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Abstract

The CEA has determined reference zones for the radioactive waste arising from the decommissioning of a linear particle accelerator (700 MeV, electrons) and a synchrotron (3 GeV, protons). The analytical approach adopted makes allowance for the design and history of the installations and the corresponding operating rules. The methodology used consisted in:

- Identifying those areas and materials where interactions could occur between the beam and the material.
- Characterising the radiation fields and determining which radioactive elements were representative of the potential activation of the materials.
- Estimating the activity per unit mass by means of semi-empirical calculations or Monte Carlo type simulations.
- Checking the radiological state of the zones by measuring the radioactivity of samples and mapping absorbed dose rates.

That process has been ratified by the safety authorities and has allowed to increase the value of 85 per cent of materials (about 3.10^7 kg), so that the production of final radioactive waste have been very limited.

1. INTRODUCTION

The waste zoning methodology used for the Saturne2 synchrotron (3 GeV, protons) will be described in this document. Charged particles (p, d and heavy ions up to ^{129}Xe) were accelerated in Saturne2 between 1979 and 1997. Dismantling began in 1998 and is due for completion in 2004. The nature, number (several hundred thousand items) and masses (around 34.10^6 kg of concrete shielding and 3.10^6 kg of metal) of the materials on the 20,000 m^2 site are such that:

- When determining disposal techniques, the facts about potential induced activation have to be known, with preference being given whenever possible to recycling rather than disposal.
- General radiological characterisation methods have to be found which can be reproduced and are statistically representative of the wide range of materials.

2. BACKGROUND AND OPERATING RECORD OF THE ACCELERATOR

The following action was taken:

- A record of the accelerator operating conditions was drawn up [1] (energies, currents and periods of beam use, types of particles etc.). The main facts to emerge are the following:
  - Of the total number of particles produced and accelerated, 73% were protons, 20% were deutons, 5% were α particles and 2% were ^3H and heavy ions.
  - The energy breakdown E of the predominant particles was as follows: 1 GeV<E<3 GeV (42%), 0.5 GeV<E<1 GeV (33%) and E<0.5 GeV (25%).
- Beam losses were localised and quantified [1]: in the particle injection and extraction zones between the successive acceleration devices or during transfer to test lines; along test lines at slits, inflectors, collimators and adjustment and diagnosis apparatus; in the vicinity of test targets and beam dump etc.
- The natures of the various types of material likely to have been activated were identified [1] and their chemical composition (major and trace elements) and isotope content were determined. The chemical compositions were obtained using AFNOR standards for metals and by neutron activation in the reactor or by I.C.P-M.S. (Inductively Coupled Plasma-Mass Spectrometer) for concrete.

3. INTERACTIONS BETWEEN BEAM AND MATTER AND RADIONUCLIDES PRODUCED

In Saturne2, the predominant activation mode was hadron reaction between particles and the nuclei of the materials. When a hadron reacts with a nucleus, numerous secondary particles are emitted (neutrons, protons, fragments of nuclei etc.) which may have sufficient amounts of energy to produce other particles in turn, after the reaction has occurred; this creates a nuclear reaction. In addition to the neutrons, protons and other fragments of nuclei emitted during this hadron reaction, secondary, unstable particles ($\pi^{\pm}$ π^0 pions) may also be created. Their lifetimes are sufficiently long for them to be taken into account in the secondary component of the radiation field. During this hadron reaction, the target nucleus is converted into a different isotope which is highly likely to be radioactive. In addition, some secondary particles that are re-emitted during the reaction may have sufficiently high energies to form radioactive nuclei themselves, by spallation or capture reactions. Direct activation by the particles (p, d, nucleus etc.) was considered as negligible on looking at activation due to neutrons. The reactions to be taken into account are of the (n,spallation), (n,xn), (n,xp), (n,γ) type.

To obtain direct activation by the beam and/or produce neutrons by means of a secondary reaction, the primary beam must have sufficient energy (threshold phenomena). For protons, for example, the threshold energy of production is 5 MeV for an iron target (reaction of the $^{56}Fe(p,n)^{56}Co$ type). For deutons, it is 2 MeV for an iron target [2].
The activation of matter (radionuclide, activity per unit mass) depends on a number of parameters:

the characteristics of the incident particles (type, fluence, energy, angle of incidence),
the effective activation cross-section,
the geometry and nature of the target material,
irradiation and decay times etc.

Table 1 shows the isotopes [2, 3, 4, 5] that are representative of the potential activation of the materials in Saturne2 (applied to other accelerators).

