Evaluation of long term radiological impact on population close to remediated uranium mill tailings storages

David Kerouanton[a*], Laure Delgove[a], Philippe Crochon[b], Patrick Devin[c], Eric Fillion[c]

[a] AREVA SGN, Nuclear Safety Department, 1 rue des Hérons, 78182 Saint Quentin en Yvelines Cedex, France

[b] AREVA NC, BU-Mines, 1 place de la Coupole, 92084 Paris la Défense, France

[c] AREVA D3S / IG, 33 rue Lafayette, 75442 PARIS Cedex 09, France

Abstract. A methodology is elaborated in order to evaluate the long term radiological impact of remediated uranium mill tailings storage. Different scenarios are chosen and modelled to cover future evolution of the tailings storages. Radiological impact is evaluated for different population such as adults and children living in the immediate vicinity or directly on the storage, road workers or walkers on the storage.

KEYWORDS: Radiological impact on population; mill tailings storages

1. Introduction

After extraction of uranium, mill tailings are stored directly on site. Such remediated storages represent a potential source of radiological impact to people located in the immediate vicinity. As a consequence, at the end of the exploitation, a radiological protection composed of waste rocks and top soil is designed to cover tailings.

Sites are monitored, radiological measurements being made in their vicinity. These measurements include the radiological background due to natural radioactivity. Therefore, the radiological impact of the site itself has to be determined by elaborating a reference scenario in order to discriminate anthropogenic contribution from background and to care that the dose rates do not overpass the regulated doses.

If this current layout should present stabilization, mining companies need to deal with long term risks such as deterioration of the radiological protection or public intrusion.

Different scenarios are chosen and modelled in order to cover future evolution of the tailings storages:

- loss of radiological protection layers (tailings visible),
- roadwork on the storage,
- children playing on tailings piles extracted ,
- a house built on the storage with and without radiological protection layers.

For each scenario, the following doses are estimated:

- external radiation dose,
- inhalation dose (due to ^{222}Rn and its daughters),
- inhalation of dust (due to the loss of the protection layers),
- ingestion of products coming from or watered with site downstream water.

Radiological impact is evaluated for different population such as adults and children living in the immediate vicinity or directly on the storage, road workers or walkers on the storage.

* Presenting author, E-mail: david.kerouanton@areva.com

2. Exposition scenarios

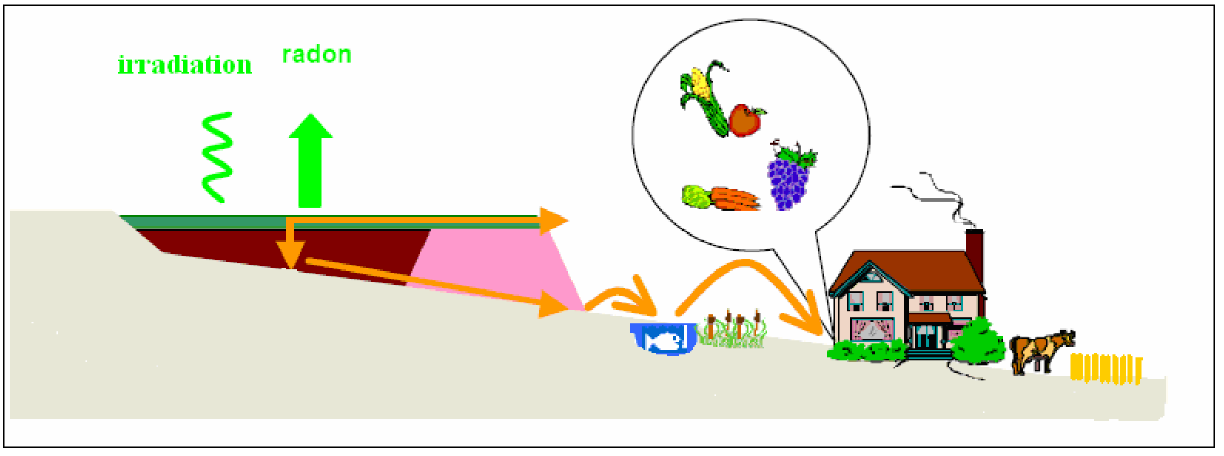
The scenarios must include normal evolution of the site but also the effects induced by potential damages such as erosion or by other potential uses of the site [1].

