A DYNAMIC FOOD-CHAIN ALGORITHM FOR ESTIMATION OF MIGRATION OF RADIONUCLIDES THROUGH THE LIVING ENVIRONMENT CONTINUOUSLY INFLUENCED BY THE ROUTINE ATMOSPHERIC DISCHARGES FROM NUCLEAR POWER PLANTS

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1 Radiological impact of routine atmospheric releases of radionuclides

A certain amount of activity (even if extremely low) is continuously released to the atmosphere from venting stacks of nuclear power plants (NPP) during their normal routine operation. The migration of radioactive isotopes in a NPP during normal operation is controlled by special purification and filtering systems and only reminder is released to free atmosphere. The level of the releases is continuously checked by online measurements. In spite of the low-level activity outputs the rigorous evidence of compliance with governmental limits and all regulations resulting from the Atomic Law of the Czech Republic have to be submitted.

All possible pathways of human body irradiation have to be taken into account, mainly:

- exposure to ***external irradiation*** from passing cloud (*cloudshine*) and from deposited material (*groundshine*)
- ***internal irradiation*** due to *inhalation* of contaminated air (including portion from *resuspension*) and due to activity intake from contaminated foodstuffs (*ingestion*)

The calculation of annual doses is based on detailed temporal and spatial modelling of the radiological situation in vicinity of NPP. Particular weather situations are weighted by annual weather statistics and the following main driving characteristics are found:

- annual-averaged ground level activity concentrations for each radioisotope in the air and corresponding values in the effective height of the release
- annual-averaged deposition rate and its time integral (total deposition on the ground) for each radioactive isotope calculated on the basis of long-termed (annual-averaged) coefficients of dry and wet deposition

2 Propagation of activity through the living environment

Once the radioactive material is released from venting stacks the admixtures are incorporated in the plume and drifted in the downwind direction. The polluted plume expands horizontally and vertically due to turbulent diffusion in the atmosphere. The radionuclides in the plume are bound in a certain physical-chemical forms (aerosols, elemental form, organically bound) and during the dispersion are removed from the plume due to several removal mechanisms. The most important are *radioactive decay* (including daughter products build-up process), *dry deposition* (gravitational setting and deposition due to contact of the contaminated plume with the ground, vegetation or urban structures) and *wet deposition* – removal by rainout (precipitation formation process inside of the plume) or by washout (interaction between falling drops and admixtures).

Within the advection and diffusion transport calculations many other factors have to be taken into account such as *thermal structure* of the atmosphere (here Pasquill-Gifford notation), surface *roughness* and other *land cover* characteristics, *orography* of the terrain, *reflection* from the ground and top of the mixing layer or inversion layers, the effect of initial *plume rise* due to vertical momentum and buoyancy, recirculation in the *wake region* of the near standing buildings. The effects are usually expressed by semi-empirical expressions derived from experimental results [1]. An approach for atmospheric modelling is given in [8]. Practical adoption for regions of NPP Temelín and Dukovany are described in [6] where more detailed discussion related to weighting of the particular weather situations by the annual weather statistics is given. In the final stage the activity dispersed in the air and deposited on the ground enters into food chains causing a certain contamination of the foodftuffs and feedstuffs. An example of the meteorological model chain calculation is shown on fig. 1. Spatial distribution of annual-average ground level activity concentration in air for nuclide C14 is shown for vicinity of NPP Temelín (release from venting stack at height 100 m).

