



Concept for Calculating Dose Rates from Activated Groundwater at Accelerator Sites

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Abstract

Licensing of particle accelerators requires the proof that the groundwater outside of the site will not be significantly contaminated by activation products formed below accelerator and target. In order to reduce the effort for this proof, a site independent simplified but conservative method is under development. The conventional approach for calculation of activation of soil and groundwater is shortly described on example of a site close to Forschungszentrum Juelich, Germany. Additionally an updated overview of a data library for partition coefficients for relevant nuclides transported in the aquifer at the site is presented. The approximate model for transport of nuclides with ground water including exemplary results on nuclide concentrations outside of the site boundary and of resulting effective doses is described. Further applications and developments are finally outlined.

I Introduction and motivation

Contamination of groundwater as a result of direct neutron radiation is a problem which arises at sites of powerful particle accelerators, as they are required for spallation sources. Such accelerators with spallation sources are in Europe in the design stage (EURISOL EUROpean Isotope Separation OnLine, ESS European Spallation Source). For licensing of such a facility it is necessary to prove that the public, the environment and the workers are sufficiently protected against the dangers of radioactivity. In Europe the Council Directive 96/29/EURATOM defines limits for protection from ionizing radiation: For members of the public the effective dose resulting from drinking water at the boundary of the supervised area has to remain < 1 mSv/y, even if all drinking water is taken from the highest contaminated location (Figure I). Accordingly, the requirements on the bottom shielding of a proton accelerator depend on the site specific hydrogeological conditions and respective activity transport. In order to guarantee on the one hand side a sufficient shielding, but on the other hand to avoid costly over shielding, a reliable method for calculation of the potential radioactive burden outside the fence of the site is required.

A complete calculation of all nuclide activation and transport processes is a complex and time consuming procedure, as shown in Ref 16. Consequently a simplified but fast to apply model, has to be developed which simulates the generation and accumulation of radionuclides in soil and groundwater and the transport of activity with groundwater in an adequately conservative manner. The sorption behavior of radionuclides on soil components and radioactive decay have to be taken into account as processes, which diminish the nuclide concentration at the fence. The model has to be adapted to specific site conditions, i.e. diverse soil compositions, ground water levels and flow rates.

In the following chapters the present state of the model is described.

II Activation of soil and groundwater

Because of very different soil and groundwater compositions activation calculations cannot be easily generalized but have to be performed in a separate step before of the transport calculations. However, the nuclide concentrations generated are proportional to the energy loss of the accelerator and to the neutron leakage of the target and depend in a well known manner on the shielding thickness. Accordingly, scaling of results to other beam losses or other shielding thicknesses is easily possible. A detailed description of activation calculations performed for soil and groundwater under a proton accelerator and a spallation target for Juelich site conditions is presented in Ref. 2. The approach used is a conventional one. In these calculations, a beam loss in the 5 MW proton accelerator of 1 W/m was taken into account. In the start-up phase of an accelerator and in certain regions substantially higher beam losses may occur, so the results presented should be taken as exemplaric only. The composition of soil and groundwater is given in Ref. 2. With these input data the activation as given in table I was calculated. Saturation concentrations are presented in Table III.

The light radionuclides are products from reactions with thermal neutrons and light elements as well as from spallation reactions of heavy elements with high energetic neutrons. The heavier produced isotopes come from the common occurring natural isotopes of the same element, like iron in soil.

III Partition Coefficients (K_d)

The understanding of the transport of radionuclides in the subsurface is of major importance for the setup of groundwater/transport models. For this purpose the

partition coefficient K_D estimates the migration potential of a specific contaminant in aqueous solution in contact to solid phases.

A Definition and Limitations

The partition coefficient (K_D) is defined as the concentration of solute in the adsorbed phase (mass of solute per unit mass of soil) divided by the concentration of the solute in the solution phase (see Eq. (1)). Therefore the units for K_D are usually given as ml/g.

$$(1)$$

where C_0 is tracer concentration in the solution before adding the soil, C is the solute concentration in the liquid phase of a soil water suspension, V is the volume of water, and M is the mass of soil. Thus, K_D is a factor related to the partitioning of a solute (or contaminant) between the solid and the aqueous phases.

Due to a great variety of parameters influencing the migration of contaminants, e.g. kind of clay minerals, suspended solids, interaction between contaminants, bacteria, physico-chemical properties of groundwater, the use of K_D values in transport modeling is always a simplifying estimation.