2.1 Reference scenario

The reference scenario (sketched on figure 1) corresponds to the normal evolution of the storage. The dose induced to the people living around the site, working or walking on the site must be evaluated for the following pathways:

- inhalation due to radon emanation from the storage ,
- irradiation from the site,
- irradiation due to irrigation of gardens with water extracted downstream of the site,
- Ingestion induced by drinking of animals, irrigation of gardens and fishing.

Figure 1: reference scenario



2.2 Altered sites

Scenarios corresponding to altered sites must be considered. They correspond to hypothetic situations linked to random natural events or intrusive human activities:

- loss of the cover (figure 2) : increased radon emanation, inhalation of radioactive dust in suspension in air, increased irradiation;
- loss of side barriers (figure 3) - landslide of a barrier generating a hole in it : increased direct irradiation of the people living around the site;
- building of a house with full basement directly on the cover (figure 4) : increased radon emanation particularly in basement, increased irradiation;
- building of a house with full basement directly this time on the mill tailings: increased radon emanation particularly in basement, increased irradiation, inhalation of radioactive dust in suspension in air;
- building of a road through the site : exposition of workers to radon, inhalation of radioactive dust and irradiation;
- outdoor games of children on excavated piles from road building : exposition to radon, inhalation of radioactive dust and irradiation.

Measurements made on sites show that the trend is a decreasing of liquid releases as the time go. This indicate that considering radioactive decrease and packing down of mill tailings, long term evolution of mill tailings can be expected to be favorable. In these conditions, considering the present source term should be conservative for long term studies.