Annual near-ground activity concentrations in air of isotope C14 [Bq/m3]

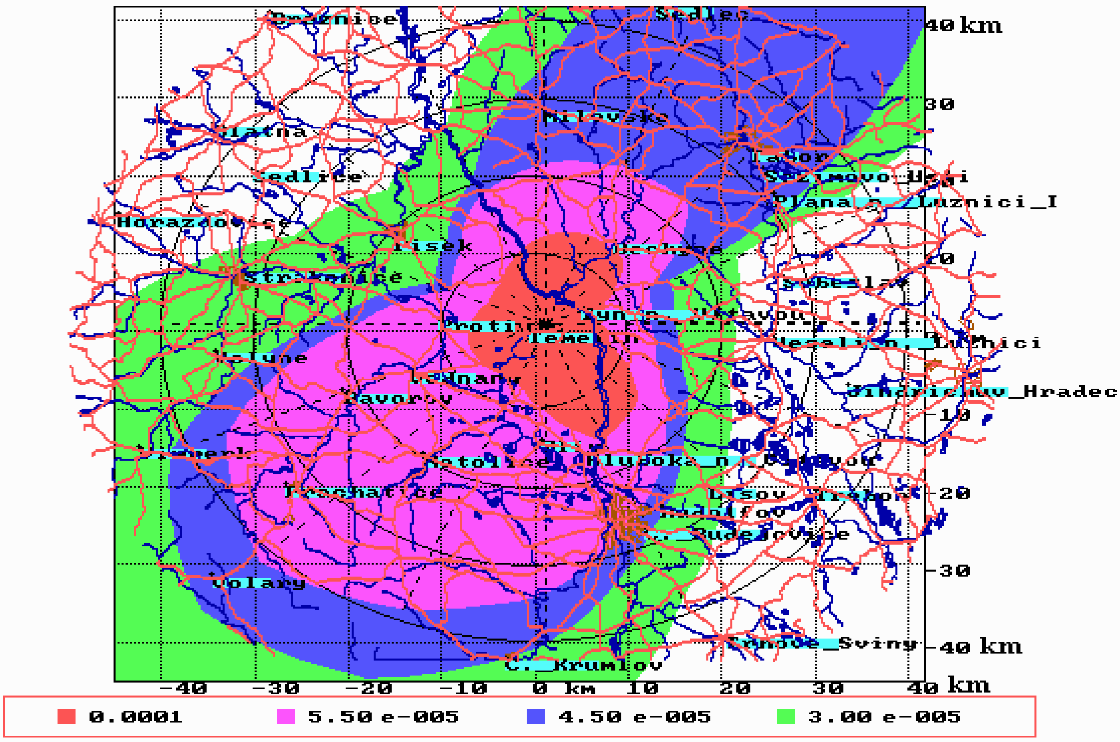

Fig. 1: Spatial activity distribution around NPP Temelin (HOSKER dispersion sch.)

Current estimations of the exposures due to different pathways comprise in general high degree of conservatism (approach of *potential doses*). As knowledge in the field grows, more precise algorithms are developed and the conservatism is decreased. The estimation is then expressed in the terms of *expected doses*. The trend is documented in the following text for case of improved dynamic modelling of ingestion pathway.

3 Dynamic food-chain modelling for the case of routine atmospheric radioactive discharges

Local dynamic model ENCONAN for transport of radionuclides through food chains was developed in [3]. Current knowledge in the field (mainly based on the European ECOSYS code) from early 90's was adopted for average Czech conditions taking into account local *consumption habits* (dependence on season and age), *agricultural production* scheme, average *agro-climatic conditions* and *phenologic characteristics* of the plants, *feeding diets* of animals, *time delays* during processing, transport and storage of foodstuffs and feedstuffs etc. The model was originally developed for incidental radioactive fallout in a certain Julian day of a year and with several extensions was implemented also in HAVAR code [7]. For normal operation of NPP an approach of equivalent number of discrete radioactive fallouts is usually used. The approach is now revised and an attempt for more detailed modelling for the case of long-termed continuous routine releases is presented.

