METHODOLOGY OF A CHEMICAL SAFETY ASSESSMENT OF RADIOACTIVE WASTE DISPOSAL WITH AN EXAMPLE PRESENTED FOR THE CENTRE DE L'AUBE LOW LEVEL RADIOACTIVE WASTE (LLW) REPOSITORY

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ABSTRACT

ANDRA (the French national radioactive waste management agency) has to insure the protection of the environment and the population all around its radioactive waste disposals. The Agency does not restrict itself to insure that there is no radiological impact: the impact of species whose first hazard is a chemical one with regard to human beings is included in the safety assessment. The methodology adopted to assess the toxicological impact is based on the methodology developed for the radionuclides. The chemical impact assessment lies in defining what has to be considered as a toxic substance, determining the inventory of these toxic substances expected to be present in the whole repository at the closure of the operational step, investigating under various scenarios the ways by which these substances are likely to be transferred through the different barriers of the repository and to reach the outlets, quantifying the transfer into the food chain and choosing criteria which could be used in the evaluation of the health effects for the exposed group.

The result of the radionuclide transfer calculation is expressed on a dose term basis, which can be compared with the current legal criterion. Concerning the chemical impact, no dose factor exists and then, the transposition of the ingested/inhaled quantity of a toxic element to a probability of effect cannot be done. Moreover, the French and European rules do not set up a chemical criterion for Nuclear Plant regarding human being protection. Therefore ANDRA has adopted indicators for ingestion and inhalation routes, based on the French drinking water regulations, on the Dutch soil regulations and on the tolerable weekly intake.

The case of lead impact in the environment of the Centre de l'Aube Low Level radioactive Waste (LLW) repository is presented as an example of application of the methodology.

INTRODUCTION

ANDRA, the French national radioactive waste management agency, has to insure the protection of the environment and the population all around its radioactive waste disposals. Within this framework, radiological impact assessment is one of its more important missions, which concerns human beings, over a short and long term period (safety assessment) and environment, over a short term period, that means during the site operational and surveillance phases (radiological and chemical parameter analysis). The Agency does not restrict itself to make sure of no radiological impact: the safety assessment for Low Level Waste repositories includes the impact of species whose first hazard is not a radiological one but a chemical one with regard to human beings.

On the basis of studies devoted to radioactivity, a methodology of approaching the question of safety and impact assessment for waste containing toxic elements has been developed, which is based on the radiological impact assessment methodology.

The chemical impact assessment includes:

- the selection of the elements to be considered in a first screening assessment;
- the assessment of the exposure levels of the exposed group. This step includes the investigation of the ways by which the toxic substances are likely to reach the environment and the food chain according to various safety scenarios;
- the impact characterisation.

These items are discussed below in the first main part relative to the methodology. The second part treats the example of the impact assessment for the lead contained in the waste disposed of in the Centre de l'Aube disposal. This example is restricted to the transfer under the reference scenario by water route, over a period during which no major climate changes are expected.

METHODOLOGY OF CHEMICAL IMPACT ASSESSMENT FOR THE SAFETY STUDIES

Selection and classification of the relevant toxic elements

In order to focus on the most relevant elements, a general list has been established, which comprises the toxic substances or elements whose impact on human beings and environment has to be evaluated regarding their potential hazard. This potential hazard depends on the intrinsic toxicity of these substances, their quantity in the whole disposal and their mobility.

- General list

The general list of chemical hazard substances to be taken into account in the impact assessment includes:

- elements considered in France as hazardous elements in the drinking water (décret 89-3, 1989) and/or in workroom air (circulaire du 19 juillet, 1982) and for which limits have been set up because of their proved effects on human health (**lead, cadmium, nickel, arsenic, cyanide, chromium, mercury, antimony, selenium, asbestos**...); as well as
- specific elements used in nuclear industry (essentially nuclear power plant) likely to induce toxic effects if they are ingested and/or inhaled (**boron**, **uranium**, **beryllium**).

This list is not exhaustive. It has been set up with the purpose of selecting the elements/substances of primary importance with regard to safety assessment. The impact of these elements/substances are investigated first. Then, according to the nature of the waste, some "new" elements may appear with a relatively high potential hazard. Consequently, they are taken into consideration, provided that useful data be available, in particular the inventory.

In the future, the list will be completed according to the practices from other countries if relevant within the context. It can be noticed that zinc is often considered. This metal has been retained in particular by the Canadians and Swiss [EUR 15687FR, 1995].