For a sophisticated modelling it is preferable to determine K_D values for the specific soil and the contaminants of concern by laboratory methods (e.g. batch methods) or, even better, in the field. Unfortunately these activities are very time and resources consuming. Often the values of the above mentioned parameters are greatly varying in the area of concern. In these cases results from laboratory and field methods are

characterizing only spots in a three-dimensional aquifer and not representing the total volume.

During a screening process it is often more effective to use K_d values from the literature and estimate critically their applicability for the specific task.

B Partition Coefficients (K_d) of Selected Elements

For the derivation of the K_d values it is necessary to analyze the chemical constituents of the groundwater which can be activated if passing an area where the groundwater is exposed to thermal neutrons and protons from a (planned) accelerator.

In addition activated elements can enter an aquifer after “production” in the shielding (concrete, soil etc.) or surrounding soil/rock of an accelerator if these elements can be mobilized e.g. by infiltration of surface water or by direct contact of shielding and groundwater. Additionally, water itself should be taken into account before starting activation calculations (Ref. 2).

Both, the composition of the groundwater and the activation products from the shield, are factors for selection of the radionuclides of concern (Table II).

Refs. 3 and 4 divide the different soil textures in 4 groups (see Table II); this is a way to handle the great variability of soil textures, mineralogical composition and chemical constituents by reducing characterizing parameters to particle size distribution and organic content:

sand soils: \geq 70 % sand sized particles

clay soils: \geq 35 % clay-sized

loam soils: \leq 80 % silt-sized particle or even distribution of sand-, clay- and silt-sized particles

organic soil: > 30 % organic matter

Most of the K_d values of Refs. 3 and 4 are compiled from literature. Where no appropriate data exists in the literature the so-called soil-to-plant concentration ratio to predict K_d based on the bioavailability of a nuclide (Ref. 5) is used. This approach is also used to calculate K_d values for some radionuclides given in Table II.

IV Activity transport in groundwater

Modeling radionuclide transport in the vadose zone and in the groundwater is computationally demanding and existing analytical solutions are not valid for complex geometries. A simplified model for a test-site near Jülich, Germany, was established for a conservative estimation of the resultant activity-concentrations and committed effective doses of different radionuclides at the boundary of the supervised area (BSA, Figure I).

A Methods

The water flow in a porous three-dimensional medium, e.g. soil or aquifer, is given by the Richards equation:

$$(2)$$

where θ is the volumetric water content [L^3L^{-3}], K the hydraulic conductivity tensor [LT^{-1}], ψ the matrix pressure head [L], z the elevation head [L] and $S(x)$ a source/sink term [T^{-1}].

For the process of solute transport through a porous three-dimensional media the advection dispersion equation (ADE) was used:

(3)

where C is the concentration [ML^{-3}], s is the sorbed concentration [MM^{-1}], ρ the bulk density [ML^{-3}], D the dispersion tensor [L^2T^{-1}], q the water flux vector [LT^{-1}] and Q^* a source/sink term [$\text{ML}^{-3}\text{T}^{-1}$].

The calculation of the radionuclide transport was done with the coupled TRACE-PARTRACE code (Refs. 6, 7). The TRACE code describes the saturated and unsaturated water flow by solving numerically the Richards equation for a finite element mesh. PARTRACE is a particle tracking code calculating the transport of solutes including sorption and decay.

For proving compliance with regulatory limits for radiation at the boundary of the supervised area (Figure 1), it is essential to model the 'worst case' in order to have a conservative estimation. Therefore the accelerator was assumed to be parallel to the groundwater flow to ensure the fastest and most concentrated transport of the radionuclides to the BSA. The bottom of the accelerator was assumed to be at the top of the groundwater zone to avoid a reduction of the radionuclides before reaching the groundwater zone induced by sorption processes in the unsaturated soil zone. Instead of an activation zone described by Ref. 8, all radionuclides were injected directly at the groundwater level, where the accelerator was located to keep the model simple. For the transport-model, the accelerator represented a continuous plane contamination source with the dimensions of 6.35 m x 300 m. The BSA was assumed to be 250 m downstream the accelerator.

