REMEDIATION AND CORRESPONDING RADIOLOGICAL IMPACT OF FRENCH URANIUM MINING AND MILLING SITES (COGEMA)

Dr Jean Louis DAROUSSIN [(1)] Sylvain BERNHARD [(2)] Daniel GRANET [(2)]
(1) COGEMA BU/DOM/QSE , BP4, 78141 VELIZY Cedex, France Tel 33 1 39 26 32 93
(Fax 33 1 39 26 27 51) E-mail jldaroussin@cogema.fr
(2) ALGADE, BP 46, 87250 BESSINES sur Gartempe, France Tel 33 5 55 60 50 02
(Fax 33 5 55 60 50 59) E-mail sylvain.bernhard@algade.com

Abstract

Environmental monitoring is described around French uranium mining and milling sites.

Main observations and evolutions of measurements are discussed highlighting some of the main issues related to uranium mines and mill remediation.

In the environment of uranium mines and mill tailings storages, measurements and doses to members of the public are low (compared to some modelisation results) and remediation improves and perpetuates this limited impact. Site remediation, the last stage of uranium mining, is a contribution to sustainable development.

Starting exploration at the end of the forties, France has produced up to 3500 tonnes of Uranium a year. Total production is more than 70000 t but, as uranium mines and mills are progressively ceasing operation, COGEMA has developed since 1991 an extensive remediation programme.

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Figure 1 Map of France showing location of main industrial sites (milling and/or heap leaching facilities and storages for tailings) – [1960 – 1980 / 1987 (or 1987)] figures are : date for start – end of operation / date of end of remediation (or date of monitoring license)

When operations stop we have to deal with three types of sites to be remediated :
underground and open pit mining sites, milling facilities and the associated mill tailings storages and non contaminated surface facilities such as warehouses and offices.

Main objective for remediation of mill tailings storages is to ensure stability for the long term.

Various studies on mill tailings have demonstrated that natural leaching of radium was limited : less than 1% of the radium content for stored mill tailings compared to 10% for fresh mill tailings. This can be explained by quick (less than 30 years) development of secondary minerals such as smectite, iron oxy hydroxydes and gypsum which trap 95 % of the total radionuclides and associated heavy metals (1, 2, 5). This can be called “self confinement” which improves the chemical stability of the tailings.

Thus remediation consists in two main works. First drainage in order to collect, for water treatment if necessary, the seepage waters resulting from settlement and compaction of the tailings. Secondly, placement of a cover to prevent any dispersion of the tailings and limit exchanges in and out of the wastes (3, 4, 5).

Environmental monitoring is adapted from the network allowing control of the impact during the operating period. At the end of remediation, a license acknowledges the work achieved and sets up the new frame of the environmental monitoring.

After remediation, remaining potential impacts to monitor are linked to the chemical (radiological and non radiological) composition of the ore and corresponding wastes.

According to the chemical composition of the ores and the corresponding chemical process for uranium recovery, the seepage from tailings may contain some metals but the chemical impact is mainly linked to a pH and sulphate control following the acid leaching.

Most of the initial radioactivity remains in the tailings (some 70% in an acid in pulp process) : potential impact is linked to radioactive quality of effluents, after possible water treatment which is compulsory as long as radium is higher than 0.74 $Bq.l^{-1}$ (or 3.7 $Bq.l^{-1}$ according to the environment) and uranium 238 higher than 1.8 $mg.l^{-1}$ (Decree 90.222). The cover prevents formation of any radioactive dust and limits readily the external exposure through gamma rays. Thickness of the cover (usually about a meter minimum) is given by tests plots which confirm a quick reduction of radon flux.