Figure 2: loss of radiological protection layers constituting the cover

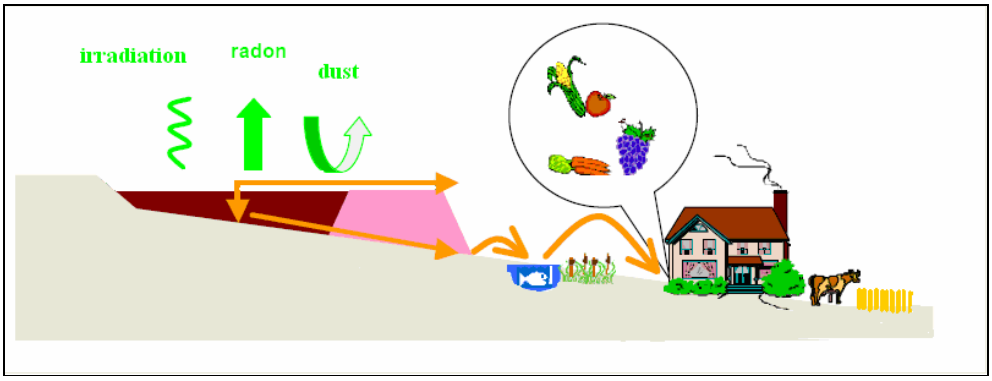


Figure 3: landslide of a site barrier

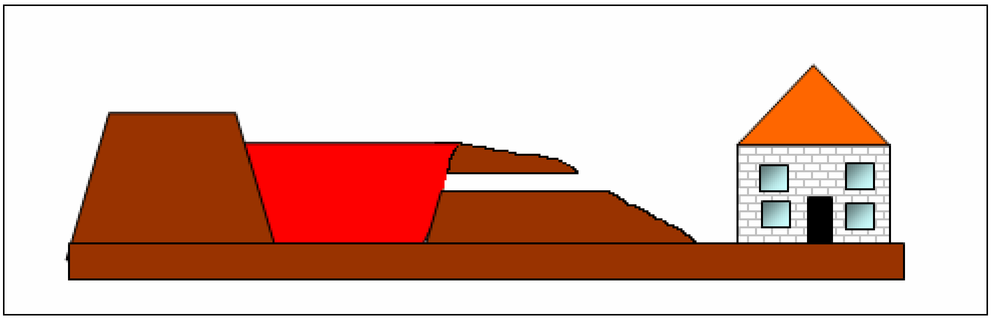
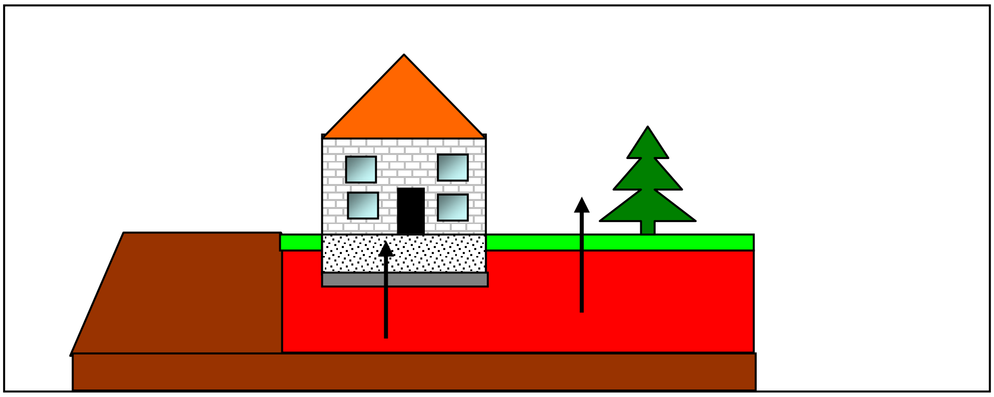