The modeling was done for a site 4 km SE of Jülich, Germany which extends about 100 m NE and 700 m NW. Based on drilling profiles (Ref. 9) a thickness of 16 m for

the aquifer was assumed. Assuming homogeneous material properties with a bulk density of 2.0 kg m^{-3} , the hydraulic gradient was set to 0.0025 m m^{-1} and the saturated hydraulic conductivity was set to 404.12 m d^{-1} (Refs. 9, 10 and 11). Thus, the Darcy-velocity has a value of 1.01 m d^{-1} which gives a mean pore velocity of 5.49 m d^{-1} , assuming a value of $0.184 \text{ m}^3 \text{ m}^{-3}$ for the saturated water content. According to the Krauthausen field experiment (Ref. 11) the dispersion length α_L for the model site was set to 3.64 m. The model was discretized by a grid of 1,120,000 cells partitioned in 51 x-nodes (spacing: 2.0 m), 351 y-nodes (2.0 m) and 65 z-nodes (0.25 m). For the front and the back of the model Dirichlet boundary conditions were imposed to represent the hydraulic gradient. No-flow boundaries were assigned to the left- and right site, the bottom and the top.

Based on the activation results, the migration of ^{14}C , ^{41}Ca , ^{45}Ca , ^{36}Cl , ^{55}Co , ^{57}Co , ^{60}Co , ^3H , ^{54}Mn , ^{24}Na , ^{32}P , ^{35}S , ^{32}Si and ^{50}V was calculated because they cover a wide range of half-lives $T_{1/2}$ and partition coefficients K_d .

B Results

The modelling showed that ^3H , ^{36}Cl and ^{14}C are the most significant radionuclides concerning their resulting activity concentration at the boundary of the supervised area (Table IV). Their small partition coefficients in combination with a relatively large half-life cause a fast migration of the radionuclides in the groundwater underlying only low decay. With a partition coefficient of 0 and its small decay ($T_{1/2} = 12.23 \text{ y}$) ^3H has the highest resultant activity-concentration at the BSA with a maximum value

of $1.14\text{E-}01 \text{ Bq l}^{-1}$. The maximum activity concentrations of ^{36}Cl and ^{14}C are smaller by a factor of 2.2 (^{36}Cl : $5.20\text{E-}02 \text{ Bq}$) and factor 10.8 (^{14}C : $1.06\text{E-}02 \text{ Bq l}^{-1}$). Within 23 days ^3H arrives at the BSA and after 161 days steady-state conditions occur and the maximum activity-concentration at the BSA is reached. Due to the retardation effect, ^{14}C needs more time to arrive at the BSA. However, due to the high half-life, the resultant activity-concentration at the BSA is not much lower than the ones for ^3H and ^{36}Cl .

^{55}Co , ^{57}Co , ^{54}Mn , ^{24}Na , ^{32}P , and ^{35}S , have a relative short half-life in combination with high K_d values such that particles are completely decayed before reaching the BSA. In contrast to this, the half-life of ^{50}V is $1.40\text{E+}17$ years and thus much higher, but the partition coefficient is 327 ml g^{-1} , being so high that the particle-plume theoretically reaches the boundary of the supervised not before 142 years after injection. Due to the small induced saturation activity concentration ($1.23\text{E-}08 \text{ Bq l}^{-1}$) the resultant maximum activity concentration at the BSA is the smallest of the investigated radionuclides.

Calculations were done on the basis of a simplified model with homogeneous properties and a continuous contamination source only for the accelerator, but not the target. The homogeneity of the model influences also the development of the particle-plume in the aquifer (Figure II; 1a, b, c). With increasing distance from the source of contamination the particle-plume disperses, rather horizontally than vertically. The centre of the highest activity-concentrations for all radionuclides is found in the horizontal middle of the model ($x = 50 \text{ m}$) and in the same depth the particles were injected. Independent from type of radionuclide, there is a strong

decrease of the activity-concentration with increasing depth (Figure II; b, c).

To ensure, that the workers, the public and the environment are protected against radiation it is necessary to determine the effective dose from the calculated activity-concentration. The committed effective dose for workers and the public (for different ages) for drinking water ingestion can be determined by the following equation 4 (Ref. 12):

(4)

where $h_{E,50}$ is the committed effective dose (Sv y^{-1}), A the activity concentration (Bq l^{-1}), B the annual consumption rate of drinking water (l y^{-1}) and DCF the dose conversion factor for the radionuclide ingested ($\text{Sv}\cdot\text{Bq}^{-1}$). The annual radiation exposition averages to about 0.004 mSv per year for an adult and about 0.02 mSv per year for a child in Western Europe. In comparison with the ingestion of nutrition, the uptake of radionuclides by drinking water is often of minor importance (Ref. 13). The DCF s for the different radionuclides were taken from the Safety Series 115/1994 published by the International Atomic Energy Agency (IAEG) (Ref. 14).

