

# **THE OLEN RADIUM FACILITY: ENVIRONMENTAL CONTAMINATION, IMPACT ON THE POPULATION AND SITE REMEDIATION**

**H. Vanmarcke, J. Paridaens and Th. Zeevaert**

**Nuclear Research Centre, SCK•CEN, Boeretang 200, B-2400 Mol, Belgium**

## **ABSTRACT**

The former radium facility at Sint-Jozef-Olen (1922-1969), located in the north of Belgium, was an economical success story between the two World Wars. In a few decades Union Minière (now Umicore) produced about half of the total amount of radium produced in the world (4.5 kg). Radium was widely used in medicine for radiotherapy and in the luminising industry until cheaper and more efficient radionuclides became available. The decommissioning and dismantling of the radium extraction installations was finished in 1983 with the construction of a heavily engineered storage facility for the radioactive waste.

In the vicinity of the radium facility environmental contamination with radium took place. The major contaminated sites are: the banks of a small river (Bankloop) receiving the liquid effluents, the former flooding zones of the same river, a waste deposit of 9 ha with mixed radium and chemical waste and a few streets with contaminated material underneath.

Recurring media coverage since 1989 of some high contaminations in Sint-Jozef-Olen resulted in a detailed radiological characterisation of the contaminated sites and an evaluation of their population impact. The population exposure from the contaminated river banks and streets is a few hundreds of  $\mu\text{Sv}/\text{year}$  depending largely on the selected residence times and exposure levels. External exposure from the waste deposit is at the moment prevented by a fence. The internal contamination from local foodstuffs is limited to a few tens of  $\mu\text{Sv}/\text{year}$  because there are at present no crops for direct human consumption on the contaminated farmland.

The principal motive to restore the sites is not the current exposure but the fact that most scenarios concerning the future use of the contaminated sites give rise to enhanced exposures to future generations. The worst scenario is the integration of the disposal site into a residential area. Exhaling radon gas would result in exposures of a few tens of  $\text{mSv}/\text{year}$ .

Umicore has drawn up a detailed plan for the restoration of the contaminated area in collaboration with the competent authorities. According to this plan a second radium disposal facility will be built in Olen to incorporate the scattered contamination. Only the contaminated farmland will not be cleaned up. For this area alternative land uses depending on the contamination level were investigated.

## **INTRODUCTION**

While looking for extractable copper ore, a mining engineer of Union Minière (now Umicore) found in 1915 important pitchblende deposits in the area of Sinkolobwe in the Katanga district of the former Belgian Congo. Radium production of the high-grade ore (about 50 % pure uranium oxide) at the Olen facility began in 1922 and 12 grams of radium were produced already within the first year.

The Umicore radium facility in Olen produced about half of the total radium production of the world that is estimated in an IAEA document at 4.5 kg (1). With the development of nuclear reactors and particle accelerators after the Second World War, cheaper and more efficient radionuclides became available and the interest for radium disappeared. The decommissioning and dismantling of the industrial installations were finished in 1983 with the construction of a heavily engineered storage facility for the radioactive waste: the grey area along the canal in figure 1 within the fence of the current Umicore (Union Minière) factory.

In 1989 and 1990, media coverage of some spots that were highly contaminated in the village of Sint-Jozef-Olen caused anxiety among the population. As the existing database, placed in the context of the more stringent radiation protection legislation, was not sufficient, the federal authorities decided to carry out a detailed assessment of the scattered environmental contamination (2-9).

## OVERVIEW OF THE CONTAMINATED SITES

The liquid effluents of the radium facility were discharged into the brook called Bankloop. The Bankloop is approximately 1800 m long from the fence of the factory to the mouth into the Kleine Nete river (see figure 1). The first 600 m of the brook, up to the canal is through the town centre, along the municipal school, the church and under the churchyard. Along both sides of the canal there is a blind arm of about 100 m caused by the construction in the 1980s of a new culvert under the canal. Then the Bankloop flows through a predominantly agricultural area. The lower reaches at the mouth into the Kleine Nete were frequently flooded in wintertime until the beginning of the 1960s when, as a result of soil reclamation work, the last 420 m of the brook was displaced a 100 m to the west.



**Figure 1.** Presence of radium at Sint-Jozef-Olen. Areas with enhanced radium concentrations from past disposal activities or caused by liquid discharges into the Bankloop are indicated in grey.