Figure 4: building of a house with full basement directly on the cover

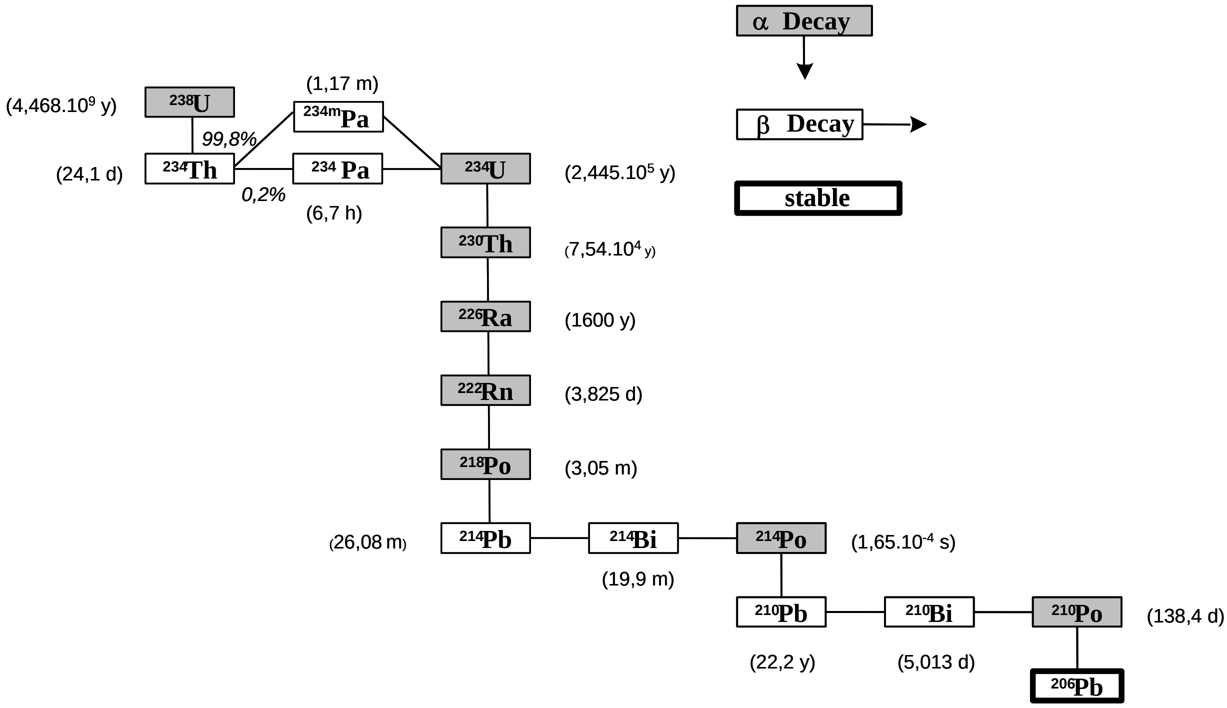


3. Dose calculation method

3.1 Equilibrium of the uranium decay chain

Natural uranium mass repartition composition is 99.27 % of ^{238}U, 0.72 % of ^{235}U and 0.01% of ^{234}U. ^{238}U chain equilibrium (figure 5) is broken after extraction and if it can be supposed that the process have the same efficiency for every isotope of uranium then the residual activity of ^{238}U and ^{234}U may remain the same.

Figure 5: ^{238}U decay chain



Due to its short half-life, ^{234}Th and its short life decay products quickly regain equilibrium. ^{234}Th decay generates ^{234}U but the number of atoms corresponds to a weak activity due to ^{234}U half-life.

Finally, mill tailings equilibrium can be considered between ^{238}U and its decay products until ^{234}U. Below ^{234}U decay chain is still at equilibrium because of ^{230}Th period.

3.2 External dose due to irradiation

Storage itself, deposit induced by irrigation and excavated piles from road building constitute the major sources of irradiation.

3.2.1 External exposure from storage

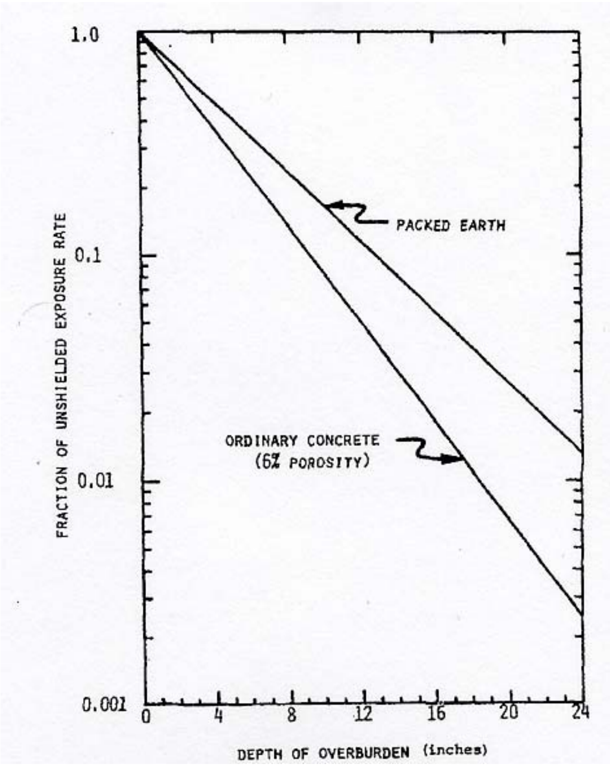
Cover and barriers are designed to attenuate efficiently irradiation due to mill tailings. Figure 6 [2] gives the gamma flux attenuation across variable thickness of earth and concrete.

External dose (Sv) due to irradiation is obtained by using the following equation:

$$D_{ext\_site} = T_{expo} \cdot \frac{\Omega}{2\pi} . e^{-\mu_{air} d} . \sum_i (DC_{ext\_\infty}(i).C_i) \qquad \text{with:}$$

T_{expo}	:	exposition time (s);
Ω	:	solid angle from which site is seen for calculation point (steradian);
μ_{air}	:	air attenuation coefficient (m^{-1});
d	:	distance from source (m) ;
DC_{ext_∞} (i)	:	radionuclide i dose coefficient for exposure to soil contaminated to an infinite depth ($Sv.s^{-1}$) / ($Bq.\ m^{-3}$);
C_i	:	radionuclide i activity concentration in mills tailings ($Bq.\ m^{-3}$)

Figure 6: gamma flux attenuation versus thickness of packed earth or ordinary concrete [2]



