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The latest development and the new extended capabilities of the GENII-LIN soil transfer model

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Abstract

Some years ago, we started developing an enhanced soil transport model, where short life nuclide contributions were accurately accounted, Teodori (2017). The aim was to extend the code capabilities to handle incidental release of contaminant to soil, by evaluating exposure since the very beginning of the contamination event, before the radioactive decay chain equilibrium is reached. In this years those new capabilities have been widely extended: the leaching model has been reworked in a more physically based manner, by using a more sophisticated formulation for the transfer rate; the soil compartment number has been increased, by introducing an intermediate layer; bioturbation by animals now also affects downward transfer of materials, by modifying the leaching constant and by void collapse; plant transfer contributes to contaminant redistribution through all soil depth by plant recycle.

Keywords: Radiation Protection; Health Physics; Soil Contamination; Numerical Simulation; Safety; Environmental impact

1. Introduction

GENII-LIN is an open source multipurpose health physics code, that has been developed at the University of Bologna to provide a reliable tool to be used for purposes such as siting facilities, environmental impact statements, and safety analysis reports. GENII-LIN is a descendant of the GENII code, a thoroughly peer-reviewed, DOE (2003), DOE (2004), and well documented, Napier et al. (1988a), Napier et al. (1988b), open source software system, which was developed at the Pacific Northwest National Laboratory (PNL) and reached maturity in the early 90s with the release 1.485. GENII-LIN has capabilities for calculating radiation dose and risk to individuals or populations from radionuclides released to the environment and from pre-existing environmental contamination. The code can handle a wide range of exposure pathways that comprehend: external exposure from finite or infinite atmospheric plumes; inhalation; external exposure from contaminated soil, sediments, and water; external exposure from special geometries; and internal exposures from consumption of terrestrial foods, aquatic foods, drinking water, animal products, and inadvertent soil intake. The radionuclide environmental concentrations are

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calculated over time up to the end of the exposure period by numerical models of appropriate transport phenomena through air, deep and surface water, deep and surface soil and biotic transport. A wide description of the software structure, the code progresses over time and the code conceptual design have been given in previous works. The code has been ported from DOS to Linux, and enhanced by adding a new modern graphical user interface built on the Qt3 libraries (Sumini et al. (2005)). The internal and external dose rate factor generators have been deeply revised to incorporate into the existing environmental pathway analysis models the more recent internal dosimetry models recommended by the ICRP (1991) and the radiological risk estimating procedures of EPA (2002) (Sumini and Teodori (2005); Teodori and Sumini (2008)). The graphical user interface has been redesigned by implementing the more recent Qt4 and Qt5 libraries, the input and output management deeply reviewed and the air transport model widely improved (Teodori and Sumini (2014)). The external dose rate factor generator has been rewritten to access the data libraries of radionuclide decay information and gamma and beta yields from ICRP (2008), to access the surface dose to organ dose conversion factors from information in ICRP (2010), and to calculate organ dose and total body effective dose following the raccomendations of ICRP (2007) (Teodori (2017)). In this paper the attention is focused on the latest significant improvements of the soil contamination model.

2. The soil contamination model

Depending on land use and occupation, the GENII-LIN code simultaneously manages up to three soil distinct main areas: residential soils, non-agricultural soils, and agricultural soils. The non-agricultural soils are used only in near-field scenarios in order to define parameters for arid and humid climate biotic transport. Immediately after the beginning of human use of the soils, the soil reverts to either residential, when the person lives there, or agricultural, when crops are grown there. Each food pathway has its own associated zone of soil, with specific transfer properties, reason why a large number of soil zones can be active in a single simulation. A single soil zone may be composed of up to 4 compartments (Figure 1). The always present surface soil is modeled as a 15 cm thick layer and is the soil portion that can exchange pollutant with the atmosphere by air deposition, irrigation, and particulate resuspension. For most far-field and many near field scenarios, this is the only portion of soil that is used. In those scenarios, where subsurface contamination is present, radionuclides may be contained in waste forms or simply distributed in the deeper layers. Radionuclides, that are simply distributed in the available subsurface soil, may be transferred to the surface soils by root uptake by plants, by physical transport by native animals, or by human activities which lead to redistribution of contaminants from deeper to surface layers (Figure 2). When the contaminants are packaged in a form, they may be released to the deep soil and made available to biotic transfer. The release process is described by the waste package decomposition model. If the deep soil overburden is greater then $0.15\ m$, one optional intermediate layer is added, located between the surface and deep soils. Any soil layer may also loose radionuclides through harvest removal, radiological decay, and leaching to deeper soil strata. The soil zones corresponding to each food type, animal type, and residential exposure are treated separately.

