 90's I introduced the method of the determination of plutonium isotopes, based on the experience gained on the training course held in 1996 in Seibersdorf and now there are enough available data for a countrywide evaluation about the most harmful artificial alpha-emitting isotopes.

4.1.1. Plutonium isotopes in Hungarian environment

The detailed description about the methods applied for Pu analysis is found in the paper Varga and Tarján (Varga and Tarján, 2008a) and Varga et al (2008b).

Following the deposition of fall-out, soil constitute the main plutonium reservoir in the terrestrial environment. The first some samplings were planned on those sites where higher concentrations were expected from the fall-out, according to the experience gained after the Chernobyl accident in the country. It is well known that the wet fall-out might bring more radioisotopes to the surface than the dry fall-out, so rainy areas and areas at higher altitude were investigated. The goal was to find the vulnerable areas in Hungary, exposed to higher activity concentrations of plutonium isotopes. While the chemical procedure takes a long time, the goal was to get and analyse samples from selected places representing the country. One of the areas investigated first was the hills of the capital, sampling was carried out in 1999, 2000, 2003 and 2004, all samples came from a 120 m circle of an undisturbed area, so they were regarded as one sampling site. The top 2.5 cm layer of the soil was investigated and Figure 2 shows the results of the $^{239+240}$Pu measurement. In 1999 the result was considered rather high and three additional control measurements were performed from the top layer. In 1999 a grass sample was also taken from this field, that had an activity concentration of 0.024±0.01 Bq/kg fresh weight. The transfer factor for the root-uptake comparing the grass with the top layer of the soil gave 0.032±0.014 (Bq/kg fresh weight)/(Bq/kg dry weight). In literature rather wide range of the transfer factors for grass can be found, namely between $5*10^{-5}$ and 0.7 with the $4*10^{-4}$ as the best estimate taken from IAEA Technical Report Series 364 (IAEA, 1994). The results from 2000, 2003 and 2004 are not significantly different from the result in 1999, even their uncertainties are overlapping. ^{238}Pu is below the detection limit of the applied procedure, calculating the isotope ratio (for ^{238}Pu using the minimal detectable activity concentration) of ^{238}Pu/$^{239+240}$Pu is less than 0.08, so the origin of the plutonium is likely the global fallout.

Vertical migration of radionuclides in the soil column is driven by convection, dispersion, diffusion and bioturbation. Convective-driven migration moves down the soil as a marked peak and shows broadening with time as a result of dispersing mixing – usually dominates in soils having high hydraulic conductivities (sandy soils). For diffusive transport the concentration maximum always remains at the surface with close to exponential decrease – typical in soils of low hydraulic conductivity. The value of 100 years as default residence half-time for upper 25 cm layer is adequate for all elements with high distribution coefficient, such as plutonium, americium, radium, lead, uranium. Figure 3 shows soil profile having good hydraulic conductivity, sampling site was same as shown on Figure 2. Among the archive samples there was a core sample from the same place just from 1991. The Figure 3 shows the two core samples, every layer of the core from 1998 contains higher activity of $^{239+240}$Pu. ^{238}Pu was not detectable from these samples, therefore the origin of Pu can not be given. However some reconstruction work started in research reactor less than 5 km far from sampling point, there is no evidence of the connection of these two fact.

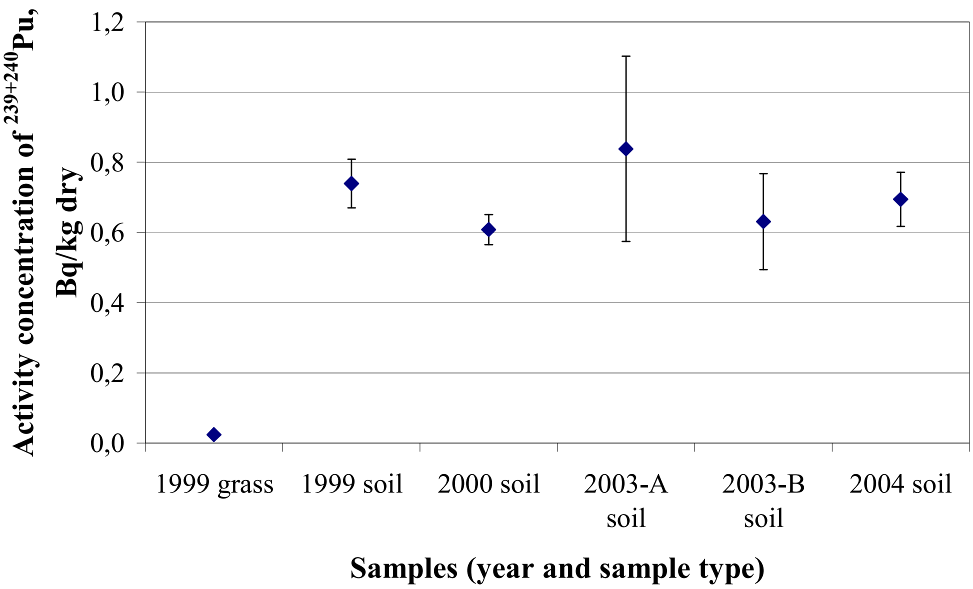

Figure 2. The activity concentration of $^{239+240}$Pu in the top 2.5 cm layer of the soil samples and the grass sample taken from the same place of Budapest (coordinates: 47.48822, 18.96173 according to WGS 84).

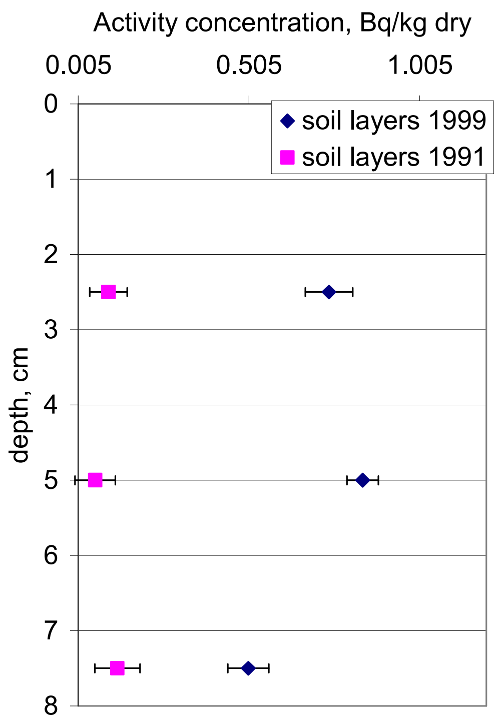

Figure 3.Activity concentrations of $^{239+240}$Pu in soil profile taken in 1991 and 1998 from the same place as Figure 2 (^{238}Pu was not detectable).

An intensive sampling campaign was carried out between March and May in 1998 in the Hungarian part of the 80 km planning zone of the Mohovce Nuclear Power Plant for getting appropriate nuclide specific data as background level for future contamination just before starting of Unit 1, in October 1998. Fall-out from Chernobyl could not be observed in these regions twelve years after the accident, the minimum detectable activity concentration could allow the precise measurement of any contamination on undisturbed areas, because the ratio of ^{238}Pu/$^{239+240}$Pu from inventory was 0.38-0.56 according to UNSCEAR 2000, but the activity concentration of ^{238}Pu remained below the detection limit. Figure 4 shows that there is a significant difference between soils taken from agricultural lands and from undisturbed area, even in the undisturbed group of samples forest generally represents slightly higher activity concentration than open areas. Undisturbed area means in our terminology forests, pastures and undisturbed areas of mountains. Agricultural area means cultivated land mainly small farms where vegetable is produced and arable lands.

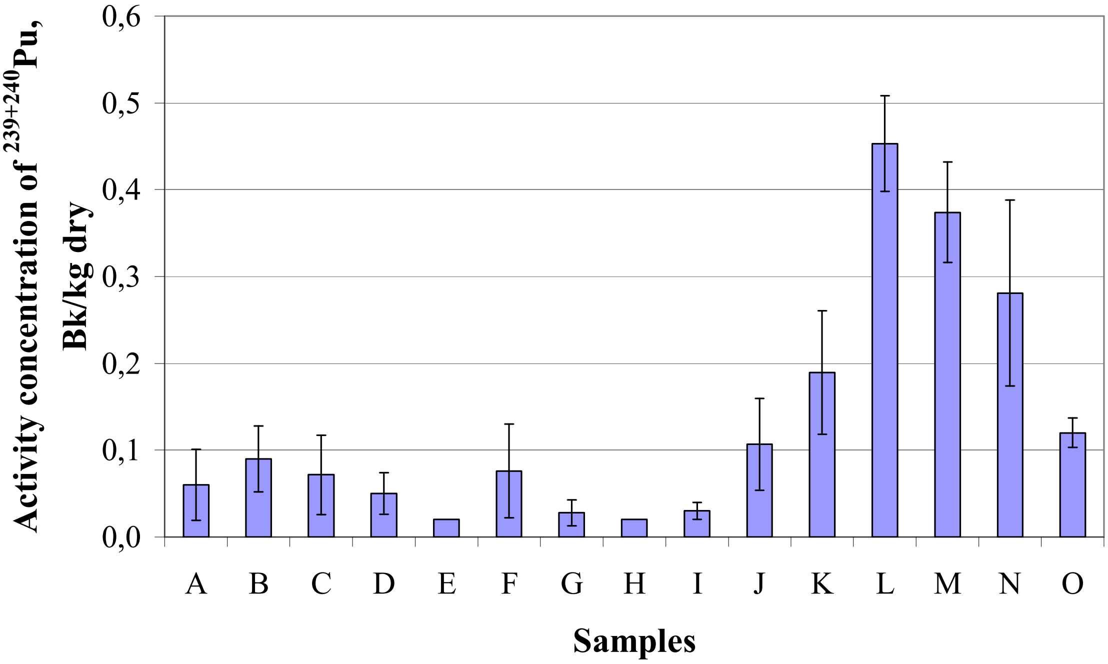

Figure 4. The activity concentration of $^{239+240}$Pu in the top 2.5 cm layer of the soil samples taken from the Hungarian part of the 80 km circle of the Mohowce Nuclear Power Plant before October 1998, samples marked A-I came from agricultural lands and J-O came from undisturbed area.

Another interesting area of the country is the feet of the Alps in the western part of the country. The climate is different from the other parts of the country, the yearly amount of rain is more than 800 mm and the average value for the country is 612 mm. Some valuable old samples originated from this area which were reserved in the laboratory of the Veterinary and Food-control Station of Vas county and plutonium isotopes were determined in them. As Table 7. shows there are some moss samples which were collected before the Chernobyl accident. In 1999 grass and soil sample-pairs were collected and the transfer factor for grass was calculated for the same year when the hilly site of Budapest was measured. The following value were obtained: 0.072±0.044 (Bq/kg fresh weight)/(Bq/kg dry weight). ^{238}Pu could be detected only in one sample; in the old part of the moss from 1981, 0.050±0.024 Bq/kg dry and the ratio of ^{238}Pu/$^{239+240}$Pu is 0.046±0.023, which is close to the ratio of the global fall-out 0.025, corrected by the decay for year 1981 according the published data in UNSCEAR 1982.

Table 7. Some results from Vas County (sampling area was the Feet of the Alps)

Date of sampling	**Sample type**	**Location latitude**	**Location longitude**	**Activity concentration of $^{239+240}$Pu±uncertainty, Bq/kg dry**
1981	moss	46.90102	16.24506	0.332±0.073
1981	old part of the moss	46.90102	16.24506	1.077±0.115
1981	moss	46.87013	16.42472	0.254±0.093
1981	old part of the moss	46.87013	16.42472	1.082±0.118
1984	old part of the moss	46.87013	16.42472	1.462±0.449
1990	soil (0-2.5cm)	46.91976	16.48878	0.282±0.039
1999	soil (0-2.5cm)	46.91976	16.48878	0.278±0.035
1999	soil (0-2.5cm)	46.85665	16.43022	0.130±0.044
1999	grass*	46.85665	16.43022	0.009±0.002
2000	soil (0-2.5cm)	47.10694	16.97060	0.147±0.083
2005	soil (0-2.5cm)	47.13903	17.09992	0.274±0.040

* result in Bq/kg fresh mass

The sampling continued and according to the results of the survey, there was a significant difference between the samples from the undisturbed area and the agricultural lands: the activity concentration of $^{239+240}$Pu from agricultural land fell in the range of 0.01-0.09 Bq/kg with the average of 0.04 Bq/kg and the activity concentration of samples from undisturbed area were in the range of 0.12-0.74 Bq/kg with 0.24 Bq/kg average. Figure 5 shows the results of the investigations from the samples collected before 2003. The difference in the average values is less than it was at the end of 90`s, namely 0.20 for undisturbed and 0.06 for agricultural land. The next bigger sampling campaign was in 2004 with the goal of obtaining data from all counties, so samples came from lower altitudes also and from the flat part of the country. Results showed that this significant difference between the undisturbed and agricultural areas of higher altitude disappeared. The average activity concentration of the soil samples were 0.051 Bq/kg with the range of 0.02-0.14 Bq/kg from 24 samples. As the map in the Figure 6 about the country shows, the soil around Nuclear Power Plant Paks had not higher activity concentration. The transfer factor for grass and soil was calculated for this region also, 0.019±0.010 (Bq/kg fresh grass)/(Bq/kg dry soil), not significantly different from the values in the other two areas.

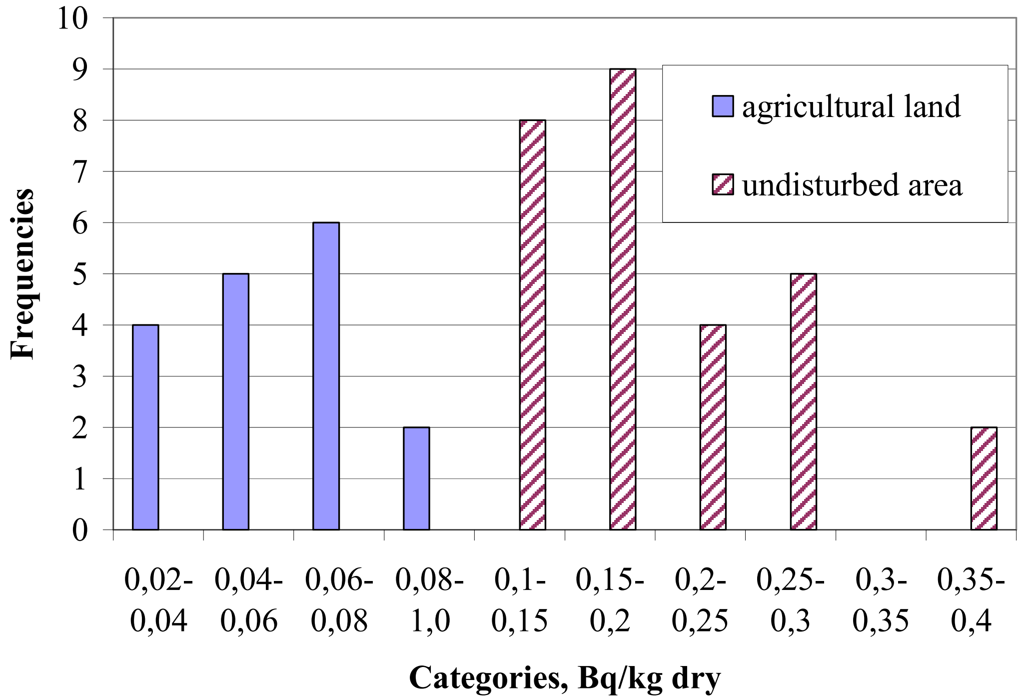

Figure 5. Distribution of activity concentration of $^{239+240}$Pu in soil taken before 2004

Moss samples were also investigated from different parts of the country. In the areas with rather high amount of rain it is easy to collect moss samples from the big stones or trees, but in the eastern, drier part of the country it is rather difficult to collect them, sometimes they are found only on the roofs of the houses. During the last few years 39 moss samples were investigated. The activity concentration of $^{239+240}$Pu in moss samples are between 0.007 and 1.299 Bq/kg fresh mass. Both median and average values are higher than in case of soils, 0.131 and 0.258 Bq/kg respectively. The map in Figure 6 also shows that some higher activity values occur in the country, but there is no trend among them. Usually the accumulation time of the moss is unknown, or only estimated, the age of the moss is not well defined, this is the reason that from the same area there are moss samples with lower and higher activities. There were only few moss samples in which the ^{238}Pu was detected, the results are listed in Table 8. From the isotope-ratio of ^{238}Pu/$^{239+240}$Pu turned out also that the plutonium found in Hungary comes from the global fall-out, the average ratio is 0.036±0.006 calculated from five samples. Four of them were collected in mountains of different parts of the country and only one came from the flat area not far from the power plant. Two moss samples from foreign countries were analysed, one from Ukraine collected in 2000 and one from Germany collected in 1998, in both of them ^{238}Pu isotope was detectable by alpha spectrometry and the ratios differ from the Hungarian mosses. The ratio 0.483±0.057 in sample from Ukraine agrees well with the ratio of Chernobyl release, and the value of 0.093±0.017 in the sample from Germany is also higher, than in Hungary as it was expected from the way of the plume in 1986. The correlation between the investigated plutonium isotopes are very good.

Figure 6. Spatial distribution of the activity concentration of $^{239+240}Pu$ in soil and moss samples in Hungary (green column: moss samples, striped column: soil samples)

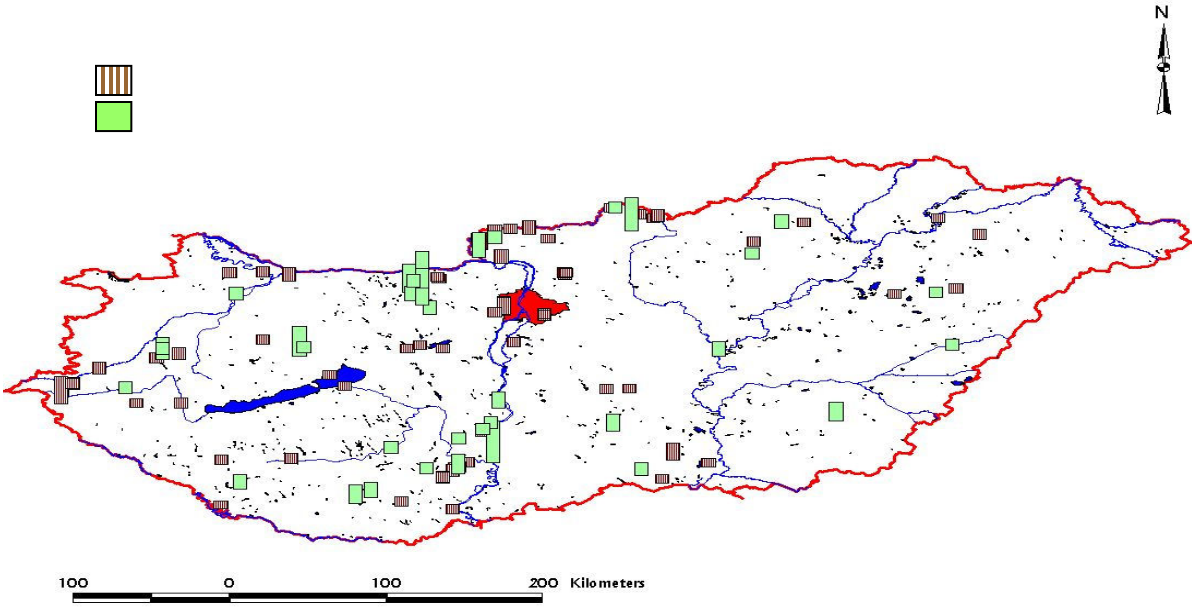

Table 8. Selected results of the moss samples

Year of sampling	County	Activity concentration of $^{239+240}Pu$, Bq/kg fresh mass	Activity concentration of ^{238}Pu, Bq/kg fresh mass	Activity concentration of ^{241}Pu, Bq/kg fresh mass
1998	Pest	0.574±0.112	0.027±0.015	0.73±0.04
1998	Pest	0.540±0.127	0.019±0.004	0.42±0.0.02
2003	Pest	1.299±0.062	0.047±0.007	2.07±0.07
2003	Borsod	0.099±0.084	0.003±0.002	0.08±0.02
2003	Tolna	0.939±0.045	0.029±0.010	0.84±0.05
2003	Komárom	1.060±0.09	< 0.04	2.39±0.19
2004	Heves	0.007±0.002	< 0.01	0.011±0.004

In last four years the activity concentration of ^{241}Pu in the archived plutonium sources was also checked by liquid scintillation spectrometry, details are described in paper of Varga and Tarján (Varga and Tarján, 2008a). The activity concentrations of ^{241}Pu were calculated back to the sampling date. The activity concentration of this isotope in soil and moss samples were between 0.04-3.74 Bq/kg with the average of 0.82 Bq/kg, and between 0.01-2.07 Bq/kg fresh mass with the average of 0.43 Bq/kg respectively. Mosses are integrators also for plutonium, they accumulate isotopes from fall-out during their long lifetime. The spatial distributions are shown on Figure 7. Significant difference could not be observed among the different soil and moss samples. The ratio of the activity concentrations of ^{241}Pu and $^{239+240}$Pu was 4.2 in 1986 from the global fall-out. The measured average ratio was 2.3 which is in quite good agreement with 2.1 the value calculated for the date of sampling taking as a reference of 4.2 for 1986. Table 9 shows some of the results in detailed form, the samples represent the general situation of the country. 3 samples of special interest were found. First one became an in-house reference material in the network, this is an ashed fodder from 1986, the ^{241}Pu/$^{239+240}$Pu activity ratio of 68.5±6.7 is close to the ratio of Chernobyl inventory, which is the range of 73-94 that was calculated from the data published in UNSCEAR 2000. The ratio of ^{241}Pu and $^{239+240}$Pu for samples of Chernobyl origin was 86±20 in Poland according to the work of Mietelski et al (Mietelski et al, 1999). The ratio measured on air filters in Austria in 1986 (Irlweck and Wicke, 1998) was lower, it ranged from 31.6-74.6 with a mean of 57.8±3.7. In fresh fallout on plants collected in Ukraine in 1986 (Buzinny et al., 1994) this ratio was lower ranging 10.2 to 40.7 with an average of 28. Low activity ratios of 26.4±4 were observed also in grass samples in Germany (Bunzl and Kracke, 1990). Hot particles and fallout from first day releases might give higher ratio close to the expected value calculated from the core inventory. According to the work of Salimen only a few percent of total transuranium deposition originates from the Chernobyl accident in Finland (Salimen, 2005).

Table 9. Some results for proving the origin of plutonium isotopes in the Hungarian environment (the ^{241}Pu results and isotope ratios were calculated always to the date of the sampling).

Year of sampling	**Type of the sample**	**County**	**Activity concentration of $^{239+240}$Pu, Bq/kg dry**	**Activity concentration of ^{241}Pu, Bq/kg dry**	**^{241}Pu/$^{239+240}$Pu from measurements**	**^{241}Pu/$^{239+240}$Pu calculated from global fall-out**
1991	soil	Pest	0.45±0.06	1.36±0.17	3.0±0.5	3.3
1998	soil	Nógrád	0.19±0.07	0.76±0.07	4.0±1.6	2.4
2000	soil	Pest	0.61±0.04	1.70±0.13	2.8±0.3	2.1
2003	soil	Tolna	0.19±0.04	0.46±0.004	2.4±0.5	1.9
2003	soil	Tolna	0.29±0.11	0.29±0.03	1.0±0.4	1.9
2004	soil	Somogy	0.020±0.008	0.043±0.004	2.2±0.9	1.8

Two samples with higher ratio was collected in 2005 around the nuclear power plant, both of them have small activities, less than the maximum permitted level even for foodstuffs (3954/87/Euratom) far below the authority limit, but the ratio shows the origin of the plutonium from the plant; one of them was a hay collected in Bács-Kiskun county and the ratio was 55.1±9.3 and the other sample was a sedge collected in Tolna county and the ratio was 18.2±3.2, which significantly differ from the 1.7 ratio of ^{241}Pu/$^{239+240}$Pu calculated from the global fall-out for the year 2005.

The activity concentration of $^{239+240}$Pu in top layer of soil from agricultural land gives information on total fall-out in the short term period, because of the cultivation. Samples from the undisturbed area might give a better estimation of the contamination in a long term period for several years, but the top layer does not always keep the whole amount of plutonium. Migration depends on the constituents of the soils and also the physical properties. If a stony soil contains small channels, plutonium will be easily transferred to the deeper layers for example in the samples from hilly part of Budapest, where 0.51 Bq/kg dry activity concentration of $^{239+240}$Pu was found in the 5-7.5 cm layer (Figure 8). In forest environment plutonium deposition is typically about 30 % higher, because of the foliar interception. The amount of plutonium retained in the top layer depends among the others on the humic acid content of the soil.

Conclusions

This study confirms that the most of the plutonium existing in the terrestrial environment is accumulated in the soil. In Hungary the plutonium contamination comes mainly from global fallout, what is confirmed by plutonium isotope-ratios.

The measured activity concentrations in Hungarian soils are at least three orders of magnitude lower than the maximum allowable concentrations corresponding to 0.01 mSv/year of external exposure obtained by Wood et al: for ^{238}Pu 80.1 Bq/kg, for ^{239}Pu and ^{240}Pu 72.6 Bq/kg and for ^{241}Pu only 19.0 Bq/kg estimating by RESRAD (Wood et al, 1999). Root uptake of plutonium is low, whereas sorption to solid matter in soils, sediments and peat is high. Generally plutonium activities in the plant samples are below the minimum detectable activity concentration. Where plutonium could be detected the transfer factor was 0.019±0.010 and 0.072±0,044 in the area where the precipitate is lower (the yearly average is 516 mm) and where it is higher (above 800 mm). The average activity concentration of $^{239+240}$Pu in the soil of pasture in Hungary is 0.098 Bq/kg with the range of 0.007-0.289 Bq/kg.

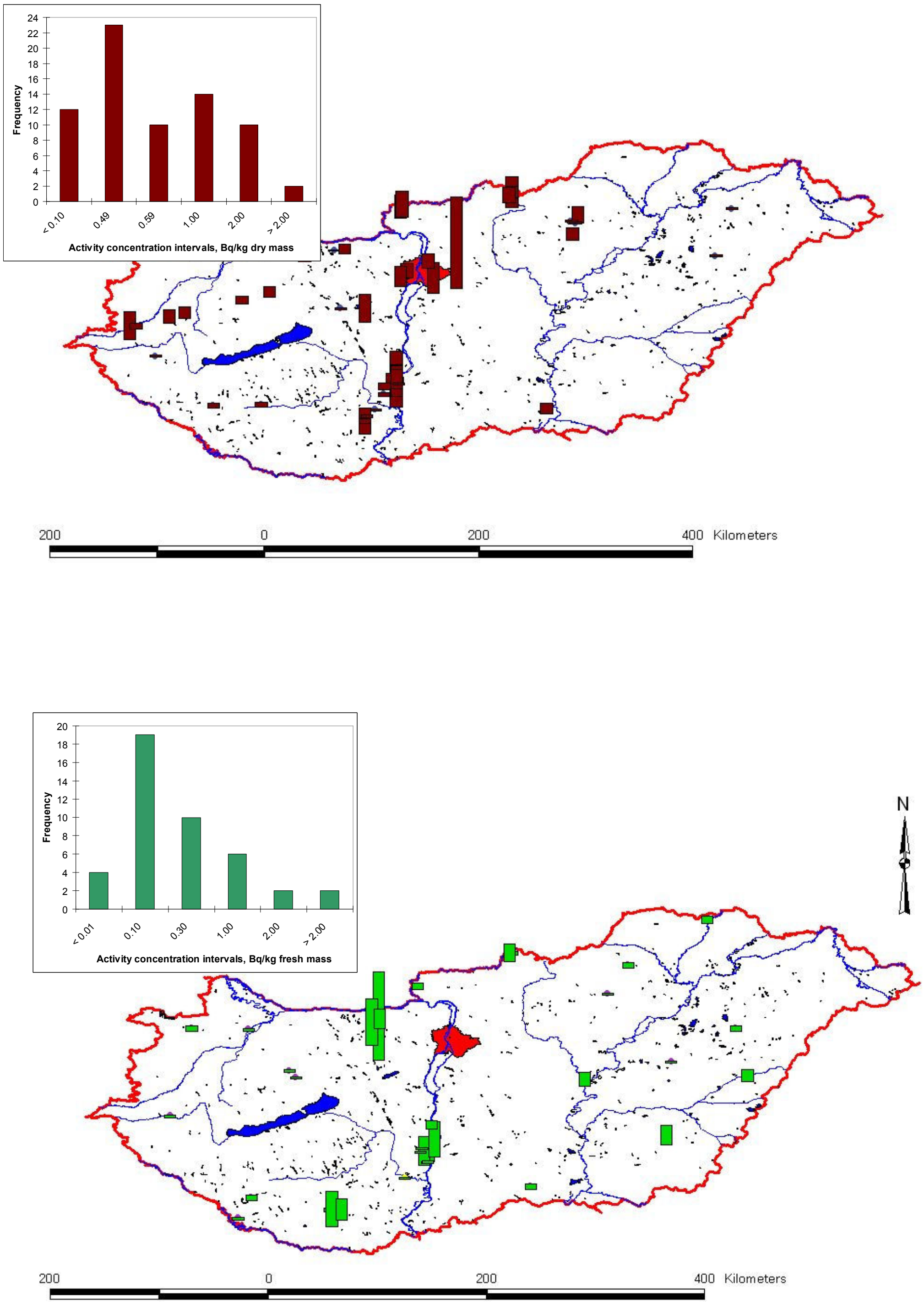

Figure 7. Distribution of ^{241}Pu activity concentrations in soil (above, concentration range 0.10-3.74 Bq/kg) and moss samples (concentration range: 0.002-2.8 Bq/kg fresh) collected since 1998 in Hungary

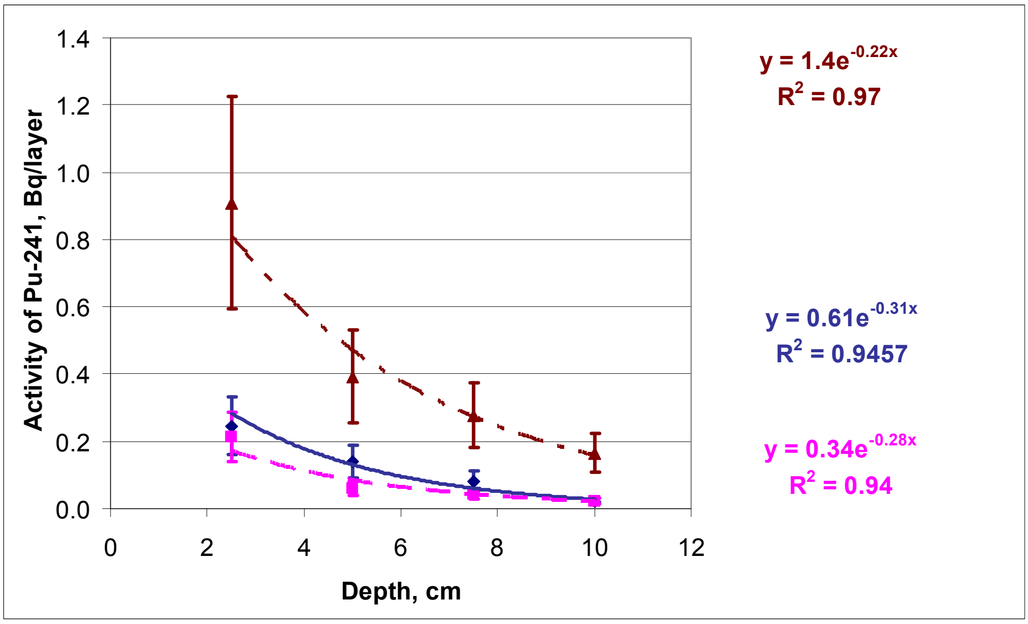

Figure 8. ^{241}Pu activity concentrations measured in three core samples collected in north part of the country. Estimation of deposition values are 58.0±5.0 Bq/m^2, 84.1±7.3 Bq/m^2, 300.6±26. Bq/m^2 respectively.