Table 1
Isotopes representative of activation of materials

Isotope	Half-life	Probable production	Isotope content(%)	Maximum effective cross-section (barn)	Possible origins
^{3}H	12.3 years	n,spallation			concrete
^{22}Na	2.6 years	^{27}Al (n,2p4n)	100	10^{-2}	aluminium, concrete
		^{23}Na (n,2n)	100	$4x10^{-2}$	concrete
^{54}Mn	312 days	^{54}Fe (n,p)	5.8	0.6	iron, steel, stainless steel, concrete
		^{55}Mn (n2n)	100	0.9	copper, aluminium
^{55}Fe	2,73 years	^{54}Fe (n,γ)	5.8	2.25	copper, aluminium, iron
		^{58}Ni (n,α)	68	0.082	stainless steel, concrete
^{57}Co	272 days	^{58}Ni (n,2n) $^{57}Ni \rightarrow$ ^{57}Co (filiation)	68	0.082	copper, stainless steel
^{60}Co	5.27 days	^{63}Cu (n,α)	69	0.03	copper
		^{59}Co (n,γ)	100	20	concrete, steel
		^{60}Ni (n,p)	26	0.16	stainless steel
^{63}Ni	100 years	^{62}Ni (n, γ)	4	14	stainless steel, steel
^{65}Zn	244 days	^{64}Zn (n, γ)	49	0.8	sheet metal, paint
^{108}Ag	127 years	^{107}Ag (n, γ) ^{108}Ag ^{108m}Ag	52	34	lead
^{110m}Ag	254 days	^{109}Ag (n, γ) ^{110}Ag ^{110m}Ag	48	90	lead
^{133}Ba	10.5 years	^{132}Ba (n, γ)	0.097	14	concrete containing barium
^{134}Cs	2.06 years	^{133}Cs (n, γ)	100	56	concrete (trace of Cs)
		^{134}Ba (n,p)	2.5		concrete containing barium
^{137}Cs	30.2 years	^{137}Ba (n,p)	11.4		concrete containing barium
		^{136}Ba- (n, γ) $^{137m}Ba \rightarrow Cs$-^{137}	7.8	0.4	concrete containing barium
^{152}Eu-	13.53 years	^{151}Eu (n,γ)	48	9000	concrete
^{154}Eu	8.59 years	^{153}Eu (n,γ)	52	3400	concrete

4 ASSESSMENT OF INDUCED ACTIVATION OF MATERIALS

The induced activation was assessed by means of Monte Carlo type simulations for concrete shielding and by semi-empirical calculations for the metal parts of the accelerators.

4.1 CONCRETE

From the operating record of the accelerator it was deduced that most of the loss points could be represented by 2 GeV proton reactions on a 1-metre diameter copper target, surrounded by concrete. This configuration (Figure 1) was modelled using the FLUKA Monte-Carlo code [6] capable of representing the generation and transport of charged and neutral particles. The code was used to calculate the neutron spectra, created by protons, in various regions of space. These spectra, along with the chemical compositions of the concrete, obtained by neutron activation in the reactor or mass spectrometry of samples [7], were introduced as input data into the DARWIN/PEPIN2 system developed at the CEA [8]. This code is used to solve the pairs of differential equations representing radioactive decay either analytically or numerically, particularly those concerning activation and spallation products.
The sequencing of the various computer codes is shown in Figure 2.
Figure 3 shows the concentration activity as a function of depth, calculated by simulation [9], in blocks of ordinary and heavy concrete.

Figure 4 shows a similar repartition of present radionuclides in heavy concrete.