● Potential hazard assessment

The first step in the impact assessment consists of the selection of the elements that have to be studied preferentially with regard to the induced chemical risk. This selection is based on the potential hazard assessment, which depends essentially on the toxicity with regard to human beings, the estimated quantity in the whole repository and the mobility of the elements within the repository, the geosphere and the biosphere. Parameters likely to be used in the characterisation of toxicity and mobility are reference doses (RfD), partitioning coefficients (Kd) and solubility.

In a first time, a water route potential hazard assessment is performed by taking into consideration the only quantity and intrinsic toxicity.

The toxicity by ingestion route is represented by the oral reference dose (oral RfD).

The estimated total amount (inventory) of the toxic elements comes from the information given by the waste Producers.

The knowledge of the ratio of the whole quantity of an element (expressed in elementary mass unit) and the reference dose for all the elements permits to sort them as a function of the potential hazard: the element for which the ratio is the highest is thought to be the most hazardous one.

Determination of the exposure levels of the population

Within the framework of safety studies, the population is taken into consideration through the most exposed man (referred as to the reference man here) living in the vicinity of the repository (directive 96/29, 1996, ICRP 46, 1985). Such a reference man is an average member of the critical group.

According to the ICRP (ICRP 81), the critical group is defined as the "group of people representative of those individuals in the population expected to receive the highest annual dose, which is a small enough group to be relatively homogeneous with respect to age, diet, and those aspects of behaviour that affect the annual doses received".

The safety approach consists in investigating the ways by which the elements initially present in the waste are likely to be transferred from the repository to the biosphere and hence can reach the reference man. These ways are described into normal development as well as accidental transfer scenarios and cover both short term and long-term periods.

These scenarios, which have been developed with the purpose of assessing the radiological impact of the repository are also used in the assessment of the chemical impact. They refer to the repository barriers, the outlets and the biosphere.

● Transfer through the repository structures to the groundwater and migration to the outlet under the reference scenario

The reference scenario considers that the waste can be leached by the rain water infiltrated through the disposal cover and structures. The elements in the leachate can reach the groundwater located under the repository and migrate to the surrounding river (outlet).

The figure 1 represents the conceptual model of the transfer of the chemical elements from the repository to the outlet whose water is supposed to serve for domestical uses (drinking, irrigation...). According to the reference scenario, the outlet is the downstream river.

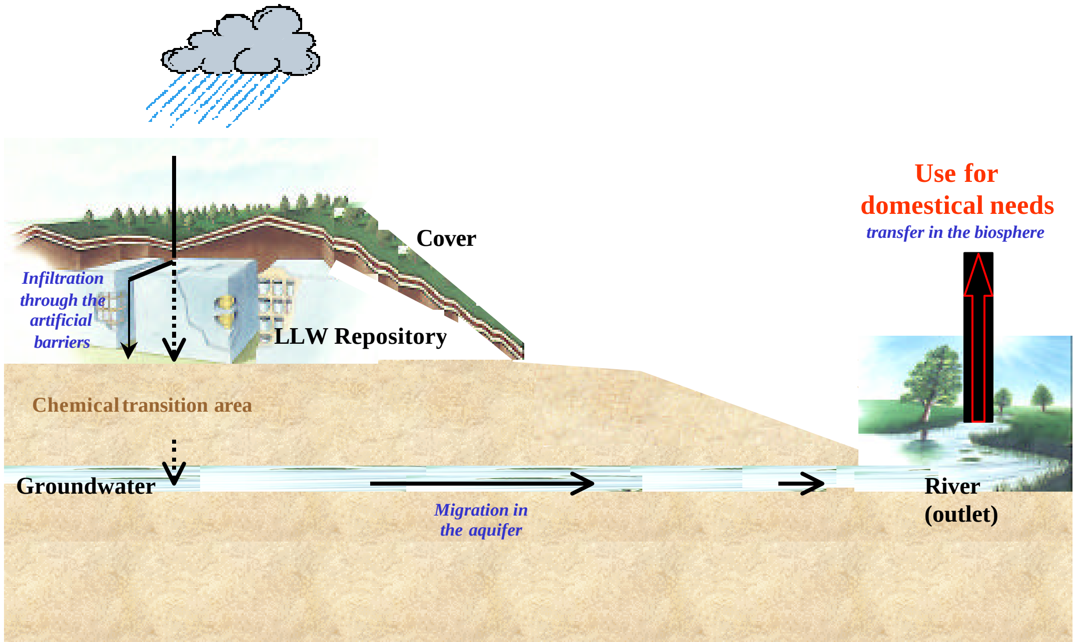

Fig. 1. Conceptual model for the migration of the element from the LLW repository to the river according to the reference safety scenario

The LLW repositories are made of concrete structures containing waste packages. Generally, the waste is also immobilised within a cement-based material. The infiltration water that may cross the disposal annually is supposed to get equilibrated chemically with these materials.