Using the higher transfer factor for prediction of the activity concentration of grass in the wet area and the another one to the other areas: the average from the calculated values is 0.0057 Bq/kg fresh mass with the range of 0.0001-0.0208 Bq/kg fresh mass. The average activity concentration of ^{241}Pu in soil is 1.22 Bq/kg. Transfer to cow milk and meat is also low, for calculation the following data are used: the consumed fodder, 16 kg dry is calculated back to fresh mass, using the Hungarian database for dry content, namely 35.6±14.2 % and transfer factors were taken from IAEA Technical Report Series 364 (IAEA, 1994). For the different kind of foodstuffs the given values were calculated to the (food Bq/kg fresh mass)/(soil Bq/kg dry mass) using dry content from my measurements. The result of the ingestion dose calculation is shown in Table 10. The situation from point of plutonium contamination considered satisfactory, the average committed effective ingestion dose for adults from $^{239+240}$Pu is $2.50*10^{-10}$ (Sv/year)/person and from ^{241}Pu is $4.87*10^{-11}$ (Sv/year)/person, altogether $2.99*10^{-10}$ (Sv/year)/person dose is obtained. This value is three orders of magnitude less, than the committed effective ingestion dose from ^{137}Cs and from ^{90}Sr in Hungary. The radiological exposure of the population was evaluated based on the results of the monitoring network; the committed effective dose due to ingestion for adults was estimated to be 0.6 μSv for ^{90}Sr and 0.3 μSv for ^{137}Cs in 2004. Dose conversion factors were taken from the Safety Series 115 published by IAEA (IAEA, 1996).

Table 10. The result of ingestion dose calculation on the basis of measured activity concentrations in soil samples taken in the country.

	$^{239+240}Pu$, Bq/kg	^{241}Pu, Bq/kg	Food consumption, (kg/year)	Consumption from $^{239+240}Pu$, (Bq/year)	Consumption from ^{241}Pu, (Bq/year)
Average activity in soil	0.098	1.22			
Beef and veal	$2.54*10^{-4}$	$5.34*10^{-4}$	0.8	$2.03*10^{-4}$	$4.27*10^{-4}$
Milk	$2.80*10^{-7}$	$5.87*10^{-7}$	58.3	$1.63*10^{-5}$	$3.42*10^{-4}$
Cereals	$7.54*10^{-7}$	$9.37*10^{-6}$	90.4	$6.82*10^{-5}$	$8.47*10^{-4}$
Root crops	$1.93*10^{-5}$	$2.40*10^{-4}$	35.4	$6.84*10^{-4}$	$8.49*10^{-3}$
Fresh vegetable	$7.20*10^{-7}$	$8.94*10^{-6}$	39.4	$2.84*10^{-4}$	$3.52*10^{-4}$
Total consumption from plutonium isotopes, Bq/year				$1.00*10^{-3}$	$1.01*10^{-2}$
Ingestion dose, Sv/year				$2.50*10^{-10}$	$4.87*10^{-11}$

The radiotoxicity of the plutonium is very high. Alpha particles emitted from plutonium isotopes have high energies and they are also heavy particles that have a route no longer than 40 μm in the tissue, that's why damage to the tissue is highly localised. About 90 % of the ionisation in an alpha track is deposited within a cylinder 0.01 μm radius, a further 9 % deposited from 0.01 to 0.2 μm. Because of this phenomena and the long half-lives of plutonium isotopes, long-term chronic toxicity is very important. In the lung or bone the latent period between exposure and potential development of solid tumors is estimated to be 20 years. When the route of intake is inhalation of small particles containing plutonium species might stay in the lungs and soluble forms are rapidly distributed. Particles with size higher than 25 nm do not result in movement of plutonium to blood. Based on mimicking iron biochemistry Pu^{4+} also forms a plutonium-transferrin protein complex (transferrin transports iron between the sites of uptake, utilisation and storage) and may be found in blood serum and then deposited in liver. The fact that the binding constant of plutonium-transferrin is in the same order as that of iron-transferrin is remarkable. Plutonium will bind readily to mineralizing tissues, like bones and the rate of incorporation depends on the age of mammals and humans also. According to ICRP model distributes 45% of dissolved plutonium is bound to bone surfaces from where it clears with a biological half-life of 50 years, and 45% is bound in the liver from where it clears with biological half-life of 20 years. The remaining 10% is assumed to pass directly through excretion after some short-term holdup in the tissues of the circulatory or urinary systems. The fractions of plutonium species taken up by the blood from the gastrointestinal tract are the following: 10^{-5} for oxides of plutonium, 10^{-4} for nitrates, 10^{-3} for other compounds; for unknown forms $5*10^{-4}$ according to ICRP 68 (ICRP, 1994).

There is no evidence for detrimental health effects due to plutonium intake by humans from low-level exposure due to global fall-out.

4.1.2. Radiocaesium in Hungarian environment

Determination of radiocaesium is carried out by gamma-spectrometry from ashed food and feed samples without chemical separation, measurements with high purity germanium gamma-spectrometers (HPGe) are sensitive enough and fulfil all requirements laid down in legislations (for example in 2000/473/Euratom).

The uncertainty analysis is unavoidable, because of the low concentrations of expected values. In the case of gamma-spectrometry using HPGe detectors the sources of standard uncertainties can be grouped according to their origin (IAEA, 2004).

Uncertainties may occur during the sample preparation are the following: drying and ashing of the sample, weighing of the sample to a sample holder, sample inhomogeneity. The last component usually does not have any role, because the whole amount of ash are measured, and the representativeness to the whole amount of sample is also could leave out because of the same reason, subsample has not been taken from the samples.

The next component is the uncertainty of energy and efficiency calibration. Energy calibration can be executed very precisely, its very small uncertainty contribution usually is not taken into account, linearity of modern ADC is considered extremely good, the function fitted to the channel-energy curve contains the second power term in the order of 10^{-9}. The efficiency calibration for a given geometry has great importance. Standards and reference materials used for calibration are in the same geometry as the sample. These are available in every laboratory of the network where gamma-spectrometry is performed. Of course the origin of these materials is the same, it has an advantage to have harmonised calibration, but disadvantage might be the same, if the source from some reason is not proper. As the intercomparison runs with invited external laboratories organised by our department showed the network have only the benefit of the harmonised calibration.

The most complex part of the uncertainty budget comes from the measurement of sample. The uncertainty has the following contributions: counting statistics, reproducibility of the geometry, self-attenuation, random and true coincidences, net peak area determination, dead time effects and decay time effects. Generally the uncertainty due to counting statistics is one of the most important ones. For the reason of decreasing the effect of the positioning, all detectors have their own sample holder frame tightly fitted onto the detector cap, but the positioning and distances are exactly the same for all of the detectors involved in daily monitoring task. They have some changeable parts depending on the diameter of the used sample holders and some other positioning rings to keep the exact distance between the sample and the top of the detector. The subtraction of instrumental background and uncertainty of corrected net peak area is obtained from the fitting. Environmental samples produce generally low count rates, therefore the random coincidence is not significant, and good pile-up rejection might be useful. Dead time effects are also negligible. Correction factor for decay between sampling and measurement is done automatically by the software, but generally never more than one month elapsed after the sample taking until measurement. For the investigated isotope decay time effect during the measurement is negligible, if not, the software can handle also. Nuclides decaying through the cascades can cause coincidence-summing especially in high efficiency semiconductor detectors. Self-attenuation correction is not taken into account, instead the network laboratories have standards of same density as the routinely measured samples.

The nuclear data also have uncertainties. In a few cases gamma emission probabilities could be the major contributor to the final uncertainty value. ^{137}Cs is not this kind of isotope, because the parameters are well known and widely investigated and used. The uncertainty of its half-life is still small compared with other sources.

Altogether 16 HPGe detectors are working continuously in the network. All of them are cylindrical, their parameters are different, depending the year of installation, n- or p-type detectors with 15-30 % relative efficiency, some of them are in ultra low background, some in low background and some only in normal cryostat. While they are installed in different laboratories in different parts of the country, the measuring circumstances are also different (building materials of the house, ventilation, stability of the electric supply), which have effect on the background of the measurement. The shieldings of the detectors are also different. From the mentioned facts come a range for the uncertainty instead of a concrete value, when the goal is to define the uncertainty of ^{137}Cs-determination at the network level, especially in case of so low activity concentration as we have in the Hungarian environment.

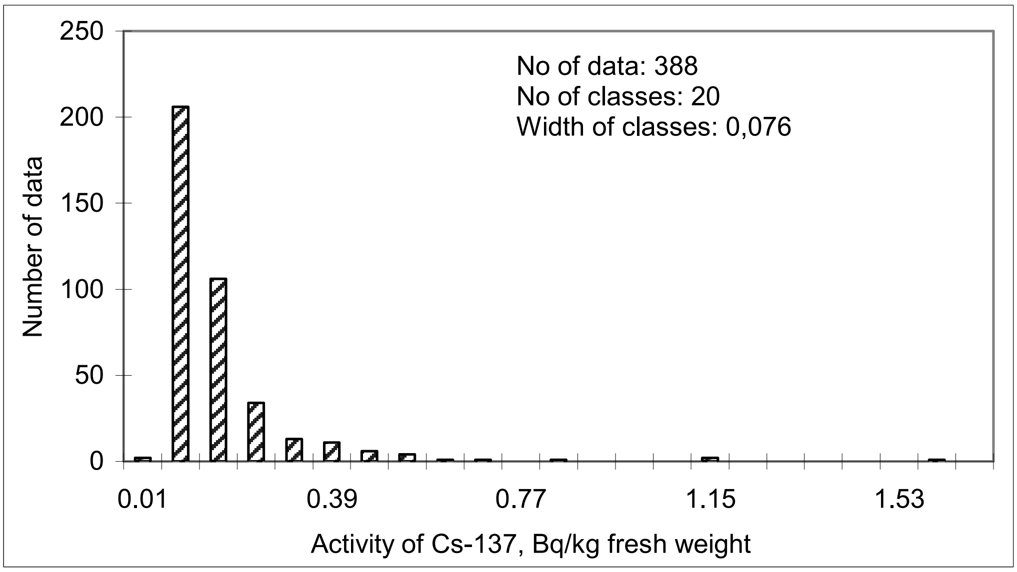

Figure 9. Distribution of the activity of ^{137}Cs in sorrel and spinach samples from the years 2000-2004

The distribution shown in Figure 9 does not follow the normal distribution. Its skewness is 4,70 and kurtosis is very high 34,4 instead of 0 and 3 respectively as in case of normal distribution. The values of arithmetic mean: 0,12 Bq/kg fresh weight, median: 0,08 Bq/kg fresh weight and mode: 0,06 Bq/kg fresh weight show also, that this concentration range of this anthrophogenic isotope does not show normal distribution. In this case robust statistic can give more reliable description about the dataset, because it is designed to cope with wide dataset, without resorting to removal of data from the dataset - robust mean: 0,092 Bq/kg fresh weight and the Median of all Absolute Distances (MAD): 0,057 Bq/kg fresh weight. Robust analysis was performed by HISTO designed by Z. Radeczki and A. Trinkl (provided by IAEA), where the algorithm was reproduced from ISO 5725-5.

The combined standard uncertainty of the measurement generally is described with the following equation, if the x_i quantities are independent:

$$u_c(y(x_1 ... x_n)) = \sqrt{\sum_{i=1}^{n} \left(\frac{\partial y}{\partial x_i} \right)^2 (u(x_i))^2} \qquad \text{Eq 4.1}$$

$u_c(y)$ combined standard uncertainty of the y measurand,

$y(x_1 ... x_n)$ the measurand function of several quantities of $x_1 ... x_n$

If the variables not independent:

$$u_c(y(x_1 \dots x_n)) = \sqrt{\sum_{i=1}^{n}\left(\frac{\partial y}{\partial x_i}\right)^2 (u(x_i))^2 + 2\sum_{i=1}^{n}\sum_{j>1}^{n}\left(\frac{\partial y}{\partial x_i}\right)\left(\frac{\partial y}{\partial x_j}\right)\mathrm{cov}(x_i, x_j)} \quad \text{Eq 4.2}$$

$cov(x_i,x_j)$ covariance between x_i and x_j.

The uncertainty components in case of gamma spectrometry are the following, as this was analysed above: stability of measuring system, energy calibration, efficiency (uncertainty of certified reference material, uncertainty of curve fitting), gamma emission probabilities, half life, random coincidences, true coincidences, dead time effect, decay time effect, self-attenuation, counting statistics, sample preparation (weighing, ashing).

The description of the uncertainty may perform taking into account the whole data coming from the countrywide network after the characterisation of the dataset. Figure 10 shows well that in such a low activity concentration for every value might be given an uncertainty range. After the curve fitting, namely the power-functions, the highest and the lowest uncertainty values were calculated and ranges are given in the Table 11 for the critical range from the point of view of dependence of the local installation circumstances and parameters of the detectors.

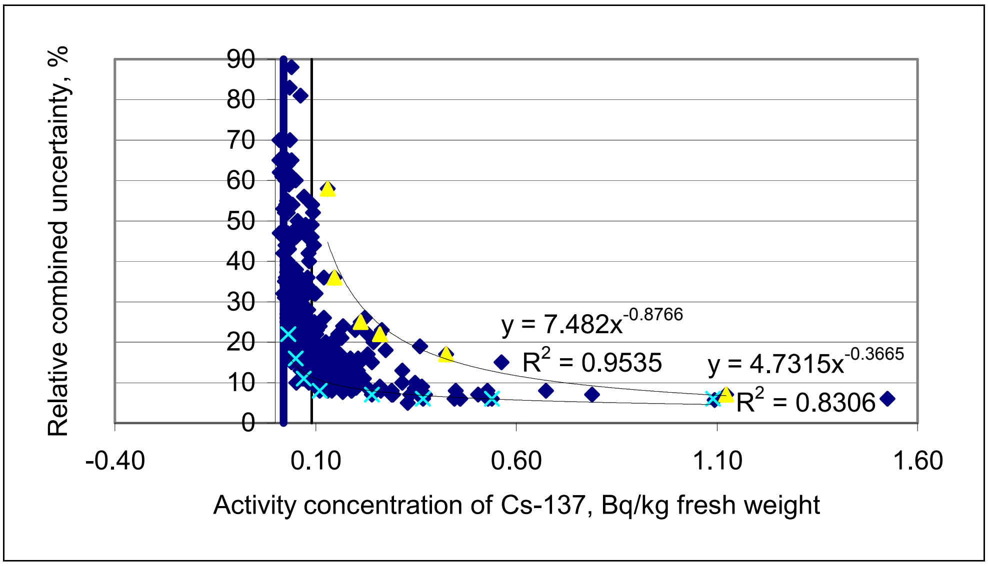

Figure 10. Relative combined uncertainty of the activity concentration of ^{137}Cs in sorrel and spinach samples from the years 2000-2004 in the network

Below 0,6 Bq/kg fresh weight the dominant component is the uncertainty of counting statistics. Above this value two main contributors are the uncertainty of calibration source and the uncertainty of efficiency curve fitting, the experimental values show that the uncertainty is not worse than 7%. Above 0,6 Bq/kg fresh weight calculation of combined uncertainty from the components taking into account the parameters of the different detectors also gives the value not worse than 7 %. Figure 10 contains another important information determined empirically, that the range of minimal detectable activity concentration spread 0,02 Bq/kg fresh weight for the better detectors to 0,07 Bq/kg fresh weight for the older ones.

From these results the conclusion could be drawn that at the reporting level, determined by 2000/473/EURATOM the network is able to give the activity of ^{137}Cs generally in food samples with uncertainty better than 10 %, which is acceptable. It is important to keep in mind that these values came from the measurements of ashed samples.

Table 11. The range of relative combined uncertainty of activity concentration of ^{137}Cs in sorrel and spinach samples on the network level

Activity concentration. Bq/kg fresh weight	Uncertainty range, %
0.1	11-56
0.2	9-31
0.3	7-21
0.4	7-17
0.5	6-14
0.6	6-12

For determining the background level data of the last five years was chosen, because the transport process governing the plant contamination is very slow nowadays and they are regarded stationary, furthermore there was no “fresh” contamination of ^{137}Cs before the investigated period, data from all the five years could treat as one homogeneous database. According to the lognormal distribution of artificial isotopes in the environment, the describing parameters are the geometric mean and geometric standard deviation, which are considered here as the upper limit of the background level. The conclusion drawn from Table 12 is that the upper limit for background level of ^{137}Cs in any kind of foodstuffs is 0.4 Bq/kg fresh weight.

Table 12: Background level for ^{137}Cs in different foodstuffs and feedingstuff in Hungary, based on the monitoring data from 2000-2005

Sample type	Number of samples	Expected value, Bq/kg	Upper limit of background level, Bq/kg
cereals	362	0.087	0.36
milk	1621	0.043	0.33
meat	1291	0.087	0.31
vegetable	979	0.042	0.21
fruit	385	0.022	0.072
feedingstuff	2056	0.22	1.10

4.1.3. Radiostrontium in Hungarian environment

Summary of the method: after usual sample preparation (drying and ashing) using $Sr(NO_3)_2$ carrier, leaching with HNO_3, some cleaning steps are included followed by precipitation of Sr as $SrSO_4$. The activity concentration is determined after ^{90}Y in-growth by low background alpha-beta proportional counter.

The following uncertainty contributors can have effect the final value associated with the result: weighing, uncertainty of determination of chemical yield, uncertainty of calibration source, uncertainty of the determination of the efficiency of the detector, uncertainty of the evaluation of the count-rate.

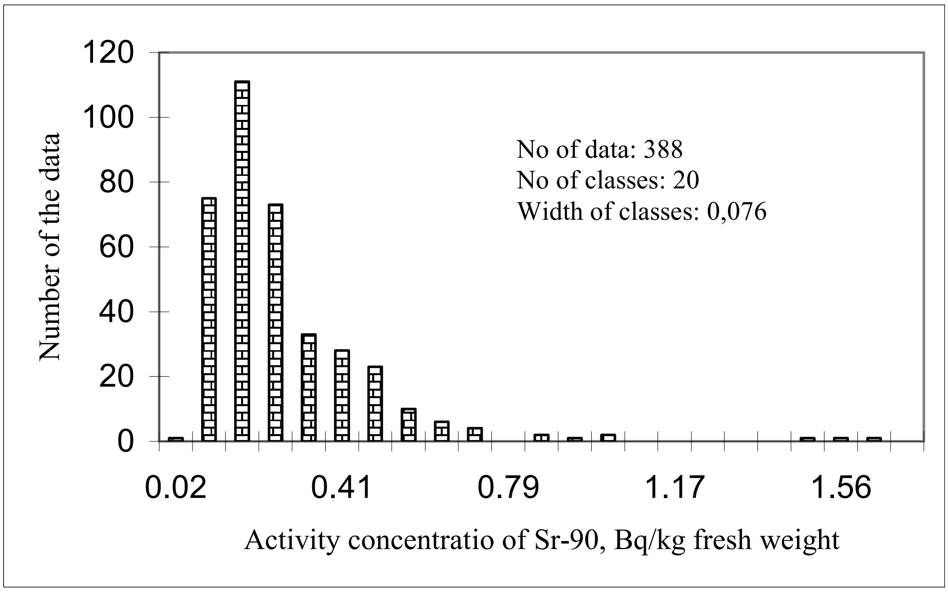

Figure 11. Distribution of the activity concentration of ^{90}Sr in sorrel and spinach samples from the years 2000-2004

The values of arithmetic mean: 0.24 Bq/kg fresh weight, median: 0.18 Bq/kg fresh weight and mode: 0.14 Bq/kg fresh weight show also, that this concentration range of this antrophogenic isotope does not follow normal distribution. The robust statistic give the following: 0.20±0.12 Bq/kg fresh weight. Robust analysis was performed by HISTO like in the previous case.

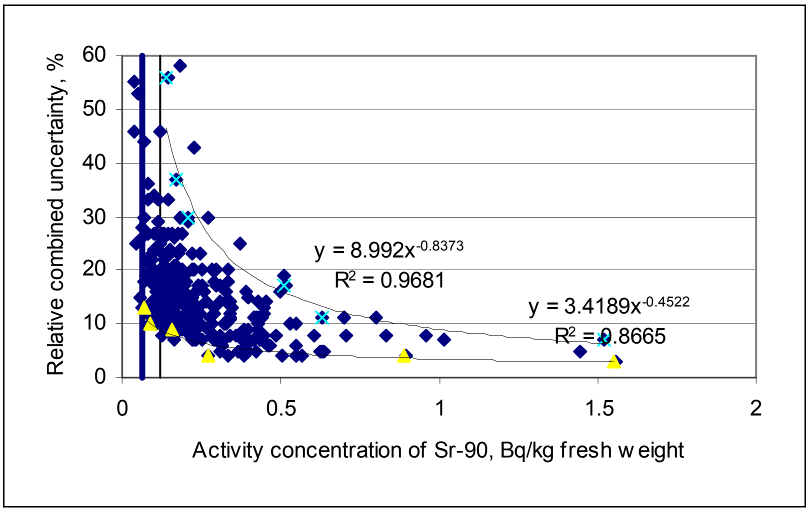

Figure 12. Relative combined uncertainty of the activity concentration of ^{90}Sr in sorrel and spinach samples from the years 2000-2004 in the network

Table 13. The range of relative combined uncertainty of activity concentration of ^{90}Sr in sorrel and spinach samples on the network level

Activity concentration, Bq/kg fresh weight	Uncertainty range, %
0.2	7-35
0.4	5-19
0.6	4-14
0.8	4-11
1.0	3-9
1.2	3-8

The uncertainty of the measurement is calculated in the same way as the equations Eq 4.1 and Eq 4.2. The following uncertainty components have importance: weighing and volume measurement during chemical preparation, chemical yield, calibration source, efficiency, counting statistics.

The distribution of dataset does not follow the normal distribution (Figure 11), however the skewness: 2.95 is close to that of the normal distribution, but the kurtosis is rather high 16.28.

In the case of ^{90}Sr the experimentally defined minimal detectable activity concentration is 0,06-0,12 Bq/kg fresh weight, depending the recovery and the ash content of the sample.

For determining the background concentrations for ^{90}Sr, the same method and same period was chosen as for ^{137}Cs. According to the data given in Table 14, the same generalisation as was applicable of ^{137}Cs could not be valid here, because the upper limits of different type of foodstuffs are not in the same order of magnitude.

Table 14: Background level for ^{90}Sr in different foodstuffs and feedingstuff in Hungary, based on the monitoring data from 2000-2005

Sample type	No of samples	Expected value, Bq/kg	Upper limit of background level, Bq/kg
cereals	336	0.11	0.36
milk	1561	0.038	0.23
vegetable	296	0.11	0.85
fruit	359	0.017	0.06
feedingstuff	1947	0.67	5.7

4.1.4. Natural isotopes in Hungarian environment

Regarding the isotopes of natural origin, in case of terrestrial isotopes their spatial distribution has high importance, which is observable on the Figure 13. In Hungarian soils uranium $^{238}U(^{226}Ra)$ concentration is in the range of 16-46 Bq/kg, ^{232}Th 22-49 Bq/kg. Uranium is widely distributed in nature, the average content of Earth's crust is about 4 mg/kg, main sources are sedimentary rocks, hydrothermal veins, pyretic conglomerate beds; it is also found in phosphate rock, lignite and monazite sands. Both ^{238}U (99.27%) and ^{235}U (0.72%) are primordial radionuclides, parent nuclides of their respective decay chain, ^{234}U is present in environment due to its production from ^{238}U, it is often present at close to secular equilibrium with ^{238}U. Radium is the heaviest member of the alkaline earth metals, the lighter members of this group play important role in plant growth and nutrition. More than 99% of natural thorium exists in the form of ^{232}Th, typical concentration 6 mg/kg in soil.

Heavy natural radionuclides, like U, Th, Ra, Pb, Po-isotopes have relatively low soil-to-plant transfer factors, uranium is the most mobile among them, because of its more complex chemical properties. U and Th accumulation of plants depend on their concentration in soil, as concentrations increase, toxic effects become of significance and the transfer factor decrease. For ^{226}Ra, ^{210}Pb, ^{210}Po linear increases in transfer factor values were obtained, depending their concentrations in soil. The monitoring results are shown in Table 15 and 16, natural isotopes have normal distribution, so upper limit was formed as expected value + 2*σ.

Somlai et al (Somlai et al, 2002) measured the ^{226}Ra content in Hungarian bottled mineral water and they found that from the 26 investigated water 85 % of them was under 0.1 Bq/l and the highest concentration was found as 3 Bq/l, which may produce the dose even more than 1 mSv/year with the daily consumption of about 1 l.

Table 15. Background level for ^{238}U(^{226}Ra) in different foodstuffs and feedingstuff in Hungary, based on the monitoring data from 2000-2005

Sample type	Number of samples	Expected value, Bq/kg	Upper limit, Bq/kg
cereals	365	0.41	1.5
milk	1334	0.088	0.21
meat	842	0.22	1.0
vegetable	981	0.18	0.83
fruit	385	0.079	0.21
feedingstuff	1254	0.95	3.2

Table 16. Background level for ^{232}Th in different foodstuffs and feedingstuff in Hungary, based on the monitoring data from 2000-2005

Sample type	Number of samples	Expected value, Bq/kg	Upper limit, Bq/kg
cereals	365	0.43	2.6
milk	1334	0.090	0.15
meat	840	0.21	0.81
vegetable	980	0.18	0.91
fruit	385	0.073	0.16
feedingstuff	1256	0.84	2.6

Regarding the most abundant radionuclide ^{40}K, the ratio in natural potassium is constant, being essential macroelement its concentration is governed by the metabolism of biota, so speaking about transfer properties has no meaning, the concentration is 347-641 Bq/kg in Hungarian soils as it is shown on Figure 14. Take the advantage of the mentioned property, ^{40}K could be used in routine work as a quality assurance monitor for differentiating between the different kinds of matrices.