The ecosystem surrounding the NPP is assumed to be continuously submerged into the contaminated environment where customary agricultural practices are applied. Root and foliar uptakes of activity into plants are modelled and contamination of foodstuffs and feedstuffs is predicted. Annual activity intakes are estimated for both direct consumption of plant products and consumption of contaminated animal products. Conservative ingestion scheme of "<u>*local production-local consumption*</u>" is still used (products produced at place x,y are here also consumed). According to the new approach the activity intake of isotope <u>*n*</u> (with exception of noble gases) is expressed as

$$A_l^{a,n}(x,y;t) \;=\; \bar{S}^n(x,y)\cdot I\mathcal{E}_l^{a,n}(t) \qquad \text{Eq.(1a)}$$

and for C14 and H3:
$$A_l^{a,n}(x,y;t) \;=\; \bar{C}^n(x,y;z=0)\cdot I\mathcal{E}_l^{a,n}(t) \qquad \text{Eq.(1b)}$$

$A_l^{a,n}$ ..... *total activity intake of isotope <u>n</u> in [Bq] for a person from age category <u>a</u> which consumes during the time period <u>t</u> contaminated product <u>l</u> (produced at place x,y);*

$\bar{S}^n(x,y)$ .... *time-averaged (during period t) deposition rate [Bq.m^{-2}.s^{-1}] of isotope <u>n</u> at (x,y);*

$\bar{C}^n(x,y;z=0)$ .... *time-averaged ground level activity concentration of <u>n</u> in air at (x,y) [Bq.m^{-3}];*

$I\mathcal{E}_l^{a,n}(t)$..... *time-integrated (within period t) activity intake of isotope <u>n</u> [m^2.s resp. m^3] due to the consumption of product <u>l</u> for person from <u>a</u> (normalised to unit deposition rate S resp. unit C);*

3.1 Foliar uptake of radionuclides

Let t^l_{veg} and t^l_{har} denote beginning of vegetation period and harvest time (in Julian days) of plant <u>*l*</u> . In the process of the continuous deposition on leaves the initial deposited activity is decreased due to weathering effects (wind, rain), radioactive decay and tissue ageing (growth dilution effect). Furthermore, the fraction of activity translocated to other parts of the plant should be taken into account. The analysis must distinguish between plants which are used totally (e.g. leafy vegetables, grass) and plant of which only a special part is used (cereals, potatoes). Translocation from leaves to the edible parts of the plant has to be accepted. This process is strongly dependent on the physiological behaviour of the isotope considered. The translocation plays important role for mobile elements (e.g. I, Cs, Mn, Te) whilst only the direct deposition onto the edible parts is assumed for the immobile elements (e.g. Sr, Ba, Zr, Nb, Ru, Ce, Pu).

Specific activity ε^n_l of nuclide <u>*n*</u> [m^2.s.kg^{-1}] at time t' after harvest (normalised to unit deposition rate) cumulated in 1 kg of the plant <u>*l*</u> during the whole vegetation period is expressed as

$$\varepsilon_l^n(t_{har}^l + t') = \left\langle \frac{1}{V_c^l}\cdot\sum_{ti=tveg}^{ti=thar} SP_i^n\cdot R^l(ti)\cdot Z^l(ti)\cdot\exp\left[-(\lambda_w+\lambda^n)\cdot(t_{har}^l - ti)\right]\right\rangle\cdot\exp(-\lambda^n\cdot t') \qquad \text{Eq.(2)}$$

V_c^l ..... *yield of fresh product <u>l</u> at harvest time [kg.m^{-2}]*
SP_i^n ..... *coefficient for total daily deposition at day <u>ti</u> ; for unit dep. rate SP = 86 400 [s]*
$R^l(ti)$ ..... *interception factor - fraction of deposition onto plant <u>l</u> in day ti; ti∈< t^l_{veg} ; t^l_{har}>; in dependence on the stage of development of plant canopy;*
$Z^l(ti)$ ..... *simple correction factor (empirical formulas) for approximation of the case of translocation of activity from leaves to other considered (edible) parts of the plant in dependence on the vegetation phases;*
λ_w ...... *loss activity rate due to weathering [d^{-1}] – a value equivalent to half-life of 25 days is here assumed;* λ^n ..... *radioactive decay rate [d^{-1}]*