Two periods are distinguished after the closure of the disposal, i.e at the end of the operational phase and after cover laying: the surveillance period (or institutional control period) and the post surveillance period (or post control period).

The surveillance phase is supposed to last about 300 years, according to safety considerations. It consists of the monitoring and the maintenance of the facility, as well as the surveillance of the environment. The environmental control is performed by the way of measurements of radioactivity and chemical parameters within the groundwater and the river located in the nearfield of the disposal, plants, soils, animal products when relevant, and air.

The post surveillance phase is a passive period during which a complete abandonment of the site may occur. Such an event is not expected to induce unacceptable consequences for the environment and the surrounding population, even if the facility is not supposed in a good working state. As a matter of fact, the repository structures are supposed to be entirely destroyed mechanically, according to the safety scenarios (such an assumption leads to the calculation of the maximum post-closure exposure levels to radioactivity).

During the surveillance period, the transfer of elements from the waste packages out of the repository is controlled by diffusion processes.

During the post surveillance phase, the transfer occurs essentially by convection. In this case, the annual amount getting to the groundwater is the product of the concentration within the infiltration water and the annual volume of infiltration water crossing the repository. For the elements whose amount is restricted by their solubility (i.e. not trace element), the transition area located between the bottom of the repository and the groundwater plays an important role : it can limit through precipitation processes, the quantity of the element likely to reach the groundwater, and hence, the outlet. Precipitation takes place in an area where a pH gradient exists: about pH 12-13 upper and pH 6-8 down, which is the measured pH of the natural groundwater.

The chemical conditions within the repository, the transition area, and even the groundwater are essential to determine the quantity of the element that can be transferred through aqueous route.

The transfer within the repository to the groundwater and the migration within the groundwater to the river is modelled by ANDRA and implemented in calculation codes. As they do not take into account the transition area, the quantity of toxic elements reaching the bottom of the disposal is directly calculated by considering their solubility within this area. The solubility is estimated by the way of calculation codes. If data are missing, experiments may be performed to determine the solubility in particular conditions.

The transfer calculation results are expressed as the concentration of the element in the water of the outlet versus time.

● Transfer in the biosphere and exposure of the reference man

The use of the water from the outlet for domestical needs implies that the potentially contaminant elements may be introduced into the food chain and may therefore contaminate the reference man through ingestion of contaminated products and inhalation of contaminated dusts in air.

The figure 2 shows the conceptual model for the transfer of the substance from the outlet to the reference man, according to an agricultural system.