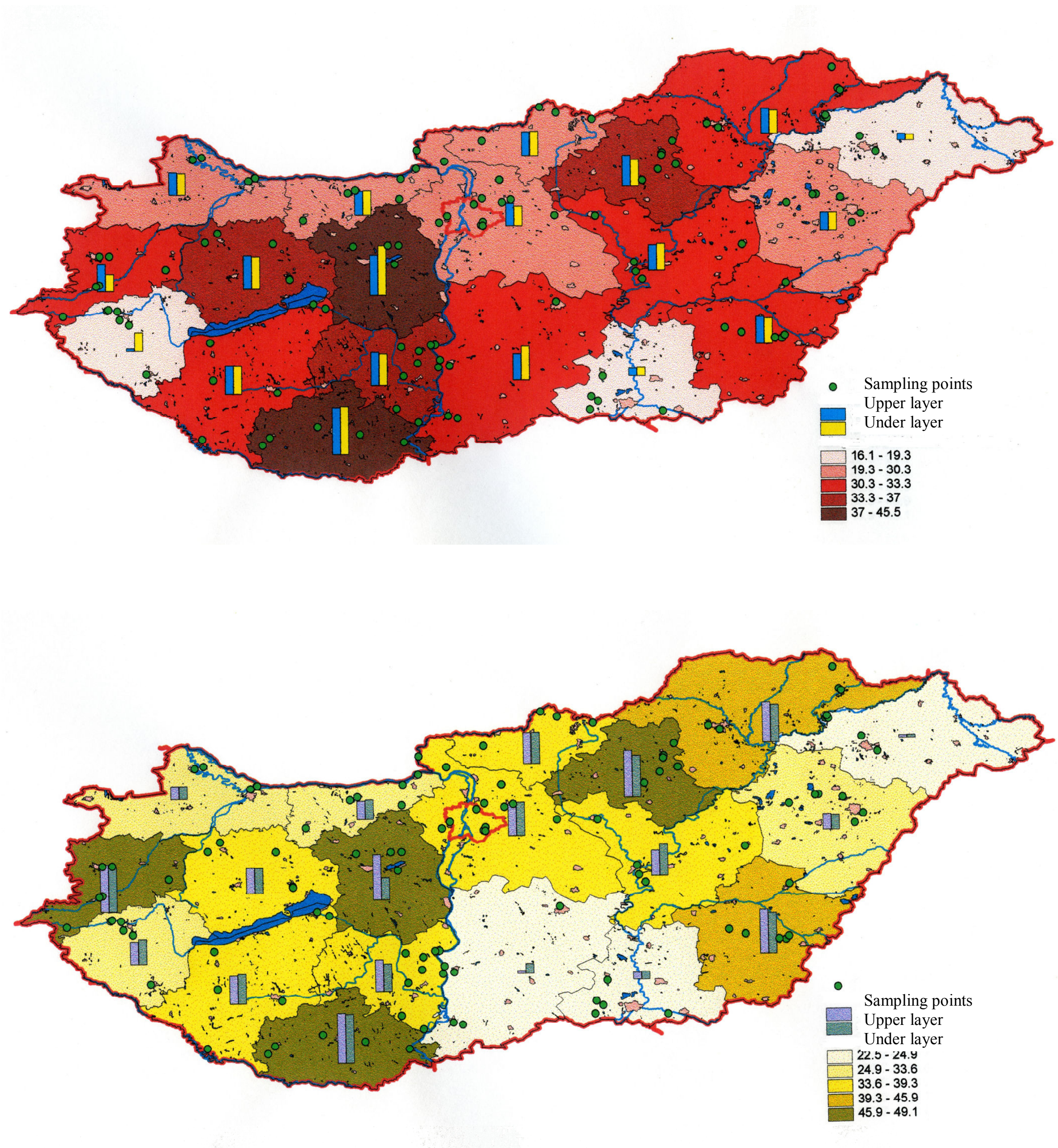

Figure 13. Spatial distribution of uranium (above) and thorium in Hungarian soils.

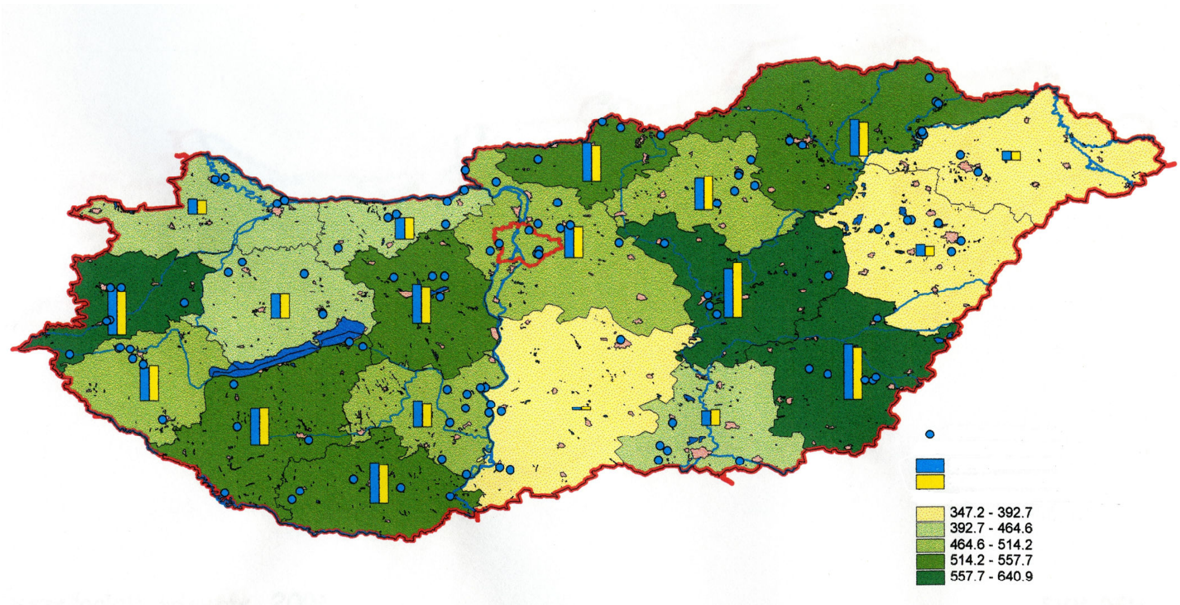

Figure 14. Spatial distribution of ^{40}K in Hungarian soils.

4.2. Recommended guideline levels for foodstuffs

My opinion is that the most important approach is in the establishing guideline level system for foodstuffs that the point of view is the immission, which means from the practice side, if once a higher concentration of an isotope was found in some kind of foodstuff there has to be a legal base of the immediate withdrawal and starting the investigation in one regulatory hand without waiting for example for the action of another authority belonging to another ministry. Foodstuffs became one of the most vulnerable and strategic substances because of their essentiality and worldwide trade and free circulation. It is time to give up the follow-up regulation system and create one for foodstuffs applicable for normal and prolonged emergency (longer than 1 year) situation, taking into account the risk as low as possible. The legislation has to be in line with ICRP main recommendations, the base of regulation should be the additional dose above the background level, therefore three stages of guideline levels are necessary:

1. background level: defined for a given country or environment for natural and artificial radioisotopes, ,
2. tolerance level applicable for normal situation: the base point is considering the totally immission situation - result of earlier releases, the exposure expected long-term, chronic type,
3. acceptance level applicable for prolonged emergency situation (longer than 1 year): when there is no shortage in any kind of foodstuff.

The type of legislation has to be based on the additional dose that is why necessary to know well the background level of different kind of radioisotopes. Time is perfect to determine and laid down these values in Hungary recently there were any accidental release affecting the state of the environment and sufficiently long time elapsed after the Chernobyl accident, as it is described in chapter 4.1.

4.2.1. Types of foodstuffs

The types of foodstuffs brought under regulation should be considered. According to the fast changes of the production of food and fast changes in the up-to-date habits and food consumption fashion, moreover increasing number of members of population living in developed countries requiring special diet, it is better to give derived limits generally for foodstuffs without picking up some recently important types. Same of the above mentioned considerations are valid for infants as well, considering that milk not the only sufficient food for them, especially those who are suffering from lactose allergy. Water and different kind of beverages regardless containing alcohol or not are qualified in this term also as foodstuffs here. Nobody can follow the consumption habits and the actual fashion of different kind of beverages, having main raw material of water consumed for example by young generation. There is no reason to pick up water and treat it separately, because it is used widely during the food production and as a raw material. Studying the available statistics of food consumption 550 kg/year consumption for adults and 200 kg/year for children younger than 1 year were accepted for the calculation according to FAO-WHO recommendation (CAC/GL 5-2006). While water and foodstuffs are treated together the annual water consumptions have to be added to these values. There is another reason to bring the water under the same regulation as the foodstuffs, in the developed countries mineral water consumption is in fashion nowadays, containing sometimes rather high concentration of natural radioisotopes according to the bedrock surrounding the well. The worldwide average consumption from water and different beverages shows a very divergent picture, it is rather hard to define a reliable consumption rate for the calculations. One of the highest water consumption values were reported in Safety Report Series No. 19 (IAEA, 2001), 0.6 m^3/year of adults and 0.26 m^3/year of infants. According to the results of different surveys in Europe and other parts of the world the real value might be much less. The results of surveys about the drinking water consumption included usually less than hundred people were not accepted here (Levallois et al, 1998; Westrell et al, 2006), the result of two other independent surveys, an American (Ryan et al) and an English (Ipsos MORI) were used for the calculation, 0.6 l/day were found as median consumption value from a sample of more than thousand people. Important to mention that WHO summarised the consumption habits in 12 classes which represents the consumption of the different populations of the world. In Hungary Ministry of Agriculture and Rural Development is responsible for the foodstuffs generally, moreover there is a requirement from European Union to have an integrated concept for the national food control system (178/2002/EC) and an integrated farm-to-table concept. This generalisation may bring some kind of careful deliberateness into the calculations. According to the last mentioned EU regulation member states should inform the Commission and each other about recorded cases of such products placed on the Community market exceeding the maximum permitted levels through the community Rapid Alert System for Food and Feed (the Hungarian contact point is our directorate).

My recommendation contains a specific degree of deliberateness, using the mentioned consumption rates and supposing the equally contaminated food for an individual. This means, that the ingestion dose of the base of the derivation is regarded as a maximum value, according to the differences in consumption habits less dose can be got. According to the Hungarian consumption habits (consumption statistic from the Statistical Yearbook of Hungary 2005, published in 2005: statistics were derived from the survey of households), supposing an average man consuming the same amount as it is in the statistics, if only the cereals are contaminated this man will have the ingestion dose one fifth than the guideline level, if vegetables (including potato) is contaminated the ingestion dose is the same as in case of cereals, if meat or milk is contaminated the dose will be one tenth of the guideline level for the individual,

In case of minor foodstuff that are consumed in small quantities, such as spices, that represent a small percentage of the total diet and hence a small addition to the ingestion dose, the guideline levels are increased by multiplication with a factor of 10; the current legislation is acceptable, minor foodstuffs are listed in the 618/89/Euratom by CN-codes (Combined Nomenclature).

4.2.2. Radionuclides of interest

Next important question under consideration is what kind of radionuclides should be regulated. There is no doubt, that the first group contains radionuclides which can be released from different nuclear installations, radiological applications and consequence of medical treatments (of course not for the person who has benefit from the treatment). The second group is natural isotopes, which could not be excluded, because of the possibility that natural isotopes can be technologically enhanced in different additives or might be a tool for a radiological attack. Malicious event would most likely be targeted at a public area. Free trade of food and feed and the inventiveness of producers give sometimes hard task for controlling authority. Regulation should prevent even the possibility of illicit actions, not allowed to make rules and regulation as a follow up action, just after a serious attack occurred. Unfortunately there are even some nuclear materials without official control according to IAEA Nuclear Security Series No. 6 dealing with combating illicit trafficking in nuclear and other radioactive material (IAEA, 2007). There is a summary in the book about the packages ranging from 0.4 g to 3 kg of highly enriched uranium were found in the period of 1993-2006. Radionuclides brought under regulation have to be a large group also from the media point of view, because members of the public, the media and political representatives have become concerned about the possible malicious use of sources and devices containing radioactive materials. There is a strong anxiety to the potential diversion of nuclear weapon material and its use in improvised, home-made nuclear devices. In all recently available regulation regarding the foodstuffs group the radionuclides according to different considerations (see above Table 3 and 4). Considering the possible legal consequences of the case of incompliance I find it important to recommend derived limit for foodstuff given in definite numerical values for each of the isotopes concerned in spite of forming any kind of groups, to be unambiguous for anybody not only for the experts. This approach can make easier the withdrawal procedure and clarifies the controversial situations.

To perform realistic calculations of radiation exposure of the population via ingestion of foodstuffs the nuclide inventory of air-borne, gaseous and liquid-form releases are required. These nuclide inventories should reflect the characteristic composition of the releases from normal operation. In Europe the following reactor types are in use: PWR – Pressurised Water Reactor, BWR – Boiling Water Reactor, GCR – Gas Cooled Reactor and AGR – Advanced Gas Reactor. The release of radioactive matter is taken into consideration as the emission of liquid, aerosol-related or gaseous matter from facility by way of planned pathways and routes; gaseous release through the stack, liquid release via waste water, machine-house water, auxiliary coolant water, main coolant water and outlet water. Determining the inventory of radioactive matter consists of the identification and determination of the radioactivity of radionuclides released a predetermined planned way, certain legal limits of detection are to be applied for the activity determinations. Continuous monitoring of the stack emissions and liquid waste from nuclear installations is the task of the owner of the nuclear installations.

In nuclear reactors different radionuclides are produced by nuclear fission, neutron capture and radioactive decay depending on the fuel type and the composition of elements in the materials exposed to the neutron flux. The main processes are

- fission, activation and decay in the fuel element,
- activation of structural materials,
- activation of the air around the reactor pressure vessel,
- activation of the primary cooling media (with additives and chemical impurities),
- transported corrosion and erosion products by the cooling media.

Obviously a large portion of the radionuclides is retained by the various cleaning systems installed inside the facility.

However the released activity of the noble gases is the highest; from the point of view of the transport through the food chain they have no effect. Tritium and ^{14}C releases have also high portion in the release. Unfortunately there is a general lack of monitoring data of ^{14}C in foodstuffs, even in plants (Hungarian monitoring network is prepared for this task).

The radionuclides compositions listed below were compiled from the publications of European Commission Radiation Protection 129 and 143, and Csom, 1997. The main considerations were the following:

- basis of the compilation was the EU discharge database,
- only those radionuclides are considered, which has larger than 0.01% contribution to the total activity.

In the samples affected by the atmospheric releases of nuclear power plants the following radionuclides may occur: H-3, C-14, Na-24, S-35, Cr-51, Mn-54, Co-57, Co-58, Fe-59, Co-60, Zn-65, Se-75, As-76, Sr-89, Sr-90, Nb-95, Zr-95, Mo-99, Ru-103, Ru-106, Cd-109, Ag-110m, Ag-111, Sb-122, Sb-124, Sb-125, Te-127, Te-129, I-131, Cs-134, Cs-135, Cs-136, Cs-137, Ba-140, La-140, Ce-141, Ce-144, Eu-154. From the liquid discharges of nuclear power plants the above mentioned radionuclides (excluding As-76, Cd-109, Te-127 and Te-129) the following radionuclides are expected in the environment: Be-7, Ca-45, Sc-46, Mn-56, Fe-55, Ni-59, Ni-63, Cu-64, Ag-108m, Sn-113, Te-123m, Ba-131, Pr-144, Pm-147, Eu-155, U-234, U-235, U-238, Pu-238, Pu-239, Pu-240, Pu-241, Am-241, Am-243, Cm-242, Cm-243, Cm-244.

In the samples affected by the atmospheric and liquid discharges of nuclear fuel reprocessing sites the following radionuclides may be expected: H-3, C-14, S-35, Mn-54, Fe-55, Co-57, Co-58, Co-60, Ni-63, Zn-65, Sr-89, Sr-90, Zr-95, Nb-95, Mo-99, Ru-103, Ru-106, Rh-106, Sb-125, I-129, I-131, Cs-134, Cs-137, Ce-144, Pm-144, Pm-147, Eu-152, Eu-154, Eu-155, U-234, U-235, U-238, Np-237, Pu-238, Pu-239, Pu-240, Pu-241, Am-241, Cm-242, Cm-244, Cm-244. This isotope composition differs from the release of power plants, the volatile radionuclides are missing and more heavy isotopes are present.

Obviously a large portion of the radionuclides is retained by the various cleaning systems installed inside the fence of the nuclear reactor. The usual consideration is that only the long-lived isotopes need to take into account since the short-lived ones will have almost completely disintegrated before reaching the food chain, but do not forget about the possible contamination of the surface of fruits and vegetables, so short lived radionuclides also have importance. Some fruits and vegetables are consumed just after gathering them without removing peels, surface contamination is not always possible to remove efficiently, for example from the surface of raspberry. However the released activity of the noble gases is the highest from the point of view of the transport through the food chain they have no effect. Tritium and carbon-14 releases have also high portion in the release. Unfortunately there is a general lack of monitoring data of ^{14}C in foodstuffs. Tritium is determined from plants in the frame of monitoring in Hungary.

Before an activity release gives an actual ingestion dose, the deposition of isotopes, their passage into foodchain and food consumption rates has to be taken into account, many processes are involved to determine ingestion dose and much depends on the nature of release and behaviour of radionuclides. According to EUR 14179 EN and EUR 19841 EN dealing with source term for a large-break loss of coolant accidents an isotope composition could be compiled for the emergency situation which might result in a prolonged exposure situation. Iodine and noble gases are always considered during the licensing studies, some countries plan to extend the list of nuclides at least to include radiocaesium, and some of them already consider an extensive list of nuclides. Behind compilation of the list are the following considerations.

- taking into account an extended list of radionuclides in the calculation of radiological consequences is possible nowadays with the available computer techniques,
- some countries already consider an extended list of radionuclides,
- it is important for public acceptance to demonstrate that all possible radionuclides are considered, even if they might give negligible contribution to public dose,
- in the absence of specific evaluations of the behaviour of substances, assuming basic pH conditions in the containment water pool as required, the chemical form can be assumed as the following: nobles gases – no chemical or physical change,

they are not included of the calculation dealing with the incorporation into foodchain, 95 % of iodine in particulate form (mainly CsI), 4.85 % elemental iodine and HI, 0.15 % organic iodine, other isotopes are presumed being only in particulate form (EUR 19841 EN),

- all fission products should be considered,
- actinides should be considered.

Table 17. PWR releases into containment (fraction of core inventory, EUR 19841)

	Gap release	Early in-vessel	Ex-vessel	Late in-vessel
Noble gases: Xe, Kr	0.05	0.95	-	-
Halogens: I, Br	0.05	0.35	0.25	0.1
Alkali metals: Cs, Rb	0.05	0.25	0.35	0.1
Tellurium group: Te, Sb, Se	-	0.05	0.25	0.005
Barium, Strontium	-	0.02	0.1	-
Noble metals: Ru, Rh, Pd, Mo, Tc, Co	-	0.0025	0.0025	-
Cerium group: Ce, Pu, Np	-	0.0005	0.005	-
Lanthanides: La, Zr, Nd, Eu, Nb, Pm, Pr, Sm, Y, Cm, Am	-	0.0002	0.005	-

Some radionuclides present in nuclear waste having half life sufficiently long potentially to permit their migration even from deep geological containment to the surface environment: above all the usually mentioned Pu isotopes and ^{241}Am, ^{129}I has high mobility in all parts of biosphere, ^{99}Tc has rather high concentration factors for fruits and vegetables, ^{79}Se may accumulate in cereals and beef, ^{237}Np has high inhalation dose factor, ^{94}Nb cause high external dose rate and ^{135}Cs has also some risk.

Do not forget about the so called natural isotopes, which usually show in environmental and food samples natural origin, they also have to be brought under regulation because of the prevention of agricultural production on sites where technologically enhanced natural isotopes occur. These isotopes might serve tools in a violent attack, especially alpha-emitting ones (Ra, Po isotopes).

4.2.3. Guideline level for normal situations – tolerance level

Although the concept of action levels for chronic exposure situation is of more general application, so far an international consensus on numerical values is desirable. ICRP gave already the basis of this consideration. Apart from the difficulty of deciding when an individual or a collective dose is small enough to be disregarded for regulatory purposes, this constraint should be regarded as the basic level of protection to be attained in normal situations. The action level represent the level of dose where action to avert exposures and reduce doses is virtually certain to be justified. In all situations the constraint is complemented by the requirement to optimise the level of protection achieved, because some probability of health effects even at small increments of exposure to radiation above the natural background can not be excluded. That is the main principle of radiation safety, is the need to ensure that all exposures are as low as reasonably achievable. The minimum value of any constraints is 0.01 mSv effective dose from a single source and single isotope, associated with trivial risk of $5x10^{-7}$ in a year which is recommended by ICRP for members of the public from single dominant sources for all types of exposure situation that might be controlled. A negligible or trivial risk is so slight, that it can be ignored from any assessments, and there is no requirement for a written risk assessment. 0.1 mSv/year is the appropriate basis of the derivation of the tolerance level of foodstuffs. There are some important considerations behind choosing this value for the reference ingestion dose level for normal situations, the level of risk attached to this value is in the order of $5x10^{-6}$, which is higher than trivial level of risk, furthermore this order of magnitude of the effective dose is in the same order of few weeks of natural background. Never forget that there is a risk of harm even from very low doses, the fact that risks are small compared to natural sources cannot be used as justification for accepting them. According to ICRP recommendations 0.1 mSv/year additional dose is the dose consequence for public exposure for prolonged components from long lived radionuclides, “it would obviously prudent to impose additional restriction on the prolonged component of the annual individual dose”. Below this dose regulatory provision will produce little or no improvement in dose reduction, because the radiation sources give rise to small individual dose and protection could be regarded as optimised. Once more 0.1 mSv/year effective dose has to be understood as additional dose above the normal background and allow exposures to multiple sources.

Being the starting point of committed effective dose of 0.1 mSv/year, I think no need to apply any protection factor, derived limit for foodstuffs allow not exceeding the general dose limit of 1 mSv for public, even there are other type of source of exposure than ingestion of food. The decision rules have to be laid down, because every measurement result (including chemical preparation) is given with its uncertainty. These rules give a prescription for the acceptance or rejection of a foodstuff based on the measurement results, its uncertainty and the specification limit, taking into account the acceptable level of the probability of making a wrong decision. An acceptance and a rejection zone have to be determined, such that if the measurement result lies in the acceptance zone the product is declared compliant and if in the rejection zone it is declared non-compliant. EURACHEM/CITAC Guide published in 2007 (EURACHEM/CITAC, 2007) gives some guidance to establish this procedure. The upper limit of the rejection zone is at the tolerance level, *TL* plus a guard band *g*, where *g* is a multiple factor of the standard combined uncertainty (u) of the measurement. For the case when the distribution of the likely values of the measurand is approximately normal, $1.64u$ will give a probability of 5% and $2.33u$ implies a probability of 1% of the wrong decision. Without having a protection factor here at this guidance level, the only acceptable rule of the acceptance and rejection zone is the case of high confidence of acceptance, it means in a concrete case, that there is only one value for the decision limit depending on the uncertainty of the given measurement result, this decision value will be another concentration in case of another measurement of food, but the upper limit will be always the same. The recommended decision rule is the following: $L\text{-}2.33u$ is dedicated to decision limit, there is the probability of 99% of good decision of acceptance. Let's see an example: if the reference level prescribed in any kind of measure for ^{99}Mo is 10 Bq/kg, there is a food sample with the measured concentration of 8(2)Bq/kg, taking into account the uncertainty, decision limit is 5.33 Bq/kg, decision will be rejection, with other words non-compliance; but in case of the determined value is 8(0.5) Bq/kg, decision limit is 8.8 Bq/kg, decision is compliance.

For the determination of numerical values of the guidance level for normal situation rounding rule of the calculated values should be established. The usual rounding used in dosimetry (according to averaging the log scale values – to 10^n if the calculated value is below $3x10^n$ and rounded up to 10^{n+1} if above) could not applicable here, because these are derived concentration values and using this rounding the governing committed effective dose might be higher, than the start point was. Keeping one eye on the conservatism during the determination of the tolerance levels preferably levels should be given for one significant digit, applying the conservative rounding rule for the second digit, rounding down for every isotopes. Despite of the sensitivity against the harm of radiation depending on the age of the person, dose conversion factors are given for age groups, only one activity concentration is supposed to given as tolerance level, the minimum of the calculated value for less than 1 year old child and for adult. Not always the value for child is the smaller, because the consumption difference sometimes cause this interesting situation.

In case of some different radionuclides were measured in a given food sample, the general relative summing law should be applied,

$$\sum_{i=1}^{n} \frac{c_i}{TL_i - 2.33u_i} < 1, \qquad \text{Eq 4.3}$$

c_i: measured activity concentration of i isotope in foodstuff,

u_i: combined standard uncertainty of the c_i (described in Eq 4.1 and Eq 4.2),

TL_i: tolerance level for i isotope,

i…n: all radioisotopes found in a given food sample,

2.33: factor for belonging to 99% probability of good decision.

There is also a social motivation for introducing this level, the tolerance level in force, everybody should have right for the safe food, as 60-70% of the income of middle class families in developing countries being spent on food, determining the health status and also the welfare status of the family.

4.2.4.Guideline levels for prolonged nuclear or radiological emergency – acceptance level

Intervention in any kind of emergency situations shall be carried out on the basis of the international recommendations for intervention and action levels in terms of the activity concentration of radionuclides in foodstuffs or water. 1 mSv or less, applies to exposure situations where individuals receive exposures that may be of no direct benefit to them but the exposure situation may be of benefit to society. Reference levels in this band would be selected for situations where there is general information on the situation and environmental surveillance or monitoring or assessment and where individuals may receive information but no training. The limits on effective dose apply to the sum of doses due to external exposures and committed doses from internal exposures due to intakes of radionuclides. 1 mSv is a general dose limit for public exposure, it can serve as the base of determining the acceptance level of different isotopes in foodstuffs. This effective dose from intake of radionuclides is sufficient for the pregnant women being sensitive group of population also according to ICRP recommendations.

Radionuclides brought under the regulation should be same for the normal situations and emergency cases, there is no reason to change the list concerning foodstuffs and feedstuffs. It should be noted, however, that although many aspects of the possible emergency scenarios resulting from a radiological attack may be similar to those arising from radiological accidents, these two types of emergencies differ in many aspects. For example the cases where elevated level of threat may exist should be handled, like during main events such as sport occasions, controlling the food sold there and the vicinity of the interested place get high priority.

According the guidance of ICRP that in circumstances where there are planned discharges of long-lived radionuclides to the environment, planning assessments should consider whether build-up in the environment would result in the constraint being exceeded, taking account of any reasonable combination and build-up of exposures. Choosing 1 mSv/year as a base of dose consequence of ingestion pathway there is a need of applying the protection factor. My recommendation is 5 for infants and 3 for adults. These factors will produce only a small degree of conservatism, because it is likely that also another source exists, namely that the threat for the children is higher to get additional dose from different kind of sources:

1. inhalation,
2. external dose,
3. ingestion of contaminated foodstuff,
4. ingestion of some soil or other contaminated substances (not food),
5. skin-contact with the contaminated material.

This approach comes from their living habits, so their protection factor should be higher than the factor of adults, ingestion of contaminated material and skin-contact is less probable for adults.

The suggested decision rule is very simple in this case, a result implies non-compliance with an upper limit of the acceptance zone is the acceptance level (AL_i) and the suggested value of *g* is 2, which is equal the commonly used coverage factor of k=2 in case of forming expanded uncertainty of a given measurement. In this case 97.5% is the probability of good decision, which is regarded appropriate, because the conservatism is covered by using protection factors during the determination of acceptance level.

In case of foodstuff containing not only one, but more radionuclide the following decision rule is suggested for making decision in line with the equation above:

$$\sum_{i=1}^{n} \frac{c_i}{AL_i - 2u_i} < 1, \qquad \text{Eq 4.4}$$

c_i: measured activity concentration of i isotope in foodstuff,

u_i: combined standard uncertainty of the c_i (described in Eq 4.1 and Eq 4.2),

AL_i: acceptance level for i isotope,

i…n: different radioisotopes found in food sample,

2: factor for belonging to 97.5% probability of good decision.

What happens with the foodstuff containing a radionuclide between the tolerance level for normal situation and acceptance level? Usually recommended action is to produce either in food industry or just in households food with higher grade of processing, because usually only a part of original activity concentration remains in foodstuff with higher preparation level. Food processing factors are published in many literature among them in IAEA Technical Report Series 364 (which is now under amendment and the new version is expected at the end of 2008). The general assumption is that above the food processing factors the radioactive decay also produce decreasing, it might be important in case of ^{131}I content of the milk, producing milk powder and storing until total decaying might serve solution in case of shortage of milk product. It is important to have clear idea about the application of these procedures, there might be some cases when during the getting rid of unwanted radionuclides the essential elements also removed. It might be necessary to issue some guidance for public as it was done for example in Finland. When this procedure does not result in sufficient decrease of the concentration, it does not go below the guideline level for normal circumstances the organised and quick withdrawal is necessary in the case when there is no lack of main foodstuffs.

4.2.5. Summary

ICRP's multi-attribute approach to the selection of dose limits necessarily includes societal judgements applied to the many attributes of risk. These judgements would not necessarily be the same in all contexts and, in particular, might be different in different societies. The recommended derivation method contains a degree of precaution with using the worst case scenario that every foodstuff is evenly contaminated. After studying the different consumption habits of the world population, there is no other way if the goal is the international legislation. The general guidance should be sufficiently flexible to allow for national or regional variations, which means here, if the government rely on the consumption statistics and would like to have a national regulation for a national average man can do it and is possible to have regulation for radioisotope content of foodstuff distinguishing between the type of foodstuffs as well. I do not support this idea, because the individual variations are so big in the consumption habits, for example if somebody has to keep a diet because of some health reason or just vegetarian from conviction. On the other hand the consumption habits of the children in different ages also varying in a wide range, moreover their habits might be even immoderate sometimes just for several weeks. I believe that it is better to have a regulation containing some prudence avoiding the extreme cases, therefore it is important to have regulation as general as possible. The requirement of the legislation also the same, compliances are avoidable regarding the individual variations, a regulation has to protect anybody of a society keeping the freedom of the individuals regarding the living habits. Any such variation in the protection of the most exposed individuals are best introduced by the use of source-related dose constraints selected by regulatory authorities and applied in the process of optimisation of protection. The recommended levels were checked by several calculations and the result is the following:

- concentration values in Table 19 represent always the maximum of ingestion dose, 0.1 mSv/year in case of tolerance level, 0.20 mSv/year for children less than 1 year old and 0.33 mSv/year for adults taking the dose of 1 mSv/year as a base and applying the protection factor of 5 for children and 3 for adults;
- supposing a man with average consumption (statistical data from the Statistical Yearbook of Hungary 2005, issued in 2006) – when only cereals are contaminated the dose of individual will be one fifth of the base of the derivation, in case of milk the situation is the same; when meat or vegetable (excluding potato) is contaminated average man will get one tenth of the dose; when only fruits or potato are contaminated average man will get 9 % of the base of the derivation.
- knowing that the above mentioned cases are just theoretical examples, it is not likely that just one type of food is contaminated after an emergency situation, the dose might vary depending on the contamination period of the year, its duration and consumption habits 5-100 % of the dose taking the base of the derivation of the given level.