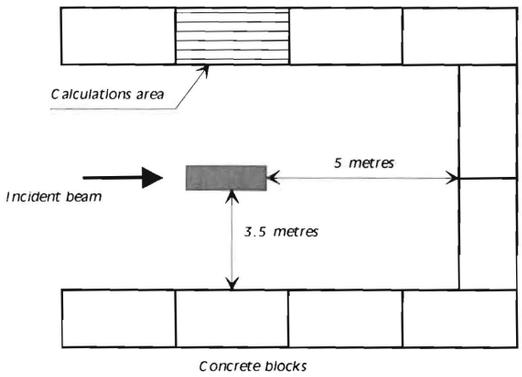

Figure 1
Geometry for calculation

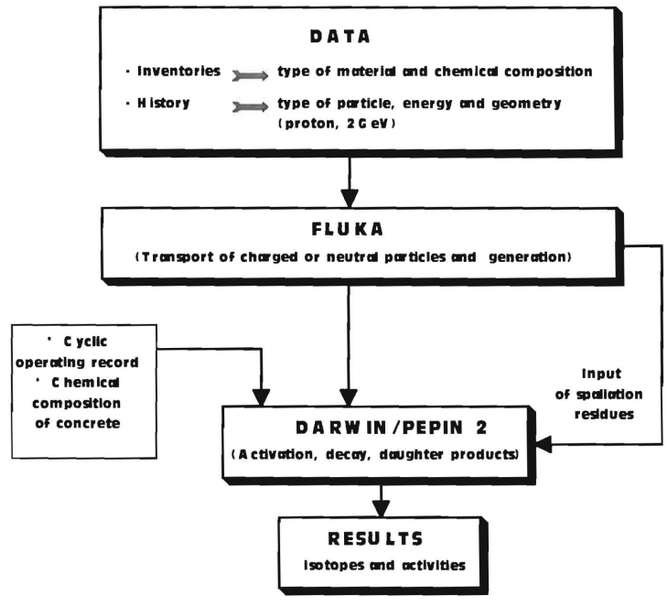

Figure 2
Sequencing of the various computer codes

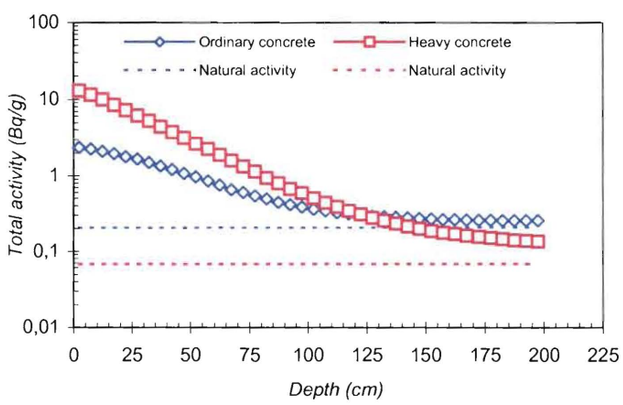

Figure 3
depth repartition of global activity in ordinary and heavy concrete (Monte Carlo simulation)

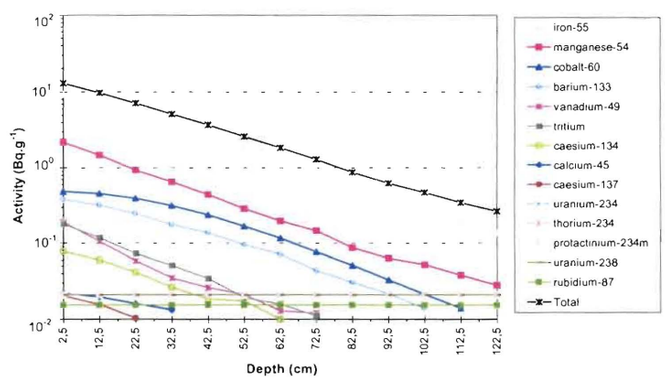

Figure 4
depth repartition of activity of present radionuclides in heavy concrete (Monte Carlo simulation)

4.2 METALS

To obtain an initial approximate result for monoenergetic neutrons, we considered that the concentration activity of a radionuclide could be calculated using the following equation, as indicated in Reference [3]:

$$A_i = \frac{N}{M_i} f_i \, \alpha_i \, \sigma_i(E) \, \phi \, [1-\exp(-\lambda_i T)] \, [\exp(-\lambda_i t)]$$

Where:

- A_i: concentration activity of radionuclide (i) ($Bq.g^{-1}$);
- N: Avogadro number (6.02×10^{23} atoms per mole).
- M_i: atomic mass of target element (g).
- f_i: concentration of target element in material.
- α_i: isotopic abundance of target element [2].
- σ_i: effective activation cross-section (10^{-4} m^2),this depends on the type of reaction, the energy of the incident neutron and the target nucleus[2].
- ϕ: neutron fluence rate (10^{-4} m^{-2} s^{-1}), this depends on the type of particles, the energy, the intensity and the target taking part in the reaction.

λ_i: radioactive constant $Ln2/T_{1/2i}$ (s^{-1})
T and t: irradiation time and decay time (s).