3.2.2 External exposure from settlings on soil

The radionuclide i annual external dose (Sv) induced by deposit on soil consecutive to irrigation of vegetable gardens can be calculated by using the following equations:

$$D_{ext\_deposit}(i) = D_{max} \cdot \left(\frac{1 - e^{-\lambda_i .365,25}}{\lambda_i} \right) . DC_{ext\_deposit}(i)$$

with:

D_{max}	:	radionuclide i maximal deposit on the ground ($Bq.m^{-2}$);
λ_i	:	radionuclide i radioactive constant (d^{-1}) ;
$DC_{ext\ deposit}$ (i)	:	radionuclide i dose coefficient for exposure to soil contaminated to an infinite depth ($Sv.s^{-1}$) / ($Bq.\ m^{-3}$).

3.2.3 External exposure from excavated piles from road building

External dose D_{ext} (Sv) due the piles can be evaluated with:

$$D_{ext} = \int DED_{ext}.dt$$

Where DER_{ext} is the gamma Dose Equivalent Rate (Sv/h) measured or calculated with an appropriate code.

3.3 internal dose due to inhalation of radon

^{222}Rn is the decay product of ^{226}Ra. In soils, ^{222}Rn gas migrates to the surface depending on geological and geotechnical characteristics of the soil.

Dose induced from ^{222}Rn inhalation is not due to radon itself because it is a gas but to its solid short life decay products (^{218}Po, ^{214}Pb, ^{214}Bi) which are also alpha emitters.

As a consequence, internal dose from radon must be calculated by using the Potential Alpha Energy (APE) of its decay products:

$$D_{inh} = T_{expo}.(K_{Rn222}.F.APE_{equilibirum}.C_{Rn222})$$

With:

T_{expo}	:	exposition time (h);
K_{Rn222}	:	^{222}Rn Conversion coefficient from [3] (1,1 $Sv.J^{-1}.h^{-1}.m^{3}$)
$APE_{equilibrium}$	:	Alpha Potential Energy concentration due to ^{222}Rn at the equilibrium (5.54 10^{-9} J/m^3 for 1 Bq/m^3 of ^{222}Rn)
C_{Rn222}	:	measured ^{222}Rn concentration (Bq/m^3)
F	:	equilibrium factor

3.3.1 Equilibrium factor

Equilibrium factor is the ratio of the measured APE and the APE when in equilibrium with its decay products. A low F factor corresponds to a just emitted radon with few generated decay products and which impact will be low.

Indoor and outdoor F factors are different because of deposit of solid decay products on the wall of homes.

Outdoor, UNSCEAR [4] recommend 0.7. Indoor, F factor is notably lower and a mean value of 0.4 can be considered [3]. On the site, radon is quickly dispersed by wind and it can be shown that F factor should not overcome 0.1.

3.3.2 Radon flux

^{222}Rn concentration can be determined from radon emitted flux. This last can be calculated with the following equation:

$$\phi_{Rn\,222} = F_{prop}.C_{Ra\,226} \quad \text{with:}$$

Φ_{Rn222}	:	radon flux ($Bq/m^2/s$);
F_{Prop}	:	proportionality factor;
C_{Ra226}	:	^{226}Ra concentration in soil (Bq/g).

In literature, a mean value for F_{Prop} of 0.5 $Bq/m^2/s$ of ^{222}Rn for 1 Bq/g of ^{226}Ra can be also considered [5].

Once emitted in soil, radon flux is attenuated by the cover which can be composed with different compacted materials.
Document [2] indicates that the radon flux is reduced by a factor e with an earth thickness of 1.5 m. This can be calculated by the following equation:

$$\Phi = \Phi_0 \quad e^{-\frac{\text{thickness}}{1.5}}$$

Figure 7 [6] gives the Half Value Layer (HVL) for different types of soil leading to a radon flux divided by a factor 2. According to document [5], a HVL value of 0.37 m relative to compacted soil can be considered.