In certain circumstances if there is a shortage in food supply or there are other serious social or economic considerations higher, but optimised action levels for foodstuffs would be proper in place. However decision to apply higher action levels specified above for emergency situation shall be subject to justification of intervention and optimisation of the elevated levels. Action levels for the withdrawal and substitution of specific supplies of foods and drinking water in case of prolonged emergency situation might be different from the above described reference levels. They shall be specified in a national emergency plan, because every government knows well the given economy, it is the matter of the optimisation of cost benefit analysis, even if it does not seem an amazing procedure from the human health point of view. When choosing the less harmful way of food supply the decision makers should know the risk of their decisions, aided by an expert group. Site specific intervention levels may be higher than the generic optimised values having consideration of site specific or situation specific factors. These may include, among the others special transport problems, the existence of hazardous weather conditions, or presence of other more harmful chemicals with more severe health detriment.

The regulations for foodstuffs and feedingstuffs are based on additional dose from all possible sources above the background level, therefore I appointed the upper level of background level for foodstuffs and feedingstuffs, taking into account the monitoring results of the term following at least twenty years of accidental release free period. The upper level of the background was defined for artificial radionuclides according to lognormal distribution and for natural radioisotopes according to normal distribution. The activity concentration above this level requires investigation. During the evaluation of the monitoring dataset of the network it is essential to have a clear idea of the uncertainties attributed to the measurements of the detection of an isotope in a network level. Table 18 contains a very brief summary of the derivation method of the recommended tolerance and acceptance level for foodstuffs listed in Table 19. Radionuclides are marked according to the Hungarian legislation, Decree of the Minister of Environment, No 15/2001. (VI.6.) on Radioactive Releases to the Atmosphere and into Waters in the Course of Using Atomic Energy and their Monitoring and in other column the most likely radionuclides from accidental releases.

Table 18. Theory of the recommended guideline level system for radioisotopes in foodstuffs

Level (in Bq/kg food)	**Method of derivation**
Background level	Natural isotopes supposing normal distribution Artificial isotopes: supposing lognormal distribution
Tolerance level intended to use for normal situation	Basis – 0.1 mSv/year, minimum(children<1 year old, adult) Decision limit = tolerance limit – 2.33*measurand combined standard uncertainty (1% probability of wrong decision)
Acceptance level intended to use for emergency situation (for infants and for adults)	Basis – 1 mSv/year, protection factor 5 for children<1 year, 3 for adults Decision limit = acceptance limit – 2*measurand combined uncertainty (2.5% probability of wrong decision)

Table 19. Recommended guideline levels for radioisotopes in foodstuffs

Radioisotope	Tolerance level, Bq/kg	Acceptance level, Bq/kg		NA[1]	NL[2]	A[3]
		Child < 1 year old	Adult			
H-3	2000	5000	10000	x	x	
Be-7	1000	3000	10000	x	p	
C-14	200	400	700	x	x	
F-18	600	1000	8000	x		
Na-22	10	30	100	x	x	
Na-24	90	100	1000	x		
Si-31	100	300	2000			
P-32	10	20	100		x	
P-33	100	200	1000		x	
S-35	200	500	3000		x	
Cl-36	30	60	400		x	
Cl-38	200	400	3000			
K-42	60	100	1000	p		
K-43	100	200	1000			
Ca-45	30	60	600		x	
Ca-47	20	50	200		x	
Sc-46	30	60	200		x	
Sc-47	50	100	800		x	
Sc-48	20	50	200			
V-48	20	40	200			
Cr-51	900	1000	10000	x	x	
Mn-51	300	600	4000			
Mn-52	20	50	200			
Mn-52m	400	800	6000			
Mn-53	800	1000	10000			
Mn-54	60	100	600	x	x	
Mn-56	100	200	1000			
Fe-52	20	50	300			
Fe-55	40	80	1000	x	x	
Fe-59	8	10	200	x	x	
Co-55	50	100	400			x
Co-56	10	20	100			x
Co-57	100	200	2000		x	x
Co-58	40	90	500	x	x	x
Co-58m	1000	3000	10000			x
Co-60	6	10	100	x	x	x
Co-60m	10000	30000	200000			x
Co-61	400	800	5000			x
Co-62m	600	1000	9000			x
Ni-59	500	1000	6000		x	
Ni-63	200	400	2000		x	
Ni-65	100	300	2000			
Cu-64	600	1000	3000			
Zn-65	9	10	100	p	x	
Zn-69	900	1000	10000			
Zn-69m	200	500	1000			
Ga-66	20	50	300		x	
Ga-67	100	300	2000	x	x	

Radioisotope	Tolerance level, Bq/kg	Acceptance level, Bq/kg		NA[1]	NL[2]	A[3]
		Child < 1 year old	Adult			
Ga-68	200	500	4000	x	x	
Ga-72	30	60	400		x	
Ge-71	2000	5000	30000			
As-72	30	60	200		x	
As-73	100	200	1000			
As-74	30	60	300		x	
As-76	30	60	200	p	x	
As-77	100	200	1000			
Se-75	10	33	100	x	x	x
Se-79	8	10	100		x	x
Br-77	500	1000	4000		x	x
Br-82	90	100	800		x	x
Rb-84	10	30	100		x	x
Rb-86	10	20	100		x	x
Sr-85	40	80	700		x	x
Sr-85m	7000	10000	70000			x
Sr-87m	1000	2000	10000			x
Sr-89	9	10	100	x	x	x
Sr-90	1	2	10	x	x	x
Sr-91	60	100	600			x
Sr-92	90	100	1000			x
Y-90	10	20	100			x
Y-91	10	20	100			x
Y-91m	3000	7000	30000			x
Y-92	50	100	800			x
Y-93	20	40	300			x
Zr-93	100	500	300			x
Zr-95	30	70	400	p	x	x
Zr-97	10	30	200			x
Nb-93m	200	400	3000			x
Nb-94	20	40	200		x	x
Nb-95	70	100	700	p	x	x
Nb-97	400	800	6000			x
Nb-98	200	500	3000			x
Mo-90	100	300	1000			x
Mo-93	40	80	100			x
Mo-99	60	100	700		x	x
Mo-101	700	1000	10000			x
Tc-96	50	100	300			x
Tc-96m	3000	6000	30000			x
Tc-97	300	600	6000			x
Tc-97m	30	70	700			x
Tc-99	30	60	600		x	x
Tc-99m	1000	3000	10000	x		x
Ru-97	200	500	2000			x
Ru-103	40	90	500	p	x	x
Ru-105	100	200	1000			x
Ru-106	4	8	60	p	x	x
Rh-103m	7000	10000	100000			x
Rh-105	80	100	1000			x

Radioisotope	Tolerance level, Bq/kg	Acceptance level, Bq/kg		NA[1]	NL[2]	A[3]
		Child < 1 year old	Adult			
Pd-103	100	300	2000			x
Pd-109	50	100	700			x
Ag-105	80	100	900			
Ag-ll0m	14	20	100	x	x	
Ag-111	24	40	300			
Cd-109	10	30	200		x	
Cd-115	20	40	300			
Cd-115m	8	10	100			
In-111	100	200	1000	x	x	
In-113m	1000	2000	10000			
In-114m	6	10	100			
In-115m	300	700	5000			
Sn-113	40	80	500			
Sn-125	9	10	100			
Sb-122	10	30	200			x
Sb-124	10	20	100	p	x	x
Sb-125	30	60	300	p	x	x
Sb-126	10	30	100		x	x
Te-123m	10	30	300			x
Te-125m	20	50	400			x
Te-127	200	400	2000			x
Te-127m	8	10	100		x	x
Te-129	400	900	6000			x
Te-129m	7	10	100		x	x
Te-131	300	700	4000			x
Te-131m	10	30	200		x	x
Te-132	7	10	100		x	x
Te-133	400	800	6000			x
Te-133m	100	200	1000			x
Te-134	300	600	3000			x
I-123	100	300	2000	x		x
I-124	2	5	30		x	x
I-125	6	10	20	x	x	x
I-126	1	3	10			x
I-129	1	3	3		x	x
I-130	10	30	200			x
I-131	1	3	10	x	x	x
I-132	100	200	1000	x		x
I-133	6	10	100	x		x
I-134	300	600	3000			x
I-135	30	60	400	x		x
Cs-129	700	1000	7000		x	x
Cs-131	700	1000	7000		x	x
Cs-132	100	200	800			x
Cs-134m	1000	3000	20000		x	x
Cs-134	6	20	20	x	x	x
Cs-135	60	100	200			x
Cs-136	20	40	100		x	x
Cs-137	9	30	30	x	x	x
Cs-138	300	600	4000			x

Radioisotope	Tolerance level, Bq/kg	Acceptance level, Bq/kg		NA[1]	NL[2]	A[3]
		Child < 1 year old	Adult			
Ba-131	80	100	900		x	x
Ba-133	10	30	200		x	x
Ba-133m	80	100	800		x	x
Ba-135m	100	200	1000		x	x
Ba-140	10	20	100	p	x	x
La-140	10	30	200		x	x
Ce-139	100	200	1000		x	x
Ce-141	40	80	600	p	x	x
Ce-143	20	50	300			x
Ce-144	5	10	80	p	x	x
Pr-142	20	40	300			x
Pr-143	20	40	300			x
Nd-147	20	50	300			x
Nd-149	200	400	3000			x
Pm-147	90	100	1000			x
Pm-149	20	50	400			x
Sm-151	200	400	4000			x
Sm-153	40	80	500		x	x
Eu-152	20	40	300		x	x
Eu-152m	50	100	800			x
Eu-154	10	20	200	p	p	x
Eu-155	70	100	1000			x
Gd-153	100	200	1000			
Gd-159	50	100	800			
Tb-160	20	40	200			
Dy-165	200	500	3000			
Dy-166	10	30	200			
Ho-166	20	40	300		x	
Er-169	70	100	1000			
Er-171	80	100	1000			
Tm-170	20	40	300			
Tm-171	200	400	3000			
Yb-175	60	100	900			
Lu-177	50	100	800			
Hf-181	20	50	300			
Ta-182	20	40	200			
W-181	500	1000	5000			
W-185	70	100	900			
W-187	60	100	600			
Re- 186	10	30	200			
Re- 188	10	30	300			
Os-185	80	100	800			
Os-191	50	100	700			
Os-191m	300	600	4000			
Os-193	30	70	500			
Ir-190	30	60	300			
Ir-192	20	50	300		x	
Ir-194	20	40	300			
Pt-191	100	200	1000			
Pt-193m	60	100	900			

Radioisotope	Tolerance level, Bq/kg	Acceptance level, Bq/kg		NA[1]	NL[2]	A[3]
		Child < 1 year old	Adult			
Pt-197	70	100	1000			
Pt-197m	300	600	5000			
Au-198	30	60	400		x	
Au-199	70	100	900			
Hg-197	100	200	1000		x	
Hg-197m	200	400	2000			
Hg-203	20	40	200		x	
Tl-200	200	500	2000			
Tl-201	400	800	4000	x	x	
Tl-202	100	200	900			
Tl-204	20	50	300			
Pb-203	200	400	1000			
Pb-210	0.04	0.08	0.6		x	
Pb-212	2	4	70			
Bi-206	20	40	200			
Bi-207	30	60	300			
Bi-210	20	40	300			
Bi-212	100	200	1000			
Po-203	1000	2000	9000			
Po-205	900	1000	7000			
Po-207	700	1000	3000			
Po-210	0.01	0.02	0.3		x	
At-211	2	5	30			
Ra-223	0.06	0.1	4			
Ra-224	0.1	0.2	6		x	
Ra-225	0.04	0.09	4			
Ra-226	0.07	0.1	1	x	x	
Ra-227	300	600	5000			
Ra-228	0.009	0.01	0.6		x	
Ac-228	40	90	1000			
Th-226	70	100	1000			
Th-227	1	2	40			
Th-228	0.09	0.1	6		x	
Th-229	0.03	0.06	0.8			
Th-230	0.08	0.1	2		x	
Th-231	80	100	1000			
Th-232	0.07	0.1	1	x	x	
Th-234	8	10	100		x	
Pa-230	10	20	400			
Pa-231	0.02	0.05	0.6			
Pa-233	30	60	400			
U-230	0.4	0.8	7			
U-231	100	200	1000			
U-232	0.1	0.2	1		x	
U-233	0.8	1	8		x	
U-234	0.9	1	8		x	
U-235	0.9	1	9	x	x	
U-236	0.9	1	9		x	
U-237	40	80	500			
U-238	0.9	1	9	x	x	

Radioisotope	Tolerance level, Bq/kg	Acceptance level, Bq/kg		NA[1]	NL[2]	A[3]
		Child < 1 year old	Adult			
U-239	900	1000	10000			
U-240	20	50	300			
Np-237	0.1	0.3	3		x	x
Np-239	30	70	500		x	x
Np-240	300	700	5000			x
Pu-234	100	300	2000			x
Pu-235	10000	30000	200000			x
Pu-236	0.1	0.3	4			x
Pu-237	300	600	4000			x
Pu-238	0.08	0.1	1	x	x	x
Pu-239	0.08	0.1	1	x	x	x
Pu-240	0.08	0.1	1	x	x	x
Pu-241	6	10	90		x	x
Pu-242	0.08	0.1	1			x
Pu-243	330	600	5000			x
Pu-244	0.08	0.1	1			x
Am-241	0.09	0.1	2	x	x	x
Am-242	60	100	1000			x
Am-242m	0.1	0.2	2			x
Am-243	0.09	0.1	2			x
Cm-242	0.5	1	30	x	x	x
Cm-243	0.1	0.2	2			x
Cm-244	0.1	0.2	3	x	x	x
Cm-245	0.09	0.1	2			x
Cm-246	0.09	0.1	2			x
Cm-247	0.09	0.1	2			x
Cm-248	0.02	0.04	0.5			x
Bk-249	10	30	400			
Cf-246	6	10	100			
Cf-248	0.2	0.4	10			
Cf-249	0.03	0.07	1			
Cf-250	0.05	0.1	2			
Cf-251	0.03	0.07	1			
Cf-252	0.06	0.1	4		x	
Cf-253	3	6	300			
Cf-254	0.03	0.06	1			
Es-253	1	3	70			
Es-254	0.2	0.4	10			
Es-254m	5	10	100			
Fm-254	60	100	900			
Fm-255	10	20	100			

1: x - Release limits for airborne effluents in Supplement 2 of the Decree 15/2001. (VI.6.)
p – Nuclides additionally reported in the annual report of Paks NPP (Bujtás, 2006)
2: x - Release limits for liquid effluents in Supplement 2 of the Decree 15/2001. (VI.6.)
p – Nuclides additionally reported in the annual report of Paks NPP (Bujtás, 2006)
3: Nuclides might be in accidental situation of PWR (Table 17)

4.3. Recommended guideline levels for feedingstuffs

Contamination of food products derived from both farmed and wild animals can represent major routes of radiation dose to humans from both routine and accidental releases of radioactivity to environment. Radionuclide activity concentrations in animal food products depend primarily on the rate of intake, gastro-intestinal absorption and turnover in tissues for most radionuclides. Fortunately, the amount of available data are greatest for the three most important radionuclides, ^{131}I, ^{137}Cs, and ^{90}Sr so it is possible to provide reliable information for these radionuclides, for many other radionuclides only few data are available. Agricultural assessment for some parameter values is substantial, for example the daily intake of different kind of animals, reviews for stable elements which are the most appropriate sources for the parameter values, rather than single values arising from radioecological literature.

Animals can be contaminated by three different routes: through the skin, by inhalation, and, most importantly, by ingestion. Uptake through the skin is not usually an important route of contamination, inhalation is potentially more important than skin absorption. The most important transfer pathway to animals is the ingestion of contaminated feed, soil and drinking water. Intake via drinking water is generally a small contributor to the total radionuclide intake and normally restricted to soon after an accident. Radionuclide intake via soil might be significant, but the availability for absorption of soil associated radionuclides is generally low. Hence, it is the ingestion of contaminated feed, processes influencing absorption and retention that determine the radionuclide content of animals.

Department of Energy in United States issued a DOE Standard under the title of " A graded approach for evaluating radiation doses to aquatic and terrestrial biota in 2002 (DOE, 2002), their graded approach distinguish assembly, screening and analysis phase. Recently ICRP established a "Framework for Assessing the Impact of Ionising Radiation on Non-Human Species" (ICRP, Draft 4a, 2007) and declared that it is necessary to develop an approach related as closely as possible to the current system for human radiological protection. On the ICRP's website a draft version is available for comments about the reference plants and animals. Plants and animals were grouped according the following reference groups: large and small terrestrial mammal, aquatic bird, amphibian, freshwater fish, marine fish, terrestrial insect, marine crustacean, terrestrial annelid, large and small terrestrial plant, seaweed. According to the determining human consumption rates from animal products in Hungary I will deal with only domestic ruminants, pig and chicken. The document draw the attention that during the derivation of guideline levels for domestic animals do not forget about protection of the animals itself, however they are raised for human consumption.

As it was mentioned earlier the practice showed that there is not enough to regulate only radiocaesium content of fodder, ^{131}I and ^{90}Sr also can cause problem. There is a rather wide database for transfer factors, it is better to give guideline levels of all isotopes, the behaviour of which was already studied, including the stable element studies.

4.3.1. Acceptance level for feedstuff

According to the European human consumption habits ruminants, pigs, poultry are considered the most important domestic animals producing foodstuffs. In case of ruminants data for cow, sheep and goat was used for the calculation with milk transfer coefficient and meat transfer coefficients. The minimum values were taken from the calculation of feedingstuffs to milk transfer of different animals and the same procedure was followed in case of feedingstuffs to meat transfer, then the minimum from the two minima was accepted as a guideline level of a given isotope for ruminants. In case of pigs the situation is unequivocal, while only meat is consumed (but sometimes other parts, like bones are cooked as well). For the guideline level of feedingstuffs of poultry meat and egg transfer were taken into account and the minimum of two calculated feedingstuffs concentration was accepted. In every case the immediate consumption of animal product assumed as a worst case.

The basis of the calculation is the recommended acceptance level of foodstuffs for adults, because the primary goal is the protection of the health of public. The theory of protection of human being and in the same time of biota seems to be justified with this acceptance level system as well. Using internal dose conversion factor of deer from ICRP Biota (ICRP, Draft 4a, 2007) and applying to cow for example for ^{137}Cs, 8.2E-3(μGy/day)/(Bq/kg) and calculating with suggested acceptance level for feedingstuff, the dose for ruminants is 3.3 mGy/year, which is not so far from the basis point of the doselimit of public of 1 mSv/year, but it is far less than the planned doseconstraint for biota, 1 mGy/day (ICRP, Draft 4a, 2007) compared to 9 μGy/day. The internal dose of a lactating cow using the maximum permitted radiocaesium activity concentration for feed from the current regulation is 240 mGy/year, which also meet the recommended requirement of 365 mGy/year for biota (data from Table 5 and Table 6), however it is rather close to it. Compared with the human dosimetric models, where mathematical and voxel phantoms are used to represent humans of different ages and sexes, with various organs, the dosimetric models for biota remain much more simplified. Internal exposure results from the accumulation of radionuclides into the tissues and organs, and from radionuclides that may pass through the gut or otherwise temporarily enter cavities within an organism, and will depend on both the physical decay properties and characteristics of the radionuclide and the biological half life of the radionuclide at either a whole body or organ-specific level. Originally the ICRP Biota recommendations were designed for the part of the environment, where humans are not present continuously, I do not see any reason why not to apply it also for the domestic animals. The short calculation above underlines the fact that there is no need for using any additional safety factor in the derivation of guideline levels for feedingstuffs, because the starting point of protection of human health serves stricter considerations, than the recommended radiological protection of animals. This fact proves the view that standards of environmental control needed to protect humans to the degree currently thought desirable would ensure that other species were not put at risk. Up to now the usual approach was considering the environment only with regard to transfer of radionuclides through the foodchain for example or as a surrounding environment, because this directly affects the radiological protection of humans. I agree that the entire environment needs to be considered, including areas where humans are absent, but only with the same degree where humans live.

4.3.2. Summary

Regulating the radioactivity content of feed only with the regulation of foodstuffs is conceivable, in the same frame and the same level of the regulation, because the basis of the calculation is the recommended acceptance level of foodstuffs for adults. There is a need here to make grouping according to animal type consuming the given feed and not the type of the feed (like alfalfa or grass or additives) because of the following reasons:

- the radioisotope content of the food of animal origin, the transfer into the tissue of animal from the fodder depend on the metabolism of given animal;
- farming habits are continuously changing depending on the price of the different kind of feed and on the possibilities of a farmer (having or not own field for producing some kind of feed), more general approach is giving the acceptance level for feed of animal type.

The primary goal is the protection of the health of public and in the same time the protection of biota also realised. Derivation of the acceptance level for feed from the acceptance level of food of animal origin gives stricter regulation, than the recommended dose levels for biota. The difference between the dose of animal via ingestion of the feed of the acceptance level is two order of magnitude less than the suggested dose limit of biota, therefore there is no need of other justification like addition rule or tolerance level, this difference is the guarantee of the proper protection of animals too.

Investigating the possible variances of consumption habits of public, generally no more than 35 % of the food is animal origin taking into account the worst case when all kind of animal product is contaminated, it means that no need to introduce tolerance level for fodder, because there is no need the stricter protection even from humans point of view in normal circumstances, it is not likely that these kind of products will determine the ingestion dose. Moreover there is a general tendency to lower the consumption of foodstuff of animal origin for some healthcare and ideological reason.

Table 20 contains a very brief summary of derivation method. The decision limits are formed the same way as in case of foodstuff. The listed activity concentrations in Table 21 have to be added the background level, speaking about additional dose, therefore the continuous monitoring of the environment is essential for knowing well and establishing the background levels for agricultural environment among them feedingstuffs also. Other parts like bone, gut and so on should be treated in the same way, this is not against the main principle of risk assessment.

Table 20. The method of the derivation for recommended acceptance level for feedingstuffs

Animal	**Method of derivation** **base: recommended acceptance level of foodstuffs**
Ruminants	Transfer from feedingstuff to meat: minimum concentration in feedingstuff from feed of cow, feed of goat, feed of sheep Transfer from feedingstuff to milk: minimum concentration in feedingstuff from feed of cow, feed of goat, feed of sheep Guideline level: minimum concentration in feedingstuff from transfer to meat and transfer to milk Decision limit = acceptance limit – 2*measurand combined uncertainty (2.5% probability of wrong decision)
Pig	Transfer to meat Decision limit = acceptance limit – 2*measurand combined uncertainty (2.5% probability of wrong decision)
Poultry	Minimum concentration in feed from transfer to meat and transfer to egg Decision limit = acceptance limit – 2*measurand combined uncertainty (2.5% probability of wrong decision)

Table 21. Recommended acceptance level of feedingstuff for domestic animals in normal situation and in case of prolonged emergency situation (1 year or longer)

Radioisotope	Acceptance level of feedingstuffs for			NA[1]	NL[2]	A[3]
	ruminants, Bq/kg	pigs Bq/kg	poultries, Bq/kg			
Be-7	7.0E+08					
Na-22	4.0E+02		2.0E+02	x	x	
Na-24	4.0E+03		2.0E+03	x		
P-32	2.0E+02	9.0E+02	2.0E+03		x	

Radioisotope	Acceptance level of feedingstuffs for			NA[1]	NL[2]	A[3]
	ruminants, Bq/kg	pigs Bq/kg	poultries, Bq/kg			
P-33	2.0E+03	9.0E+03	2.0E+04		x	
S-35	1.0E+03				x	
Cl-36	3.0E+03				x	
Cl-38	2.0E+04					
Ca-45	1.0E+03	1.0E+05	1.0E+04		x	
Ca-47	5.0E+02	4.0E+04	5.0E+03		x	
Cr-51	3.0E+05			x	x	
Mn-51	3.0E+05	3.0E+05	1.1E+06			
Mn-52	1.0E+04	1.0E+04	5.0E+04			
Mn-52m	4.0E+05	4.0E+05	1.7E+06			
Mn-53	7.0E+05	7.0E+05	2.0E+06			
Mn-54	4.0E+04	4.0E+04	1.0E+05	x	x	
Mn-56	7.0E+04	7.0E+04	2.0E+05			
Fe-52	2.0E+03		3.0E+03			
Fe-55	8.0E+03		1.0E+04	x	x	
Fe-59	1.0E+03		2.0E+03	x	x	
Co-55	2.0E+04		2.0E+04			x
Co-56	6.0E+03		6.0E+03			x
Co-57	1.0E+05		1.0E+05		x	x
Co-58	3.0E+04		3.0E+04	x	x	x
Co-58m	6.0E+05		6.0E+05			x
Co-60	6.0E+03		6.0E+03	x	x	x
Co-60m	1.0E+07		1.0E+07			x
Co-61	3.0E+05		3.0E+05			x
Co-62m	6.0E+05		5.0E+05			x
Ni-59	1.0E+04				x	
Ni-63	4.0E+03				x	
Ni-65	4.0E+03					
Zn-65	7.0E+01	2.0E+02	8.0E+02	p	x	
Zn-69	7.0E+03	2.0E+04	8.0E+04			
Zn-69m	7.0E+02	2.0E+03	8.0E+03			
Se-75	9.0E+03	1.0E+02	1.0E+02	x	x	x
Se-79	9.0E+03	1.0E+02	1.0E+02		x	x
Sr-85	1.0E+04	7.0E+04	2.0E+04		x	x
Sr-85m	1.0E+06	7.0E+06	2.0E+06			x
Sr-87m	2.0E+05	1.0E+06	3.0E+05			x
Sr-89	2.0E+03	1.0E+04	3.0E+03	x	x	x
Sr-90	2.0E+02	1.0E+03	3.0E+02	x	x	x
Sr-91	1.0E+04	6.0E+04	2.0E+04			x
Sr-92	2.0E+04	1.0E+05	3.0E+04			x
Y-90	1.0E+03					x
Y-91	1.0E+03					x
Y-91m	4.0E+05					x
Y-92	1.0E+04					x
Y-93	4.0E+03					x
Zr-93	5.0E+06		2.0E+07			x
Zr-95	6.0E+06		2.0E+07	p	x	x
Zr-97	3.0E+06					x
Nb-93m	3.0E+07		4.0E+07			x
Nb-94	2.0E+06				x	x
Nb-95	8.0E+06		1.0E+07	p	x	x

Radioisotope	Acceptance level of feedingstuffs for			NA[1]	NL[2]	A[3]
	ruminants, Bq/kg	pigs Bq/kg	poultries, Bq/kg			
Nb-97	7.0E+07		8.0E+07			x
Nb-98	3.0E+07		4.0E+07			x
Mo-90	5.0E+04		2.0E+04			x
Mo-93	5.0E+03		2.0E+03			x
Mo-99	3.0E+04		1.0E+04		x	x
Mo-101	5.0E+05		2.0E+05			x
Tc-96	5.0E+03		9.0E+02			x
Tc-96m	5.0E+05		9.0E+04			x
Tc-97	1.0E+05		1.0E+04			x
Tc-97m	1.0E+04		2.0E+03			x
Tc-99	1.0E+04		1.0E+03		x	x
Tc-99m	1.0E+05		3.0E+04	x		x
Ru-97	8.0E+04	3.0E+05				x
Ru-103	2.0E+04	9.0E+04		p	x	x
Ru-105	4.0E+04					x
Ru-106	2.0E+03			p	x	x
Rh-103m	5.0E+07					x
Ag-105	1.0E+06					
Ag-ll0m	1.0E+05			x	x	
Ag-111	4.0E+05					
Cd-109	2.0E+03		1.0E+03		x	
Cd-115	4.0E+03		2.0E+03			
Cd-115m	1.0E+03		7.0E+02			
Sb-122	2.0E+04					x
Sb-124	1.0E+04			p	x	x
Sb-125	3.0E+04			p	x	x
Sb-126	1.0E+04				x	x
Te-123m	5.0E+03		8.0E+02			x
Te-125m	7.0E+03		1.0E+03			x
Te-127	3.0E+04		5.0E+03			x
Te-127m	1.0E+03		2.0E+02		x	x
Te-129	1.0E+05		1.0E+04			x
Te-129m	1.0E+03		2.0E+02		x	x
Te-131	7.0E+04		1.0E+04			x
Te-131m	3.0E+03		5.0E+02		x	x
Te-132	1.0E+03		2.0E+02		x	x
Te-133	1.0E+05		1.0E+04			x
Te-133m	1.0E+04		2.0E+03			x
Te-134	5.0E+04		8.0E+03			x
I-123	5.0E+03	1.0E+05	1.0E+04	x		x
I-124	8.0E+02	2.0E+03	1.0E+02		x	x
I-125	5.0E+02	1.0E+03	1.0E+02	x	x	x
I-126	2.0E+02	7.0E+02	5.0E+01			x
I-129	8.0E+00	2.0E+02	1.0E+01		x	x
I-130	9.0E+02	1.0E+04	1.0E+03			x
I-131	5.0E+01	7.0E+02	5.0E+01	x	x	x
I-132	2.0E+03	7.0E+04	5.0E+03	x		x
I-133	2.0E+02	7.0E+03	5.0E+02	x		x
I-134	8.0E+03	2.0E+05	1.0E+04			x
I-135	1.0E+03	2.0E+04	2.0E+03	x		x
Cs-129	1.0E+04	1.0E+04	4.0E+04		x	x

Radioisotope	Acceptance level of feedingstuffs for			NA[1]	NL[2]	A[3]
	ruminants, Bq/kg	pigs Bq/kg	poultries, Bq/kg			
Cs-131	1.0E+04	1.0E+04	4.0E+04		x	x
Cs-132	1.0E+03	1.0E+03	4.0E+03			x
Cs-134m	4.0E+04	4.0E+04	1.1E+05		x	x
Cs-134	4.0E+01	4.0E+01	1.0E+02	x	x	x
Cs-135	4.0E+02	4.0E+02	1.0E+03			x
Cs-136	2.0E+02	2.0E+02	5.0E+02		x	x
Cs-137	7.0E+01	6.0E+01	1.0E+02	x	x	x
Cs-138	9.0E+03	9.0E+03	2.0E+04			x
Ba-131	1.0E+04		1.0E+04		x	x
Ba-133	3.0E+03		2.0E+03		x	x
Ba-133m	1.0E+04		9.0E+03		x	x
Ba-135m	1.0E+04		1.0E+04		x	x
Ba-140	1.0E+03		1.0E+03	p	x	x
Ce-139	3.0E+06				x	x
Ce-141	1.0E+06			p	x	x
Ce-143	9.0E+05					x
Ce-144	2.0E+05			p	x	x
Pm-147	2.0E+07					x
Pm-149	9.0E+06					x
W-181	1.0E+06					
W-185	2.0E+05					
W-187	1.0E+05					
Pb-203	1.0E+04					
Pb-210	1.0E+01				x	
Pb-212	1.0E+03					
Po-203	2.0E+06		4.0E+04			
Po-205	2.0E+06		3.0E+04			
Po-207	9.0E+05		1.0E+04			
Po-210	9.0E+01		1.0E+00		x	
Ra-223	3.0E+02					
Ra-224	5.0E+02				x	
Ra-225	5.0E+02					
Ra-226	8.0E+01			x	x	
Ra-227	4.0E+05					
Ra-228	5.0E+01				x	
Th-226	3.0E+04					
Th-227	1.0E+03					
Th-228	2.0E+02				x	
Th-229	2.0E+01					
Th-230	6.0E+01				x	
Th-231	3.0E+04					
Th-232	3.0E+01			x	x	
Th-234	3.0E+03				x	
U-230	2.0E+02		8.0E+01			x
U-231	3.0E+04		1.0E+04			x
U-232	3.0E+01		1.0E+01		x	x
U-233	2.0E+02		9.0E+01		x	x
U-234	2.0E+02		9.0E+01		x	x
U-235	2.0E+02		1.0E+02	x	x	x
U-236	2.0E+02		1.0E+02		x	x
U-237	1.0E+04		5.0E+03			x

Radioisotope	Acceptance level of feedingstuffs for			NA[1]	NL[2]	A[3]
	ruminants, Bq/kg	pigs Bq/kg	poultries, Bq/kg			
U-238	2.0E+02		1.0E+02	x	x	x
U-239	3.0E+05		1.0E+05			x
U-240	1.0E+04		3.0E+03			x
Np-237	3.0E+04				x	x
Np-239	6.0E+06				x	x
Np-240	6.0E+07					x
Pu-234	1.0E+07		6.0E+07			x
Pu-235	1.0E+09		6.0E+09			x
Pu-236	2.0E+04		1.2E+05			x
Pu-237	2.0E+07		1.0E+08			x
Pu-238	6.0E+03			x	x	x
Pu-239	6.0E+03			x	x	x
Pu-240	6.0E+03			x	x	x
Pu-241	5.0E+05		2.0E+06		x	x
Pu-242	6.0E+03					x
Pu-243	3.0E+07		1.0E+08			x
Pu-244	6.0E+03					x
Am-241	2.0E+02		2.0E+03	x	x	x
Am-242	2.0E+05		1.2E+06			x
Am-242m	5.0E+02		2.0E+03			x
Am-243	5.0E+02		2.0E+03			x

1: x - Release limits for airborne effluents in Supplement 2 of the Decree 15/2001. (VI.6.)
p – Nuclides additionally reported in the annual report of Paks NPP (Bujtás, 2006)
2: x - Release limits for liquid effluents in Supplement 2 of the Decree 15/2001. (VI.6.)
p – Nuclides additionally reported in the annual report of Paks NPP (Bujtás, 2006)
3: Nuclides might be in accidental situation of PWR (Table 17)

5. Radioecological vulnerability

When considering the radioecological impacts of artificial radionuclides it is important to be able to make forecast the way in which radioactive releases of different types and magnitudes will behave in specific ecosystems over a variety of time scales. High activity, acute releases of radionuclides, such as those from nuclear accidents, may result in a different range of impacts within specific ecosystems than low activity, chronic releases, such as those from radioactive waste repositories. Understanding and forecasting these impacts are difficult when considering complex ecological systems, but this can be facilitated by the construction and use of reliable models, based on data from as wide a variety of sources as possible. Such models, however, must be validated if they are to be used in making long-term assessments.