The neutron fluence rate (ϕ) is the hardest to calculate since it depends on a wide variety of parameters. To estimate it, empirical relationships [10] were used once their validity for Saturne2 had been checked [4] for different proton energy ranges E. Thus the fluence of the secondary particles produced by a proton with an energy E > 1 GeV reacting on a thick metal target (copper, iron etc.) can be obtained by solving the following equation:

$$\phi(\theta) = 1/[2R^2(\theta + 35E^{-1/2})^2] \quad \text{hadrons}.10^{-4}\ m^{-2}$$

Where R is the distance (m) from the target and θ (degrees) is the diffusion angle of the particle.
In the calculations, we used maximum effective cross-section σ for the monoenergetic neutrons produced (the most conservative hypothesis).
Since some of the radioactive isotopes produced have short half-lives, ranging from a few minutes to around one hundred days, they were assumed to have a negligible effect on the overall activity per unit mass.
For example, the activity per unit mass values (Bq/g) obtained for the predominant radioactive elements in the most highly activated areas (extraction areas SD2 and SD3 of the 3 GeV proton beam between the accelerator and the test lines) were:

Metals	Isotopes (activity per unit mass in Bq/g)
Copper	^{60}Co (80)
Aluminium	^{22}Na (89), ^{54}Mn (48)
Steel	^{54}Mn (124), ^{57}Co (33), ^{60}Co (19), ^{63}Ni (34)
Lead	^{108}Ag (19), ^{110m}Ag (602)
Iron	^{54}Mn (171)

5 REFERENCE WASTE ZONING

On the basis of the theoretical estimates described above, it was possible to determine "interest zones" as a function of the calculated activity values.
The installations were divided into "zones with no added radioactivity", "non-contaminating zones" and "contaminating zones". The definitions of these zones are given below [11]:

Zones with no added radioactivity (ZNAR): zones in which no radioactive substances have ever been produced, treated, handled, used, held, stored or transported or in which no devices emitting particles that could lead to activation have ever been used.

Non-contaminating zones (NCZ): zones in which radioactive substances are or have been produced, treated, handled, used, held, stored or transported, but in which there are no contaminating radioactive substances or devices emitting particles liable to activate the waste generated.

Contaminating zones (CZ): zones characterised by the presence of contaminating radioactive substances or the emission of particles likely to activate the waste generated.

In the case of Saturne2, the added radioactivity is due only to the activation of materials by the radioactive fields produced by the particle beams (beyond the threshold energy of production, compare § 3). There is no residual contamination.
The case of activation differs from that of a contaminated environment in that there is no absolute containment barrier that makes it possible to say that from a given point, there is no added radioactivity. We have assumed that there are two kinds of activated object:

Items of which at least one side is activated to a considerable extent, in other words, whose activation concentration is at least equal to the activity per unit mass values given in circular [11], i.e. 1/10 of the values laid down by EURATOM directive 96/29. Zones containing items in this category will be considered as contaminating zones.

Items whose sides are exposed, or potentially exposed, to particle beams, and whose activation is negligible, in other words, whose activation concentration is less than the activity per unit mass values given in [11]. Zones containing items in this category shall be considered as non-contaminating zones.

Finally, geographical limits have been set to circumscribe the influence of particle beams:

The thicknesses of walls constituting accelerator shielding have been calculated in such a way that, in the most severe beam energy and intensity conditions, the equivalent dose rate produced by neutron and γ rays, either direct or diffused, does not exceed 7.5 µSv/h. The corresponding fluence rates can be calculated from these equivalent dose rates. The International Commission on Radiological Protection[1] [12] recommends 0.7 $n.s^{-1}.cm^{-2}$ per µSv/h for an average neutron spectrum of up to 10 MeV and 0.1 $n.s^{-1}.cm^{-2}$ per µSv/h for neutrons with energies of a few hundred MeV. (The amount of neutrons that are not slowed down by the shielding and whose energy levels do not drop below 10 MeV is negligible).

A fluence rate of around 5 $n.s^{-1}.cm^{-2}$ does not cause any significant activation of the materials beyond the protective blocks.

Figure 5 shows the reference zones determined previously for waste (green, zones with no added radioactivity- yellow, non-contaminating zones- red, contaminating zones).