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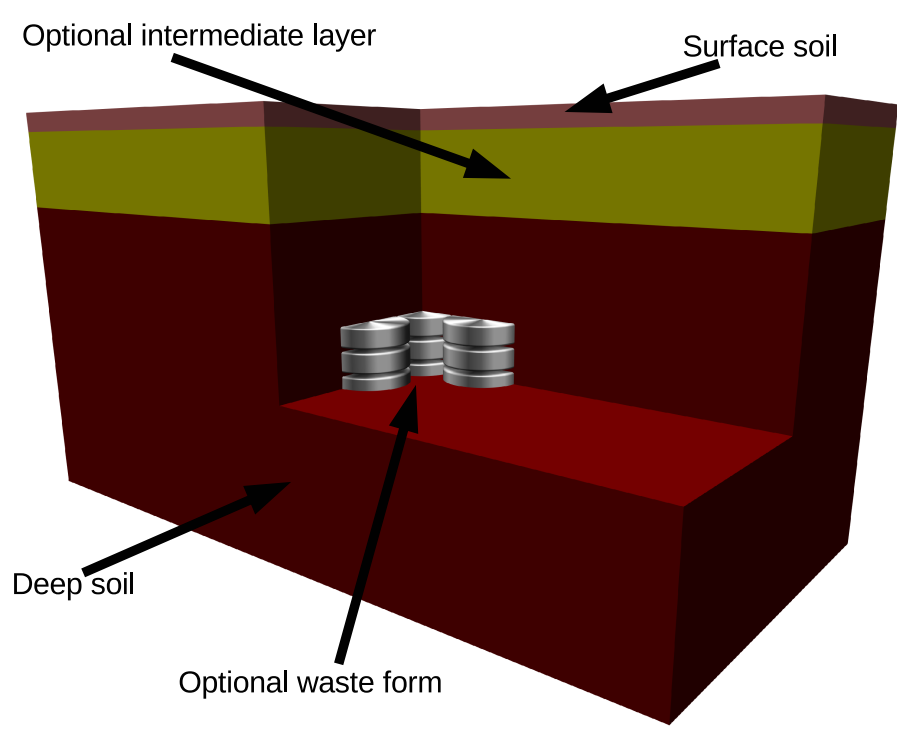
Figure 1: The soil compartment model in GENII-LIN
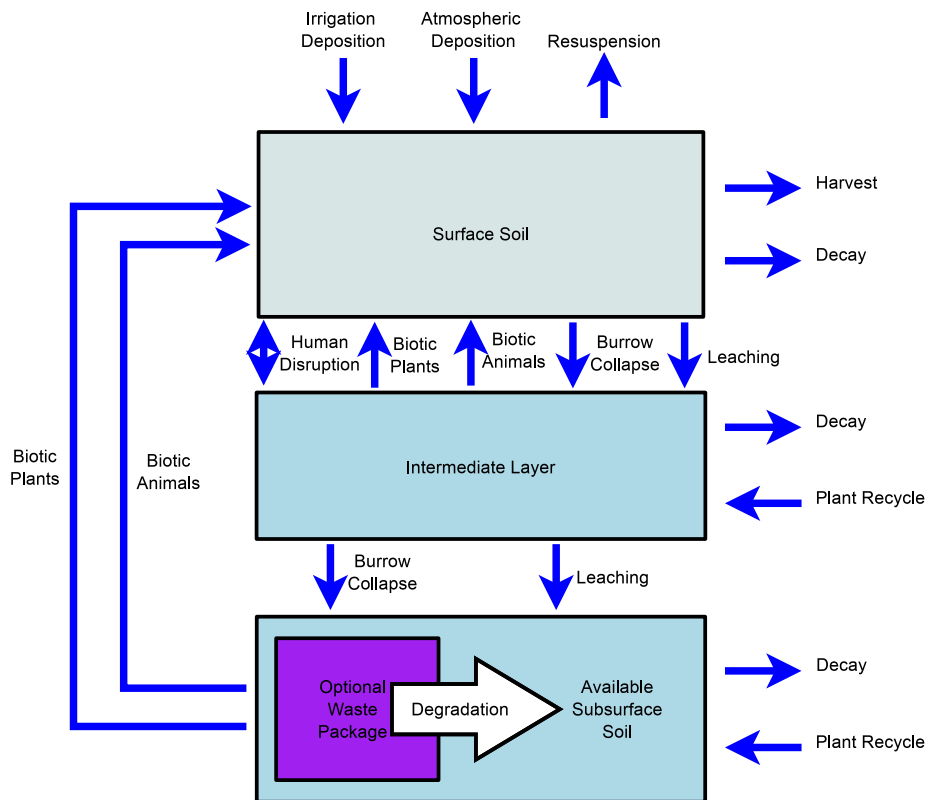
Figure 2: The soil transfer model in GENII-LIN