5.1. Transfer factors

Soil-plant transfer is an important link for the entry of radionuclides into the human food chain and transfer factor is widely used as a parameter for predicting plant uptake in radiological assessment models. In general, the transfer of radionuclides from soil to plant is relatively low compared to direct deposition on the foliage, there might be 2-3 order of magnitude difference between them in case of wheat (Scott, E.M. Ed, 2003). The uptake of radionuclides via roots may become a relevant pathway only for radionuclides with half-life at least 1 year. The main problems with this factor is that speciation is not taken into account instead of the total activity of soil is used. In cases where the determination of the soil activity is difficult, an aggregated transfer factor is applied (Bq/kg plant)/(Bq/m^2 soil). Although there are many exceptions, in general, transfer factors are higher for sandy soils than for soils rich in clay, and transfer factors for generative plant parts (grains, tubers) are lower than those for vegetative plant parts (leaves, stems). Transfer factors have considerable variability as well as spatial and temporal distribution of the soil-plant system. The processes affecting plant uptake for any radionuclide would thus be controlled by two sets of factors: those associated with the physiological requirements of the plant, and the physico-chemical factors influencing the distribution of radionuclides between the soil solid phase and soil solution. The uptake of radionuclides as well as that of nutrients - from soil via root is influenced by soil type, pH, sorption capacity, clay content, organic matter and management practice, but it is hard to quantify them separately, usually an empirically derived soil-plant transfer factor is applied. There are some general assessments:

- Soil to plant transfer of ^{137}Cs has been found to increase with increasing organic matter content, whereas the opposite trend has been observed for ^{90}Sr, for Pu organic matter content did not have an effect.
- Soil pH (3.9-8.4) did not affect radiocaesium transfer, whilst transfer factor for radiostrontium decreased with increasing pH.
- The nutrient status of the soil has important effects on radiocaesium and radiostrontium transfer through interactions with the analogues K and Ca.
- Soil moisture content also appears to have an important effect on Cs and Sr uptake.

The advantages of using transfer factors that refers to the soil horizons, from which nutrients and radionuclides are mainly taken up:

- The variations of transfer factors for specific soil horizons is significantly lower compared to other definitions, notably in case of a non-uniform vertical distribution of radionuclides in soil.
- Transfer factors for specific horizons offer a conceptual advantage for predictive modelling, because there is an evidence that they will stay fairly constant in the future.
- Transfer factors for specific horizons are a direct measure of the availability of radionuclides for uptake by fungi or green plants. However it is very difficult to localise fine roots and fungal mycelia in situ in a species-specific manner. In the case of radiocaesium the isotopic ratio $^{134}Cs/^{137}Cs$ originating from Chernobyl could have been used for that purpose in Hungary about 15 years ago, when the amount of the former was detectable.

Remarkable fact is that plant roots strongly modify their soil environment while taking up their nutrients.

Sheppard et al (Sheppard et al, 1997) assessed the variability of transfer factor values for 23 naturally occurring elements for a single plant species, sweet lowbush blueberry (Vaccinium angustifolium) across a wide geographic range in Canada. The transfer factor values of various elements were correlated and cluster analysis showed groupings that reflected the periodic table of elements, which is not surprising because of the similarity in the chemical properties.

Speaking about the transfer factors never forget that it is the net result of two distinct processes, first one has geochemical nature and is governed by fixation and sorption ability of soils and the concentration of competitive ions, the second one is the physiological process of the plant related to the transfer of ions from the soil solution into the plant root, which also depends on the concentrations of other ions. There are several elements which have great importance from the point of view of plant contamination through the surrounding soil. One of the determining processes is the root uptake, the order of the availability of different elements generally is the following in decreasing order Sr > I > Ba > Cs,Ru > Ce > Zr,Nb > Pu.

Organic content of the soil usually inhibit the bounding of metallic elements to the soil, while sorption of caesium on clay is highly preferred process, there are several competitive reaction also for example high concentration of potassium ions could decrease this sorption. Usually plant-soil transfer factors are classified according to four main textural soil groups, organic, sand, loam and clay. Rather big variations were found within soil groups in the literature. A study carried out in the frame of SAVEC project (SAVEC, 2001) showed that clay and organic matter content and exchangeable potassium content explained 59% of variability in transfer values and pH had no statistically significant effect.

For the determination of most vulnerable area in Hungary from the soil-plant transfer I chose the evaluation of the ^{90}Sr results, because the chemical behaviour of strontium is not dependent on the time elapsed between the contamination and the investigation like in the case of caesium. Strontium has several stable isotopes ^{88}Sr (82.58%), ^{86}Sr (9.86%), ^{87}Sr (7.0%), ^{84}Sr (0.56%), its natural abundance in earth crust is 388 ppm. The most important long-lived isotope is beta-emitting ^{90}Sr isotope with half-life of 28.8 year. Behaviour of this element is similar to calcium, the amount of different anions in soil-solution play an important role in the sorption process of strontium. The distribution coefficient (amount of ^{90}Sr in solid phase / amount of ^{90}Sr in soil-solution) increases assuming the same anion-concentration according to the following order: $Cl^- < NO_3^- < SO_4^{2-} < F^- < HCO_3^- < CO_3^{2-} < PO_4^{3-}$.

Transfer factors (amount of ^{90}Sr in plant in Bq/kg fresh / amount of ^{90}Sr in soil in Bq/kg dry) were determined from the database of 10 years of monitoring. Two sampling points were constantly monitored in every county, and the range of the transfer factors are 0.04-0.7, the number of transfer factors were 59 close to lognormal distribution with kurtosis of 1.5 and skewness of 0.7, expected value is 0.11. According to Dixon test there is only one outlier in the population, 0.701. Studying the possible reasons of this high value Geographical Information System was used, with the soil maps with 1:100 000 measure, containing organic content, soil type, pH, land cover data layers. It turned out that this one is the only place in the country, where the soil type is clearly sand, other agricultural area are covered some kind of mixed soil. According to the transfer factors this sandy area in Hajdú-Bihar county could be regarded as a vulnerable area of the country in terms of agricultural production. The correlations with pH, organic content were also investigated, but in Hungarian circumstances there were no significant effect observed.

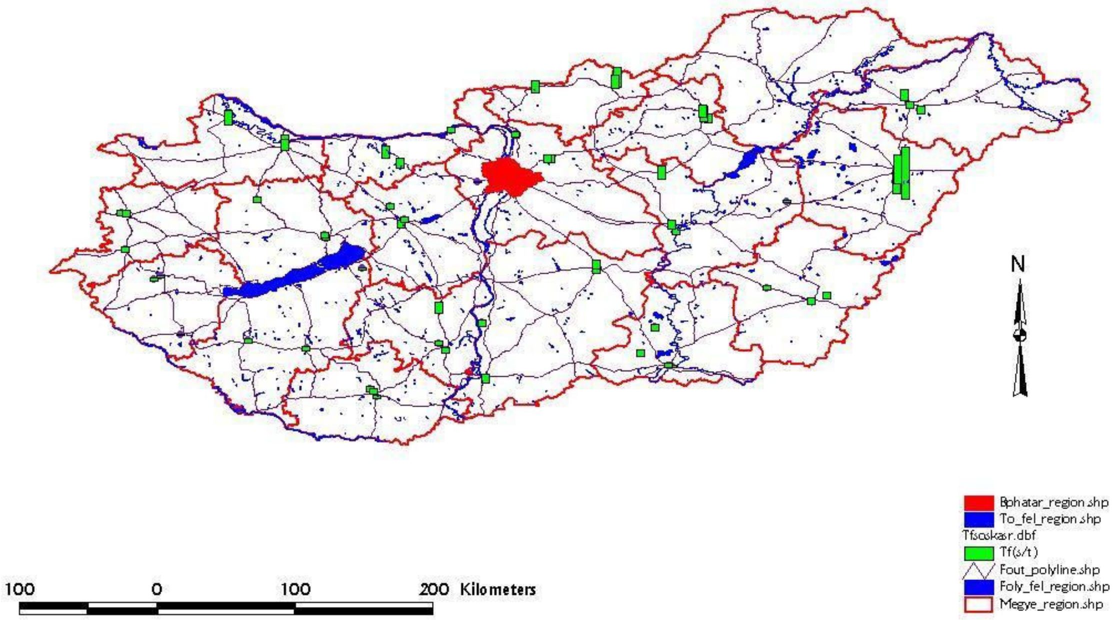

Figure 15. Spatial distribution of transfer factors of ^{90}Sr, results of 10 years of monitoring in the same places (derived from 1215 plant-soil sample pair)

The most important transfer pathway to animals is the ingestion of contaminated feed, soil and drinking water. Among them the ingestion of contaminated feed and absorption and retention determine the radionuclide content of animals. From the detectable artificial isotopes ^{137}Cs is the proper one for investigating the transfer between the fodder and food of animal origin, according to my interest. It has sufficiently long half-life and chemical properties also allow this investigation: alkali metal, reactive, easily soluble, it is the second most electropositive chemical element and has the second lowest ionization potential (after francium), bioavailability is the highest among the artificial isotopes on this stage of the foodchain, all amount entered into the gastrointestinal tract will be absorbed as well as ^{3}H, ^{14}C, ^{35}S, ^{99}Mo and I isotopes (IAEA TECDOC in preparation). Milk and fodder samples were taken monthly in the same time from the same farm in every county year by year. The fluctuation was studied by calculating the interval of ^{137}Cs activity concentration in milk from basic statistical parameters (minimum, maximum and median value) of specific activity database measured in fodder, using the well known equation of:

$$c_m = Q_F F_m c_g e^{-\lambda t} \qquad \text{Eq 5.1}$$

c_m: concentration of ^{137}Cs in milk, Bq/l

Q_F: the amount of feed consumed by the animal, 16 kg dry weight/day

F_m: transfer coefficient: the fraction of the animal's daily intake of the radionuclide that appears in each litre of milk in equilibrium, 0.0044 day/L

c_g: specific activity of ^{137}Cs in feed, Bq/kg dry weight

λ: decay constant of ^{137}Cs, 1/year

t: time interval between sampling and reference time, year

Regarding the dairy management factors Q_F value was taken from IAEA Safety Series No. 57 (IAEA, 1982). The composition of the consumed fodder during a year was taken into account by experimental way: the transfer coefficient used for the calculation is the result of the study carried out in the middle of 90's, when this coefficient was calculated from the database of our network containing the results of five years, namely 0.0044 day/L. Figure 16 shows the result of the comparison of calculated and observed concentration of ^{137}Cs in milk in 2001 as an example. Lines represent the calculated activity concentrations from the minimum, median and maximum values of the feedstuff dataset, as the equation shows. To make the figure more compact 4 series was formulated from the measured activity concentrations according to the quarters of the year, points are represents the observed values. Seasonal differences could not be observed, there is no separate group on the figure, which was expected from the small activity concentration range of the input data. Every year there was a very good agreement between the calculated minimum, median and maximum values of activity concentration of ^{137}Cs in milk and the observed value. The aim of this small vulnerability study was to find out the borders of the fluctuation regarding the transfer from fodder to milk, and to control the conditions of cow-keeping in the country. From the results it turned out that this part of food chain is stable in Hungary year by year from this point of view.

Applying the analogue model for fodder and meat, transfer factor of 0.02 day/kg was used and activity concentration were calculated from the same way, from the minimum, median and maximum concentrations of the fodder, lines shows these levels and points are the observed values on the figure as on the previous one. Conclusion from the result of Figure 17 could be drawn immediately, deer, roe deer and wild hog need more attention. This result comes from the living area of these wild animals. It is obvious that the radioactive contamination usually remains longer in forest environment than the other type of environments. There is no self-cleaning process or only very slow in the forest ecosystems.

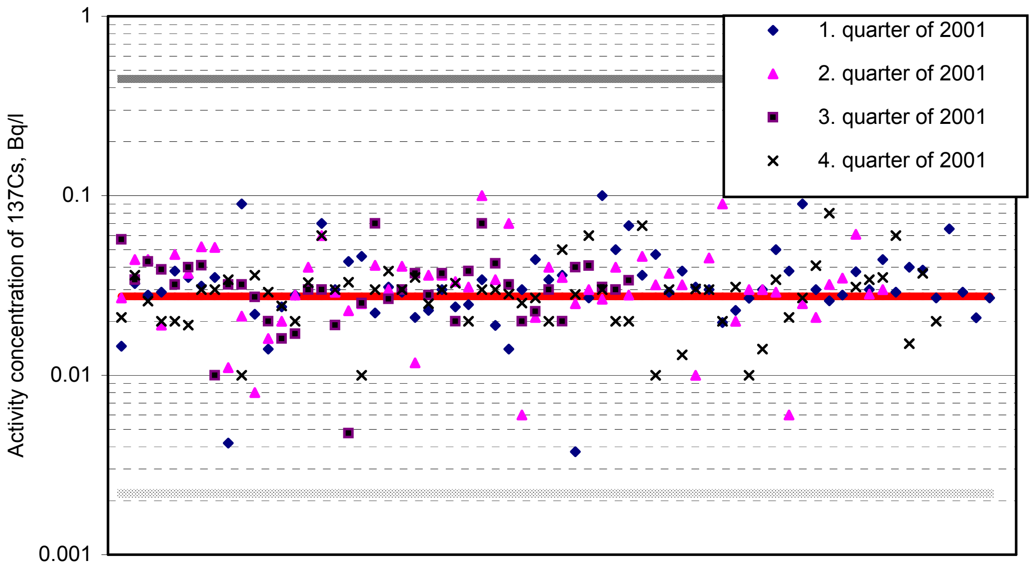

Figure 16. The predicted and observed activity concentration of ^{137}Cs in milk

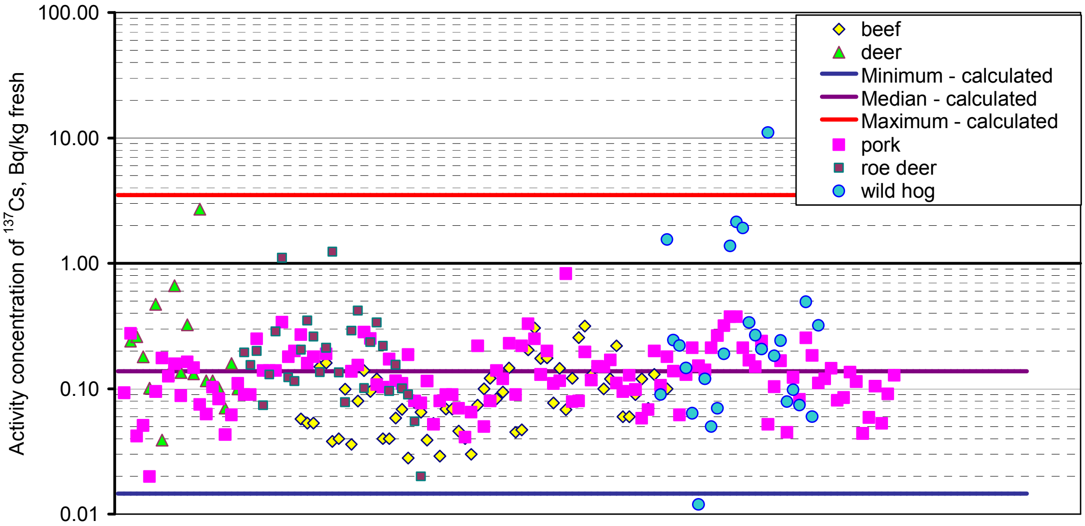

Figure 17. The predicted and observed activity concentration of ^{137}Cs in meat

The SAVEC project (SAVEC, 2001) provided an opportunity to re-evaluate post-Chernobyl data in Central European countries, namely Poland, Czech Republic and Hungary (Sanchez et. al., 2001), to select and verify data issued in statistical yearbooks and annual reviews of monitoring networks. For the general population ^{137}Cs intake estimated from whole body counting data showed a good correlation with intakes estimated from annual average consumption and from isotope content of different foodstuffs. The results regarding the transfer factors agreed well with values from other European countries. From investigations of SAVEC project and the above mentioned facts comes that in Hungary no area could be pointed as a vulnerable area regarding the caesium transfer from fodder to animal, therefore the food of animal origin across the country. As it was expected the meat of wild animals require special interest, especially the meat of wild hog. Because of the Hungarian way of life it was not possible to designate any vulnerable group of people in the society level, only very few people are regarded such kind, who lives or works in the forest. Nowadays 57% of the forest area of Hungary is in the governmental ownership, 42% is private and 1% belongs to the associations.

5.2. Residence time

The Network has developed a rather large database since the 80's. Big effort was made to validate data of former periods. Although annual reports are available from the second half of the 60's these early data give only a rough estimate about the activity concentration of isotopes, because of the techniques and methods available that time. Fortunately some samples are preserved in county stations, so these samples were measured by up-to-date equipment and methods and data were re-evaluated. Based on this practice the long-term countrywide averages were formulated.

Milk, fodder, sorrel-spinach, meat, wheat, apple databases were extended in this way, ^{137}Cs and ^{90}Sr content of the samples were re-evaluated; Figure 18 shows the annual national average of specific activity of ^{137}Cs in fodder and milk as well as ^{90}Sr in milk. It shows that the specific activity of ^{137}Cs increased 2-3 orders of magnitude in the year of Chernobyl accident and after 4-7 years, depending on the food type the value went back to the level before the accident. The results give the similar figures in the case of other sampling media too. So called ecological half life of these foodstuffs could be determined for caesium in two time steps, for example for the milk the decrease of the activity concentration within the first 4 years after the Chernobyl accident gives 0.4 years half-time, while the second period could be described with 29.4 years half-time.

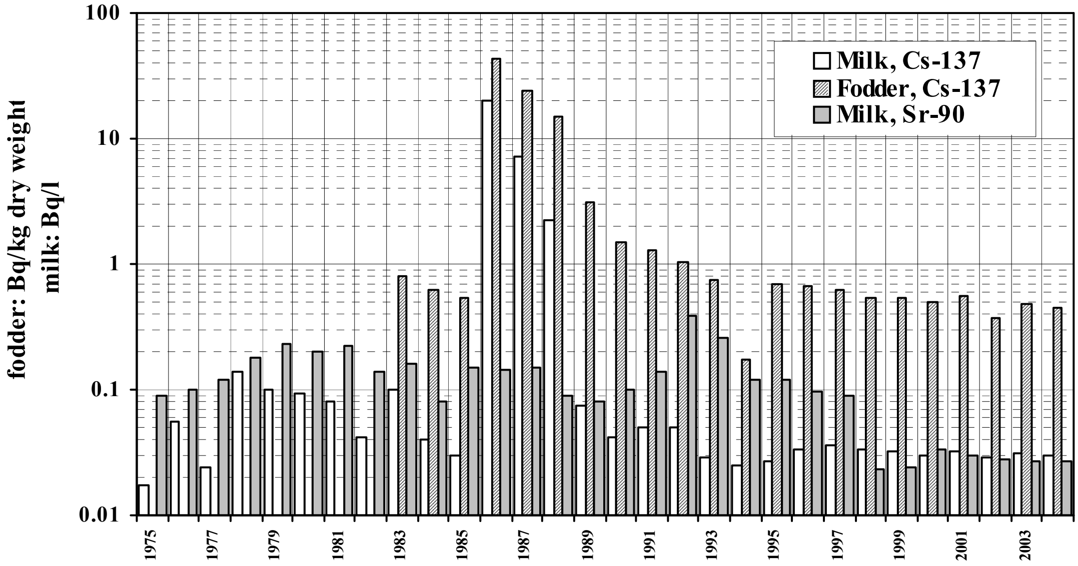

Figure 18. Changes of activity concentrations of ^{137}Cs and ^{90}Sr in milk and fodder

Table 22 gives the ecological half-time for the main foodstuffs and fodder. This big difference between two stages could be explained with the different models determining the activity concentration in different media. While the radioactive material entering the atmosphere are dispersed by atmospheric turbulence and transported by the wind, the principal processes involved in transport in terrestrial environment include dry and wet deposition, interception by vegetation, and loss of radionuclides from plants due to weathering, resuspension, translocation and uptake from soil. Although the activity retained by the vegetation is eventually weathered off to soil, the fraction initially intercepted is an important quantity and the retention factor is controlled mainly by the chemical form of the contaminant. Plant surface is negatively charged, so the absorption of cations are more effective than anions. According to the results of Müller and Pröhl (Müller and Pröhl, 1993) the differences among plants are of minor importance compared to the variations among radionuclides. In the first several years following the Chernobyl accident the governing process was the deposition connected with foliar uptake and the resuspension also had a rather significant contribution, then the root-uptake and resuspension became the determining processes, which transport usually much smaller amount of radioactive isotopes. The long half-time is connected to the caesium interaction with the clay content of the soil, fixation of radiocaesium in interlayer positions and a slow diffusion and binding to frayed edges of the clay content of soil, generally the exchangeable caesium was decreased by factor of 5 within a decade.

Table 22. Effective half-time of ^{137}Cs in different foodstuffs and feed in Hungary

media	half-time of "quick" decrease, years	half-time of "slow" decrease, years
milk	0.4	29.4
wheat	0.6	27.7
vegetable	0.9	12.3
fruit	0.4	7.9
fodder	0.8	13.3

SAVE project gave fast half life of 1 year and slow one of 10 years, SAVEC (SAVEC, 2001) project determined these in Poland as 0.71 year for the fast process and 15.4 years for the slow, all results refer to rye grass. Results given in the Table 22 are consistent with these findings. The reason of this phenomena is that the sorption process of the caesium during the first year after the fresh fallout is governed by the phenomena that caesium is present in the environment adhered to small particles in easily reachable form for plants then after the sorption process caesium is present on the so called frayed edges of sites of minerals, from where getting into the soil solution is a much slower process.

The direct comparison of the Austrian and my calculation is not possible, because the time interval involved in the investigation is different. They do not differentiate between the two time-period of the decreasing, which is essential according to my opinion, because of the different governing mechanisms determining the isotope content of the foodstuff, this is the reason why Schwaiger et al (Schwaiger et al 2004) found other half-times, namely from 1.6 to 7.8 years

Table 23. Comparison of effective half-time in Austria and Hungary according to the ratio of fruit half-time to other foodstuffs.

media	ratio of fruit half-time to other foodstuff	
	in Hungary	in Austria
milk	0.3	0.2
wheat	0.3	0.4
vegetable	0.6	0.6

5.3. Analogues

The bioavailability of the hazardous materials depends on their physical, chemical properties, furthermore the way on of release and on the biogeochemistry of the environment at the location of immission. Self-cleaning capacity of the receptor medium also has a determining role. The movement of radionuclides between the different phases among the different parts of the ecosystem was already studied a lot, but is still not well understood in every detail. Finding a proper analogue either for a process, or for a distinct part of the environment or for an isotope often leads to better, more reliable description of the studied system. The use of analogues is not an accurate way of modelling, but it could be used if no data are available in the frame of screening models. Thorough knowledge is required of the system for which an analogue is sought to derive parameter values. Relevant processes and features have to be known, such as time scales of these processes, physical, chemical and biological properties of the environment and relevant media.

5.3.1. Analogue isotopes

Application of analogue isotopes is the most common form and is often used without any specific justification or even recognition that an analogue is being used. Short-lived fission products whose environmental behaviour has been extensively studied in the context of reactor accidents or routine discharges are used as analogues for long-lived isotopes of relevance for solid waste disposal, like data of ^{131}I for long lived ^{129}I and similarly well known ^{134}Cs or ^{137}Cs for the long lived ^{135}Cs.

Stable isotopes whose environmental behaviour has been extensively studied in the context of chemical toxicity are used as analogues for less common and less studied radioactive isotopes found in radioactive discharges or disposed as wastes.

The uptake of ^{75}Se or ^{79}Se by plants will be strongly determined by the stable selenium status of the soil. Similarly, soils may be low in stable iodine and chlorine, in particular, for iodine the fraction of a radioisotope entering the blood is taken up by the thyroid. This process is enhanced in iodine-deficient conditions. For chlorine, homeostatic requirements mean that the half life of retention in the bodies of animals varies in inverse proportion to the level of the stable element in the diet.

In general the behaviour of isotopes of the same element is identical, except for light elements especially in case of hydrogen. In particular, equilibration of a short-lived isotope in environmental media may be strongly influenced by its physical decay, whereas equilibration of a long-lived isotope may be almost entirely determined by biogeochemical transfer processes.

5.3.2. Analogue elements

The purely chemical properties of elements follow fairly well established patterns that can be used as a basis for identifying potential analogues. Elements in the same group of the periodic table usually exhibit similar chemical behaviour, they have the same number of external electrons available to form chemical bonds. In case of essential macro-elements for plants occurring in soil, the uptake and transfer of a chemically similar element will depend on the lack or excess of the essential one. There is a need to examine the biochemistry of the elements, despite of their chemical similarity they could have significantly different functions in biota.

The most commonly used analogue element-pairs are K and Cs, Ca and Sr, Ba and Ra, Re and Tc. Ba, Ca and K are regarded as biogenic elements indicating the influence of biogenic processes.

Ca is often used as analogue element for Sr, for example the correlation coefficient between Ca and Sr in suspended particulate matter was found 0.9986 according to the work of Varnavas et al. (Varnavas et al. 2001). Table 24. shows another example for good correlation of the dissolved ^{90}Sr and stable Ca content of the fraction less than 10 kDa (one Dalton is the mass of one Hydrogen atom) after two-step ultrafiltration of surface water (Matsunaga et al., 2004).

Table 24. Fractional distribution of dissolved ^{90}Sr and stable Ca in the fraction less than 10 kDa after two step ultrafiltration of the surface water (Matsunaga et al. 2004.).