Assuming annual drinking-water ingestion (child) of 240 l y^{-1} in combination with the activity-concentrations, which were calculated at the boundary of the supervised area, the committed effective doses are shown in Table IV. Among these, a change in significance-ranking compared to the resultant activity concentrations at the BSA can be observed. ^{36}Cl is the most significant radionuclide to be protected against. However for this particular case, the orders of magnitude of the committed effective doses for all radionuclides are negligible small and these doses will not endanger the public. The reason for the negligible small resultant activity-concentrations and

committed effective doses is that the induced saturation activity concentrations of the different radionuclides were also that small, neglecting the target area. The results given here should be taken as exemplary only and not as generally representative. This is because they depend strongly on site and accelerator specific parameters such as beam loss, shielding efficiency, ground water flow rate/distance to the fence, assumptions on local losses etc.

V Conclusion and outlook

A simplified model on groundwater activation and transport is proposed, which is conventional concerning activation, but reduces the complex problem of activity transport with ground water to the following main parameters:

- groundwater flow velocity/distance to the site boundary
- halve life of the nuclides
- K_d values of nuclides in the respective soil
- dispersion behaviour of the soil

On basis of this simplified model a fast estimation of groundwater activations is possible, which allows an assessment of shielding adequateness.

The prove of a sufficient conservatives of the approach remains to be performed. A report on the whole approach, covering all aspects more detailed, will be published soon (Ref. 15).

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VI References

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Table I: Activities [Bq/m³] in Juelich soil and groundwater calculated for a continuous irradiation of 9 month (accelerator beam loss 1 W/m, bottom shielding thickness below accelerator: 0.8 m; neutron leakage conditions in the target area as in Ref. 2.

Nuclide	Beam Line/Accelerator		Target	
	Soil	Ground Water	Soil	Ground Water
H-3	3.95E-03 ± 1.32E-03	3.02E-03 ± 1.01E-03	1.16E+05 ± 3.86E+04	1.01E+05 ± 3.36E+04
Be-7	6.12E-04 ± 2.04E-04	4.97E-04 ± 1.66E-04	9.32E+03 ± 3.11E+03	7.55E+03 ± 2.52E+03
C-14	2.72E-04 ± 9.05E-05	2.75E-04 ± 9.15E-05	7.91E+03 ± 2.64E+03	8.30E+03 ± 2.77E+03
Na-22	5.34E-04 ± 1.78E-04	8.76E-07 ± 2.92E-07	8.13E+03 ± 2.71E+03	1.50E+02 ± 5.00E+01
Na-24	1.94E-04 ± 6.45E-05	7.91E-06 ± 2.64E-06	6.71E+03 ± 2.24E+03	1.04E+03 ± 3.47E+02
Si-32	2.73E-06 ± 9.10E-07	1.22E-08 ± 4.06E-09	4.16E+01 ± 1.39E+01	1.85E-01 ± 6.15E-02
P-32	1.59E-05 ± 5.30E-06	6.29E-06 ± 2.10E-06	8.96E+02 ± 2.99E+02	1.27E+03 ± 4.25E+02
S-35	9.65E-06 ± 3.22E-06	1.13E-05 ± 3.78E-06	3.30E+02 ± 1.10E+02	1.47E+03 ± 4.90E+02
Cl-36	1.52E-04 ± 5.05E-05	1.20E-03 ± 4.00E-04	2.55E+04 ± 8.50E+03	1.56E+05 ± 5.20E+04
Ca-41	7.46E-05 ± 2.49E-05	1.47E-05 ± 4.92E-06	1.13E+03 ± 3.78E+02	1.37E+03 ± 4.57E+02
Ca-45	7.13E-04 ± 2.38E-04	9.99E-07 ± 3.33E-07	1.42E+05 ± 4.74E+04	9.29E+01 ± 3.10E+01
Sc-46	4.05E-04 ± 1.35E-04	9.75E-12 ± 3.25E-12	4.23E+04 ± 1.41E+04	1.48E-04 ± 4.95E-05
V-50	4.79E-06 ± 1.60E-06	1.23E-11 ± 4.12E-12	7.89E+01 ± 2.63E+01	2.22E-04 ± 7.40E-05
Mn-54	1.26E-04 ± 4.22E-05	2.33E-10 ± 7.75E-11	2.01E+04 ± 6.70E+03	3.02E-02 ± 1.01E-02
Fe-55	2.33E-03 ± 7.75E-04	4.40E-09 ± 1.47E-09	3.74E+05 ± 1.25E+05	6.84E-01 ± 2.28E-01
Fe-59	2.79E-05 ± 9.30E-06	5.58E-11 ± 1.86E-11	3.65E+03 ± 1.22E+03	6.26E-03 ± 2.09E-03
Co-55	4.52E-08 ± 1.51E-08	8.19E-11 ± 2.73E-11	6.87E-01 ± 2.29E-01	1.25E-03 ± 4.15E-04
Co-57	3.32E-07 ± 1.11E-07	3.12E-10 ± 1.04E-10	5.07E+01 ± 1.69E+01	9.99E-03 ± 3.33E-03
Co-60	3.83E-08 ± 1.28E-08	2.72E-10 ± 9.05E-11	5.07E+00 ± 1.69E+00	4.32E-02 ± 1.44E-02
Zn-65	9.44E-06 ± 3.15E-06	1.43E-09 ± 4.77E-10	1.04E+03 ± 3.45E+02	1.58E-01 ± 5.25E-02