In the period before 1960 radium contaminated waste products were deposited on different locations in the vicinity of the former radium facility. Location D1 in figure 1 was formerly a low-lying area. Between 1955 and 1960 the difference in level was removed with residues of the cobalt production, debris of a former building of the radium production and with a limited amount of radium extraction residues. Radium contamination is also located under the present municipal (IOK) and industrial (UM) disposal site. This contamination is now buried under more than 10 m non-radioactive waste and therefore cannot be detected with a gamma detector. On different places in the village of Sint-Jozef-Olen and in the neighbouring city of Geel, radium contaminated substances were used as paving materials. Nine or ten stretches of road and several isolated points were identified during the different gamma surveys. The two most contaminated roads are shown on figure 1, namely, the *Kapellekensstraat* and the *Grensstraat*. They were restored on behalf of the municipal authorities in the late 1990s and the contaminated materials were removed to the D1 dump.

## BANKLOOP, FIRST 1400 M

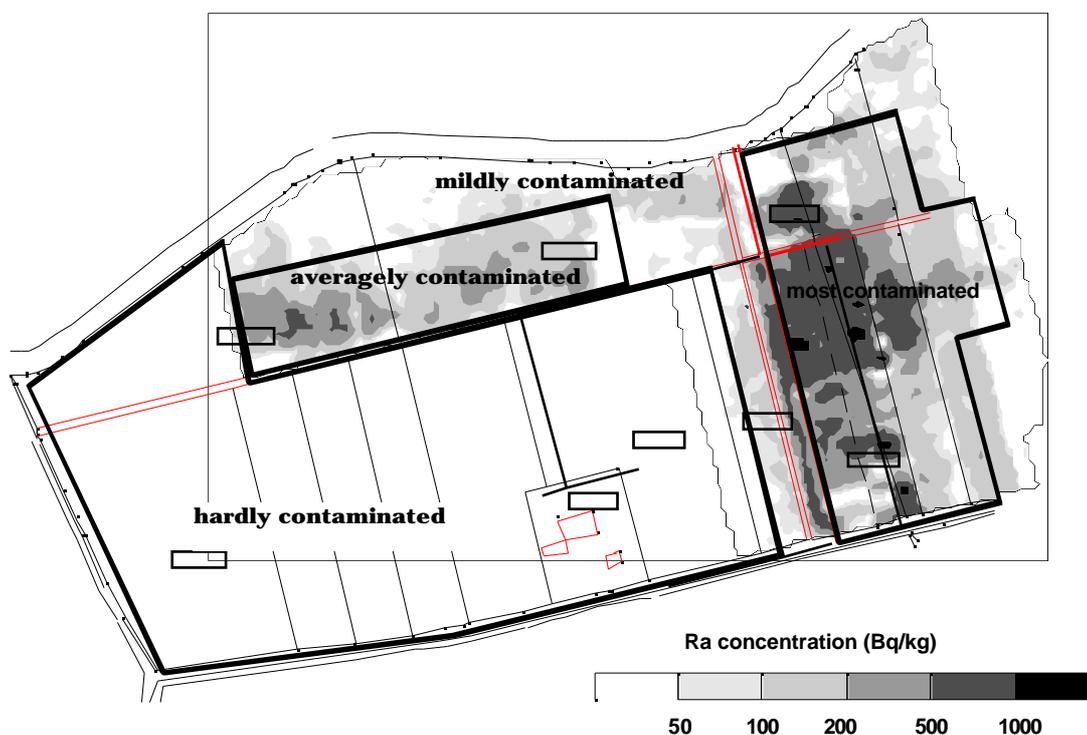
The Bankloop flows through a region that is characterised by a sandy soil and drains an area of 7.7 km<sup>2</sup>. The radium contamination of the Bankloop was mapped from the fence of the Umicore factory to the merging with the Kleine Nete river. Gamma dose rates were measured every 10 m at right angles to the Bankloop with portable detectors. Measurements were done in the middle of the brook, at the borders and at both banks every 2 m until the background level was found or obstacles prevented access. In addition, 15 borings were made along the Bankloop to gain an insight into the depth profile of the radium contamination.