Sampling place	**^{90}Sr, %**	**Ca, %**
Sahan River, March 1996	80	97
Sahan River, August 1996	98	97
Glubokoye Lake, August 1996	97	98
Kiev Reservoir, April 1998	97	99

The counter-example is that an increase in soil strontium concentration over 4 orders of magnitude was accompanied by increase in tissue strontium burdens of earthworms according to the work of Morgan et al. (Morgan et al. 2004), while tissue calcium burdens appeared to be maintained at fairly uniform levels, irrespective of soil calcium concentrations. The contrasting behaviour suggest that differences exist in ability of earthworms to physiologically control tissue burdens of these two analogue elements and this may relate to differences in the solubility of sequestration products of strontium and calcium. In conclusion the risk posed by radiostrontium intrusion into terrestrial ecosystems is compounded in calcium-poor locations.

The availability and distribution of radiocaesium in soil-plant system is incorporating the effects of soil K status, namely the exchangeable K and clay content and time, these are the major factors controlling the bioavailability of caesium. They are chemical analogues, therefore they behave as competitors for plant and soil sorption sites (Absalom et al. 1999.). At K concentrations up to 1mM, the plant and soil solution concentration ratio is decreasing more than 2 orders of magnitude with increasing K concentrations. Above 1mM K, this concentration ratio is almost unaffected by K supply. These results predict that K fertilisation is a successful countermeasure for producing cereals (Smolders et al. 1997.). Transport pattern occurring in the bean plants following the foliar uptake for periods longer than 1 day is similar between K and Cs (Levi, 1970). It is likely Cs to be taken up by K transport systems on plasma membranes of root cells, it is a multiphases process mediated by carriers and ion channels. Uptake at low external (in the surrounding soil) K concentrations (< 10-12 mg/l) is dominated by carrier-mediated high-affinity transport system, which can operate against an electrochemical gradient requiring energy. Conversely, at higher external concentration of K^+ (>12-40 mg/l) the electrochemical gradient is diminished or reversed, and uptake has dominantly channel-like properties (Zhu 2001.).

Several studies demonstrated that Ba behaves geochemically very similar to ^{226}Ra for example in the ocean and that the oceanic $^{226}Ra/Ba$ ratio is constant nearly everywhere. According to Staubwasser et al. (Staubwasser et al 2004) the observed relationship between Ba (mol/g) and ^{226}Ra (mol/g) in the open ocean is:

$$[^{226}Ra]_{sw}=4.75x10^{-9}[Ba]_{sw}-3.58x10^{-20} \quad \text{Eq 5.2}$$

The two exemptions from this relationship are

- parts of the deep ocean bottom water, where an imbalance is caused by unusually high diffusion rates of ^{226}Ra as a result of very low sedimentation rates,
- some near shore environments, where any combination of river discharge, groundwater discharge and estuarine desorption from suspended matter may alter the $^{226}Ra/Ba$ ratio from the open ocean value.

This similarity of the two elements allows that ^{133}Ba could be used as a tracer in laboratory experiments for example to quantify the uptake kinetics by sediments in aqueous suspensions since it is an easily measurable gamma-emitter and can serve as a good analogue of the environmental behaviour of ^{226}Ra (Barros et al. 2004).

Since lanthanides are often used as oxidation state analogues for actinides, their distribution can reflect long-term behaviour of the radioactive transuranic elements. Exemptions to the typical lanthanide pattern are cerium and europium with their 4+ and 2+ oxidation state, next to the 3+ oxidation state common for all lanthanides. This property gives the possibility to use cerium as analogue of plutonium isotopes, forming hardly insoluble CeO_2 and PuO_2. There are several studies where thorium serves as an analogue for Pu^{4+}, and La^{3+} as an analogue for Cm^{3+} and Am^{3+}. More over the precipitation of lanthanides are closely linked with iron and manganese. Nd^{3+}, redox-invariant ion gives more adequate analogy for Pu^{3+} and Am^{3+}, which have solubility at least one order higher than Pu^{4+}. The solubility of multivalent actinides, especially plutonium are often investigated by oxidation-state analogy, which considers that lanthanides and actinides having same oxidation states will behave chemically similar. The speciation of the elements has a significant role, for example in case of the plutonium isotopes, due to their different processes of origin or formation. In case of high temperature weapon explosions plutonium oxides get into the environment forming highly refractory oxides, they remain mainly in the forms, how they are injected into the environment, above all they have nearly no interaction with the surrounding media. For example recently found plutonium in the Hungarian environment originated from global fall-out determined by the nuclear weapon tests. Nuclear processing plants and laboratory research never form refractory oxides, however plutonium is released mainly in reduced form into the environment and the oxidation process takes about 1-3 days until the equilibrium state is reached. While Pu^{4+} is the most abundant form bounded to the particulate matter, Th^{4+} is the proper analogue for the ecological studies in this country, because there are no other sources available where the other oxidation states should be taken into account. The final reservoir both of species are sediments and soils. Direct correlation was observed between Pu^{4+} and Th^{4+} in lake studies, $Pu(OH)_4$ has strong affinity for sediments and suspended particulates, Pu^{4+} sorption is the limiting factor in plutonium transport processes (Choppin 1998).

5.3.3. Analogue parameters

Soil distribution coefficient, K_d values can vary significantly with soil type. Where knowledge of soil conditions is not available, a generic soil K_d value can be adopted. This may be an average over soil types, a value for the soil type expected to maximise doses, or simply a value for the soil type for which data are most extensive. In the lack of the proper K_d for a soil can be used for a sediment with same characteristic like pH, Eh.

In case of plants some analogues may seem relatively obvious, e.g. between pasture grass and forage. Generic data for grain might be expected to provide a good analogue for rice, but the growing conditions are so different. For example PATHWAY model uses a single transfer factor value of 0.01 (Bq/kg plant)/(Bq/kg soil) for all crops and soil types, while ECOSYS has crop-specific transfer factor values that are independent of soil type. In this case the higher uncertainty of the predicted values should be taken into account. During the parametrisation of the model, the results observed for ryegrass appeared satisfactory for a variety of crops for example for grass and cereals (Absalom et al. 1999.).

Phylogenetic effects at the level of plant family and order have been observed for C, Ca, Cs, Cu, K, Mg, Ni and Zn, from this comes a hypothesis, by analogy with the behaviour of Ca and other minerals, that phylogenetic attributes of a species can be used to estimate the soil to plant transfer of many radionuclides. Phylogenetic effects are less strong for Cd, Cr, and Na, and hardly absent for N, P and Pb (Beresford et al. 2004.).

When making comparisons between animals of different types, consideration also has to be given to the mass of the animal. Conventionally, transfers to animal products have been expressed through the use of transfer factors that are the ratio of the concentration in the product to the rate of intake of the radionuclide. For unit rate of intake, the concentration in a particular product tends to be higher for animals of smaller mass, though this effect may be counteracted by more rapid metabolic turnover in smaller animals. In the other case the comparable products from different animals, meat from different animals, cow's milk and goat's milk are also of limited use, because for most assessments the animals considered are quite different from each other.

Although analogies may be drawn between different animal types, this is likely to involve the use of allometric relationships or more properly biological scaling, which is the effect of size on biological variables. Many biological phenomena scale as quarter powers of the mass., and not simply the application of transfer factors for one animal type to another. However this still leaves a requirement for a potentially large number of radionuclide-reference organism transfer values.

5.3.4. Process indication by analogues

I made a study regarding the freshwater ecosystem, fish, plant samples, soil samples at river bank and sediment core samples from the river were investigated. Most of these samplings were carried out in 2005 and 2006 along the Danube river. Some of the results of the survey is used during the investigation of riparian ecosystem as well. First of all I was curious what kind of relation was between the isotope content and sediment particle size. Only macro size distribution of samples were studied by wet sieving in the lack of ultrafiltration system. Regarding the whole experiment the maximum of all isotope was expected in the smallest particle size fraction, only two of them ^{137}Cs and ^{210}Pb follow the well known rule, but the results gave the maximum in the particle size range of 63-150μm for six isotopes. According to the isotope content of different fractions the isotopes could be grouped into two parts, as it is demonstrated on Figure 19. ^{228}Ac, ^{212}Pb, ^{208}Tl, ^{234} Th, ^{214}Pb, ^{214}Bi, descendants of ^{232}Th and ^{226}Ra, they are measured with their short lived daughter products. The figure shows well the equilibrium between ^{214}Pb and ^{214}Bi isotopes having half-life of the same order. The main source of ^{210}Pb (and ^{210}Po) in the environment is from the decay of ^{222}Rn gas evolved from the soil into atmosphere. These radionuclides deposit on the ground in association with aerosols due to washout and sedimentation. It is also striking that short lived ^{222}Rn behave similarly, in keeping with the distribution of parents ^{238}U and ^{232}Th, while ^{210}Pb differs significantly from their distribution. ^{210}Pb expected to behave the same manner as the others being part of ^{238}U chain, but it showed similarity with ^{137}Cs. Studying the possible reasons of this phenomena chemical similarity could be excluded, because Cs is alkali element, Pb is so called post-transition element, their oxidation state, solubility, reactivity and all chemical behaviour is completely different. The only reason of their similar behaviour is their common physical “origin”, that is a physical process connected with the mobility of fine particles being both of them “airborne” isotopes, not like the direct ^{238}U and ^{232}Th descendants. For the ^{137}Cs this statement is obvious, but the case of ^{210}Pb needs some explanation. Being the daughter of ^{238}U, expectation is the same for the place of enrichment with its parent isotopes. Moreover in the decay series there is ^{222}Rn, a noble gas isotope which can escape from the place where the others were before, and in turn daughters of radon will be deposited onto fine particles of the aerosol, just like ^{137}Cs and from this point the pathway of the fine particles is the same, both of them get into the sediment samples during the deposition of fine particles.

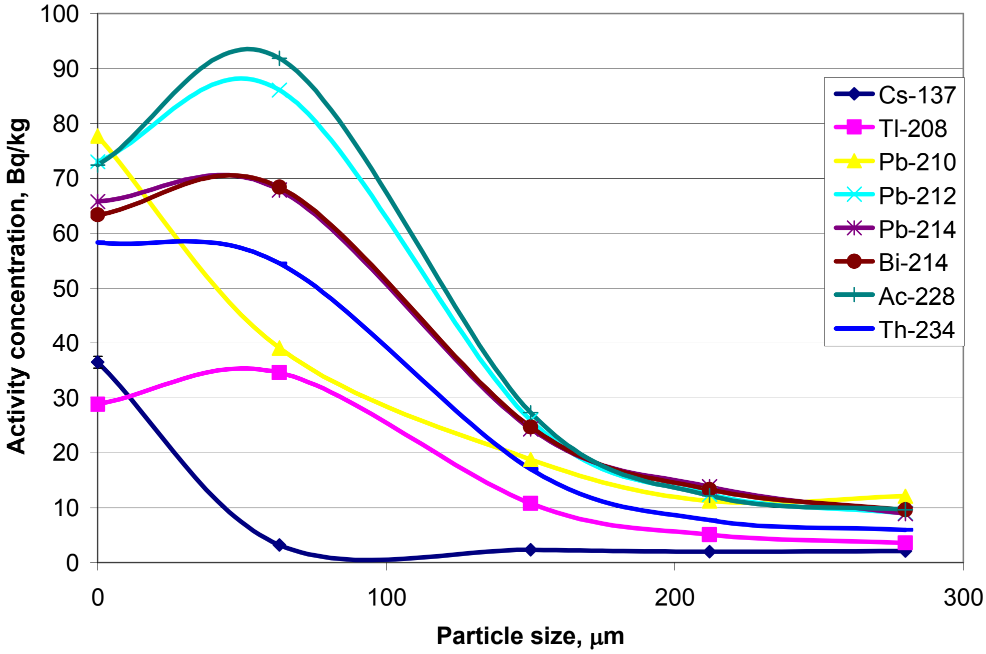

Figure 19. Activity concentration measured by gamma-spectrometry in sediment sample divided into five fractions by wet sieving, using series of 63µm, 150µm, 212µm, 280µm sieves.

The analysis of sediment core samples confirmed the same behaviour of this two dedicated isotopes, as it is demonstrated on Figure 20. This figure shows the activity concentration of ^{137}Cs and ^{210}Pb in different layers of the same sediment core. The sampling was carried out under water, in a shallow section of the Danube river. The concentration ratio of two isotopes is fairly constant, 1.80(3) as it comes from Figure 21, where the uncertainties also indicated. Because of the continuous washing up by water there is no other determining process which could cause differences between the behaviour of these two isotopes, just the convection of the particles by the water and their deposition. Figure 22 shows the correlation between ^{137}Cs and ^{210}Pb content for all of 40 sediment samples collected during 2005 and 2006, while the next one (Figure 23) contains only those data points where the measurement uncertainties were less than 15%.

The situation is different if the subject of analysis is a soil core sample. The correlation between ^{137}Cs and ^{210}Pb totally disappears, there are some layers where ^{137}Cs activity concentration is higher and there are some where they are smaller than ^{210}Pb. The half lives of the two isotopes are in the same order of magnitude and in this environment there is enough time for chemical processes to take place which will result totally different activity concentrations because of their chemically different behaviour (Figure 24). The soil sample was collected from the river bank of the Danube river, but the soil was covered with plants usually growing in the fields not in the wetlands.

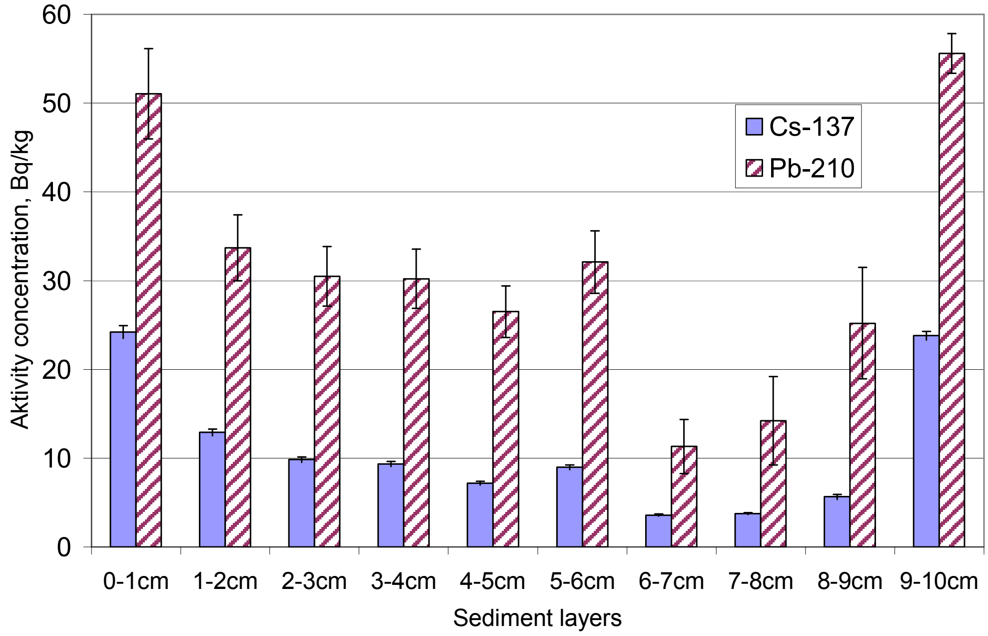

Figure 20. Activity concentration of ^{210}Pb and ^{137}Cs in sediment core sample from the Danube

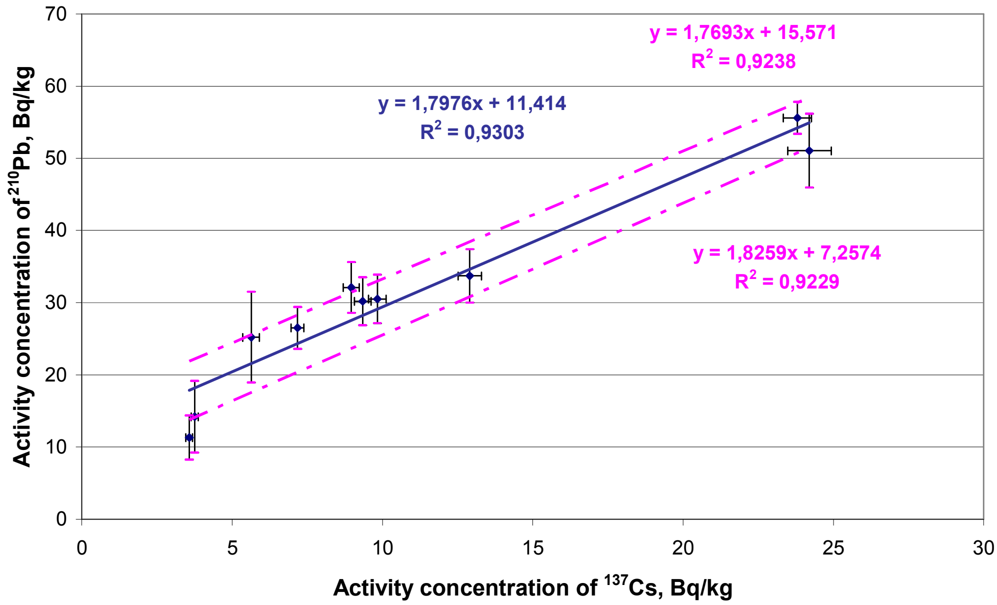

Figure 21. Correlation between activity concentration of ^{210}Pb and ^{137}Cs in the same sediment core sample as in the Figure 20

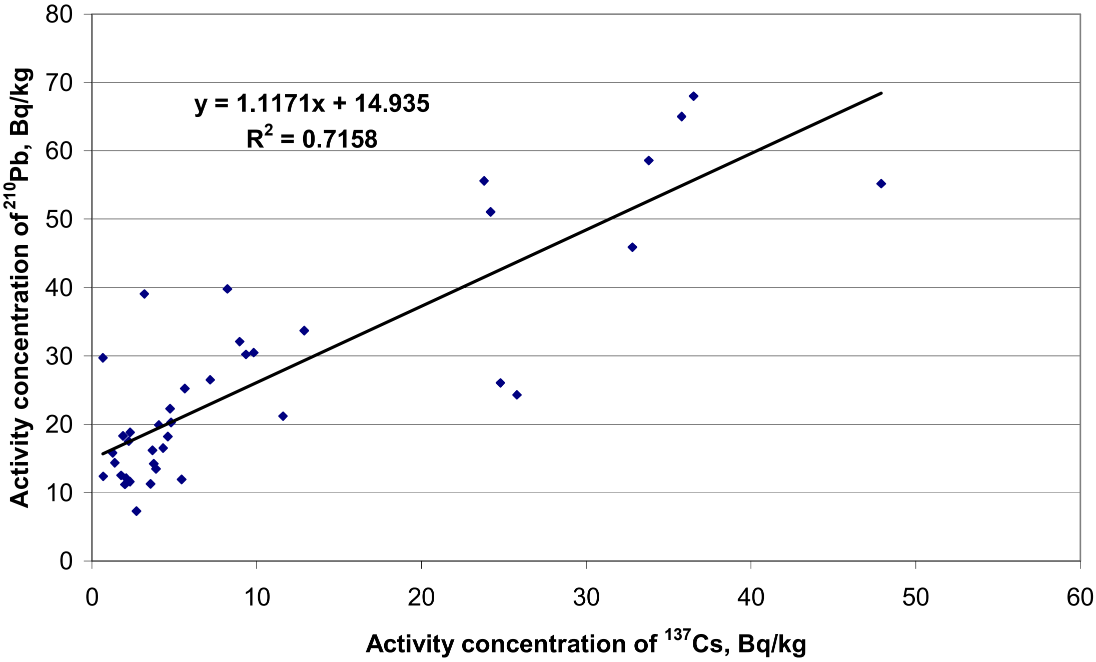

Figure 22. Correlation between activity concentration of ^{210}Pb and ^{137}Cs for all sediment samples collected during 2005 and 2006.

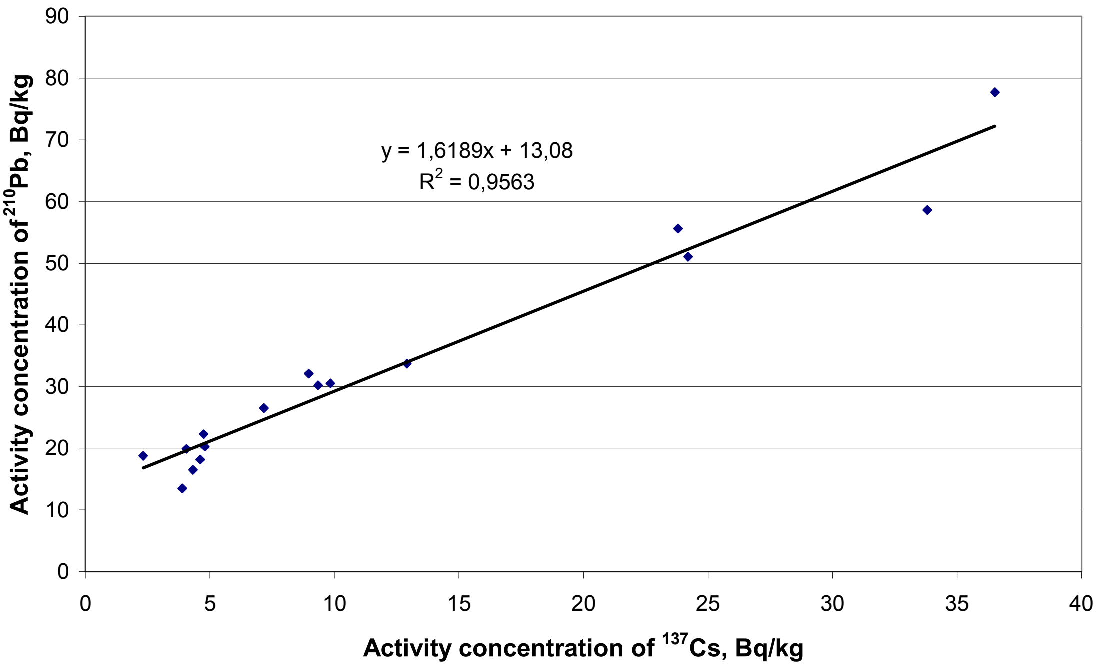

Figure 23. Correlation between activity concentration of ^{210}Pb and ^{137}Cs for sediment samples with less than 15% relative standard uncertainty.

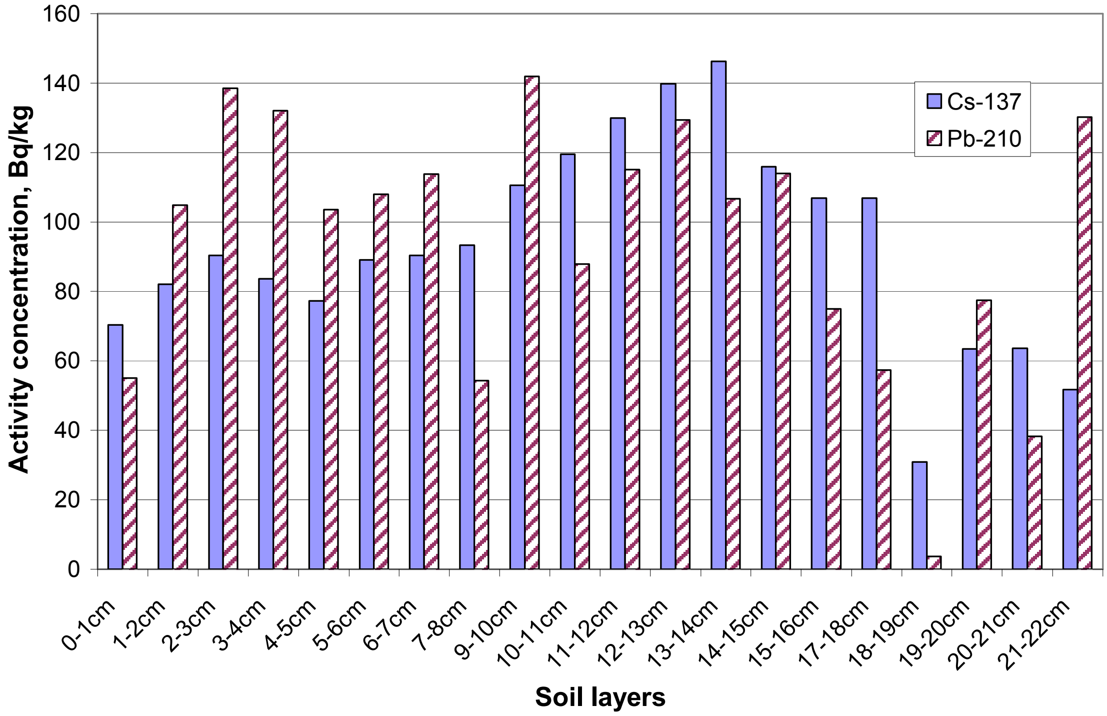

Figure 24. Activity concentration of ^{210}Pb and ^{137}Cs in the soil core sample from the river bank

Further investigation was performed on the 10 years database of the network. It was necessary to take into account the relative standard uncertainty of the results because of the generally low concentrations. As the Table 25 shows in case of soil and sorrel this limited dataset could give a better relationship between these two isotopes.

Table 25. Correlation between ^{210}Pb and ^{137}Cs in different samples

	From whole database (min 10, max 2000 samples)	From reduced database - data with relative standard uncertainty < 10%
Soil	0.12	0.33
Sorrel	0.13	0.77
Green part of moss	0.37	0.37
Bottom part of moss	0.97	0.97
Sediment	0.97	0.97
Mixed samples (sediment and soil)	0.93	0.93
Tobacco leaves	0.79	0.79

The relationship of these two isotopes is a good indicator of the deposition process itself. In case of river-sediment samples being under the water the concentration profile is governed mainly by the deposition and the situation is the same for bottom part of moss samples, while on the mixed samples containing sediments and soil parts, which is under the water only during the high water even flood these two isotopes show worse correlation.

Moreover in case of soil core sample the correlation was worse, as there are more processes forming the concentration profile, like different chemical and geochemical properties of soil. During the lifetime of the soil there is enough time for chemical processes and for example the effect of the organic matter on concentration values. The reason of the similar behaviour of these isotopes in the mentioned sampltypes is both of them could be regarded airborne isotopes, ^{137}Cs coming from global fall-out, while ^{210}Pb is the solid state, long-lived daughter isotope of ^{222}Rn. Processes are the same but the volumetric scale is different, as ^{137}Cs comes from above the troposphere and ^{210}Pb deposition is a near surface phenomenon. Potentially resuspension is a persistent source for radionuclides in air, subsequent to their deposition on the ground. Resuspension refers to the removal of deposited material from the ground to the atmosphere as a result of wind, traffic, soil cultivation and weathering. When an extended area is evenly contaminated the good correlation between ^{137}Cs and ^{210}Pb will show the effect of resuspension process well. In case of leafy vegetable both of the phenomena have significant role, consequently the correlation is also rather good.

With the help of evaluation of the results of sunflower sample collected in 2003, the achievements of my analogue investigations could be summarised, the biogeochemical processes could be appointed. This plant is big enough to cut and treat different parts of plant separately, namely stem, leaves, disk and seeds. Necessarily the soil sample was taken also during the sampling there, but in the following analysis the results of soil measurements play role only for checking the availability of different radionuclides, like a reservoir for elements (for example ^{137}Cs content was 190(4)Bq/kg). ^{137}Cs content of leaves were found higher than expected, 75(1)Bq/kg, this result is not fit either to the ^{137}Cs-^{40}K curve or ^{137}Cs-^{210}Pb curve. From the above mentioned phylogenetic effects of plants comes, that K and Cs being alkali elements will behave in the same way in the soil-plant system, that means ^{40}K and ^{137}Cs moves together which will give information for root uptake process of plant. In case of the leaves of this sample 29% of ^{137}Cs contamination came from root uptake. From the result of my study about the behaviour of ^{137}Cs and ^{210}Pb comes that knowing ^{210}Pb content of a plant ^{137}Cs contamination from resuspension process is possible to estimate, so resuspension process resulted 26% of ^{137}Cs contamination of leaves. The remainder part, 44% attributable to “fresh” deposition, which is possible because of sunflower has long vegetation period. The base of the prediction is shown on the Figure 30 and given as following with the uncertainties of the parameters:

for root uptake: $C_{Cs\text{-}137} = 0.0108 \pm 0.0004\ C_{K\text{-}40} - 2.659 \pm 1.064$ Eq 5.3

for resuspension: $C_{Cs\text{-}137} = 0.4522 \pm 0.0090\ C_{Pb\text{-}210} + 4.073 \pm 0.937$ Eq 5.4

It is emphasized that the overall assessment must be supported by field measurements to produce site-specific data.

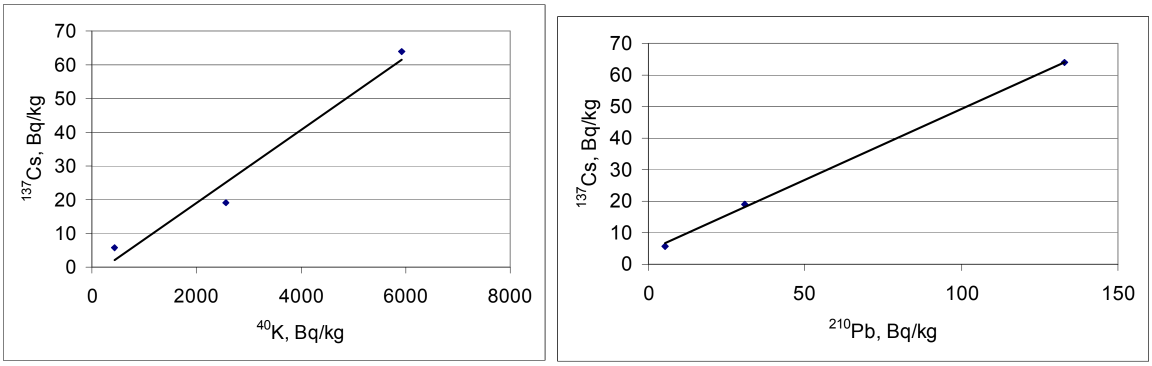

Figure 30. The result of measurement by gamma-spectrometry of sunflower sample from Galgagyörk (2003), isotope-pair results of seeds, stem, disk are on the figures.