3. Leaching

Leaching from upper soil layers into deeper soils is modeled by means of a soil removal rate constant, $\lambda_{i,i+1}^r \; \left(y^{-1}\right)$, which is a non-radiological decay constant accounting long-term percolation of deposited radionuclide r out of the layer i to the layer $i+1$. As proposed in Muller-Lemans et al. (1996), we have

$$\lambda_{i,i+1}^r = \frac{Q_{i,i+1}^w + K_{d,i}^r Q_{i,i+1}^s}{V_{w,i} + K_{d,i}^r m_{s,i}} \tag{1}$$

In equation 1

- $Q_{i,i+1}^w \; \left(\frac{m^3}{y}\right)$ and $Q_{i,i+1}^s \; \left(\frac{kg}{y}\right)$ are the water and solid matter flux from layer i to layer $i+1$;

- $K_{d,i}^r \; \left(\frac{m^3}{kg}\right)$ is the distribution coefficient of radionuclide r in layer i;

- $V_{w,i} \; \left(m^3\right)$ is the volume of water in layer i; and

- $m_{s,i} \; (kg)$ is tha mass of solid material in layer i.

The denominator in equation 1 can be expressed in terms of the total volume $V_{t,i}$ of the layer:

$$\lambda_{i,i+1}^r = \frac{Q_{i,i+1}^w + K_{d,i}^r Q_{i,i+1}^s}{V_{t,i}\left(\theta_i + K_{d,i}^r \rho_i\right)} \tag{2}$$

where θ_i is the is the soil volumetric water content and $\rho_i \; \left(\frac{kg}{m^3}\right)$ is the soil bulk density. Now, under the Hypothesis of one dimensional flow, after dividing both numerator and denominator by the layer cross sectional area, we obtain

$$\lambda_{i,i+1}^r = \frac{q_{i,i+1}^w + K_{d,i}^r q_{i,i+1}^s}{z_i\left(\theta_i + K_{d,i}^r \rho_i\right)} \tag{3}$$

where

- $q_{i,i+1}^w \; \left(\frac{m}{y}\right)$ is the rate of water volume flow per unit area from the layer i to the layer $i+i$;

- $q_{i,i+1}^s \; \left(\frac{kg}{m^2 y}\right)$ is the rate of solid matter flow per unit area from the layer i to the layer $i+i$; and

- $z_i \; (m)$ is the layer thickness.

In the earlier releases of the code, leaching affected only the surface layer and leached material was moved out of the layer and lost by the system. Here we wanted to rework the model framework in a more physically based manner, by using a more sophisticated formulation for the transfer rate. The soil removal rate constant defined by equation 3 replace the one suggested by Baes and Sharp (1981), which was used in the previous soil model.

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4. Biotic transport and harvest removal

The code estimates the transport of radioactivity through soil layers by plants and animals by means of embedded transfer models. These models are built on those ones primarily developed by McKenzie et al. (1986) to estimate potential radiation dose to individuals from biotic transport of activity at low-level radioactive waste burial sites. Biotic pathways involve translocation by plant root systems and by burrowing insects and small mammals. Plant examples comprise grass, shrubs, and trees; insect and mammal examples comprise pocket mice, badgers, moles, harvester ants, termites and earth worms. Flora and fauna activity results in the transport of soil components upwards against the force of gravity and against the downwards flow of water. The burrowing activity, which results in excavation of soil, all of which is deposited on the surface soil layer, is accounted by the simplified model expressed by eq. 4:

$$Q_{s_{ir}} = \sum_{j=1}^{a} C_{s_{ir}} \frac{M_{ji}}{\rho}. \tag{4}$$

In it:

- $Q_{s_{ir}}$ is the quantity of radionuclide r yearly moved to the surface from soil stratum i $\left(\frac{Bq}{m^2 \times y}\right)$;

- a is the number of animal species considered;

- $C_{s_{ir}}$ is the concentration of radionuclide r in the soil stratum i $\left(Bq/m^3\right)$;

- M_{jn} is the mass of soil yearly moved from the soil stratum i to the surface by animal j $\left(\frac{kg}{m^2 \times y}\right)$;

- ρ is the soil density $\left(\frac{kg}{m^3}\right)$.