Everywhere, except at the displaced mouth, enhanced dose rates were observed, mostly confined to a narrow strip of 5 to 10 m wide on one or both sides of the brook. The surface area of the contaminated banks was calculated from the gamma measurements. The most contaminated layer is often at a depth between 20 and 40 cm because most of the contamination occurred more than 50 years ago. In order not to miss contaminated ground buried under a few tens of cm of cleaner ground, the action level should be set rather low, for instance at 0.15  $\mu\text{Sv/h}$ , which is about twice the local background. The total amount of contaminated ground and sediment for this action level was estimated at 9700 m<sup>2</sup>, of which 6250 m<sup>2</sup> lies in the residential area (8).

## FORMER FLOODING ZONES OF THE BANKLOOP

Enhanced dose rates were observed in the former flooding zones of the Bankloop, which were in the beginning of the sixties transformed into farmland. The site is at present part of a large dairy farm. The contamination of an area of about 42 ha of farmland was measured by means of gamma dose rate measurements in the points of a 10 by 10 meter grid, laid over the area. From these data, soil radium concentrations were calculated (10).

The whole area was then divided into four plots taking into consideration both practical and radiological aspects (figure 2). Physical borders such as the omnipresent ditches often were taken as a practical boundary of a plot. On the other hand, the division in some cases was also based purely on differences in soil radium concentration. Finally, we ended up with average soil radium concentrations of 40, 140, 380 and 1040 Bq/kg in the four plots respectively. For practical purposes, we will refer to them as hardly contaminated (plot I), mildly contaminated (plot II), averagely contaminated (plot III) and most contaminated (plot IV).



**Figure 2.** Overview of the division into four plots of the contaminated farmland. Plots I, III and IV each consist of a single piece of land. Plot II consists of two pieces, one situated to the right of plot IV and the other between plot I and plot IV and above plot III. The total area is about 42 ha.

For each plot, relatively simple dose calculations were performed using different scenarios such as a residential one and growing of crops for human or animal consumption. The most relevant exposures were taken into account, namely radon inhalation, external gamma exposure and radium ingestion through the food chain. Table 1 shows the results of the calculated annual doses to the population in each of the different scenarios and for each of the four plots.

**Table 1.** Overview of the annual doses to members of the critical group (mSv/year), for four plots with different radium concentrations. The rejected scenarios are printed in bold italics.

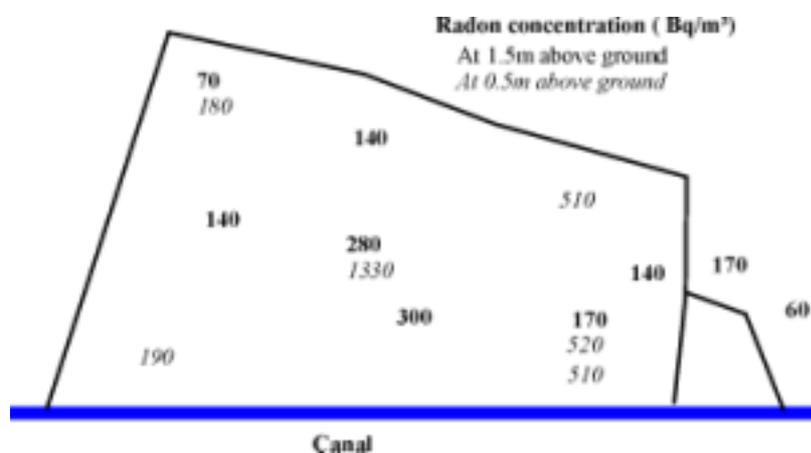
Scenario	Plot I 40 Bq/kg hardly contam.	Plot II 140 Bq/kg mildly contam.	Plot III 380 Bq/kg averagely contam.	Plot IV 1040 Bq/kg most contam.
Residential	<b>0.25</b>	<b>1.3</b>	<b>3.8</b>	<b>10.5</b>
Crops human consumption	0.035	<b>0.12</b>	<b>0.34</b>	<b>1.0</b>
Crops animal consumption	0.0008	0.003	0.007	<b>0.02</b>

The residential scenario involves a family living in a dwelling constructed on the contaminated land and growing their own crops. Radon exposure indoors is by far the most important contributor to the dose. The next two scenarios consider farming but not living on the land. The dose here is the dose to members of the public as a result of direct consumption of the crops in the first case or of indirect consumption of milk or meat for example in the second case. Notice the difference of a factor 50 between the two scenarios.

Our advise to the land owner and the authorities was to reject the residential scenario everywhere, the growing of crops for human consumption on about half of the surface (plots II, III and IV), and the growing of crops for animal consumption on about 7.5 ha (plot IV). It must be pointed out however that the latter was only due to the practical problem of physically separating some very heavily contaminated spots from the less contaminated land within the most contaminated plot. It was interesting to see that the expected doses due to milk production were fairly insignificant on all four plots, even on the most contaminated one.