Table II: Selected K_d values [ml/g]

element	soil type			
	sand	silt	clay	organic
Be	250	800	1.300	3.000
Ca °	5	30	50	90
C #	5	20	1	70
Cl	0.8	0.25	4.4	11
Co °	60	1.300	550	1.000
Cu	30	105	159	369
Eu	236	825	1.255	2.908

Au	30	105	159	369
H	0	0	0	0
Mn °	50	750	180	150
P °	5	25	35	90
Si °	35	110	180	400
Na	76	265	403	934
S	14	49	75	174
Va	327	1.141	1.737	4.024

Ref. 3, ° Ref. 4, *italic*: calculated K_d values using the "soil-to-plant concentration ration" (CR) from Ref. 5

Table III: Modeled radionuclides

nuclide	half-life (y)	partition coefficient (ml g ⁻¹)	induced saturation activity concentration (Bq l ⁻¹)
¹⁴ C	5.73E+03	7	2.75E-01
⁴¹ Ca	1.03E+05	5	1.47E-02
⁴⁵ Ca	4.47E-01	5	9.99E-04
³⁶ Cl	3.00E+05	0.8	1.35E+00
⁵⁵ Co	2.00E-03	30	8.19E-08
⁵⁷ Co	7.45E-01	30	3.12E-07
⁶⁰ Co	5.27E+00	30	2.72E-07
³ H	1.23E+01	0	3.02E+00
⁵⁴ Mn	8.55E-01	50	2.33E-07
²⁴ Na	1.71E-03	76	7.91E-03
³² P	3.91E-02	5	6.29E-03
³⁵ S	2.40E-01	14	1.13E-02
³² Si	1.72E+02	35	1.22E-05
⁵⁰ V	1.40E+17	327	1.23E-08

Table IV: Modeling results

nuclide	concentration-ratio	activity-concentration	travel time until BSA is reached	travel time until steady-state conditions occur	Committed effective dose	
	C_{max}/C_{input} (-)	$C_{a,max}$ (Bq l ⁻¹)	$t_{t,BSA}$ (d)	$t_{t,steady}$ (d)	Child (<1 y) (mSv y ⁻¹)	Adult* (mSv y ⁻¹)
¹⁴ C	3.85E-02	1.06E-02	859	9785	3.55E-09	1.47E-09
⁴¹ Ca	3.84E-02	5.66E-04	618	7085	1.63E-10	2.58E-11
⁴⁵ Ca	6.16E-06	6.15E-09	663	3055	1.62E-14	1.05E-15
³⁶ Cl	3.86E-02	5.20E-02	114	1316	1.22E-07	1.16E-08
⁵⁵ Co	----- concentration below numerical threshold -----					
⁵⁷ Co	----- concentration below numerical threshold -----					