3.2 Root uptake of radionuclides

Let us assume that the isotopes are well mixed within root zone. In general, the root uptake of activity is calculated from the concentration of activity in the soil using equilibrium transfer factors which give the ratio of activity concentration in plants (fresh or dry weight) to soil (dry soil). Continuous specific deposition [s] of radionuclide n during vegetation period of plant l normalised to unit deposition rate leads to the following resulting deposition on the ground at harvest time

$$\Omega_l^n(t_{har}^l) \;=\; \Omega_l^n(t_{veg}^l)\cdot\exp\left[-\lambda_{ef}^n\cdot(t_{har}^l-t_{veg}^l)\right] + \sum_{ti=tveg}^{ti=thar} SP_i^n\,.\,(1-R^l(ti))\cdot\;\exp\left[-\lambda_{ef}^n\cdot(t_{har}^l-ti)\right]$$

$$\lambda^n_{ef} \;=\; \lambda_f + \lambda_m + \lambda^n \qquad \text{Eq.(3)}$$

where the normalised deposition on the ground for time of the beginning of vegetation period t^l_{veg} in the year (M+1) after starting of NPP operation can be approximated by formula

$$\Omega_l^n(t_{veg}^l) = \frac{86400}{\lambda_f+\lambda_m+\lambda^n}\cdot\left[1-\exp(-(\lambda_f+\lambda_m+\lambda^n)\cdot(t_{veg}^l+M\cdot 365)\;)\right] \qquad \text{Eq.(4)}$$

Besides of radioactive decay the deposited activity on the ground which is available for further root transport is decreased by the effects of migration of isotopes out of the root zone (rate of migration λ_m in [d^{-1}] is provided from experiments) and fixation of radionuclides in soil in the form of immobile substances (different fixation rates λ_f in [d^{-1}] are used for Cs and Sr, for other elements the fixation is assumed of minor importance). When using the idea of equilibrium transfer factors, then the specific activity ${}^R\varepsilon^n_l$ of nuclide n [$m^2.s.kg^{-1}$] (normalised to unit deposition rate) cumulated in 1 kg of the plant l during the whole vegetation period due to root transport has form

$${}^R\varepsilon_l^n(t_{harl}^l) = \;\Omega_l^n(t_{har}^l)\cdot BV_l^n\;/\;PH_l \qquad \text{Eq.(5)}$$

BV_l^n .... *soil to plant transfer factor of nuclide n for plant type l [Bq.kg^{-1}plant / Bq.kg^{-1}soil];*
PH_l .... *effective root zone (surface weight of contaminated soil) in [kg/m^2] - depends on ploughing practices (depth of 0.25 m and 0.1 m are assumed for arable soil or pasture soil, respectively; different values are applied for different cuts of forage);*

3.3 Activity intake for case of direct consumption of the products

Provided that the daily consumption rate PD_l^a [$kg.d^{-1}$] of product l by a person from the age category a is constant, the integral normalised activity intake in period < t_{del}^l ; t_{con}^l > is given for the case of direct product consumption (leafy vegetable spring + autumn, fruit vegetables, root vegetables, cereals, potatoes, fruit) as

$$I\varepsilon_l^{a,n}(t_{con}^l) \;=\; \left[\;\varepsilon_l^n(t_{har}^l) + {}^R\varepsilon_l^n(t_{har}^l)\;\right]\cdot\;PD_l^a\cdot\frac{1}{\lambda^n}\cdot\left[\exp(-\lambda^n\cdot t_{del}^l)+\exp(-\lambda^n\cdot(t_{con}^l-t_{har}^l))\right] \quad \text{Eq.(6)}$$

t_{del}^l denotes time delay between harvest and beginning of consumption of the product l due to transport, processing and storage; t_{con}^l means end of consumption in a year.

3.4 Activity intake from consumption of contaminated animal products

Somewhat more complicated modelling scheme is introduced for this case. Let us assume 3 types of the main animal products b : milk, meat (beef, pork, poultry) and eggs. From the products are produced various foodstuffs p(b) (for example from the milk are produced the foodstuffs p : fresh milk, cream, cheese, milk dry, milk condensed, curd, others – with various specific time delays for consumption).