Figure 7: radon flux attenuation versus earth thickness

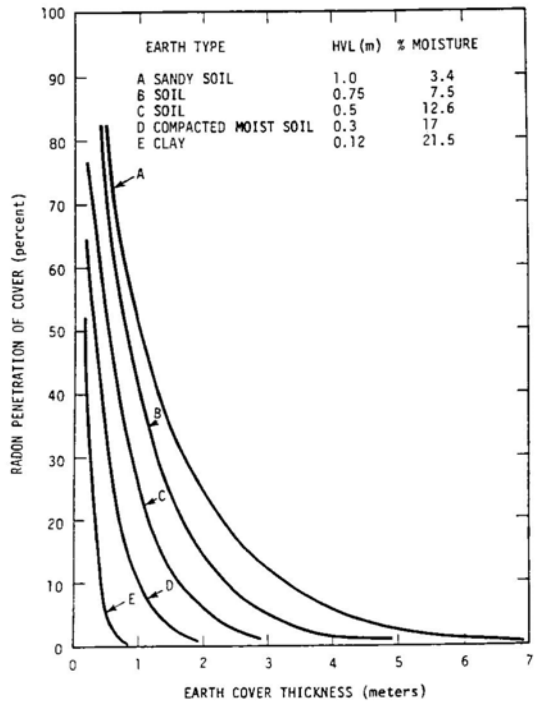
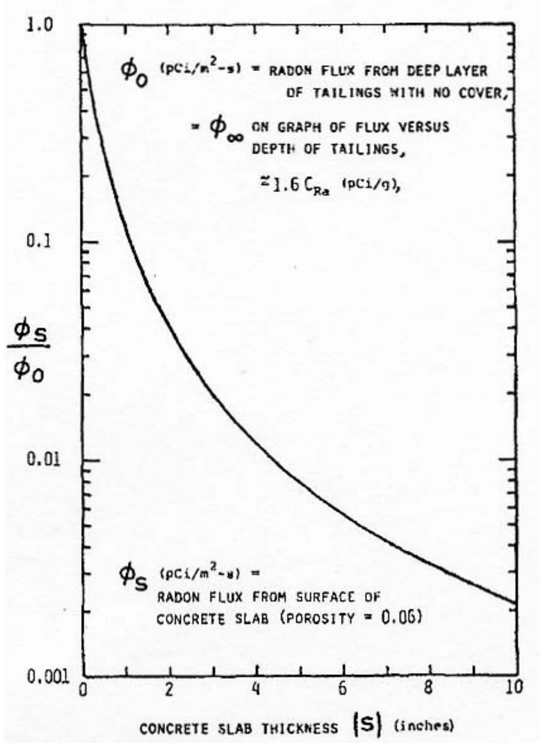


Figure 8 [2] gives the fraction of flux emanating from concrete surface versus concrete slab thickness. A thickness of 10 cm of concrete leads to the emanation of 1.2 % of the original flux.

Figure 8: radon flux fraction emanating from concrete versus concrete slab thickness



3.3.3 Radon concentration

Once the radon flux at the surface is quantified, ^{222}Rn concentration can be determinated at any point on the site or outside of the site by using an atmospheric dispersion code such as COMODORE [7].

The Atmospheric Transfer Coefficient (ATC in s.m^{-3}) characterizes the dispersion. It corresponds to the transfer function between the activity emitted and the activity concentration at the calculation point.

ATC is defined as the ratio of the time integrated concentration and the integral of source function:

$$ATC = \frac{\int \chi.dt}{\int Q.dt} \qquad \text{with :}$$

χ(x,y,z,t) : concentration of the pollutant at point (x,y,z) at time t (Bq.m^{-3}),

Q : total amount of pollutant emitted (Bq),

χ(x,y,z,t) is the solution of the turbulent diffusion equation which can be modelled by simple gaussian model such as PASQUILL or DOURY models [7]. By this way, the dilatation of puffs emitted from the site can be assessed as a function of the distance and the concentration of radon can be determined.