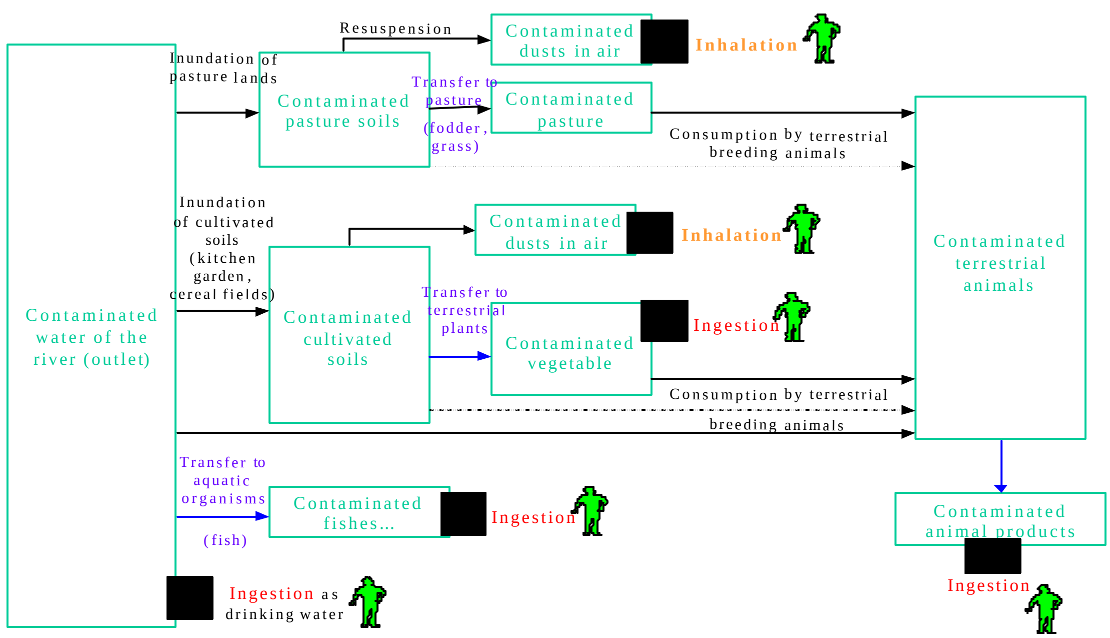

Fig. 2. Transfer routes of chemical elements from the outlet of the repository to the reference man according to an agricultural system

The biosphere pathways are modelled and implemented in a calculation code named Aquabios, which considers that the various compartments are in equilibrium with the others.

The partition of an element between two compartments, as for example between the soil and the plant, is illustrated by a transfer factor (FT), which is the ratio of the plant concentration of the element and the concentration in the soil, at the equilibrium. The calculation requires therefore the knowledge of the transfer factor values for all the elements in an appropriate valence and all the compartments.

The results are presented on a table which gives the concentration of the element within each compartment (soil, animal products, vegetables…) and the mass of the element ingested and inhaled by the reference man, according to given alimentary customs, as a function of time. These results represent the exposure levels to be compared with relevant indicators in order to determine the impact for the reference man.

Indicators for the chemical impact assessment

Radiological impact can be assessed by the knowledge of the exposure dose, which comprises the contribution of every radionuclide by every exposure route (ingestion, inhalation and external exposure). The dose is the product of the activity and the dose factor. The dose is then expressed in Sievert. The dose factor depends on the isotope, the chemical form and the exposure route. The acceptable dose set up by the Euratom Directive is 1 mSv per year (directive 96/29, 1996).

Concerning the chemical impact, the French and European rules do not impose a criterion for Nuclear Plant regarding human being protection. That is the reason why ANDRA has adopted indicators for valuing the chemical impact based on the following considerations:

- the concentration of toxic elements in the outlet-water should not exceed the limit value admitted in the drinking water (first criterion);
- the concentration of the toxic in the soil should not exceed the value above which a soil is considered as polluted soil (second criterion);
- the quantity of a toxic element ingested and inhaled by the reference man should not exceed the value above which health effects are expected (third criterion), or, for stochastic effects, the associated risk must be acceptable.

The toxic element maximal concentrations allowed in the drinking water are mentioned in the 89-3 decree dated January 1989. This decree will be replaced in 2003 by the transcription in French law of the 98/83/CE European directive dated November 1998 (directive 98/83, 1998).

The soil concentration criterion corresponds to the limits set up in Holland (i.e. for Netherlander soils). Investigations are carried out in France and some data should be suggested before long.

The third one is chosen from literature data regarding the different concentration thresholds and the associated effects on human beings resulting from chronic and/or acute exposures.

APPLICATION TO LEAD

Potential hazard assessment for the toxic elements included in the radioactive waste devoted to the Centre de l'Aube repository

The potential hazard assessment by ingestion route of the toxic elements recorded in the inventory of the Centre de l'Aube Low Level radioactive Waste repository has shown that lead is the element with the highest ratio of the estimated quantity over the toxicity (RfD). It is up to 10^{11} kg, the ratio corresponding to the less hazardous element being 1,2 10^{6} kg (for beryllium). The classification of the toxic elements according to the quantity and the oral route toxicity is the following:

$$Pb > Sb > Cr > Cd > B > Ni > Be$$

In spite of a relatively low quantity, chromium appears to be the third element of importance. The ratio is a conservative ratio calculated by dividing the total quantity of chromium, irrespective of chemical form, by the RfD related to Cr(VI), which is the more toxic valence.

The majority of the lead present in the waste comes from screens used for the protection of the workers against ionising radiation. The quantities of metallic lead in a LLW radioactive repository are thus very high. As an example, estimated lead mass in the Centre de l'Aube exceeds the one of the other recorded toxic elements for more than a factor 1000.