The last several years 25 samples from undisturbed areas of different parts of Hungary were analysed for gamma-emitting isotopes by HPGe gamma-spectrometry and for plutonium isotopes by alpha-spectrometry. The correlation coefficient between ^{234}Th and $^{239+240}$Pu isotope content of these soils was 0.29, which indicates that the origin of the plutonium isotopes is fall-out and not resuspension. Thorium isotopes are always lithogenic origin, they can serve as tracers for resuspension. In case of resuspension better correlation is expected. The investigation of the moss samples gave good correlation coefficient 0.94 between ^{234}Th and $^{239+240}$Pu. While thorium exists only in 4+ oxidation state, the good correlation means that plutonium in the Hungarian environment is mainly also in 4+ oxidation state, this explains the similar chemical behaviour and bioavailability of these two species of the two actinide elements in the environment, which determine the following relationship with the uncertainties of the parameters coming from the experiments:

$$[^{239+240}Pu]_{moss}=(0.021\pm0.001)[^{234}Th]_{moss}-(0.355\pm0.042) \quad \text{Eq 5.5}$$

5.3.5. Limitations in use of analogues

As with any other choice of parameter values for modelling, decisions on using analogues must take into account the assessment context and particularly the level of realism or conservatism of the assessment. The best analogue for a realistic assessment might not be the best for a conservative assessment. An analogue could only be proven to be valid by comparing its behaviour in the conditions of interest, while confidence in the validity of an analogue will increase as the quality of the justification increases, there will always be some residual uncertainty.

The timescales for experiments or observations on short-lived isotopes may be limited by radioactive decay and so might not reflect all aspects of environmental behaviour in the long-term. While some care is needed in cases where there are large differences in half-life isotope analogues can normally be assumed to be more reliable than element or media analogues.

More fundamentally, although isotopes of an element have the same chemical behaviour, this chemical similarity does not necessarily translate into similar behaviour in the environment. This effect is demonstrated by the absorption of common elements from the atmosphere, most plants show higher ratios of $^{12}C/^{13}C$, $^{14}N/^{15}N$ and $^{16}O/^{18}O$ than found in the atmosphere. These differences tend to be more important for lighter elements because the relative differences are larger, except for hydrogen these isotopic differences will in general be very much smaller than most other uncertainties in the model system.

For element analogue, chemical similarity does not necessarily translate into similar behaviour in the environment. Key considerations include variations in valence and ionic radius. Thus, in the case of the lanthanides, there is a consistent trend across the series from predominantly 2+ through to 4+, and this trend can be reflected in trends in environmental behaviour. A problem of element analogue is that the initial distribution of elements in the environment can affect the behaviour of the radionuclides being modelled. If the soil is naturally or as a result of past activities poor or rich in a particular element that is important as plant or animal nutrient, then the uptake and transfer of chemically similar radionuclides released to the environment will be affected.

Sometimes the difficulty of the determination of an isotope leads to using an analogy. The chemical similarity of radium and strontium is obvious being both alkali earth elements. From the radiological point of view ^{226}Ra, ^{228}Ra and ^{90}Sr has the main importance due to their long half life, but unfortunately neither these isotopes is an intensive gamma-emitters, so there is a need for radiochemical separation before the measurement as well as in case of ^{90}Sr. ^{228}Ac, daughter of ^{228}Ra has a well identifiable gamma-line and because of the difference between the half-life of ^{228}Ra (5.75 years) and ^{228}Ac (6.1 hours), secular equilibrium is supposed between them in the nature, so ^{228}Ac is applicable as an analogue for ^{90}Sr isotope.

5.4. Riparian ecosystem

Riparian ecosystem is one of the ecosystems which may be most vulnerable, describing with long residence time of radionuclides (woody areas) and with high transfer factors (covered continuously by water) as well. The extent of riparian vegetation in Hungary is small it is present only along the rivers and around the lakes, but it has high biological value, the goal of protection of riparian ecosystems should get some priority.

Riparian ecosystems serve as the primary link between terrestrial and aquatic ecosystems and in many respects they are symbolic of large-scale landscape change because of their sensitivity to fluvial disturbances. They also are the ultimate expression of ground water and surface water interactions. In riparian zone vegetation and bird populations are regarded most valuable; riparian ecosystems can be viewed as a scale phenomenon of from minute details to largest scale. Generally all riparian ecosystems are considered to be at risk to decline and the perception of historical region-wide loss prevails.

Ground water is an important source of water for agriculture, industry and public supply. The recharge to the aquifers commonly comes from precipitation at higher elevations in mountains adjacent to the aquifers. The effects of ground water withdrawal can be local or on a large scale. Wells in an aquifer are typically pumped for an extended period.

Floods have a major role in shaping the species composition and productivity of riverine riparian vegetation. Floods create openings in typically dense riparian assemblages, transport seeds and vegetative propagates, saturate flood plains to encourage seedling growth and deposit nutrient rich sediments that promote germination and recruitment. In alluvial reaches, major floods cause bank scour and remove at least some of the riparian vegetation. Plants may be uprooted individually in response to detrital loading on their upstream sides, or whole terraces may be removed during lateral avulsions, channel widening or meander propagation. In particular, mesquite bosques developed on terraces may be removed during flood induced channel meandering. Flood control, a common justification for dams, reduces peak discharges and the amount of water available to riparian systems on floodplains as well as potentially changing the seasonality of flooding. Long-term flood control may cause conversion of formerly mesic floodplains to xeric shrublands, conversely reduced flooding, coupled with lower variability in daily discharge greatly benefits established riparian species on the streamward side of floodplains, because formerly scoured channel margins become stable habitat for new plant establishment. The response of riparian vegetation to flow regulation depends on the wide spectrum of possible water use and flood control, which ranges from complete diversion of surface flow to dam releases with flood control. After construction work a period of geomorphic adjustment to the changed flow and sediment transport regimes are expected followed by an adjustment in riparian vegetation. Flow diversions strongly affect riverine riparian ecosystem by reducing recharge to shallow aquifer systems. Water use of agriculture also lowers river flows, particularly during the summer period, thereby stress riparian plants in the season when they need water most. The Figures 25 and 26 show the timedependency of the runoff of Danube and the distribution shows that the frequency weighed average runoff is 2281 m^3/s with the range of 844-7860 m^3/s, only 211 was higher than 5000 m^3/s from 10359 observations. Flood has not too strong effect to riparian zone in Hungary.

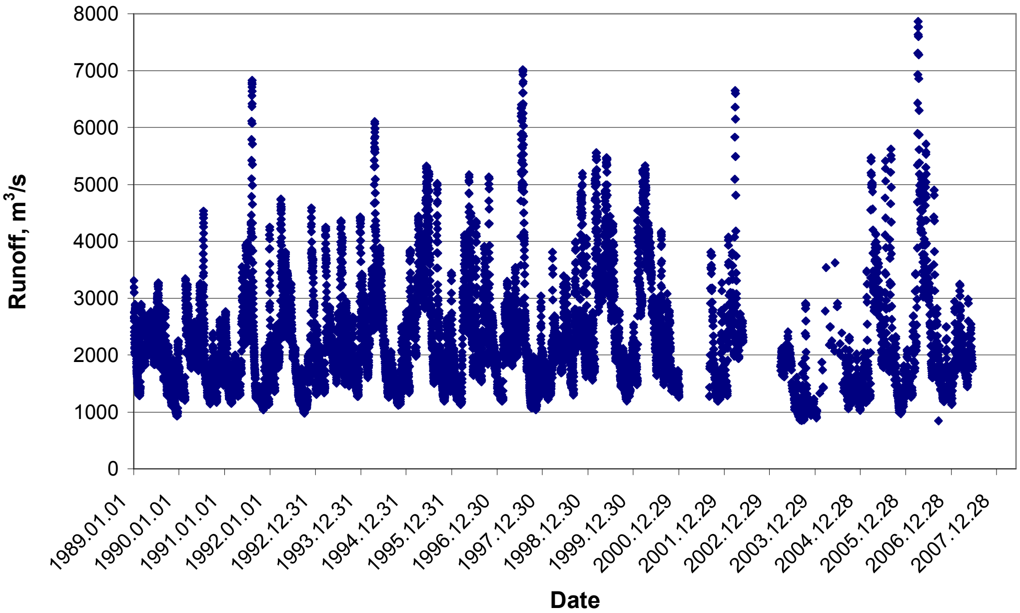

Figure 25. Flow rate of Danube at Dombori from1989-2007, average is 2281 m^3/s (with the range of 844-7860)

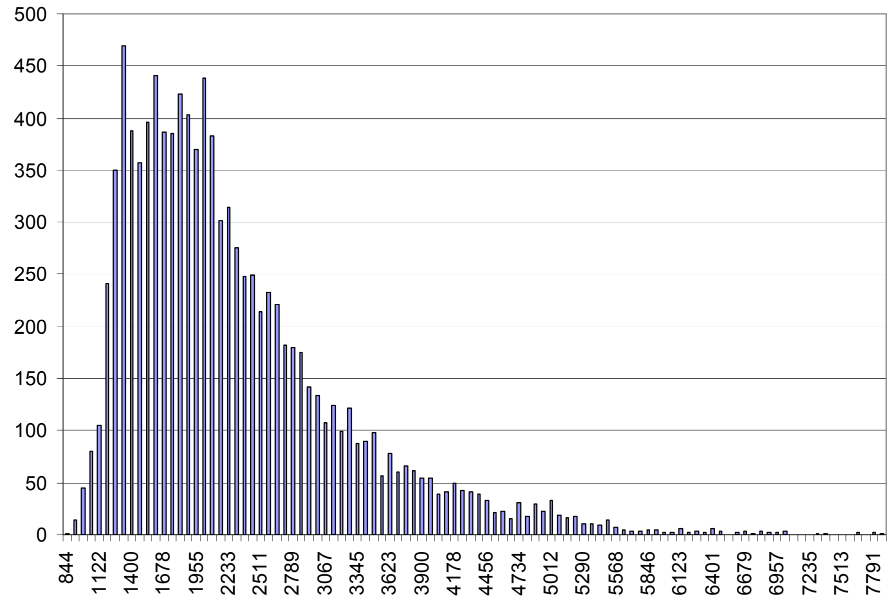

Figure 26. Distribution of the flow rate of Danube 1989-2007

Regarding riparian vegetation, riparian trees are sensitive to changes in ground-water levels. If lowering occurs slowly enough in response to drought or to steady pumping out, roots may be able to elongate quickly enough to keep pace and allow the tree to survive. The rate of the change is the most important factor in plant survival, not only the absolute depth of water. Trees in bosques are known to become stressed when ground-water level decline to 13.7-15.2 m below land surface. Mesquite can withstand relatively large fluctuation.

Figure 27 shows that from the theoretical concentration at the entry point of radioactive liquid discharge into the river the concentration decreases very fast according to the following model:

$$C_w^{\max}(x) = \frac{A_i}{QT_i}(1-f_p)\exp\left(-\frac{(\lambda+k_1)}{v}\right)x \qquad \text{Eq 5.6}$$

x	distance downstream, m
A_i	amount of radioactivity discharged (of isotope i), Bq
Q	volumetric flow rate of river, $m^3 s^{-1}$
T_i	time over the discharge takes place, s
f_p	particulate-sorbed fraction, dimensionless
λ	decay constant, s^{-1}
v	advection coefficient (mean water velocity), $m s^{-1}$
k_1	rate of transfer of radionuclides to bed sediments, s^{-1}

$$k_1 = \left(\frac{sK_d}{1+sK_d}\right)\frac{v_p}{d} \qquad \text{Eq 5.7}$$

s suspended solids concentration, mg l^{-1}

K_d distribution coefficient, l kg^{-1}

v_p settling velocity of suspended solids, m d^{-1}

d river mean depth, m

The parameters are the following:

s: 13 mg l^{-1} suspended solid in water,

v_p: 1.5 m s^{-1} ,

d: 5 m,

f_p: 0.06,

K_d: 5E+13 l kg^{-1}

Even if the theoretical discharge is in the order of 10^8 Bq in one second it is impossible to catch it by measurements, and because of the huge dilution it will cause no harmful effect for the ecosystem of river or riparian zone. On the Figure 27 he activity concentration of river water in downstream are shown at average velocity 1.15m/s (2283 m^3/s), high flow 5 m/s (7860 m^3/s) and small flow (844 m^3/s), the curve of average and high flow goes together only small flow is different.

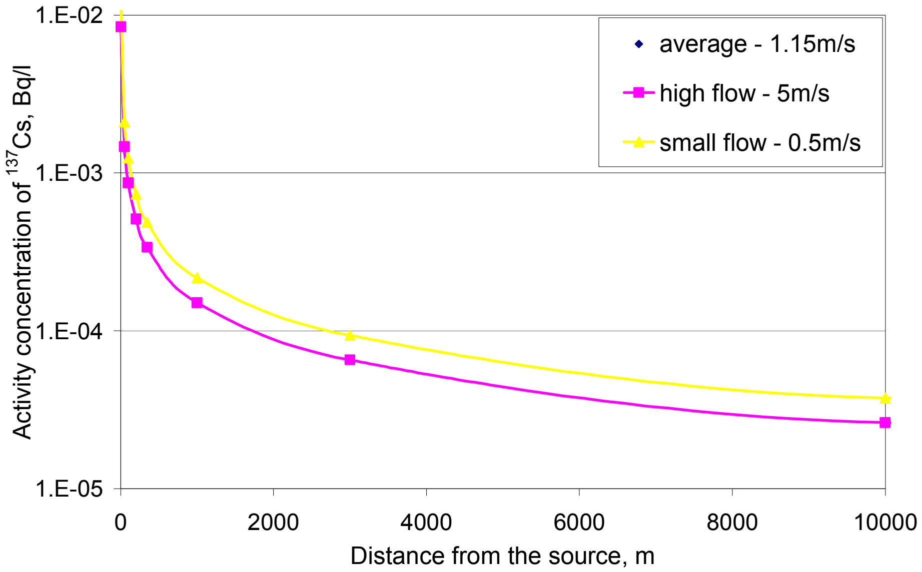

Figure 27. Decrease of the activity concentration of river water in downstream

The problem of the riparian zone will arise if the contamination comes from the deposition and not the stream driven, because the residence time of the water here is very long and the chemical surrounding will help the dissolution of any contaminant to increase their bioavailability.

Radionuclides in growing wood originate from either the initial atmospheric deposits that entered the plant by foliar absorption or root uptake determined by the riparian area under water. The relative contribution depends on the type of tree, the season at time of deposition and the time elapsed after the deposition, with root uptake being dominant pathway for uptake of growing wood over the long term, moreover a strong age dependency could be observed. Conservative approach is applying pine tree data for deciduous forests. Generally forest is a sensitive against the fall-out having large surface of the leaves. In forest the weathering is more complex due to the canopy structure, since several vegetation layer, such as crown, trunk and understorey are involved. The loss of radionuclides from the crown may be the input to understorey vegetation, which results in a delay in the loss of radionuclides from vegetation to soil. The activity in leaves can be added to the organic horizons of "soil" with a time step of one year in case of deciduous trees for the purposes of long term dose assessment to biota living on and in forest soil. In case of coniferous forest this approach leads to meaningless conservatism. For the determination of the necessary depth of soil or mixed soil-sediment samples collected together with plants, I determined the rootmass of the small trees, as on Figure 28 is shown, conclusion is, that 20 cm depth is enough for sampling in riparian zone also, because 94% of total rootmass is in the upper 20 cm layer. The values of Figure 28 were obtained with a birch tree younger than 2 years collected from the riparian zone of Danube.

Grazing by domestic and wild animals has direct and indirect effects on riparian ecosystems. Livestock heavily use riparian areas because of their high productivity, particularly of herbaceous species and readily available water and shade. Effects that are considered negative include consumption and trampling of native plant seedling, soil compactions, destabilization of channel banks, increase in streamflow sediment concentrations and sometimes displacement of wildlife. Despite claims that livestock grazing diminishes riparian vegetation, numerous reaches that sustained livestock had increases in riparian vegetation of about the same magnitude as ungrazed reaches.

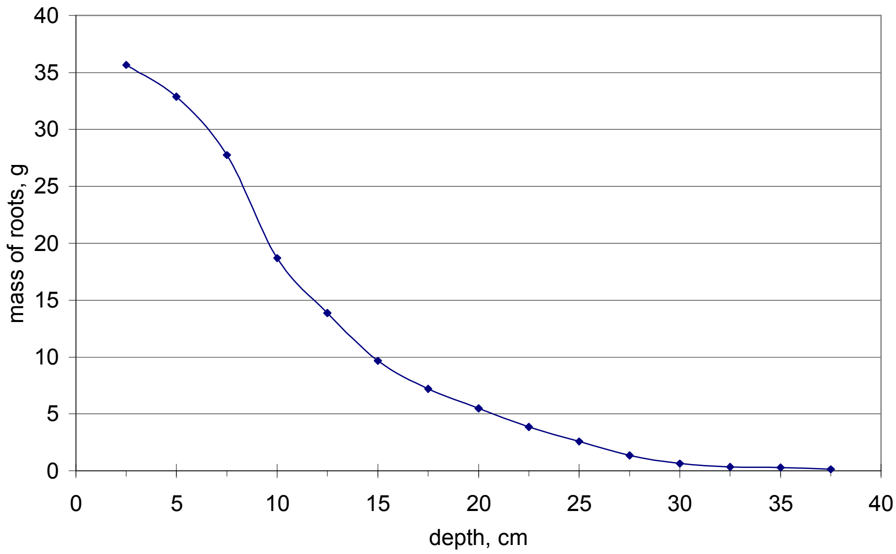

Figure 28. Distribution of root-mass of birch tree

When considering radiation doses to animals living in wetland ecosystems, several factors related to their physiology and ecology indicate that frogs are one of the most sensitive organisms. In temperate climates, frogs hibernate for almost six month (Stark et al. 2003) in the winter period by digging themselves into the soil or into the sediment of a shallow pond or stream. Their home range is small, approximately $200m^2$ and therefore a frog can spend its life of about 10 years, in the same wetland area. They are poikilothermic animals, their body temperature is the same as their close surroundings. At lower temperatures their metabolism is slower and as a consequence the biological half-life of contaminants in their organism is longer. For every 10^oC decrease in temperature, the biological half-life is increased by factor of two. Frogs have thin and permeable skin, consisting of a living cell layer. In addition, frogs have an external embryonic development and lay their eggs without shells or amnion directly in the water that makes them potentially sensitive to exposure to ionizing radiation.

Bioconcentration factors defined as the ratio between the activity concentration of ^{137}Cs in frogs and soil and in frogs and water were taken from Stark et al. (Stark et al 2004).

$$BCF = \frac{A_{organism}}{A_{soil}} \text{, range: 0.006-7.0} \frac{Bq/kg \ fresh \ weight}{Bq/kg \ dry \ weight} \qquad \text{Eq 5.8}$$

$$BCF = \frac{A_{organism}}{A_{water}} \text{, range: 2500-19000} \frac{Bq/kg \ fresh \ weight}{Bq/l} \qquad \text{Eq 5.9}$$

In the sitespecific analysis a kinetic/allometric method is more useful as an alternative for estimating BCF ratio, because the range of the above given bioconcentration factors is too wide.

The kinetic methods for estimating bioconcentration have been developed in the last several decades. The most familiar is the first order kinetics to estimate time-dependent radionuclide concentrations in organism, however many of the kinetic approaches used still rely on the use of single concentration ratios (moreover equilibrium is supposed) to estimate radionuclide concentration in biota. The compartment modelling approach can be used as part of dynamic calculations of contaminant migration in the environment.

There is one more alternative method, the allometry, which is the study of size and there is an observed regularity in a large number of physiological parameters, which relate to body mass. Allometry, the biology of scaling is the study of size and its consequences. Regardless of this variety, there is an observed regularity in large number of physiological parameters which relate to body mass. There are several allometric equations that relate body mass to many parameters, including ingestion rate, lifespan, inhalation rate, home range. The most common form of allometric equation is the power function. While these equations were derived from empirical observations, there is a growing body of evidence to suggest that these relationships have their origins in the dynamics of energy transport mechanisms. The daily rate of consumption by an animal, the metabolic rate is known to scale to body mass to the 3/4 power the exponent of body weight larger for juveniles than adults. There is considerable debate about the value assigned to power function, most of the discussion focuses on values 3/4 versus 2/3 for the exponent. Mentioning just some of them: White and Seymour (White and Seymour 2005) state that a rigorous analysis available for mammals and birds both indicate a basal metabolic rate proportional to $M^{2/3}$, they do not rely on data collected to date. Without trying to resolve the debate, the question that needs to be considered from a radioecological perspective is – how important is the selection of this value. The following figure (Figure 29) shows well the sensitivity a general equation to this object of debate, namely across eight order of magnitude of input data the relative difference between the expression using two exponent in the output data is 5.62, while across four orders of magnitude 2.15 and across two orders of magnitude which is the most relevant question during the radioecological studies is only 1.47. This fact should be considered during the evaluation of the overall uncertainty of a given model, comparison of the component uncertainty will give the answer of importance of this exponent. Probably it may not be of crucial importance deciding whether 3/4 or 2/3, commonly used is the first one.

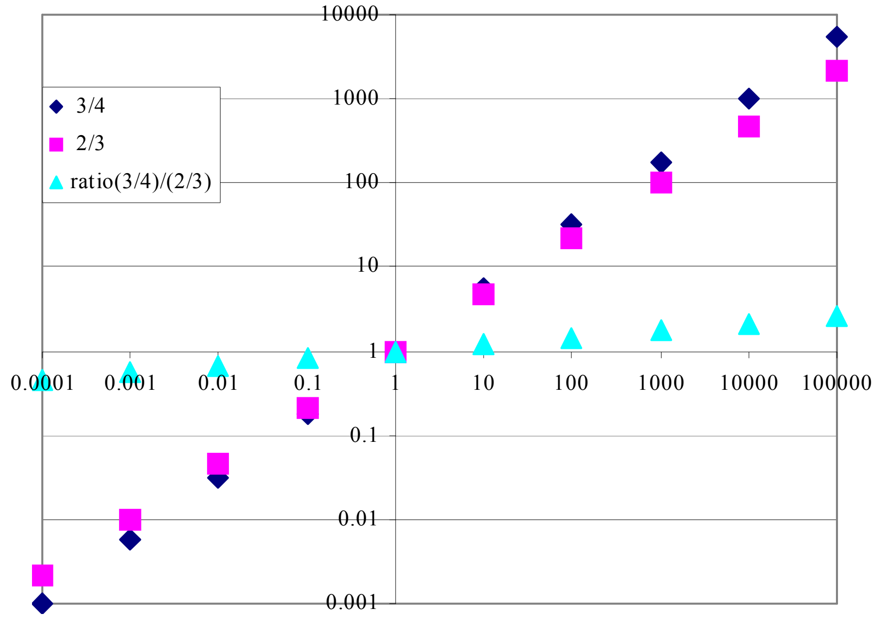

Figure 29. Dependence of the allometric estimate on choice of exponential parameter

The best way is for modelling radioecological processes to combine these methods according to the goal of the protection-calculation. The following description gives a possible combination of the methods:

First of all food intake rate might be calculated from basic metabolic rate functions (Wicker and Schultz, 1982) with the help of allometric approach:

$$r = \frac{A}{dc} 0.017 M^{0.75} \qquad \text{Eq 5.10}$$

r:	intake rate, kg/d,
A:	active to basal metabolic rate,
d:	fraction of energy ingested that is assimilated and oxidized,
c:	caloric value of food, J/kg,
0.017:	empirically derived constant (unit conversions)
M:	animal mass, kg
0.75:	allometric scaling factor, which relates body mass to intake rate.

This function can be then inserted into a single compartment kinetic equation:

$$\frac{dq}{dt} = R - kq \qquad \text{Eq 5.11}$$

When the initial activity is 0 in the compartment, the system can be solved for q(t) as

$$q(t) = \frac{R}{k}(1 - e^{-kt}) \qquad \text{Eq 5.12}$$

q: amount of radionuclide in the compartment, Bq,

t: time, d,

R: input rate, Bq/d,

k: loss rate constant from the compartment, $k = k_{bio} + k_{phys}$ 1/d,

If $R = rC_{food}a$

r: food intake rate, kg/d,

C_{food}: activity concentration in forage, Bq/kg,

a: assimilation fraction to animal (dimensionless),

One more simplification possibility, if $kt \to \infty$, $e^{-kt} \to 0$,

then $q \to \dfrac{rC_{food}a}{k}$.

The combination of kinetic and allometric version of intake calculation is the following:

$$q = \left(\frac{A}{dc}\right)\frac{Ca}{k} 70 M^{0.75}(1 - e^{-kt}) \qquad \text{Eq 5.13}$$

The Table 26 shows that it is better to use the real concentrations, determined by accurate measurement, which is easily executable with up-to-date equipments and sample preparation techniques. Never forget that the models have to be attached with great importance to prediction, when there are no direct data available. Going back to the frogs, they are protected animals, the usual sample preparation technique (ashing) is not the best way for getting information about their dose.

Table 26. Measured and calculated activity concentrations of ^{137}Cs in watershed, first part of food-chain (for the calculation AQUASCOPE (Smith et al, 2005) was used)

Matrix	Measured activity concentration	Calculated from measured sediment activity concentration
Predatory fish, Bq/kg fresh	0.36±0.06	3.45(0.69-17.3)
Water of Danube (from 20.7 l) mBq/l	1.4±0.6	5.79(1.16-28.9)
Soil-sediment, Bq/kg	3.5±0.6	

Frog being poikilothermic animals, their body temperature is the same as their immediate surroundings, the biological half-life constant 0.011 at 20°C, 0.0057 at 10°C and 0.0028 at 0°C (Reiche et al, 1970). Some dynamic approach could be brought into this combined model, from the above mentioned fact and from changes of the daily average temperature in Hungary. For description of these changes in radioecology mainly power functions are used. Taking into account that the consumption of frog is 0.6g from food, calculating with predatory fish as worst case and 0.06g (Loman, 1979) from soil or sediment and assimilation factor for ^{137}Cs is 0.74 (Duelmann et al, 1986), the curve on the Figure 30 shows the temporal changes of ^{137}Cs amount coming from its food living in the environment described in Table 26. In the radioecology for taking into account the seasonal variances usually a periodic (sinus) or polynomial mathematical description is applied, here the periodic one gave no perfect result, because the amplitude. The result is the following polynom describing the amount of radionuclide in this single compartment, which is now the protected frog:

$$q=-5E\text{-}1.3t^5+2E\text{-}9t^4+3E\text{-}7t^3-0.0002t^2+0.0102t+4.6845, \qquad \text{Eq 5.14}$$

q amount of ^{137}Cs in frog, Bq

t the day of the year, starting with first of January.

Since frogs spend a relatively short time in the water (during spawning) and the water activity concentrations are low, the radiation dose from water constituted a minor part of the total radiation dose that a frog receives annually. If all essential parameters are present, such as breeding pond, food items and hibernation sites, a frog can spend its entire lifetime in a rather small area. In the case that a frog moves, dispersal distance is likely proportional to home range size and is usually less than 2 km. Supposing that frog lives as long as 10 years, it will accumulate from this environment just from its food 15 Bq of ^{137}Cs, using DCF value of 4.3E-6(Gy/y)/(Bq/kg) (Coppelstone, 2001) and the body mass of 30g, this will result 2.15E-4 Gy/y internal dose, which is only 0.06% of the suggested biota guide level of 1 mGy/d for terrestrial animals (DOE, 2002).

Above described model combines the all advantages of the three types of modelling for reaching the goal, more realistic dose assessment for biota.

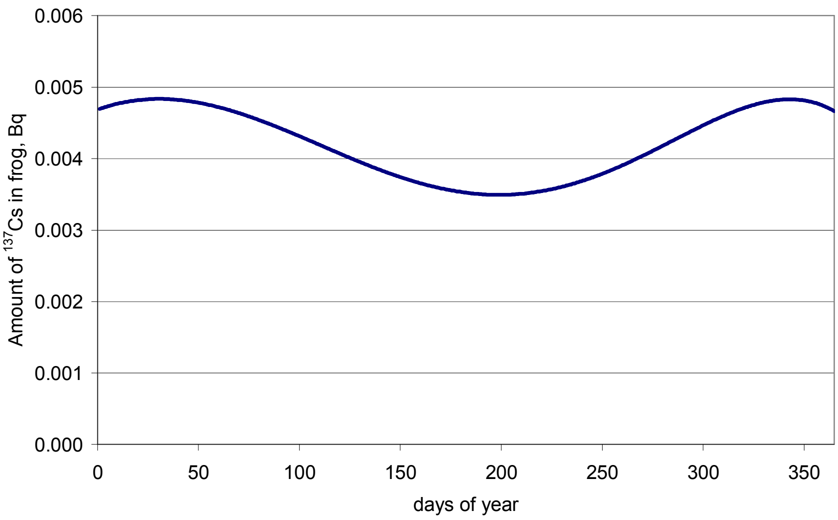

Figure 30. Changes of the ^{137}Cs content of a frog assimilating from its food during the year

5.5. Summary

Investigation of transfer factors, residence time and analogues could help whether or not to dedicate an area, an ecosystem vulnerable from radioecological point of view.

For the determination of most vulnerable area in Hungary from the soil-plant transfer I chose the evaluation of the ^{90}Sr results among the detectable artificial isotopes, because the chemical behaviour of strontium is not dependent on the time elapsed between the time of contamination and the investigation. Caesium is not proper for such kind of investigation, because there is a process to retard caesium in frayed edges of clay. Plutonium isotopes are also not sufficient for this purpose, because its mobility is very low and the activity concentration in Hungarian soils is low and there are territories where it is not possible to detect them in the soil. There were only 3 grass sample with detectable $^{239+240}$Pu isotope content which is not enough to perform a countrywide study.