### D1 DUMP

Mixed radium and chemical wastes were deposited on an area of 9 ha, designated as the D1 dump. The radon concentrations in open air on and near the D1 disposal site were measured monthly during one year with alpha track detectors. The locations and the annual average radon concentrations are indicated in figure 3. The average radon concentration at 1.5 m above ground is 180 Bq/m<sup>3</sup>, with monthly averages up to 500 Bq/m<sup>3</sup>. The radon concentration at 0.5 m is 2 to 4 times higher indicating a high exhalation rate. At a short distance of the D1 dump in the prevailing wind direction, an average radon concentration of 170 Bq/m<sup>3</sup> was measured in a garden of a residential dwelling (2,3).

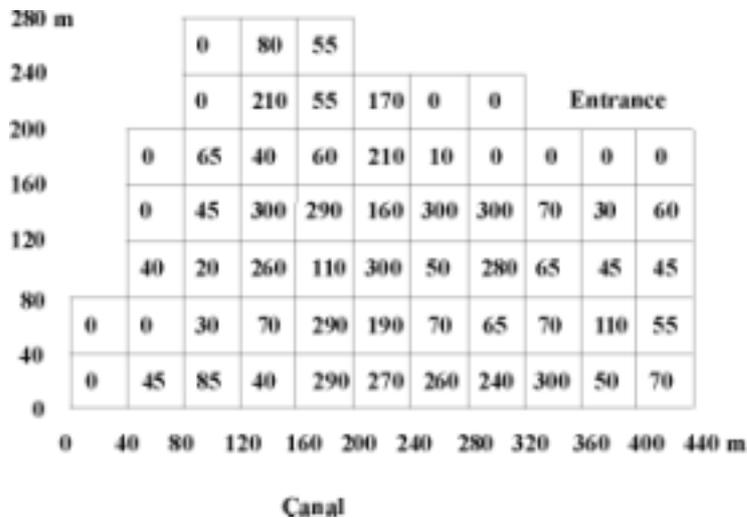


**Figure 3.** Annual average radon concentrations at and in the vicinity the D1 dump measured at 1.5 m above ground (bold) and at 0.5 m above ground (italics).

Despite the sometimes dense vegetation, gamma measurements were performed every 25 m. The maximum, mean and median values of the grid measurements were 150, 2.8 and 1  $\mu$ Sv/h. The dose rate varies often over several orders of magnitude over distances of a few metres (2,3).

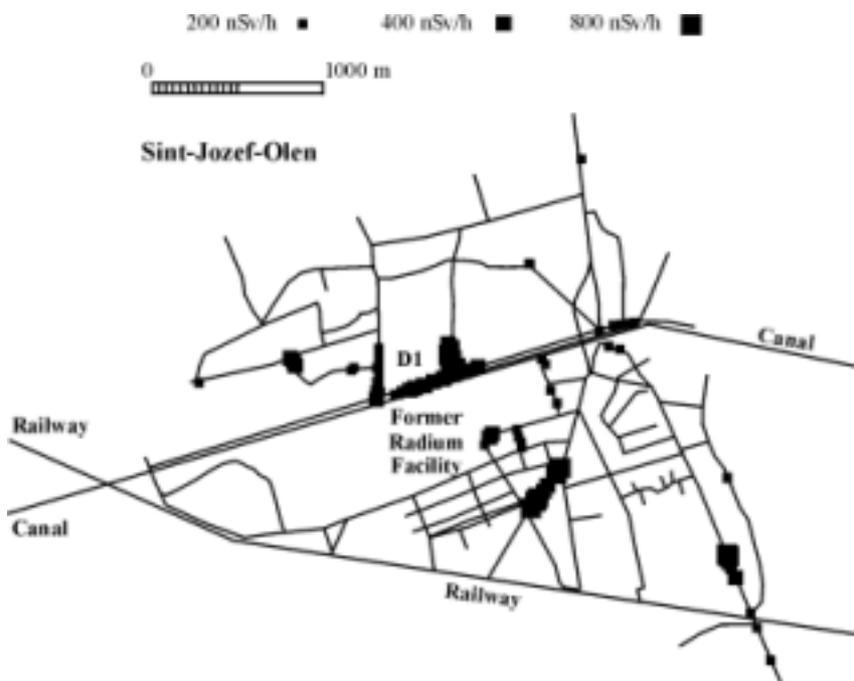
To gain an insight into the depth profile of the radium contamination of the D1 dump borings were made every 40 m, down to a depth of 3 m. The dose rate profile was measured by lowering a NaI detector in the borehole. The amount of contaminated material was calculated from the dose rate measurements assigning an area of 40 by 40 m to each measurement and selecting a threshold of 0.5  $\mu$ Sv/h. The results of the 61 borings are shown in