At the time being, the mobility of every toxic has not been taken into account when calculating the potential hazard because several data are missing yet. In the future, Kd values for artificial (cement matrix) and natural (underground water and rock) materials, will be used to determine whether the above classification is reliable or not with regard to the potential risk for human beings in the case of potential releases in the environment.

Determination of the exposure levels of the population

● Transfer through the repository structures to the groundwater and migration to the outlet under the reference scenario

In the case of lead impact assessment, a simple assumption is made which considers the metal as entirely corroded. Specific studies carried out on sorption capacity of cement and natural matters have shown that lead is not very movable within the repository when associated with concrete because of a high distribution factor (Kd about 2000 L/kg, Thuret, 1999) and because the diffusion of oxidising species (dissolved O_2), does not occur. As a matter of fact, corrosion tests of a lead sheet put into a cement matrix core sample has shown that corrosion seems to be limited by diffusion processes. The same experiment making use of lead sheets placed into a basic solution of pH 13 shows that lead is rapidly and widely corroded. The lead concentration of the solution quickly reaches the solubility limit. These experiments indicate that metallic lead is highly corroded in a basic environment, as long as the diffusion of Pb^{2+} and dissolved O_2 species can occur. When the diffusion of oxidising species is hindered, the corrosion stops, and hence lead mobility is very low.

Therefore, in the safety assessment, the quantity of lead likely to be transferred from the waste to the biosphere is considered negligible during the surveillance phase, which is supposed to last about 300 years. That means that, as long as the repository barriers are thought to conserve their physical and chemical characteristics, lead from the disposed waste is not expected to be present in the river.

The safety scenario considers that, from the end of the surveillance phase, the characteristics of the repository are altered. In other words, after 300 years, the artificial materials are supposed to be entirely destroyed (in a powder state). Then only the geosphere should provides confidence that quantities of the element will not reach the outlet and be transferred through the food chain.

The concentration of lead in the groundwater is controlled by the minimum of its solubility within the transition area. That means that the annual quantity of lead transferred to the groundwater does no exceed the product of the minimum solubility within the transition area (about 130 µg/L, see figure 3) and the volume of infiltration water having crossed the disposal per year likely to be in contact with lead. For the calculation, assumption is made that only about 10% of the volume of infiltrated rain water can be in contact with the metallic lead whose repartition within the whole repository is not homogenous.

The solubility of lead has been measured in many solution mixtures containing various proportions of water equilibrated with cement and water taken from the Centre de l'Aube groundwater. The solution with pH up to 12 units consists of concrete water only, while the one with pH 8,5 represents the natural groundwater only. The pH of the water within the transition area is fixed by the concrete water as long as it represents more than 20 percents of the mixture.

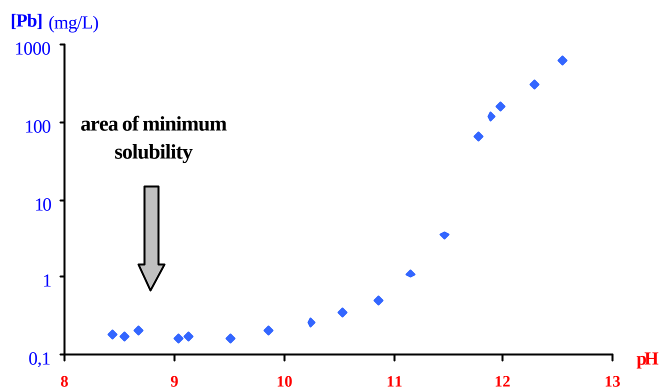

Fig. 3. Minimum solubility of lead within the transition area

The concentration of lead in the river is then assessed by the modelling of the transport of the element to the river. The calculation code used is MODFLOW/MT3D.

The calculation is made by considering a distribution factor of lead on the Centre de l'Aube groundwater natural materials of 100 L/kg. This value is quite consistent with those measured through batch experiments (the Kd of lead on the sand from the Centre de l'Aube groundwater is comprised between 150 and 730 L/kg, Thuret, 1999).

The calculated lead concentration in the river as a function of time is shown on table I.