[1] Protection quantities in publication at the starting date of Saturne2

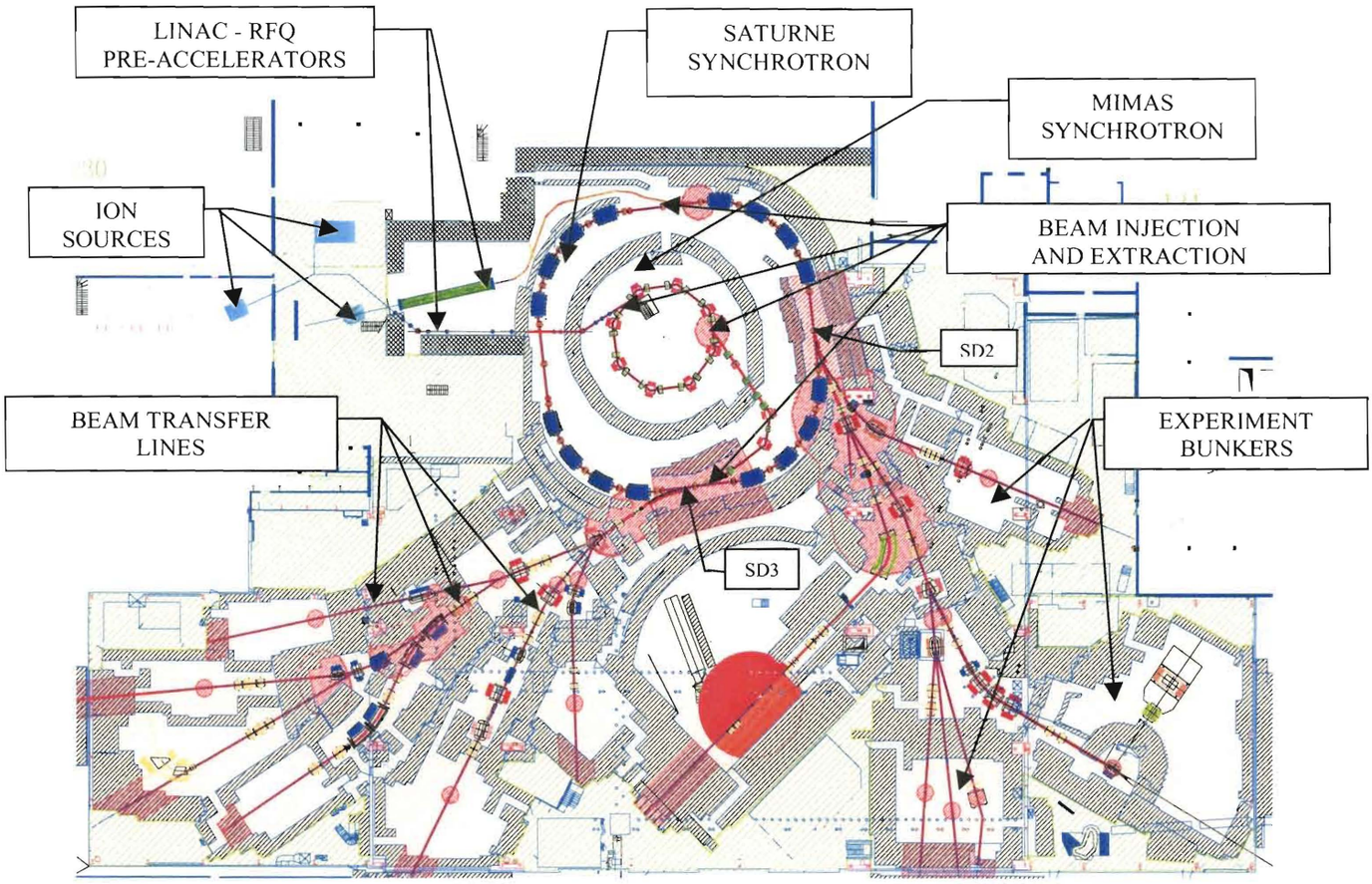

Figure 5
Reference waste zones

6 CHECKING THE RADIOLOGICAL STATE OF ZONES BY MEANS OF SAMPLING AND ANALYSIS

The zones shown above are based on theoretical studies only. It is therefore important to check their validity statistically by means of sampling.
The "with no added radioactivity" nature of the zones was confirmed by surface checks. The detection limits, obtained using a polyradiameter and the associated probes, for wipe tests over 10^{-2} m^2 with a yield of 30%, were: 5.10^2 Bq/m^2 for β radiation, 1,4. 10^4 Bq/m^2 for γ radiation and 1,5. 10^4 Bq/m^2 for x-rays.