Soil fauna activity also results in physical and biochemical conversion of soil and water, physical and biochemical conversion of soil components, and easier transport processes through voids and macropores, reason why the soil removal rate constant (3) is affected by animal activity by means of $q_{i,i+1}^{w}$ and $q_{i,i+1}^{s}$ parameters.

Collapse of burrows results in transport of earth from upper soil layers to deeper ones. Under steady-state conditions, assuming that the number of burrows per unit area is constant over time, voids compaction and other processes must cause an equally large soil material flux in the opposite directions.

To simulate this phenomenon, voids in the soil strata created by animal burrowing activity are removed at the end of each year to simulate cave-in of burrows. Radioactivity in each soil stratum is adjusted as the voids are removed according to the following expression:

$$Q_{r,i} = C_{r,i-1} V_{i-1} \tag{5}$$

where

- $Q_{r,i}$ $\left(\frac{Bq}{m^2 y}\right)$ is the rate of activity flow per unit area from layer $i-1$ to layer i;

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- $C_{r,i-1}$ $\left(\frac{Bq}{m^3}\right)$ is the concentration of nuclide r in layer $i-1$; and

- V_{i-1} $\left(\frac{m^3}{m^2}\right)$ is the volume of soil per unit area yearly moved from soil layer $i-1$ to soil layer i.

The burrow collapse effectively mixes all radioactivity that has accumulated on the soil surface into the uppermost soil stratum. Equation 5 is assumed to be a reasonable approximation of biotic processes.

The transfer of activity by plants from the layer i to the surface is estimated as follows:

$$Q_{P_{ir}} = \sum_{l=1}^{p} C_{s_{ir}} B_{v_i} R_{li} \frac{B_l}{K} \tag{6}$$

where

- $Q_{P_{ir}}$ is the quantity of radionuclide r yearly moved from soil stratum i to the surface $\left(\frac{Bq}{m^2 \times y}\right)$;

- p is the number of plant species considered;

- B_{v_r} is the soil-to-plant transfer factor $\frac{\left(\frac{Bq}{g}\right)_{plant}}{\left(\frac{Bq}{g}\right)_{soil}}$;

- B_l is the yearly total biomass production of plant l $\left(\frac{kg}{m^2 \times y}\right)$;

- R_{li} is the fraction of roots of plant l in soil stratum i.

A portion, or all of the annual biomass production is assumed to be recycled. When plant material is recycled, the contaminant burden returns to the soil. By denoting with w_l the fraction of plant l biomass returned to surface soil, the rate of activity per unit area transfered to the surface soil stratum is:

$$Q_{P_{ir}}^{S} = \sum_{l=1}^{p} C_{s_{ir}} B_{v_i} R_{li} \frac{B_l}{K} w_l \quad \left(\frac{Bq}{y \times m^2}\right) \tag{7}$$

At the end of each yearly time step, a second portion of the biomass production is assumed to be recycled, because some plants reach end of life. Radionuclides returned to the soil strata through end of life recycling are redistributed in proportion to the plant biomass in each layer. All above-ground contributions from end of life plant recycling are added to the soil surface. Radionuclides are transferred by end of life recycling from the plants to the soil stratum i, according to the following expression:

$$R_{ri} = \sum_{l=1}^{p} C_{rl} b_{li} f_l \tag{8}$$

where

- R_{ri} $\left(\frac{Bq}{m^2 y}\right)$ is the activity of radionuclide r returned to soil layer i as a result of the recycling of plant biomass;

- C_{rl} $\left(\frac{Bq}{kg}\right)$ is the concentration of nuclide r in plant l;

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- b_{li} $\left(\frac{kg}{m^2}\right)$ is the biomass of plant l in soil layer i; and

- f_l $\left(y^{-1}\right)$ is the fraction of biomass of plant l, which is recycled yearly.

Harvest removal is a discrete process and occurs at the end of each calculation year. A quantity of each radionuclide, expressed by calculated vegetation concentration from root uptake, multiplied by the harvested yield (an input parameter) is subtracted from the soil layers. The amount of subtracted material is normalized by the root penetration factor.

5. Deposition rates

For scenarios where air deposition is considered, the deposition rate for unit area is given by

$$R_{ar} = C_{ar}v_{dr} \tag{9}$$

In it:

- R_{ar} $\left(\frac{Bq}{m^2 y}\right)$ is the activity of nuclide r deposited for unit time and unit area on surface soil;

- C_{ar} $\left(\frac{Bq}{m^3}\right)$ is the concentration of nuclide r in air; and

- v_{dr} $\left(\frac{m}{y}\right)$ is the deposition velocity of nuclide r.