The cover will also reduce infiltration; a proper topography and revegetation limit erosion

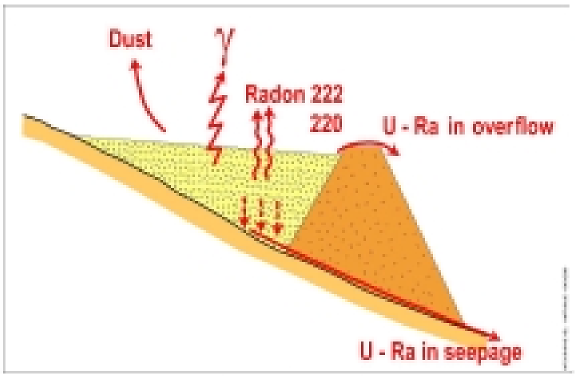


Figure 2 Main potential radiological impact of a mill tailings storage facility before remediation

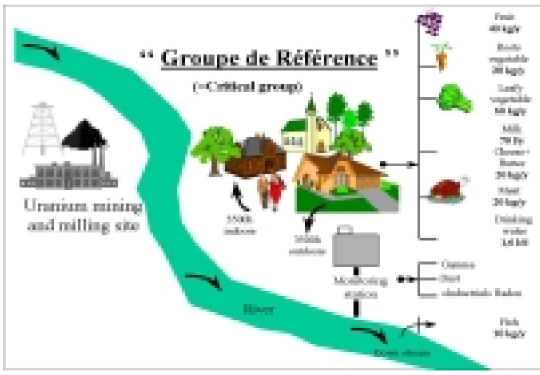


Figure 3 Conceptual design of a monitoring network and evaluation of effective dose.

Monitoring is the direct mean to measure and assess the real impact of a facility. The network includes measuring stations in the environment (near residential areas, up and downstream) and on the site. Type of sampling or direct continuous measurement depends on the element tracked.

Monitoring covers the different pathways discussed previously.

- Air quality measurement are done by site alpha dosimeter developed by ALGADE (6) which allow simultaneous measurement of radon 222 and radon 220 potential alpha energy (PAE) and long lived alpha emitters in dust. The three measures are made on a continuous monthly basis and, for impact assessment purpose, ALGADE has developed calculations, based on radon isotopic signature, to evaluate separately the "industrial radon" coming from the storage site and the natural radon which is not influenced by mining and milling (7, 8).
  Gamma dose rate is recorded by a three months exposure of a TLD.
- Water is analysed for uranium 238, radium 226 and other elements according to site specificity. Water discharged after treatment is continuously controlled for pH and continuously sampled every week. Environment water monitoring is a weekly or monthly sample up and down stream. Evaluation of the impact is based on samples of the domestic water distributed to the " groupe de référence" (or critical group) of the population.
- Sampling of the most characteristics components of the local food chain is undertaken every year and analysed for uranium 238, radium 226 and lead 210.

The monitoring network is particularly comprehensive around industrial sites grouping mines, mills and uranium mill tailings storage facilities.

Results

Systematic control by multielements analysis of discharges showed only a few specific problems (Al, pH or Fe due to acidification).

Following is a presentation, for radionuclides, of different examples of real continuous measurement results, plotted on curves, collected before and after remediation.

As the cover placement is progressing Potential Alpha Energy of Radon 222 and gamma dose rate show a decrease near Bessines mill tailings storage site (fig. 4).

Three of the selected monitoring stations shown on figure 4 are in the vicinity ("La Chataignière" being a residential area) and "Bessines site East" is located on the industrial site of Bessines (140 ha). The site used to group underground and open pit mines, mill and heap leaching facilities as well as the corresponding tailings storage impoundments (a ring dyke limited one and an open pit for a total area of 50 ha and 20 millions tons of mill tailings and heap leaching wastes).

Milling stopped mid 1993 and a first layer of the cover was spread in January 94 in the area of "Bessines site East" (see the gamma dose rate curve). As the cover progresses on the tailings, 12 months average curves for PAE of Rn222 show a small but significant decrease all through 1997.

Note that seasonal variations are large with peak concentrations of the radon daughter concentration during summer period.

On Ecarpière site, progress of the cover is perfectly seen on the gamma dose rate (and Rn222 PAE) curves for the stations located on the site (fig. 5).

As the cover progresses, the decrease of radon flux impacts directly the radon daughter concentration on the tailings storage site ("Ecarpière storage dyke East") and is less significant in the immediate vicinity of the mill site (11 millions tons of mill tailings and heap leaching wastes on a tailings area of 73 ha, total area including mill and mines = 240 ha). After remediation (finished in 1995), the impact of the storage is indistinguishable in the villages neighbouring the site.

Such measured Rn 222 PAE datas are correlated to radon flux. They are better to use as input parameters for impact assessments to the population - see total exposure results following – or for modelisation such as ExternE exercises.