Feeding diets of animals are combined from the basic feedstuffs l (potatoes, wheat, barley, beet, maize (silage), perennial forage: 1. cut, 2. cut, 3. cut). There could be incorporated also some waste products from the foodstuffs processing (eg. whey for the pig diets). Daily intake of normalised activity of radionuclide n [$m^2.s.d^{-1}$] in day t' into animal body due to consumption of feedstuffs l again accounts for foliar and root uptake of activity

$$^{SUM}A^n(t') = \sum_{(l)} \left[\mathcal{E}^n_l(t') + {}^K\mathcal{E}^n_l(t') \right] \cdot PDK_l \qquad \text{Eq.(7)}$$

PDK_l is daily feeding rate [$kg.d^{-1}$] of the feedstuffs l . For description of the further transport of activity of isotope n into animal product b is used equilibrium transfer factor F_b^n [$d.kg^{-1}$ or $d.litre^{-1}$] which expresses the fraction of daily intake of contaminant n which appears in a unit of animal product b. Specific normalised activity of nuclide n [$m^2.s.kg^{-1}$] in 1 kg or litre of animal product b which is produced in a day t' is given by

$$\mathcal{E}^n_b(t') = {}^{SUM}A^n(t') \cdot F^n_b \qquad \text{Eq.(8)}$$

The relation is valid only for equilibrium state, without consideration of short-termed changes in feeding quantities or in animal weight in fattening. More precise solution based on more complex compartment metabolic models should be adopted in the future steps of analysis when appropriate data will be available. Time integrated intake of specific activity of contamitant n for a person from age category a during period from consumption beginning *tkp* to a certain day t due to consumption of foodstuffs p (produced from animal product b) can be written as

$$I\mathcal{E}^{a,n}_{p,b}(t) = \int_{tkp}^{t} \mathcal{E}^n_b(t') \cdot PD^a_b \cdot f1^a_{p,b} \cdot dt' \qquad \text{Eq.(9)}$$

tkp implicitly comprises all possible time delays for consumption; PD_b^a is daily consumption rate [$kg.d^{-1}$] of animal product b (in all related foodstuffs p(b)) for a person from age category a ; $f1^a_{p,b}$ means fraction of consumption of p in the initial value of b for age category a.

3.5 Total activity intake into a human body

Total time integrated normalised intake of activity of nuclide n for age category a due to both direct consumption and consumption of contaminated animal products is schematically written as

$$I\mathcal{E}^{a,n}_{TOT}(t) = \sum_{(l)} I\mathcal{E}^{a,n}_l(t) + \sum_{(b)} \left\{ \sum_{(p)} I\mathcal{E}^{a,n}_{p,b}(t) \right\} \qquad \text{Eq.(10)}$$

4 Overview of the main features of the food-chain algorithm

Ingestion scheme: Basic model from [3] for the Czech territory was adopted. Conservative "local production – local consumption" model is assumed. Local database has been created in [6] taking into account all phenomena mentioned above in the chapter 3. The process of data acquisition expresses an effort for assembling of realistic local-specific characteristics and time dependencies including various delays. For update of some items was used an experience from the knowledge base of the RODOS system [5].

Specific pathways for isotopes C14 and H3: The transfer of tritium and carbon-14 between atmosphere and terrestrial environment is more complex and simplified evaluation is performed using Eq.(1b). It is based on assumption that living environment components come into rapid equilibrium with the C14 or H3 in the atmosphere. For plants the important role plays foliar uptake during the photosynthesis process. According to the US NRC Reg. Guide 1.109 the concentration of the isotope X in vegetation (X=C14, H3) is calculated by assuming that there is known its equilibrium ratio f^X to the natural component. Concentrations of the nucleus in the plant *l* is in relation to the corresponding value in the near-ground layer of air derived from the annual near-ground activity concentration of X. In the model are used recommended values f^{C14}=1.0 and F^{H3}=0.5. A simple constant value of the specific activity ε_l^X is then used for each day of the vegetation.