For scenarios where water pathways are considered, the deposition rate from irrigation for unit area is given by:

$$R_{wr} = C_{aw}I \tag{10}$$

In it:

- R_{wr} $\left(\frac{Bq}{m^2 y}\right)$ is the activity of nuclide r deposited for unit time and unit area on surface soil layer;

- C_{aw} $\left(\frac{Bq}{m^3}\right)$ is the concentration of nuclide r in water; and

- I $\left(\frac{m^3}{m^2 y}\right)$ is the irrigation rate for unit area.

6. Manual Redistribution

Due to human activities on a site, material may be transfered from the deeper soil or contained waste compartments to the surface soil. This process is modeled simply by introducing a manual redistribution factor, which relates the resultant surface soil concentration, in Bq/m^3, to the initial subsurface concentration, in Bq/m^3.

7. Waste form decomposition

To account for the release of nuclides from waste containers and waste forms in deep soil, a simple waste availability model is implemented, based on the relationship proposed by McKenzie et al. (1982, 1983). In this relationship, the quantity of waste released to soil is defined by the expression:

$$Q_W^r = -\lambda_W W_0^r \tag{11}$$

In equation 11:

- $Q_W^r \left(\frac{Bq}{m^2 y}\right)$ is the activity yearly released for unit area to deep soil layer;

- $W_0^r \left(\frac{Bq}{m^2 y}\right)$ is the activity per unit area contained in the waste form;

- $\lambda_W = \frac{ln2}{T_{W,\frac{1}{2}}} \left(y^{-1}\right)$; and

- $T_{W,\frac{1}{2}} \ (y)$ is the package half life.

This simple model is based on the hypothesis that the waste form thickness fills the deep soil layer thickness.

8. The equations of the model

After collecting equations 3, 4, 6, 7, 9 and 10, and after denoting by λ^r the radiological decay constant of radionuclide r, the transfer of activity A, per unit area for the first layer, is given by

$$\frac{d}{dt}A_1^r = -\left(\lambda^r + \lambda_{1,2}^r\right)A_1^r + \sum_{i=2}^{3} Q_{s_{ir}} + \sum_{i=2}^{3} Q_{P_{ir}}^S + R_{ar} + R_{wr} \tag{12}$$

For the intermediate layer the tranfer of activity is:

$$\frac{d}{dt}A_2^r = -\left(\lambda^r + \lambda_{2,3}^r\right)A_2^r + \lambda_{1,2}^r A_1^r - Q_{s_{2r}} - Q_{P_{2r}} \tag{13}$$

Finally, for deep soil, the transfer of activity is

$$\frac{d}{dt}A_3^r = -\left(\lambda^r + \lambda_{3\rightarrow}^r\right)A_3^r + \lambda_{2,3}^r A_2^r - Q_{s_{3r}} - Q_{P_{3r}} + \lambda_W W_0^r e^{-(\lambda_W + \lambda^r)t} \tag{14}$$

In order to describe the evolution of isotopic changes in the soil layers, this set of linear equations need to be coupled with the Bateman equations describing the time evolution of nuclide concentrations undergoing serial or linear decay chain. This is achieved through the GENII-LIN generalized decay chain processor, which provides the activity of any member of a decay chain as a function of time from any initializing condition. Recently enhanced variants of the processor provide the total activities of chain members for conditions of continual input of nuclides to the system and non radiological removal to a sink. The resulting system is integrated sequentially over one-year time intervals up to cover the period of interest, from the beginning of contamination to the end of the exposure period. At the end of each step, the previously described discrete processes of end of life plant recycle, burrows collapse and harvest removal occur and are accounted.

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Table 1: Soil contamination at the beginning of the exposure.

Initial contamination from Th^{232} and Th^{228}			
Nuclide	Activity $\frac{Bq}{m^3}$		
	Surface Soil	Intermediate layer	Deep Soil
Th^{232}	0	0	1×10^6

Table 2: Dose by pathway from one year exposure to ground.

Effective Dose Equivalent by pathway (Sv)			
		GENII-LIN	ReSRAD
Internal	Inhalation	5.5×10^{-05}	–
	Soil Ingestion	2.4×10^{-11}	–
External	Surface Soil	7.2×10^{-09}	–
	Intermediate layer	3.8×10^{-15}	–
	Deep Soil	4.0×10^{-08}	4.15×10^{-08}