^{60}Co	2.70E-04	7.32E-11	4184	28407	9.49E-16	5.97E-17
^3H	3.80E-02	1.14E-01	23	161	1.76E-09	4.94E-10
^{54}Mn	----- concentration below numerical threshold -----					
^{24}Na	----- concentration below numerical threshold -----					
^{32}P	----- concentration below numerical threshold -----					
^{35}S	----- concentration below numerical threshold -----					
^{32}Si	3.09E-02	3.77E-07	4626	51383	6.60E-13	5.06E-14
^{50}V	3.86E-02	4.76E-10	38480	435653	**	3.90E-16

BSA: boundary of supervised area (250 m downstream the accelerator), C_{max} : maximum resultant concentration at the BSA, C_{input} : initialized concentration of the contamination source, $C_{\text{a,max}}$: maximum resultant activity-concentration at the BSA, * equivalent to workers, ** no value found in the literature

Figure I: Limits for protection from ionizing radiation (Ref. 1) according to European Council Directive 96/29/EURATOM

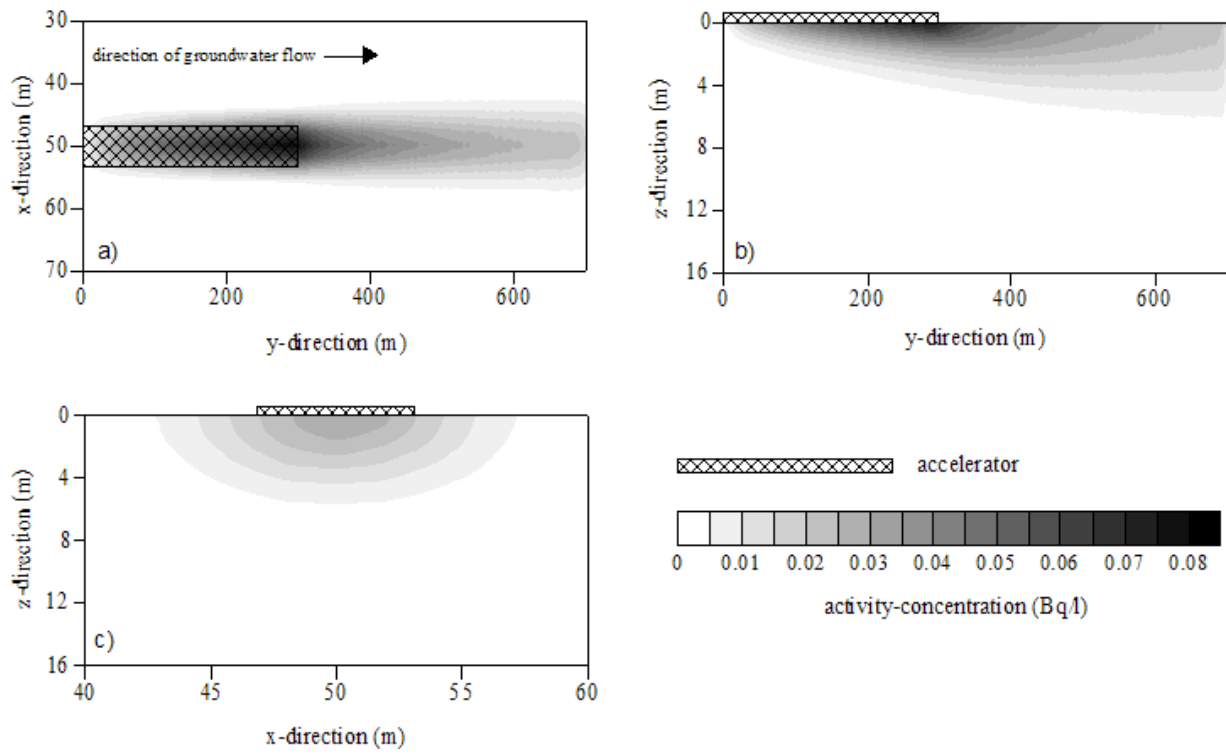


Figure II: Resultant activity-concentration plume of ^3H for a homogeneous model with a continuous contamination after steady-state conditions occur. a) x-y-view ($z = 0$ m), b) y-z-view ($x = 50$ m), c) x-z-view ($y=550$ m (boundary of the supervised area)).