According to the methodology, ExternE studies involve large populations (up to 2000 kms away from the site) and variations in the source term radon flux give a wide range of results (9). Implementation by CEPN (10) shows that measured natural radon flux in the vicinity of the site has, in Europe, a much larger collective impact than radon coming from uncovered tailings (ratio of 250000). After remediation the calculated impact due to radon is nil.

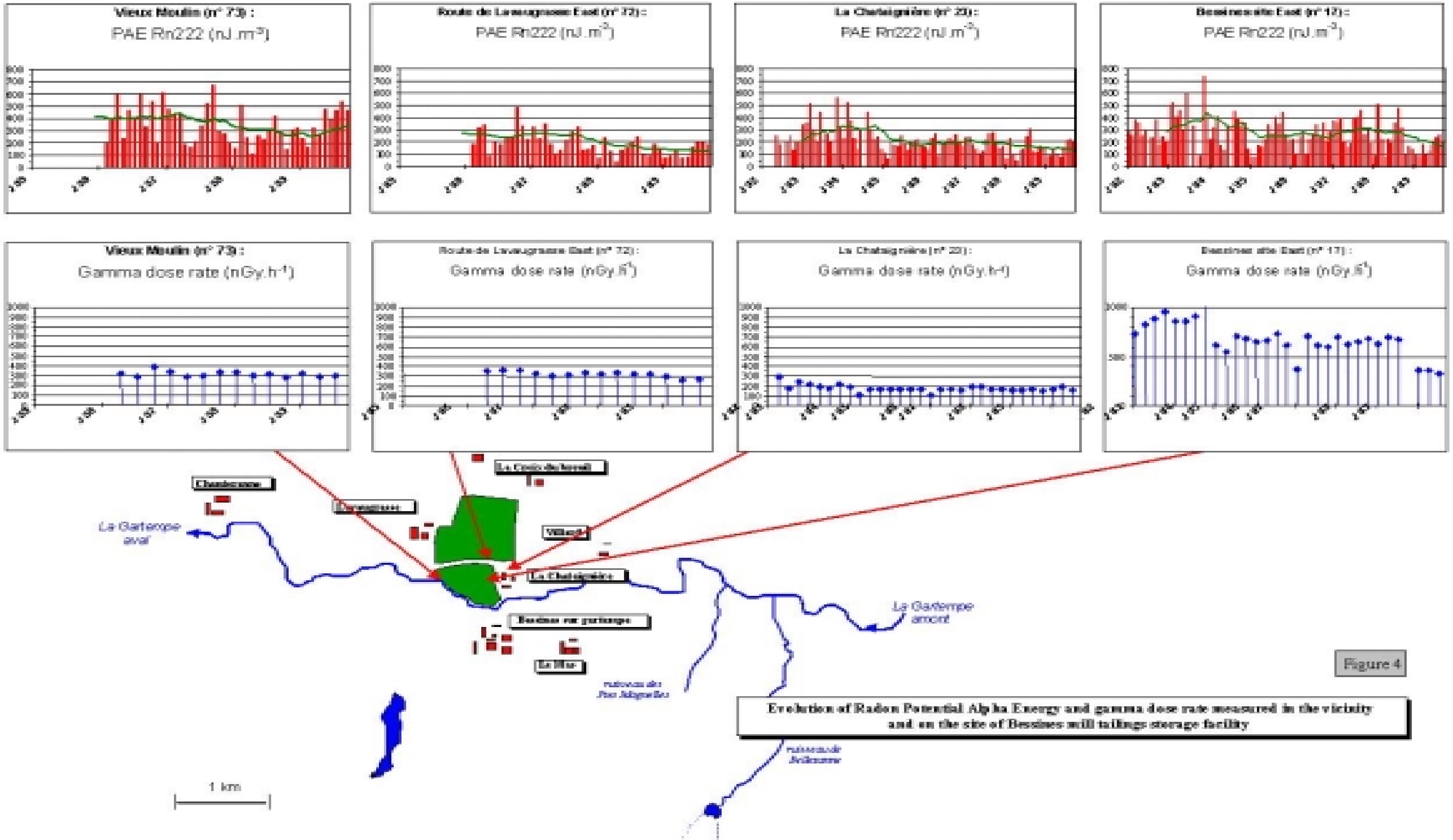


Figure 4

Evolution of Radon Potential Alpha Energy and gamma dose rate measured in the vicinity and on the site of Bessines mill tailings storage facility