9. Test case

Here we want to display the new soil model capabilities. For this purpose we analyze a hypothetical residential scenario, where a receptor is one year exposed to 50 cm thick contaminated soil layer buried 50 cm below the ground. At the beginning of the exposure, the deep soil layer is assumed uniformly contaminated with Th^{232}. Being the overburden greater than 15 cm, the code adds an intermediate layer 35 cm thick:

$$Th^{232} \xrightarrow[1.405 \times 10^{10} y]{\alpha} Ra^{228} \xrightarrow[5.75y]{\beta-} Ac^{228} \xrightarrow[6.25h]{\beta-}$$

$$Th^{228} \xrightarrow[1.91y]{\alpha} Ra^{224} \xrightarrow[3.63d]{\alpha} Pb^{212} \xrightarrow[10.64h]{\beta-} Bi^{212} \begin{cases} \xrightarrow[61m]{.3593\ \beta-} Po^{212} \xrightarrow[3 \times 10^{-7} s]{\alpha} Pb^{208} \\ \xrightarrow[61m]{.6407\ \alpha} Tl^{208} \xrightarrow[3.1m]{\beta-} Pb^{208} \end{cases}$$

(15)
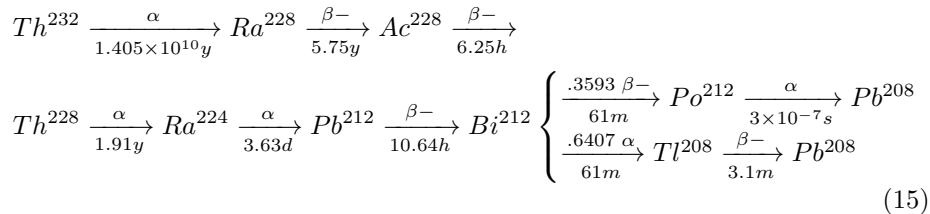
The input to the code is shown in table 1. No daughters need to be added, when they are not present at the beginning of the contamination scenario. The code itself adds them, after reading the Master Nuclide Library. Table 2 reports the dose by pathway. The code extimates radioactivity biotic transport from deep soil layer to surface soil, from the beginning of the contamination up to the end of the exposure period, reason why the receptor receives dose from direct exposure to ground surface contamination, suspended activity inhalation, and inadvertent soil ingestion. In the third column we reported the dose assessed by running the ResRAD-onsite code, Kamboj (2018). ResRAD does not consider nuclide transfer to upper layers, however the computed deep soil contribution to the external dose is very close to the deep soil contribution extimated by running GENII-LIN.

The computed dose by nuclide is reported in table 3. The embedded generalized decay chain processor calculates the activity of any member of the decay

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Table 3: Dose by radionuclide from one year exposure to ground. Dose from Bi^{212} comprehends doses from Po^{212} and Tl^{208}

	Effective Dose Equivalent (Sv)				
Nuclide	Inhalation	Ingestion	External	Internal	Annual
Th^{232}	5.40×10^{-05}	1.90×10^{-11}	1.70×10^{-11}	5.40×10^{-05}	5.40×10^{-05}
Ra^{228}	2.50×10^{-07}	4.50×10^{-12}	1.40×10^{-17}	2.50×10^{-07}	2.50×10^{-07}
Ac^{228}	1.10×10^{-09}	2.60×10^{-15}	4.00×10^{-08}	1.10×10^{-09}	4.10×10^{-08}
Th^{228}	3.50×10^{-07}	5.80×10^{-14}	3.40×10^{-13}	3.50×10^{-07}	3.50×10^{-07}
Ra^{224}	3.30×10^{-08}	5.00×10^{-14}	2.60×10^{-12}	3.30×10^{-08}	3.30×10^{-08}
Pb^{212}	1.90×10^{-09}	4.60×10^{-15}	3.30×10^{-11}	1.90×10^{-09}	1.90×10^{-09}
Bi^{212}	2.60×10^{-10}	2.00×10^{-16}	6.80×10^{-09}	2.60×10^{-10}	7.10×10^{-09}
Total	5.46×10^{-05}	2.36×10^{-11}	4.69×10^{-08}	5.46×10^{-05}	5.47×10^{-05}

chain (15) as a function of time, in this vein each nuclide contribution to dose is accurately accounted.

In Teodori (2017) and Teodori (2019), we compared dose from direct exposure to soil contamination calculated by using GENII-LIN code with dose calculations made by running widely used international Monte Carlo codes: MCNP and PHITS. In other words, we compared GENII-LIN exposure model with site specific models. We found good agreement for different nuclides and different contamination scenarios. Here we wanted to compare GENII-LIN with a similar multipurpose health physics code. Calculations show that neglecting continuos upward transfer from plants uptake and bioturbation may lead to understimate significantly the dose to receptor.