Effects of seasonal consumption: More realistic seasonal scenarios can be treated in consumption scheme of a certain products. Considerable differences were found for some nuclides in relation with the average annual consumption. Sensitivity analysis was done for case of green vegetables where the total annual consumption was assumed to be realised within one month after harvest. For example the total activity intake of I131 was about one order higher than for the case of consumption during the whole year.

Estimation of various feeding practices: A certain hypothetical scenario for the 3 cuts of forage and corresponding feeding rates for dairy cows and cattle in fattening are shown on the fig. 2. The combination of indoor fattening in a stable and continuous pasture of the fresh grass can be studied. Let grazing regime starts in a Julian day *tkrm* and lasts for the whole vegetation period of the forage-1st cut until harvest time *thar1*. In the following days free grazing continues, but the complementary feeding from the already harvested forage-1st cut is realised (see continuous linear decreasing on the fig. 2) until the day *tx1*. After this day only continuous grazing takes place again until the harvest time *thar2* of the forage (2. cut), etc.

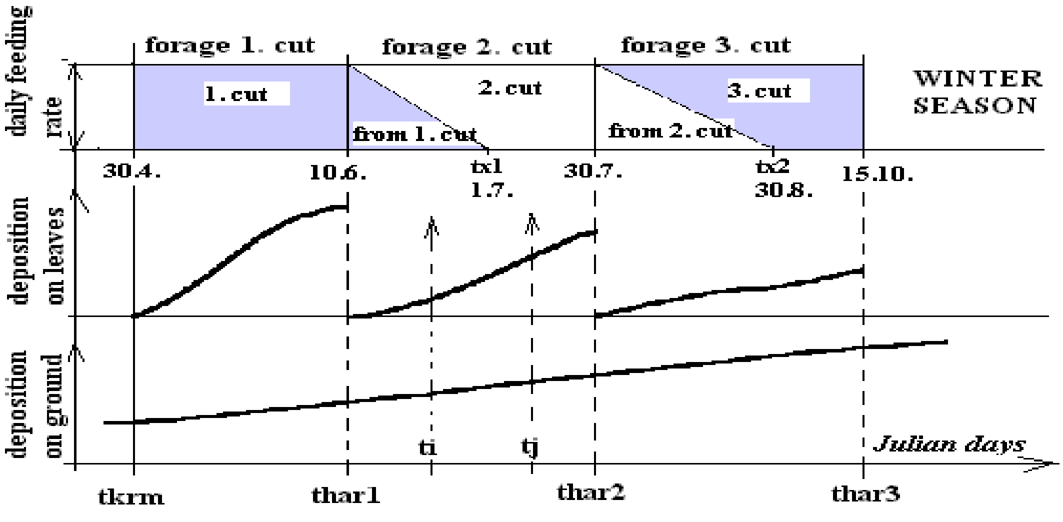

Fig. 2: Hypothetical feeding scenario for cows and cattle for 3 cuts of forage

The whole range of a year is divided by days *tkrm, thar1, tx1, thar2, tx2, thar3* into 7 intervals where the corresponding detailed modelling of deposition on leaves and ground is applied (in the winter season 2/3 of hay was obtained from the 1st cutting cycle and 1/3 from the 2nd cycle) . Let us notice that purely continuous grazing grass model is reached when enter on input *tx1= thar1* and *tx2=thar2.* More complicated formulas are derived separately for each of the time interval for calculation of specific activities in the grass and for resulted time integrals of the activity intake. Further discrimination for isotopes carbon-14 and tritium is introduced. This detailed modelling was adopted also into the code HAVAR [7], where the day of accidental radioactive fallout considerably influences the magnitudes of doses.

5 Results and areas of applicability, comparative and validation studies

The more precise values of the total normalised annual activity intake $I\mathcal{E}_{TOT}^{a,n}(t=1year)$ of each radionuclide <u>*n*</u> for age category <u>*a*</u> given by Eq.(10) represent the main result of this new approach of food-chain algorithm. In the following table 1 are included results for adult category for several isotopes from the given spectrum of annual releases of nuclides predicted for the routine operation of a typical WWER 1000 nuclear power plant.