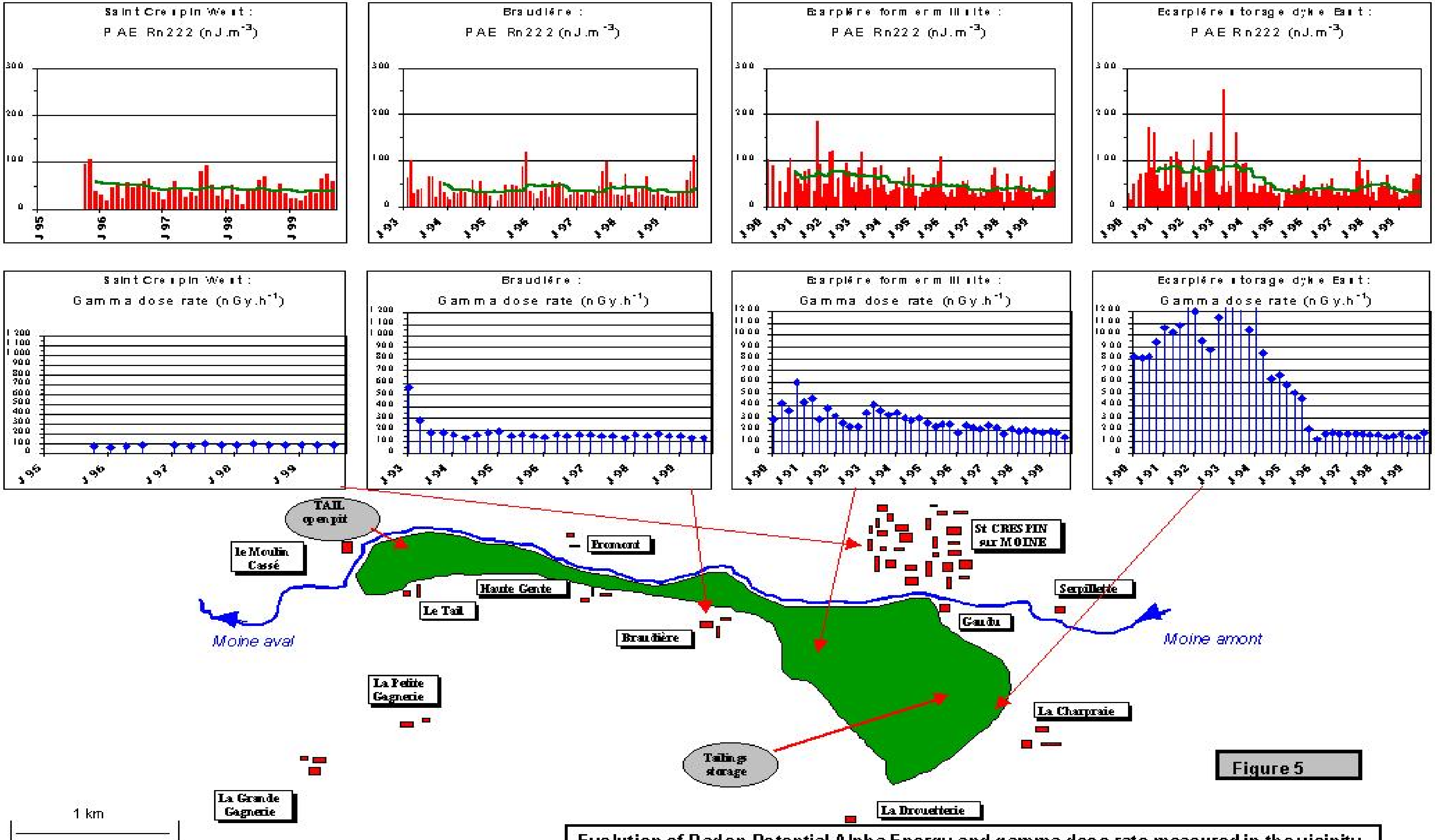


Figure 5

Evolution of Radon Potential Alpha Energy and gamma dose rate measured in the vicinity and on the site of Ecarpière mill tailings storage facility

Gamma dose rate may be influenced by natural geological environment as shown by pluri-annual gamma monitoring around Ecarpière (fig. 5).