10. Conclusions

In this years we have revised and enhanced the GENII-LIN soil transfer model, whose capabilities have been extended to cover a wider range of exposure and contamination scenarios. The most significant improvements, we have introduced, are here summarized:

- short life nuclide contributions are now correctly accounted;

- the soil compartment number has been increased, by introducing an intermediate layer;

- the leaching model has been reworked in a more physically based manner, by using a more sophisticated formulation for the transfer rate; leaching is no longer limited to soil surface, and leached material is no longer lost, but transfered to deeper soils;

- bioturbation by animals also affects downward transfer of materials, by modifying the leaching constant and by void collapse;

- plant transfer contributes to contaminant redistribution through all soil depth by plant recycle.

Though still under an intensive test phase to check stability and reliability, the new soil transfer module is simple to use, low resource consuming, perfectly

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working and well interfaced with the other modules of the GENII-LIN computational framework, whose capabilities are remarkably extended. We decided to develop a transfer model, which falls between the two main categories of equilibrium models and dynamic ones, Owen Hoffman et al. (1988). Equilibrium parameters have been used to set up a compartment and multilayer dynamic model, where transfer among compartment and layers is simulated according to first order kinetics equations. By choosing approriate parameters, the model may be applied and adapted to a large variety of sites, where root uptake, plant recycle, bioturbation and leaching are actively involved in the transfer of radionuclides, permitting the estimation of human exposures and doses in case of nuclear and radiological accidents, and for routine calculations.