Tab. 1: *Normalised annual activity intake of isotopes due to ingestion pathway – adults*

After start of NPP operation	*r a d i o n u c l i d e*							
	H3$^{(*)}$	**C14**$^{(*)}$	**Co60**	**Ni63**	**I131**	**Cs134**	**Cs137**	......
in **1st year** $^{(**)}$	0.12E+5	0.22E+6	0.19E+9	0.18E+9	0.51E+7	0.25E+9	0.29E+9	....
in **31st year** $^{(***)}$	0.12E+5	0.22E+6	0.26E+9	0.62E+9	0.51E+7	0.27E+9	0.36E+9	....

$^{(*)}$ ... normalised to unit of near-ground act. concentr. in air [m^3]; other nuclides: normalised to unit deposition rate [m^2.s]; $^{(**)}$ ... M=0 in Eq.(4) ; $^{(**)}$ ... M=30 in Eq.(4) ;

Real spatial distribution of the annual intakes of activity around NPP are found according to Eqs.(1). For example the distribution for annual intake of C14 is calculated when multiplying the values from fig. 1 by values for C14 from table 1. The results are submitted to consecutive estimation of radiological impact on population, particularly:

- Annual <u>*effective and equivalent doses*</u> are estimated. Committed doses from internal irradiation due to ingestion pathway are included. The corresponding doses in [Sv/y] are determined by multiplying the real annual intake of isotope by dose conversion factors enumerated in the Reg. no. 184/97 of the State Office for Nuclear Safety which conforms with the Czech Atomic Law. The method was used for purposes of various verification studies in the Safety Reports for the Czech NPPs. Profound analysis of compliance of the calculated conservative potential doses with obligatory regulatory limits is documented.
- The results from tab. 1 and detailed spatial distribution of annual near-ground activities and deposition rates (for release from both venting stack (100 m height) and engine buildings (45 m)) for the whole spectrum of discharged radioisotopes were delivered to TGM University in Brno for purposes of the EIA evaluation process according to the latest US EPA regulations [9]. It represents an alternative independent way of safety analysis in comparison with the dose evaluation concept mentioned above. <u>*Risk of cancer estimation*</u> is here based on hypothetically postulated Linear, No-Threshold (LNT) model.

Some other particular cases were analysed. For example the real discontinuous irrigation by contaminated water (12 events during vegetation period) was simulated by equivalent continuous spraying during the whole vegetation period. The results confirmed <u>*mutual adequacy*</u> of food-chain models for continuous and accidental deposition used in [6] resp. [7]. Extensive <u>*comparative studies*</u> of the presented algorithm were performed against the European code PC CREAM [2] and the results are documented in the part III of [6]. Similarly, the verification of the values from table 1 was performed. Due to a certain restriction on the PC CREAM input data the Czech local data had to be somewhat simplified. On the other hand this fact supports the idea on <u>*development of own local codes*</u>.

6 Prospects of the future development

Three main topics seem to be in the centre of interest when proceed from estimation of conservative potential doses to the more realistic expected doses:

- Adoption of more realistic ingestion scheme instead of the "*local production-local consumption*" one. For some cases of feeding and fattening the equilibrium transfer factor concept should be substituted by realistic compartment metabolic models.

- Introduction of multi-regional concept, when the whole territory is split into a certain number of so called *radioecological zones* according to the differences in climate, phenological characteristics, agricultural production and feeding regimes, consumption habits, soil types (strongly influencing soil to plants transfer factors) etc.
- Model parameters improvement, which includes reconstruction of various items on fine spatial grid (site specific data, soil type, land use, elevations, agricultural production, population, more detailed meteorological statistics etc.)

Utilisation of experience from the European project RODOS dealing with accidental releases is expected in future. The customisation of its food-chain and dose module for the Czech Republic [5] has been started on the basis of co-operation of the Czech institutes SÚRO and ÚTIA with outstanding foreign partners. At the same time a harmonisation of the important topic with EU policy will be achieved.

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