Table I. Evolution of the calculated lead concentration within the river during the post surveillance phase, according to the reference scenario (µg/L)

time after the surveillance period (years)	C_{river} **(µg/L)**
0	0
1000	9,6E-06
2000	5,6E-05
3000	1,2E-04
5000	3,4E-04
10000	1,7E-03
20000	6,4E-03
30000	1,1E-02
50000	1,9E-02
100000	2,4E-02
200000	2,4E-02

● Assessment of the exposure levels of the reference man

According to the fact that lead will not be corroded as long as the concrete barriers of the repository will keep their initial characteristics (physical and chemical characteristics) and that dissolved lead is quite strongly sorbed on cement based materials, ANDRA assumes that no lead will reach the outlet during the surveillance phase.

Then, the lead impact calculation and assessment concern the post-surveillance phase, which is supposed to start about 300 years after the closure of the repository.

The partition of lead in every compartment of the biosphere during the post surveillance phase has been calculated with the Aquabios calculation code. The values of the transfer parameters within the biosphere are the ones defined for the radioactive lead (^{210}Pb) (see table II).

The variation of the calculated concentration of lead in the cultivated soil is shown on table III.

Table II. Major values of the transfer parameters of lead used in the biosphere model

Kd soil (L/kg dw)	**Soil-plant transfer factors (kg dw/kg fw)**		**Foliar uptake factors (m2/kg fw)**		**Plant to animal product transfer factors (d/kg dw)**	
2000	- cereals	1.3E-02	- cereals	7.9E-02	- beef	2.0E-03
	- fruits	1.6E-03	- fruits	3.0E-02	- dairy products	2.0E-03
	- leaves	3.2E-03	- leaves	2.8E-02	- pork	6.5E-02
	- roots	4.0E-03	- roots	2.2E-02	- poultry meat	2.3
	- potatoes	2.0E-03	- potatoes	5.1E-02	- eggs	1.4
	- legume folder	2.0E-02	- legume folder	1.2E-01	- milk	2.0E-04 (d/L)
	- grass	5.0E-03	- grass	1.2E-01		

Table III. Evolution of the calculated lead concentration (mg/kg dw) in the cultivated soil (kitchen garden) irrigated with the river water

time after the surveillance period (years)	C_{soil} (mg/kg dw)
0	0
1000	4,8E-06
2000	3,1E-05
3000	7,6E-05
4000	1,4E-04
5000	2,5E-04
10000	1,5E-03
20000	6,3E-03
30000	1,2E-02
40000	1,7E-02
50000	2,0E-02
100000	2,6E-02
120000	2,6E-02
200000	2,7E-02

Figure 4 shows the contribution of ingested food and fluid and inhaled particles to the total amount of lead incorporated by the reference man. Terrestrial plants and drinking water are shown to contributed to 88 percents of the total dose of lead.

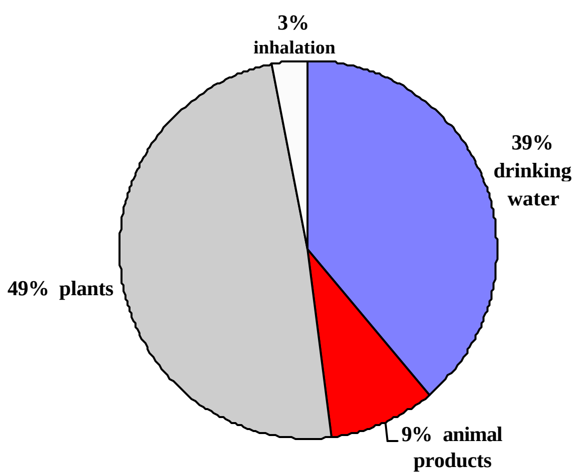

Fig. 4. Contribution of inhalation, plant and animal product ingestion and drinking water to the total amount of lead incorporated by the reference man

Chemical impact assessment

● Concentration in the water

The lead concentration in the river does not exceed the guideline value admitted in the drinking water in France (50 µg/L and 10 µg/L in the future).

● Concentration in the soil of the kitchen garden

The concentration of lead in the cultivated soil does not exceed the guideline value admitted in Holland (85 mg/kg), because no criterion exists in France in the time being.