For the long term effect of the releases on the agricultural production the radioecological vulnerability criteria were appointed. ^{90}Sr transfer between soil and plant gives an appropriate discrimination for the vulnerable agricultural areas. The absorption of ^{137}Cs in the animals gives the most vulnerable point of the feed and animal transfer. The isotope ratio of ^{241}Pu/$^{239+240}$Pu gives information even at low concentration levels whether the contamination came from nuclear installation, which can determine the emphasis of further survey. Among the analogues the correlation between ^{210}Pb and ^{137}Cs gives information for the ratio of resuspension of artificial isotope among the root uptake and fresh fall-out.

6. Criteria for managing of monitoring network

6.1. Monitoring Schedule

The frequency of sampling and measuring the samples was defined to obtain the best representative information about the specific radiological situation. The following aspects are taken into account year by year:

- role of agriculture in the national economy,
- changes in the structure of agriculture and food consumption,
- geographical distribution of production,
- features of territorial units and administrative units,
- organisation of public administration,
- capacity of radiochemical laboratories,
- obtaining a reasonable data set for statistical evaluation.

On one hand there is a need to produce the same type of data every year in order to investigate the long-term tendencies, on the other hand monitoring schedule has to follow the changes and sometimes the actual consumption-structure, too. Also the network should be prepared for answering the questions of the general public, it means that some kinds of foodstuff consumed less than 5% in a year are also involved in the yearly monitoring schedule (Technical Report Series No. 295, 1989). Consumption data come from the evaluation of an appropriate questionnaire (Yearbook of Household Statistics, from years of 1985 to 2003). Due to this compromise each laboratory has to collect and analyse more than hundred samples in a year from different places of the respective region in the frame of the routine monitoring. Samples are collected from harvested products from open-air production and only the consumable parts of the plants are measured. From the publication of these idea (Varga et al 2006) the whole network reorganised, but the sampling schedule remain same, the base of the collection is the county. Some aspects of the sampling schedule might be of interest:

- Milk is collected monthly from the same farm year by year, milk products, like cheese and milk powder are sampled 6 times a year in every county;
- Cereals: annually 3 samples are collected from different locations in the county;
- Potato has an important role in Hungarian cuisine, 3 samples are collected;
- Meat: 6 beef, 6 pork, 5 poultry samples are collected, fish from fresh water lakes or rivers is collected once a year; however the consumption rate is low of the latter (1.6 kg/year per person), there was a need from the public to control it,

- Vegetables: leafy vegetables like spinach and sorrel are sampled 2 times (end of spring and autumn) 3-3 samples form different places of the county; from green paprika, tomato, green beans, green peas, maize, root vegetables, onion, garlic 2 samples are collected from each;
- Fruits: apple has the highest production and consumption rate - 4 samples are taken per year, from different kind of berries 1 sample is gathered;
- Collecting pair of samples is important from the ecological point of view, like milk-fodder, grass-soil, lucerne-soil, sorrel-soil;
- The network collects samples, controlling the semi-natural and natural environment also, like meat of wild animals, moss, mushroom picked up in a forest or grazing land;
- Living in temperate zone the production periods of different kind of foodstuffs are taken into account in the yearly sampling program and also in the monthly program issued month by month to monitoring stations.

Bioindicators are also involved in normal sampling programme to get information during the whole year. In this way the network is able to provide data about the specific activity of different radionuclides in different medium, time dependency and spatial distribution of radionuclides in soil, plants, foods and feeds, in order to give base to issue export allowances, to assess ingestion doses and to give support to decision-makers in the case of an incident or accident at a nuclear facility. Being responsible for the whole extent of foodstuffs monitoring of import food also became one of the task of the network.

6.2. Methods applied in the network

All methods applied in the network are harmonised from the sampling to the final dataevaluation, this approach gives some freedom for the analyst, but several crucial point should be kept in everybody's mind. Most of the samples are processed by ashing at 450^{o}C, however, soil samples are air-dried, moss samples are dried at 105^{o}C. In the network screening methods and nuclide selective methods are applied. Generally speaking screening is a type of analysis aimed at eliminating from further consideration factors that are less significant for the protection or safety point of view, in order to concentrate on the more significant factors.

<u>Screening methods:</u>

1. Gross-beta measurement: This method has been applied for 30 years in the network. Nowadays this simple counters are gradually replaced with low-background alpha-beta counters coupled to high capacity sample changer.

2. Gross-alpha measurement: The ashed food and plant samples are measured. Four laboratories have the above mentioned ultra low-background alpha-beta counter operating with P10 gas. The typical limit of detection at 95% confidence level is 0.3-0.7 Bq/kg fresh weight depending on the sample type.
3. Gamma spectrometry by NaI(Tl) scintillation detectors is kept in routine work for checking fresh samples and measuring ashed samples also for the first quick information. For emergency situation these detectors are calibrated for measuring larger amounts of samples in fresh weight, mainly food and feed samples in 400 cm^3 Marinelli beakers. However this method is able to give specific activity of gamma-emitting isotopes, it is considered a screening method because of the way as it is used and the limitation of this technique.

<u>Nuclide selective methods:</u>

1. Determination of ^{90}Sr: After usual sample preparation (drying and ashing) using $Sr(NO_3)_2$ carrier, leaching with HNO_3, some cleaning steps are included followed by precipitation of Sr as $SrSO_4$. The specific activity is determined after ^{90}Y in-growth by low background alpha-beta counter. The typical minimum detectable activity is 10^{-1} or 10^{-2} Bq/kg fresh weight in cereals, fruit, vegetable and milk.
2. Gamma spectrometry by HPGe detectors: 9 of the county laboratories have gamma-spectrometers with HPGe detectors. On the site of Department of Radiochemistry of Central Agricultural Office, Food and Feed Safety Directorate, there is a low-background counting room, its walls are made from 1 m barite-concrete, 7 gamma spectrometer systems work there, 2 of them are portable for the in-situ measurements and they are also applied for homogeneity tests of in-house reference materials in low background shielding. Main features of the systems: coaxial detectors both n-type and (8) p-type (8) with the resolution of 1.8-2.2 keV, 9 detectors have an efficiency of 30%, 4 have 25-27% and 3 have 15%, 6 of them are able to work from 10 keV. Modular systems are preferred and InterWinner software is used for spectrum-evaluation. The typical limit of detection is in the orders of magnitude 10^{-1} or 10^{-2} Bq/kg fresh weight in meat, fruit, vegetable, cereals and milk depending on the sample type, the isotope and measuring systems; for soil this value is mainly one order higher. Certified reference materials (CRM further on) are available with same density and same geometry as ashed samples from National Office of Measures or from IAEA. For emergency situation there are other reference materials with the matrix of 1 g/cm^3 density in different geometries prepared from QCY48 and QCB40 certified reference materials.

3. Alpha spectrometry by PIPS detectors: This technique together with the analytical chemistry background is available in two county laboratories and in the central laboratory. The usual collecting time of alpha-spectrum is one week, it is lengthy but necessary to achieve the required sensitivity. J LaRosa and et. al. (1992) published a combined method for determination of Pu-isotopes, ^{90}Sr and ^{144}Ce isotopes in environmental samples, this method gave the basis of the method used in the network. Limit of detection is 0.02 Bq/kg for $^{239+240}$Pu and 0.04 Bq/kg for ^{238}Pu in air-dried soil.

   Uranium isotopes and ^{210}Po are also determined from the natural radioactive isotopes.
4. Liquid scintillation technique: The tritium content of leafy plants and water of Danube is determined by this technique. Samples are gathered from Bács and Tolna counties, from the vicinity of the Nuclear Power Plant of Paks. Sample preparation is carried out by a special combustion technique. Low diffusion plastic vials and Ultima Gold LLT cocktail are used for determination of tritium by Quantulus 1220. With the applied method has the minimum detectable activity of 0.3 Bq/kg fresh weight in plants and water also.

   ^{241}Pu is also routinely determined by LSC using the source prepared for alpha-spectrometry.

The listed methods are routinely involved in the monitoring program, which is of high importance in case of emergency, because the method is in everyday use can be reliable in the conditions when the quick and reliable analysis and response is essential.

6.3. Quality assurance on the network level

Laboratories are accredited according to MSZ EN ISO/IEC 17025, participation in proficiency tests and intercomparisons is required. Central laboratory takes part in international runs, mainly organised by AQCS of IAEA and the laboratory became a member of Analytical Laboratories for Measurement of Environmental Radioactivity (ALMERA) Network co-ordinated by IAEA.

Intercomparison runs are organised for the network regularly, once or twice a year. Mainly natural samples were chosen, from which we could collect large amounts, and their activities were about or slightly above the typical natural levels, for example mud from lake Balaton, red cinders, humus and so on. After homogenisation and test required in the case of in-house reference material the samples were distributed among the laboratories. Participation in the intercomparison has been mandatory for the stations of the network. Moreover, voluntary participants were generally recruited, including international laboratories as well. In the 90's the results were evaluated according to basic statistical methods and the recommendation of Organisation and Analysis of Data, Protocol for the Food Analysis Performance Assessment Scheme (FAPAS). From 2000 proficiency test are evaluated according the suggestions of Analytical Quality Control Service (AQCS) of International Atomic Energy Agency (IAEA), taken into account the deliverables of INT/1/054 project on Preparation of in-house Reference Materials and Organisation of Proficiency Test Rounds.

Regarding the intercomparisons it was an important outcome, no significant difference occurred among the participating networks. For example the laboratories of the foreign networks and the members of different Hungarian networks did not formulate separate groups. This was the case for all kinds of samples (i.e. red cinder, red mud, humus). This is all the more interesting, because networks usually apply the same standards and CRM's and these sets are generally different among networks. Being Collaboration centre of IAEA the network has more possibility to get well characterised reference materials, which helps a lot in the good laboratory practice.

The main technical requirement is to have uncertainty of a measurement at least so small, that the spatial variation of the radionuclide could be followed. The network made and makes effort to reach this goal, to have reliable dataset describing the situation of the country well.

Annual meetings are organised for the network, where each problem is discussed, some exercises are held and solutions are harmonised. These events are important not only from techniqual point of view, regarding to having harmonised radioanalytical methods, but from the point of view of team-building also, which must be a part of the everyday work. Again in the case of emergency situation, easier to communicate and rely on the collaborator who is well known among the team and the personal contact with each other also should be perfect inside the team.

In normal case, keeping an eye on 2000/473 Euratom, and recommended tolerance level for foodstuffs (1. Chapter) and the mentioned uncertainty evaluation given in chapter dealing with background determination, the capacity of the network is the following:

- determination of radiostrontium: 2000 sample pro year,
- gamma-spectrometry by HPGe detectors: 3500 sample pro year,
- gross alpha measurement: 2000 sample pro year,
- alpha-spectrometry (U-isotopes, Pu-isotopes, ^{241}Am): 100 sample pro year,
- tritium: plants - 50 sample pro years, water – 300 sample pro year

- the number of the analysis of ^{241}Pu is determined by the capacity of the source preparation for alpha-spectrometry, there is no limit from the side of liquid scintillation spectrometry.

In nuclear emergency the normal requirements for sensitivity could not be fulfilled, usually keeping two orders of magnitude worse is enough for the decisions and suggestions for countermeasures. During the trainings and the intercomparisons this responses of the laboratories are also tested and reporting the results with associated uncertainty is also required. Without these kind of checking the reliability of the results is questionable during an emergency situation. The following capacity should be achieved with extra staff for the administration and sample preparation:

- determination of radiostrontium: 100 sample pro week,
- gamma-spectrometry by HPGe detectors: 500 sample pro day,
- scintillation gamma-spectrometry: 2000 sample pro day,
- gross alpha measurement: 80 sample pro day,
- tritium in water 50 sample pro day.

6.4. Summary

For the managing the countrywide network dealing with the control of foodstuff and agricultural production the following criteria has to be kept in mind:

- there is a need of the independent control from any economical organisation and interest, because the food supply for public and the agricultural production is strategic question; only the governmental control is acceptable taking into account the need of the society;
- the location of the laboratories has to be taken account the total coverage of the country, no problems could be arised from the sample-transport or providing the result even in emergency situation;
- keeping the economic efficiency and high quality of work in sight the control of the radioactivity has to be based on the infrastructure of general food control system, the monitoring schedule has to be issued for controlling the inland production, export and import products;
- the yearly revised monitoring program has to contain above the controlling the foodstuffs, samples giving information of the agricultural environment among them the forest and wild animal management, stationary elements for providing the long

term tendencies and the same time it is essential to give floor for the changes in the field of production and consumption habits;

- harmonisation of analytical work and the quality assurance system is essential;
- the rapid methods applicable in emergency situation has to be kept in everyday use beside the nuclide selective measurement techniques providing more accurate results and better minimum detectable activity concentrations;
- keeping of the staff well-trained is important, with courses, intercomparisons and proficiency tests.

According to my vulnerability study in this work the monitoring program could be extended with the controlling more carefully the forest ecosystem because of the wild animal management and the slow clearing process of this ecosystem, which means sampling different elements of a given foodchain in the same time and location. The other ecosystem which is not controlled in detail is the fresh water ecosystem connected with the riparian zone. Fish is the only sample type collected and measured regularly from these ecosystems**.** The foodchain regarding the agricultural production is appropriate, keeping sample pairs coming from the same time from the same location representing different parts of foodchain in monitoring schedule is necessary. According to my practice it is important to leave some free capacity in normal monitoring program. This part should be at least 10 % of the total amount the samples, because every laboratory has to be some spare capacity for occasional investigations or just for specialities in production of the county which could not be foreseen at the time of issuing the monitoring program.

7. Summary

15 years of practice in monitoring of foodstuff and agricultural environment gave the base of the PhD theses. After thoroughly studying the legislation in force regarding the radioisotope content of foodstuffs and feedstuffs either relating for European Union or international guidance I feel it necessary to complete the follow-up regulations with the multilevel regulation applicable in normal circumstances and prolonged emergency situation (longer than 1 year). The radioactivity of foodstuffs and feedstuffs has to be regulated in connection with each other. The recommended levels are the following:

- tolerance level is applicable for the whole population in normal circumstances, this level is derived from 0.1 mSv/year effective dose, choosing the minimum value from the activity concentration of foodstuff calculating for children and for adults taken into account the different age-dependent parameters. Based on the tolerance level the decision limit was determined depending on the combined standard uncertainty of the isotope specific measurement;
- acceptance level is applicable in case of prolonged emergency situation (longer than 1 year) for the whole population but it contains separate values for children less than 1 year old and for adults. The base of the derivation is 1 mSv/year effective dose applying the protection factor of five for children and three for adults;
- upper limit of the background: regarding the above recommended levels and the base of their derivation means additional dose above the background level, it is essential to determine the upper limit of the activity concentrations in foodstuffs and feedingstuffs coming from natural background with the investigating such a period which follows at least twenty years of accidental release free term;
- feedstuffs: applying the acceptable level for feedstuffs deriving from the recommended acceptance level of foodstuffs gives enough safety both for population and for animals themselves. During the derivation the animal specific transfer parameters were taken.

Supporting the decision making procedure in the field of agriculture I appointed those processes which determine the vulnerability of the terrestrial foodchain. ^{90}Sr transfer for soil-plant system is applicable to indicate the vulnerable agricultural area. The gastro-intestinal absorption of ^{137}Cs gives the most vulnerable part of the feed-animal transfer. The ration of $^{241}Pu/^{239+240}Pu$ is very sensitive and well applicable to indicate the origin of the isotopes from the nuclear installations even in very small concentration range. This can be a key subject for the planning of further surveys. Analogue elements and parameters are appropriate for the estimation of the necessary radioecological parameters and indication of processes in the environment with degree of certain prudence. In the terrestrial environment the riparian zone is the most vulnerable ecosystem, I estimated the accumulation of the ^{137}Cs isotope in the frog by combining the advantages of allometric and kinetic approach.

On the base of above mentioned investigations the criteria were summarised for managing the monitoring network having task of controlling the elements of foodchain, especially foodstuffs and factors influencing the agricultural production.

New scientific results:

1 thesis: I elaborated the guideline level system for regulating the radioactive isotope content of food and feedstuff, which takes into account the all possible sources, among them releases from the nuclear installations both during the normal operation and accidental situations. I recommend the application of the age-independent tolerance level based on the risk of $5x10^{-6}$ in normal circumstances. In case of prolonged emergency situation (longer than one year) the acceptable levels are applicable for two age groups (children below one year and adults) based on the additional dose of 1 mSv/year. Decision limits were defined for each levels depending on the measurement combined standard uncertainty. In case of more radioisotope found in the food or feedstuff the addition rule has to be applied.

2. thesis: The radionuclide content of food and feedstuffs has to be regulated connected with each other. Derivation of the acceptance level for feed from the acceptance level of food of animal origin gives stricter regulation, than the recommended dose levels for biota. The difference between the dose of animal via ingestion of the feed of the acceptance level is two order of magnitude less than the suggested dose limit of biota, therefore there is no need of other justification like addition rule or tolerance level, this difference is the guarantee of the proper protection of animals too.

3. thesis: The regulations for foodstuffs and feedingstuffs are based on additional dose from all possible sources above the background level, therefore I appointed the upper level of background level for foodstuffs and feedingstuffs, taking into account the monitoring results of the term following at least twenty years of accidental release free period. The upper level of the background was defined for artificial radionuclides according to lognormal distribution and for natural radioisotopes according to normal distribution. The activity concentration above this level requires investigation. During the evaluation of the monitoring dataset of the network it is essential to have a clear idea of the uncertainties attributed to the measurements of the detection of an isotope in a network level [1, 2].

4. thesis: For the long term effect of the releases on the agricultural production the radioecological vulnerability criteria were appointed. ^{90}Sr transfer between soil and plant gives an appropriate discrimination for the vulnerable agricultural areas. The absorption of ^{137}Cs in the animals gives the most vulnerable point of the feed and animal transfer. The isotope ratio of $^{241}Pu/^{239+240}Pu$ gives information even at low concentration levels whether the contamination came from nuclear installation, which can determine the emphasis of further survey. Among the analogues the correlation between ^{210}Pb and ^{137}Cs gives information for the ratio of resuspension of artificial isotope among the root uptake and fresh fall-out [3, 4, 5, 6, 7].

5. thesis: For the managing the countrywide network dealing with the control of foodstuff and agricultural production the following criteria has to be kept in mind:

- there is a need of the independent control from any economical organisation and interest, because the food supply for public and the agricultural production is strategic question; only the governmental control is acceptable taking into account the need of the society;
- the location of the laboratories has to be taken account the total coverage of the country, no problems could be arised from the sample-transport or providing the result even in emergency situation;

- keeping the economic efficiency and high quality of work in sight the control of the radioactivity has to be based on the infrastructure of general food control system, the monitoring schedule has to be issued for controlling the inland production, export and import products;
- the yearly revised monitoring program has to contain above the controlling the foodstuffs, samples giving information of the agricultural environment among them the forest and wild animal management, stationary elements for providing the long term tendencies and the same time it is essential to give floor for the changes in the field of production and consumption habits;
- harmonisation of analytical work and the quality assurance system is essential;
- the rapid methods applicable in emergency situation has to be kept in everyday use beside the nuclide selective measurement techniques providing more accurate results and better minimum detectable activity concentrations;
- keeping of the staff well-trained is important, with courses, intercomparisons and proficiency tests [8, 9].

<u>Scientific papers in connection with theses</u>

[1] B. Varga, S. Tarján: Reporting of Uncertainty in Environmental Monitoring of Radionuclides, Combining and Reporting Analytical Results, Eds A. Fajgelj, M. Belli, U. Sansone, The Royal Society of Chemistry, ISBN: 0854048480, ISBN-13:9780854048489, Cambridge, (2007) 195-204.

[2] Tarján S, Varga B, Sebestyén T: A ^{222}Rn minőségbiztosítási szemszögből, III. Magyar Radon Fórum, ISBN 963 6420 99 8, Veszprém (2006) 51-57.

[3] B. Varga, S. Tarján: Determination of ^{241}Pu in environmental samples, Applied Radiation and Isotopes 66 (2008), 265-270.

[4] B Varga, S Tarján, N Vajda: Plutonium isotopes in the Hungarian environment, Journal of Environmental Radioactivity 99 (2008) 641-648.

[5] B. Varga, E Leclerc, P. Zagyvai: The role of analogues in radioecology, Journal of Environmental Radioactivity folyóiratba beadott

[6]IAEA TECDOC: Handbook of parametervalues for the prediction of radionuclide transfer to humans in terrestrial and freshwater environments – megjelenés alatt
2. Definitions, data analysis and use of analogues
2.1. Definitions and units - S.Fesenko, N.Sanzharova, M.Vidal, A.Shubina, Y.Thiry, E. Reed, B. Varga
2.4. Use of analogues - E. Leclerc, K. Tagami, S. Uchida, B. Varga

[7] IAEA Technical Report Series 364 amendment in preparation - Handbook of parametervalues for the prediction of radionuclide transfer in temperate environments – contributor – megjelenés alatt

[8] B. Varga, S. Tarján, M. Süth, B. Sas: Radionuclide Monitoring Strategy for Food-Chain in Hungary, Journal of Environmental Radioactivity, 86 (2006) 1-11.

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8. Összefoglalás

A PhD dolgozat alapját mintegy 15 évnyi monitoring munka végzése, szervezése adta. Alaposan áttekintve a jelenlegi, élelmiszerek és takarmányok radioaktív izotóptartalmára vonatkozó nemzetközi és európai szabályozást, szükségesnek látom az élelmiszerek radiológiai biztonságára vonatkozó, hatályos, baleseti helyzetet követő rendelkezéseket kiegészíteni egy többszintű, az élelmiszert és takarmányt egyidejűleg szabályozó határértékrendszerrel, amely a következőkön alapul:

- tolerálható szint a lakosság teljes körére alkalmazandó, normál, nem baleseti helyzetre vonatkozó, izotóponkénti koncentrációértékeket jelöl ki a 0.1 mSv/év effektív dózisból származtatva, minimumát véve a gyermekek és a felnőttek táplálékára kiszámolt értéknek. Az egyes izotópkoncentrációk meghatározásakor kapott mérési összbizonytalanságot felhasználva az általános tolerálható szint alkalmazásához eseti döntési szintet határoztam meg;
- elfogadható szint a lakosság körében alkalmazandó, az egy év alatti gyerekekre és felnőttekre külön megállapított értékekkel; az elhúzódó baleseti helyzetre (hosszabb, mint 1 év) vonatkozó izotóponkénti aktivitáskoncentráció határérték 1 mSv/év effektív dózisból származtatott koncentráció, felnőttekre hármas illetve egy év alatti gyerekekre pedig ötös „védelmi tényező” alkalmazásával;
- háttér határkoncentráció érték: mivel a fenti két szint, valamint a származtatás alapját képező effektív dózisok is a háttérsugárzásból származó terhelés feletti szintet képviselik, ezért szükséges egy hosszabb, legalább húsz év „friss kibocsátástól mentes” időszak utáni periódus vizsgálata a környezetben várható természetes és mesterséges izotópok határkoncentrációinak kijelölésére, a különböző élelmiszerekre és takarmányra egyaránt;
- takarmányokra elégségesnek tartom csak elfogadható szint megállapítását, ami az élelmiszerekre javasolt elfogadható szintből származtatott, az állati szövetekre vonatkozó egyedi átviteli tényezők alkalmazásával. Számítást végeztem arra, hogy az emberek védelmével teljesül az állatok védelme is.

A baleseti döntéselőkészítést is elősegítendő, a mezőgazdaság biztonsága szempontjából kijelöltem azokat a transzportfolyamatokat, amelyek a tápláléklánc érzékenységét meghatározzák. A sérülékeny termelési területek kijelölésére alkalmas a ^{90}Sr talaj-növény transzport folyamat átviteli tényezője. A takarmány-állat átvitel legérzékenyebb elemét a ^{137}Cs izotóp felszívódása jelenti. A ^{241}Pu/$^{239+240}$Pu izotóparány rendkívül érzékenynek bizonyult a nukleáris létesítményből származó kibocsátás jelzésére, még rendkívül alacsony koncentráció tartományban is, ami a további felmérések tervezésének súlypontjára meghatározó lehet. Az analóg elemek, paraméterek megfelelő körültekintéssel alkalmasak a szükséges radioökológia paraméterek hiányának pótlására, valamint a környezetben lezajló folyamatok indikációjára. A szárazföldi mérsékeltövi ökoszisztémák közül a mocsaras területek a szennyezésekre legérzékenyebben reagáló rendszerek, ezért becslést végeztem a békák szervezetében felhalmozódó ^{137}Cs tartalomra vonatkozóan allometrikus illetve kinetikus modell ötvözésével.

A fenti elemzések alapján felállítottam egy általános kritériumrendszert a tápláléklánc egyes elemeinek, elsősorban az élelmiszerek illetve a mezőgazdasági termelést befolyásoló környezeti tényezők vizsgálatával foglalkozó monitoring hálózat számára.

1 tézis: Kidolgoztam egy irányadó határértékrendszert az élelmiszerek radioaktív izotóptartalmára, amely minden lehetséges szennyező forrást figyelembe vesz, köztük a nukleáris létesítmény normál üzeme illetve a baleseti körülmények során a tápláléklánc szennyezettségét befolyásoló radioizotópokat. Az $5x10^{-6}$ kockázathoz rendelhető, korcsoport-független tolerancia szintet normál körülmények között javasolom alkalmazni. Az 1 mSv/év dóziskorlát betartását garantáló, két korcsoportos (egy év alatti gyerekekre és felnőttekre) elfogadhatósági szint viszont egy évnél hosszabban tartó baleseti helyzetre alkalmazandó. Mindkét szintre megállapítottam a mérési bizonytalanságtól függő döntési szinteket, valamint több izotóp együttes jelenlétére érvényes összegzési szabályt.

2. tézis: Az élelmiszerek és a takarmányok radioizotóp tartalma együtt szabályozandó, mivel az állati eredetű táplálékok esetében az elfogadható szint takarmányokra visszaszármaztatása szorosabb megkötést jelent, mint az eddigiek során javasolt határértékek az állatok dózisterhelésére. Nincs szükség tolerancia szint és külön összegzési szabály megállapítására, mivel a legalább két nagyságrend különbség a takarmányok radioizotóp tartalmából származó dózis és az állatokra javasolt elfogadható dózis között garanciája a védelem megfelelőségének.

3. tézis: Mivel a lakosság dózisterhelésére vonatkozó szabályozás a lehetséges összes mesterséges forrásból származó többletdózison alapul, az adott környezetre jellemző koncentráció értékek ismeretére van szükség, minimálisan 20 év baleseti kibocsátástól mentes periódus utáni időszak országos monitoring ellenőrzéseit véve alapul. A háttérre vonatkozó határkoncentrációk megállapításánál a mesterséges izotópokra a lognormál eloszlást, míg a természetes eredetűeknél a normál eloszlást célszerű alkalmazni. A határkoncentrációt meghaladó mért értékek kivizsgálást igényelnek. Mivel egy országos hálózatban több laboratórium dolgozik együtt, az eredmények országos szintű értékelésénél tisztában kell lenni az adott izotóptartalom elemzésének hálózati szinten meghatározott mérési bizonytalanságával [1, 2].

4. tézis: A kibocsátás hosszú távú, mezőgazdasági termelés szempontjából fontos hatásának elemzésére radioökológiai érzékenységi kritériumokat jelöltem ki. A sérülékeny termelési területek kijelölésére alkalmas a ^{90}Sr talaj-növény transzport folyamat átviteli tényezője. A takarmány-állat átvitel legérzékenyebb elemét a ^{137}Cs izotóp felszívódása jelenti. Rendkívül kis koncentrációértékeknél is információt szolgáltat a ^{241}Pu/$^{239+240}$Pu izotóparány annak eldöntésére, hogy nukleáris létesítményből származott-e a kibocsátás, ami a felmérések tervezésének súlypontját határozza meg. Az analógiák közül a ^{210}Pb és a ^{137}Cs korrelációja következtetni enged a mesterséges izotóp reszuszpenziójának mértékére a kihullás illetve a gyökéren keresztüli felszívódás folyamatai mellett. [3, 4, 5, 6, 7]

5. tézis: Az élelmiszerek és a mezőgazdasági környezet monitoringjával foglalkozó országos laboratórium hálózat működtetésének alábbi kritériumait határoztam meg:

- mivel az élelmiszerellátás és a hozzá kapcsolódó mezőgazdasági termelés országos szintű stratégiai kérdés, ezért az ellenőrzésnek függetlennek kell lennie minden gazdasági szervezettől és érdektől, kizárólag a társadalom érdekét szolgáló állami irányítás fogadható el;
- a laboratóriumok elhelyezkedése biztosítéka legyen a területi lefedettségnek úgy, hogy baleseti helyzetben se okozzon gondot a mintaszállítás és az eredményszolgáltatás;
- a gazdaságosság és a minőségi munka fenntarthatósága érdekében az élelmiszerellenőrzés teljes vertikumára kiépített rendszer infrastruktúrájának

kihasználásával és figyelembevételével elkészített monitoring tervet kell kiadni mind a hazai, az export és az import termékek ellenőrzésére vonatkozóan;

- az évente felülvizsgált monitoring tervnek tartalmaznia kell az élelmiszerek radioaktivitásának felmérésén kívül a mezőgazdasági környezetre információt adó mintákat, beleértve a vadgazdálkodást; állandó elemeket a hazai termelés monitorozására a hosszú távú tendenciák megadására, valamint teret kell adni a fogyasztási és termelési szokások módosulásából adódó változásoknak;
- az analitikai módszerek és a minőségbiztosítási rendszer harmonizálása;
- a gyors, baleseti helyzetben alkalmazandó mérési metodikák napi gyakorlatban tartása a normál körülmények között megkívánt, nuklidszelektív, kedvezőbb kimutatási határt biztosító méréstechnika mellett;
- munkatársak folyamatos tréningben tartása képzéssel, körvizsgálatokkal és jártassági tesztekkel. [8, 9]

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