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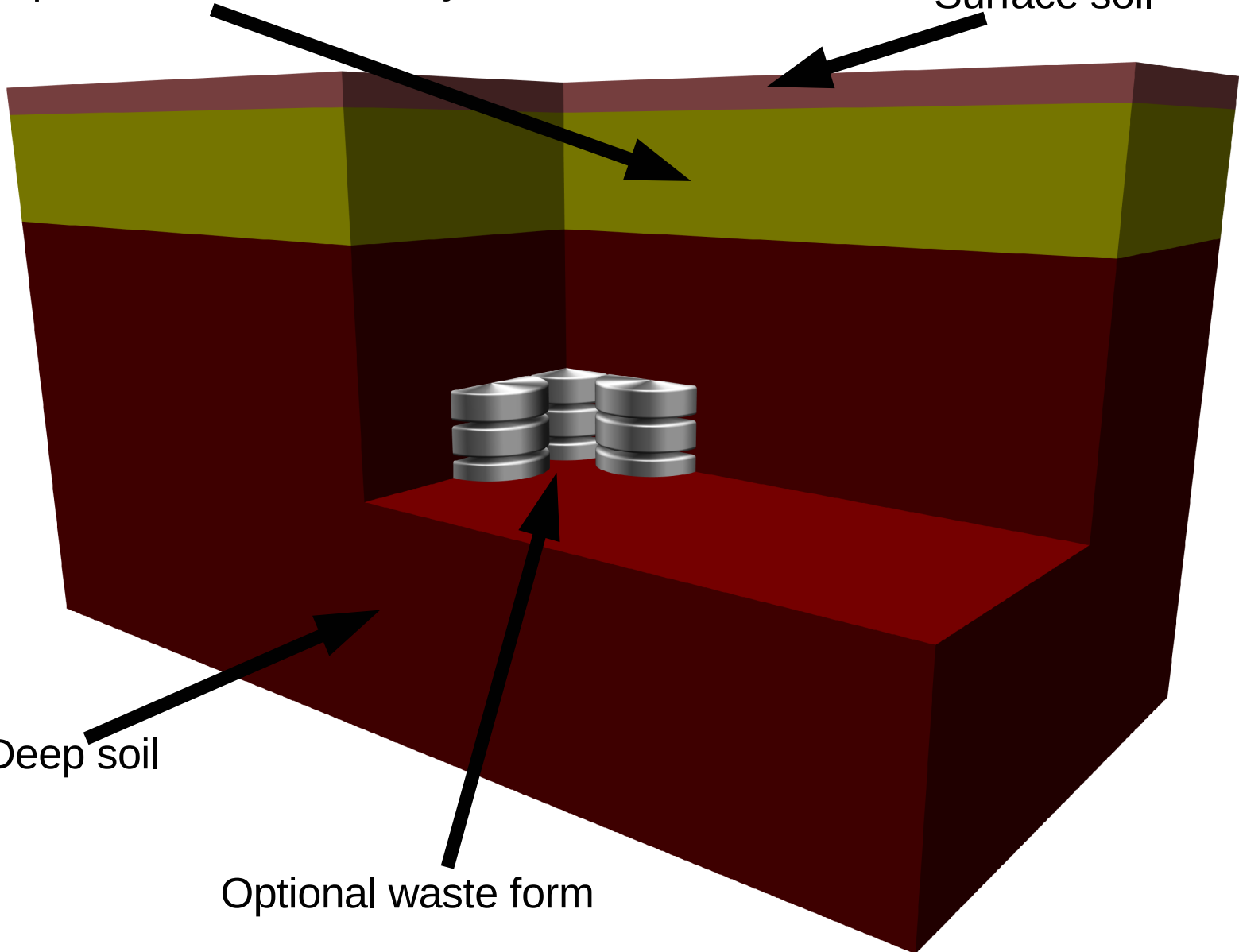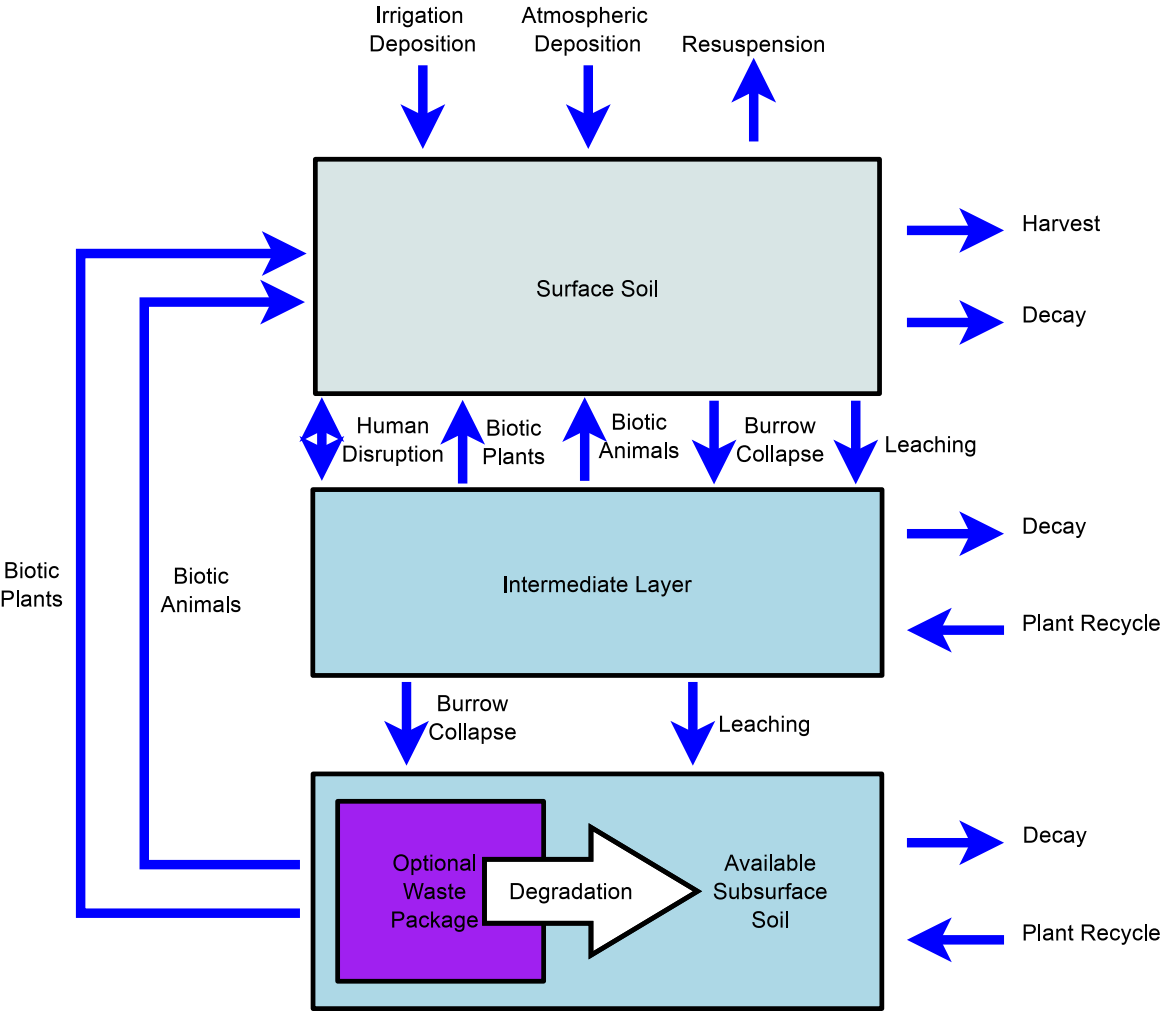
Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Francesco Teodori: Conceptualization, Methodology, Software, Data curation,, Visualization, Software, Validation, Writing, Reviewing and Editing,