● Criterion regarding the reference man

The quantity of lead added to the annual amount of ingested and inhaled lead by the reference man (164 mg/year) is negligible. It represents less than 0,2% of the reference lead intake, with regard to the CIPR 23 (ICRP 23, 1985) data on the reference man (0,44 mg/day from food and fluids and 0,01 mg/day from airborne). The daily intake following the chronic exposure to lead from the Centre de l'Aube repository is much lower than the Provisional Tolerable Weekly Intake PTWI (25 µg/kg/week according to WHO, 1993, or about 91 mg/year)

CONCLUSION

On the basis of studies devoted to radioactivity, a methodology of approaching the question of safety and impact assessment for waste containing toxic elements has been developed, which is based on the radiological impact assessment methodology. This process is entirely innovating for toxic waste disposals because it deals with human health chemical risks over a more or less long-term assessment period. The impact study for non-radioactive waste disposal does not include the calculation of the quantity of the hazardous elements from the waste likely to reach the environment over a long-term period. The impact assessment is generally limited to tens years. The management of such non-radioactive disposals.

Within the framework of radioactive waste management, the calculations performed to evaluate the exposure levels of the most exposed man living in the surroundings of the Centre de l'Aube repository show that the risk associated is quite negligible.

As a matter of fact, the annually ingested lead is about 0,025 mg, to be compared with the provisional tolerable intake of 91 mg/year. The associated risk corresponds to the ratio of the exposure level and the PTI, that is 0,025/91, or $2{,}8\ 10^{-4}$.

It must be noticed that the lead exposure levels resulting from other industrial facilities, the automobile circulation and natural erosion are not taken into account.

The assessment presented here is in keeping with a general safety analyse and hence, it is supposed to be conservative. The results correspond consequently to the highest expected impact values.

The uncertainties are quite wide but the assumptions made for the exposure level calculation lead to the maximum impact. If the maximum impact is acceptable, it is admitted that the safety of the repository is ensured.

PERSPECTIVES

One of ANDRA's perspectives is to apply the impact assessment methodology to the other convenient toxic elements recorded in the waste devoted to the Centre de l'Aube repository (including chemically toxic radionuclides with a long half-life, like uranium).

It is shown that the transition area, which is not taken into account within the framework of radionuclide transfer assessment, has an important influence in limiting the quantity of elements likely to reach the aquifer, the river and then the biosphere (and hence the man). Data about the solubility of the toxic elements of interest within this area have to be determined.

A major problem involves the use of a trace element calculation code to evaluate the exposure levels of the reference man to toxic elements, which are not necessarily in trace quantity.

The validation of this procedure will be done through comparison exercises with other codes specifically devoted to chemical pollutants.

Moreover, ANDRA continues its investigations regarding the chemically toxic inventory in order to limit the long-term risk for the exposed population.

The methodology presented for radioactive waste containing toxic elements is consistent with the risk assessment methodology developed in the USA in 1983 by the Sciences Academy.

REFERENCES

(1) Décret 89-3 du 3 janvier 1989 relatif aux eaux destinées à la consommation humaine à l'exclusion des eaux minérales naturelles.

(2) Circulaire du 19 juillet 1982 relative aux valeurs admises pour les concentrations de certaines substances dangereuses dans l'atmosphère des lieux de travail.

(3) Directive 96/29 Euratom du Conseil du 13 mai 1996 fixant les normes de base relatives à la protection sanitaire de la population et des travailleurs contre les dangers résultant des rayonnement ionisants.

(4) Publication n° 46, Annals of the ICRP, Radiation Protection Principles for the Disposal of Solid Radioactive Waste, Vol. 15, n°4, Pergamon Press (1985).

(5) Directive 98/83/CE du Conseil du 3/11/1998 relative à la qualité des eaux destinées à la consommation humaine

(6) B.THURET, M-O.GALLERAND, S.MATRAS, "Sorption du plomb sur différents matériaux", Revue Française de Génie Civil. Volume 3 – n°7-8/1999, 757-774 (1999).

(7) ICRP n° 23, Report of the Task Group on Reference Man, Pergamon Press (1974).

(8) Evaluation of certain food additives and contaminants: forty-first report of the Joint FAO/WHO Expert Committee on Food Additives, World Health Organisation, Geneva (1993).

(9) Annals of the ICRP Publication, Radiation Protection Recommendations as Applied to the Disposal of Long-lived Solid Radioactive Waste, Editor: J.VALENTIN, Stockholm, Sweden, Pergamon (1981)

(10) Etude d'évaluation des éléments toxiques présents dans les déchets nucléaires, Rapport de la Commission Européenne EUR 15687FR, Collection Science et technique nucléaire (1995)