St Crespin Ouest and Braudière are located on both side of a major EW fault channelling river Moine and separating gneiss in the north and uraniferous leucogranite in the south : this geological distinction gives nearly a two fold difference in the gamma dose rate of corresponding stations on figure 5.
This is an illustration of the difficulty to evaluate the added gamma dose rate (and consequently the total added exposure) due to industrial sites.

Water seeping out of the mill tailings storage and underground mine water overflow of Ecarpière are collected and still need to be treated. Water release is of a good quality with no impact on the river (fig. 6).

Water management involves a good knowledge of the different types of water : in Ecarpière, seepage water from the mill tailings are pH 6-7, Ra226 0.5 – 1 $Bq.l^{-1}$ and U238 0.1-0.3 $mg.l^{-1}$. Iron rich mine overflow (#100 $mg.l^{-1}$) oxidise quickly, with a pH drop from 5-7 to 3-4 ; Ra is ~1.5 $Bq.l^{-1}$and U ~0.4 $mg.l^{-1}$. Water quality after treatment (lime, barium chloride and flocculent) is good and has not impact on the river Moine.

Evolution of water quality in the flooded open pit of Le Tail (fig. 7) is an example of natural improvement due to return to natural reduced underwater equilibrium.

At the end of mining (early 1991), water was allowed to flood Le Tail open pit and the water table stabilised in 1993. Located in a fractured zone rich in pyrite, initial oxidation contributed to a drop in pH and concentrations in uranium and radium up to 1 $Bq.l^{-1}$ Ra226 and 4 $mg.l^{-1}$ U. After leaching of free sulphur minerals and establishment of chemical equilibrium, pH, U and Ra stabilise to natural local concentrations allowing free release to the environment.