The "non-contaminating" or "contaminating" nature of the zones was validated on the basis of:

- Samples taken by core drilling to a depth of 0.4 m in the concrete, at 50 points around the Saturne ring. The sampling points, sampling method and measurement results are described in document [1]. The measurement results, obtained in the laboratory by γ spectrometry, confirm the orders of magnitude calculated in §4. They are systematically lower than the values obtained by modelling. Figure 6 gives the activity per unit mass values (Bq/g) obtained for extraction zone SD2 [13].
- Experiments carried out [4] to determine the activation of various metals. Thus, disks (diameter 0.05 m, thickness 0.002 m) of copper, aluminium, iron and stainless steel were placed in extraction zone SD3 and bombarded by the secondary particles emitted for 1.8×10^5 s. The values obtained for the predominant radioactive elements (except those with short half-lives) made it possible to validate the semi-empirical formulas and confirm the orders of magnitude calculated theoretically.

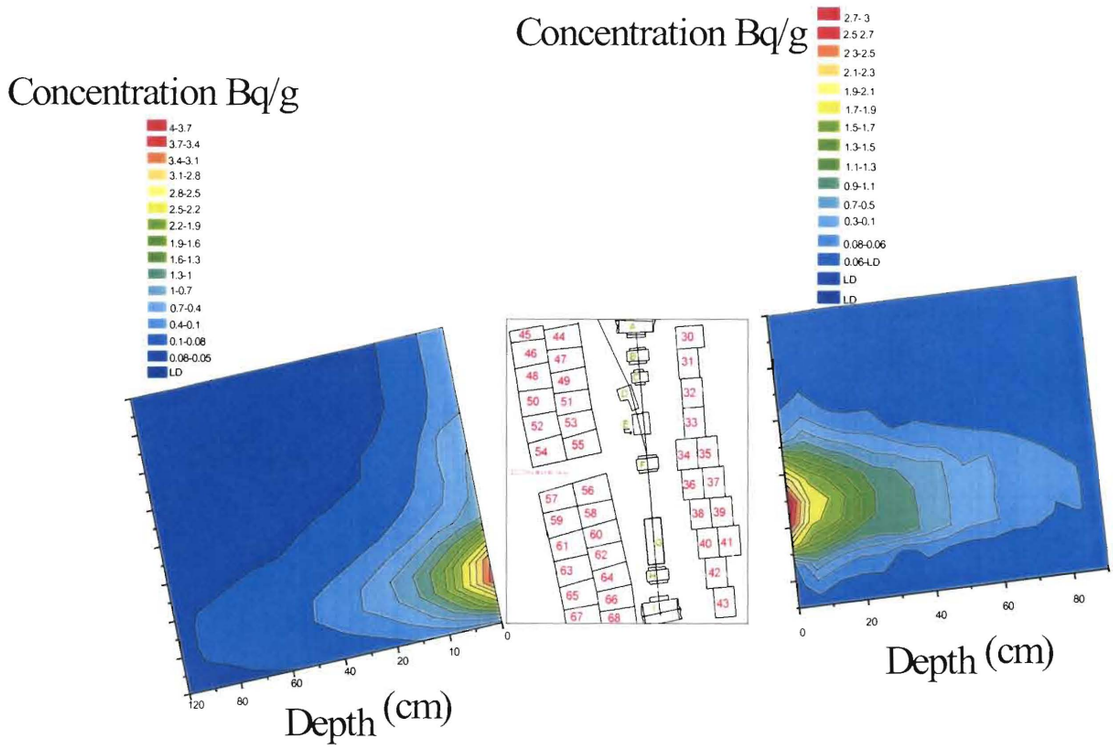

Figure 6
Activity concentration as a function of depth in SD2
(samples)

7 CONCLUSION

When applied strictly, the methodology adopted to determine the waste zones of the Saturne2 synchrotron, can be used to establish zones for conventional and other radioactive waste zones fairly accurately, without resorting to an inordinate amount of sampling and analysis, given the high volumes and masses of the items concerned. This approach, when supplemented by the creation of standard spectra and transport functions to estimate the levels of activity of the accelerator components, leads, after validation by the safety authority, to the checking, sorting (according to nature and level of activity) and rigorous management of waste generated in an accelerator. Thus, it was possible to recycle almost 85% of the material:

In the public works sector in the form of concrete blocks (crushed and used as ballast for roads),

In smelters or, in the case of metals, re-used in the construction of new accelerators.

Less than 15% is considered as ultimate waste and sent for disposal by means of the appropriate techniques.

This methodology can be applied to other accelerators, as was the case of a linear particle accelerator (700 MeV, electrons) at CEA/Saclay.

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