In the case of a house built on the storage, exposition to ^{222}Rn in the basement must be considered. The concentration of ^{222}Rn can be determined from the following equation:

$$\frac{d}{dt} C_{Rn222}(t) = C_{Rn222}(t=0) - (\lambda + T).C_{Rn222}(t)$$

Which solution is:

$$C_{Rn222}(t) = \frac{\phi}{h.(\lambda + T)}.\left(1 - e^{-(\lambda+T).t}\right)$$

With:

T	:	basement ventilation rate (s^{-1});
λ	:	^{222}Rn radionuclide i radioactive constant (s^{-1});
Φ	:	emanation flux of ^{222}Rn (Bq. $m^{-2}.s^{-1}$)
h	:	basement height (m).

3.4 internal dose due to inhalation of dust

Without cover or in case of road building, the wind can lick directly the mill tailings surface leading to airborne radioactive particles and inhalation dose to the people. Dust particles are composed of uranium isotopes and their decay products.

The internal dose (Sv) due to inhalation of dust can be determined by using the following equation:

$$D_{inh\_dust} = \beta_{inh}.T_{expo}.C_{dust\_air}.\left[\sum_i DC_{inh\_i}.A_{m\_i}\right]$$

With :

β_{inh}	:	respiratory rate ($m3.h^{-1}$);
T_{expo}	:	exposition time ($h.year^{-1}$);
C_{dust_air}	:	dust concentration in air (Bq. $m^{-2}.s^{-1}$);
DC_{inh_i}	:	radionuclide i inhalation dose coefficient ($Sv.Bq^{-1}$);
A_{m_i}	:	radionuclide i activity concentration in mill tailings ($Bq.g^{-1}$);

Dust concentration in air can be evaluated with:

$$C_{dust\_air} = \frac{D}{F_{dilution}} \qquad \text{with :}$$

D	:	dust concentration in air ($g.m^{-3}$);
$F_{dilution}$	:	dilution factor;

The dust concentration is extracted from literature (table 1).

Table 1: Dust concentration for the different scenarios

Scenario	Dust concentration in air (µg/m^3)
building site	1000
Storage	500
Indoor	10
Garden	20

3.5 internal dose due to ingestion

People living around or directly on the storage can drink contaminated water pumped downstream of the site. This water can be used to irrigate vegetable garden and animals can also drink it. Eaten fishes are also supposed to be taken in the same downstream water.

Then, calculation is based on the radionuclides concentration in water. From the irrigating rate ($l.m^{-2}.j^{-1}$), annual deposit on soil is determined. Deposited activity enters the superficial layers of the soil and transit through the plants towards the animals.

In case of children playing directly on the site, a small quantity of soil can be considered to be eaten.

Ingestion dose due to plants and animal meat consumption can be deducted by the use of code such as COMODORE [7].

3.5 Sensitivity

Dose assessment is made by using mean or standard values for the different parameters involved in the models detailed above. In order to take into account the range of values on which the parameters can vary, a sensitivity study is carried out.

4. Conclusion

A methodology is elaborated in order to evaluate the radiological impact of remediated uranium mill tailings storages. Different scenarios are chosen and modelled to cover future evolutions of the tailings storages.

Radiological impact is evaluated for different population such as adults and children living in the immediate vicinity or directly on the storage, road workers or walkers on the storage.

Equation and methods are detailed.

REFERENCES

[1] Doctrine en matière de réaménagement des stockages de résidus de traitement de minerai d'uranium. Groupe de travail du Ministère de l'Aménagement du Territoire et de l'Environnement – Direction de la Prévention des Pollutions et des Risques (DPPR) (in French).

[2] SCHIAGER KEITH, J. Analysis of radiation exposures on or near uranium mill tailings piles. Radiation data and reports, vol. 15, n°7 (1974)

[3] INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION, publication 65 Protection against Radon-222 at home and at work (1993)

[4] UNITED NATION SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION. Ionising Radiation : sources and biological effects (1993)

[5] UNITED STATES NUCLEAR REGULATORY COMMISSION. Scientific basis for risk assessment and management of uranium mill tailings. Uranium mill panel study (1986)

[6] GUETAT, P, MONTFORT, M., AMEUR, A. Etude d'impact du site minier de l'Ecarpière. Rapport CEA-DAM (in french);

[7] Notice utilisateur relative au logiciel COMODORE Sud-Est. ALTRAN_EILIS_NAK_DT_07_0028B (in french)