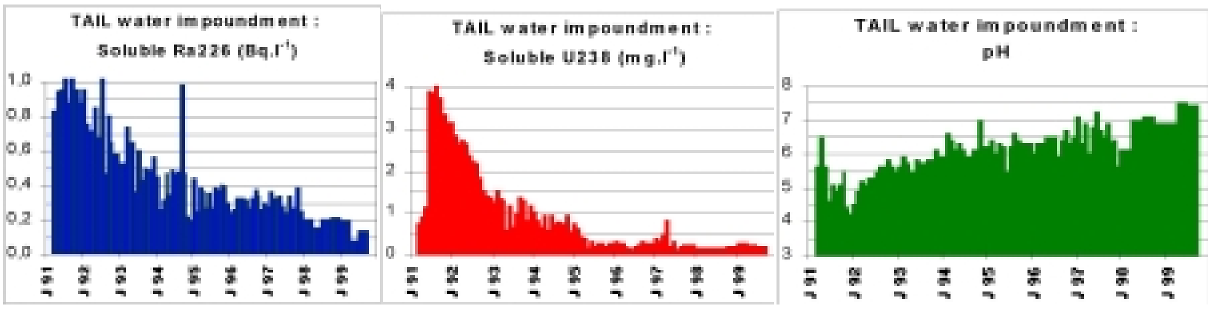
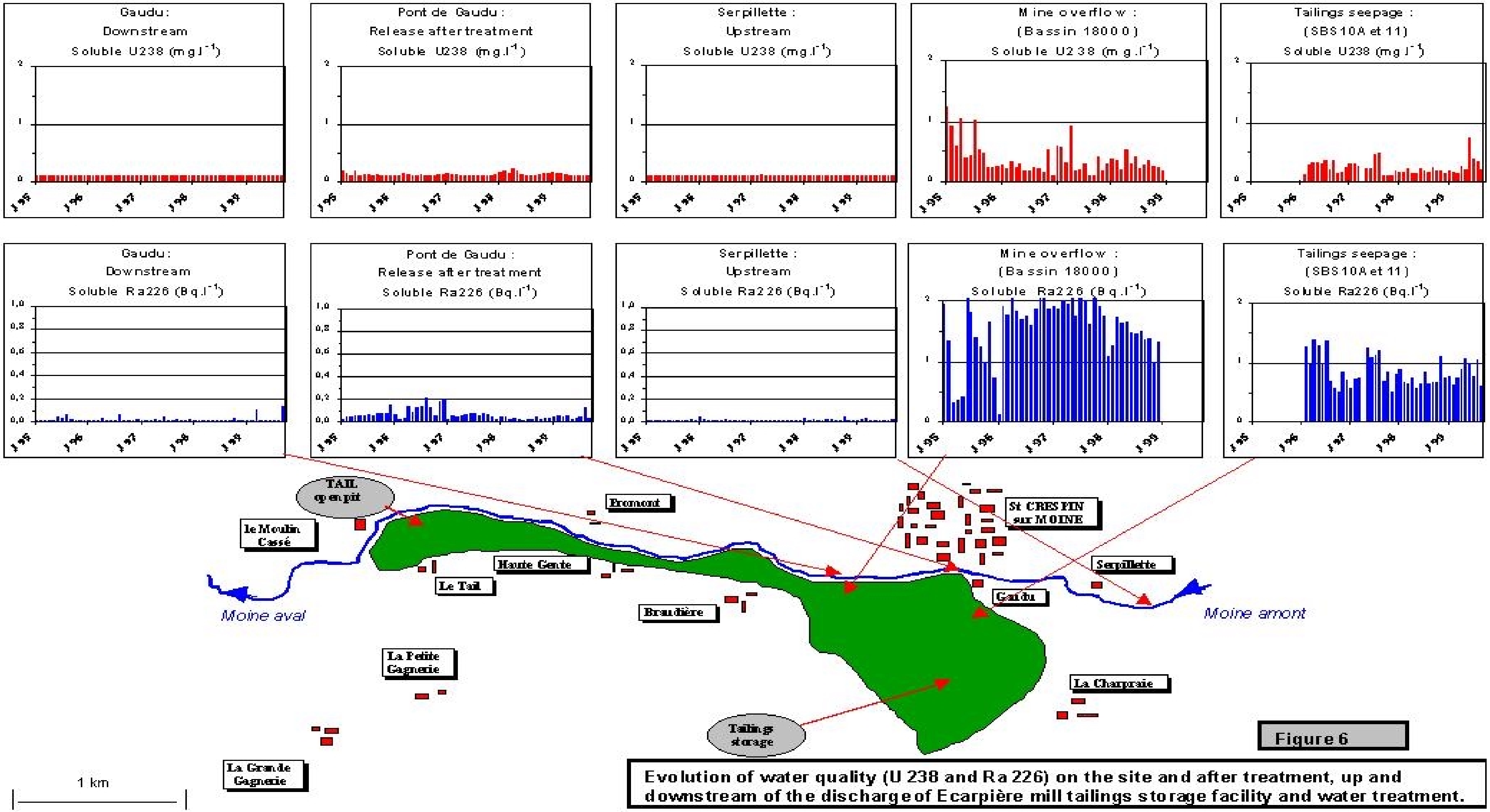


Figure 7 Natural improvement of an open pit mine water quality (Le Tail – see location near Ecarpière on figure 6**)**



Evolution of water quality (U 238 and Ra 226) on the site and after treatment, up and downstream of the discharge of Ecarpière mill tailings storage facility and water treatment.

In the frame of the future implementation of European Directive 96/29 (ED) (ED takes into account ICRP 60 recommendations) we have calculated the Total Annual Added Exposure for a realistic exposed group.

Under current French regulation (Decree 90.222) compliance is achieved if the ATAER (Annual Total Added Exposure Rate), based on a maximum annual exposure of 5 mSv, is less than 1 (4, 5). Following the ICRP 60 recommendation, ED 96/29 has fixed 1 $mSv.year^{-1}$ as the new limit : exposure should be evaluated using a realistic scenario for the exposure of the " groupe de réference " (or "critical group).

Following is a proposal of a detailed implementation of ED. In any case, it should be adapted to site specificities with the difficulty to choose the right indicators specially concerning the food chain. But it is also difficult to understand by the public and needs many analysis.

EXPOSURE SCENARIO of the "GROUPE DE REFERENCE" and RADIOLOGICAL IMPACT
Proposal for detailed realistic calculation of the added* annual effective dose in $mSv.year^{-1}$

$$\Delta A.t_1.10^{-6} . 1 + \Delta B.(t_1 + t_2).10^{-6}.0,39 + C_1.t_1.10^{-6}.1,1 + C_2.t_2.10^{-6}.1,1 + \Delta D.(t_1 + t_2).d.10^{-3} . 1,4.10^{-2}$$

Air

$$+ \Delta_1 Ra.b_1.\ 2,8.10^{-4} + \Delta_1 U.b_1.\ 4,5.10^{-5} + \ldots$$ Water

$$+ \Sigma_i (\Delta_i\ Ra.b_i).2,8.10^{-4} + \Sigma_i (\Delta_{i.}U.b_i).4,5.10^{-5} + \ldots$$ Food

ΔA = Gamma dose rate ($nGy.h^{-1}$) ΔB = Potential Alpha Energy Radon220 ($nJ.m^{-3}$)
$C_1\ C_2$ = Potential Alpha Energy due to industrial Rn 222 indoor (1) and outdoor (2) ($nJ.m^{-3}$)
ΔD = radioactive dust in the air (Long lived α emitters Bq alpha)
$\Delta_1 Ra$ = Radium Concentration in ingested liquid ($Bq.l^{-1}$)
$\Delta_i Ra$ = Radium Concentration in product i ingested ($Bq.kg^{-1}$ fresh weight)
$\Delta_1 U$, $\Delta_i U$,....= Uranium Concentration in ingested water ($Bq.l^{-1}$) and products i ($Bq.kg^{-1}$) (details for "i" in fig.3)

t_1 = 3500 h spent indoor $d = 0,8\ m^3.\ h^{-1}$
t_2 = 3500 h spent outdoor $b_1 = 1,6\ l.d^{-1} \times 365$
b_i = annual consumption for product "i" of the food chain (kg of fresh weight)
With 1 ; 0,39 ; 1,1 ; $1,4.10^{-2}$; $2,8.10^{-4}$ et $4,5.10^{-5}$ being equivalent dose coefficients for each of considered radionuclides
* *"added dose" means that each exposure is due to the difference (Δ) between that of the station where the "Groupe de Réference" is located and the station representative of the natural background.*

As example, on the base of sampling and measurements done on seven monitoring stations in the vicinity of Bessines industrial site, the **following table gives the result of annual effective dose for the different pathways according to the proposed detailed scenario**.

Bessines 1998	Dose (mSv)			Added dose (mSv)	
	Background	La Chataignière	Average 7 stations (Residential areas)s	La Chataignière	Average 7 stations
Gamma	0.60	0.56	0.70	0.00	0.10
Long lived α	<0.04	<0.04	<0.04	0.00	0.00
Industrial PAE Rn indoors	-	0.47	0.17	0.47	0.17
Industrial PAE Rn outdoors	-	0.19	0.09	0.19	0.09
Domestic water	0.28	0.28	0.28	0.00	0.00
Food chain	0.31	0.55	0.43	0.24	0.12
Total added effective dose				**0.90**	**0.48 (0.11 to 0.90)**

Background for gamma, dust, domestic water and food chain is the average of three stations representative of the natural radioactivity in the same geological context as Bessines : but they are not giving the measures on the site before the start of mining and milling thus giving probably an overestimation of the resulting added exposure.

In 1998, total effective dose of the 12 background monitoring stations implemented around mining sites in France range from 1.4 to 4.3 $mSv.year^{-1}$ (average 2.5 $mSv.year^{-1}$) which is much larger than the calculated added effective dose due to the remediated sites.

Conclusion : After uranium mining and milling, remediation perpetuates a limited radiological impact to the public, contributing to sustainable development.

The results of several years of monitoring show a direct influence of the cover on the tailings for stations located on site. The impact due to gamma exposure or radon is rapidly decreasing and the total impact to the public is limited.

We must keep in mind the difficulty to assess a 1 mSv added effective dose due to the natural variability of radioactivity and the resulting problem to apply it to U mining and milling. Although ways should be explored to simplify the scenarios for the implementation of ED 96/29 to the mining context, a detailed test shows that, after the remediation work done so far, the impact on the public satisfies the proposed limit.

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