figure 4. From the data in figure 4, a waste volume of 101 200 m<sub>3</sub> is calculated. Rounding up the depths to the nearest half metre, for instance 170 cm becomes 2.00 m, results in a waste volume of about 120 000 m<sub>3</sub> (4,8).



**Figure 4.** The maximum depth of the contamination of the D1 dump is for each boring indicated in cm. The values are derived from the borehole measurements assuming a threshold dose rate of 0.5  $\mu$ Sv/h.

## ROADS



**Figure 5.** Map of the contamination of the roads in the vicinity of the former radium extraction plant in Olen.

All the roads of Sint-Jozef-Olen and those of some adjacent areas suspected of being contaminated were monitored with a van equipped with four NaI detectors. The results of this survey are shown in figure 5. About 5 % of the 11 000 measurements are above the investigation level of 0.2  $\mu$ Sv/h. They were subjected to a detailed foot survey with automatic hand-carts especially designed for this purpose. Every 5 m the dose rate was registered in the middle of the road and at a fixed distance to the left and the right of the middle. The volume of contaminated paving was estimated from these measurements and amounts to 1100 m<sub>3</sub> (2,8).

## SUMMARY OF THE IMPACT ON THE POPULATION

The current exposure of the population was estimated at (8):

- *external exposure* from the contaminated river banks and paving: ~ 0.1 mSv/year depending largely on the selected residence times and exposure levels (external exposure from the D1 dump was prevented a few years ago by the erection of a fence);
- *internal exposure* from local foodstuffs: ~ 0.01 mSv/year (there are at present no crops for direct human consumption on contaminated land);
- *radon exposure* in the dwelling adjacent to the D1 dump: 4 mSv/year (the fence prevents radon exposure on the D1 dump itself).

The principal argument in favour of restoring the sites is not the current population exposure, but the fact that most of the scenarios concerning the future use of the contaminated areas envisage enhanced exposures to future generations. The worst scenario is the integration of the D1 dump in a residential area. The radon concentrations in the dwellings could reach very high values resulting in doses to the residents in excess of 50 mSv/year (see table 3).

The waste volumes to be removed depend on the action levels selected. A summary of action levels and waste volumes is given in table 2.

**Table 2.** Overview of the proposed action levels and the resulting waste volumes.

Contaminated site	Action level Gamma dose rate at the surface	Waste m <sub>3</sub>
D1 dump	0.2 µSv/h, to a depth of 1 m	120 000
	0.4 µSv/h, if digging deeper than 1 metre and then covering with clean soil	+ 5 000 for vegetation
Bankloop	0.15 µSv/h	9 700
Streets	0.2 µSv/h	1 100
<b>Total (rounded)</b>		<b>135 000</b>
Chemical waste at the D1 dump		82 000

## DIFFERENT RESTORATION OPTIONS FOR THE D1 DUMP

SCK undertook a radiological assessment of five options to restore the D1 dump on behalf of Umicore (7).

1. Leaving the situation as it is.
2. In-situ restoration without digging out the waste.  
Two solutions are proposed, 2a and 2b, differing only in surface area and height of the storage facility.
3. In-situ restoration and excavation of the waste.  
Two solutions are proposed. In option 3a the chemical waste is separated from the radioactive waste and removed to the nearby industrial discharge of Umicore. In option 3b all the waste remains on site.
4. Construction of a storage facility next to the municipal and industrial discharge (500 m from the D1 dump).  
The D1 dump is completely excavated and levelled up with clean soil. The waste is transported to a nearby area, and the radioactive waste gets covered by the chemical waste. The waste is then capped with a multilayer cover and outfitted with a geo-membrane under and around the storage facility.
5. The removal of the chemical waste to an industrial discharge and of the radioactive waste to the national agency for radioactive waste (NIRAS•ONDRAF).

Options 2a, 2b, 3a, 3b, and 4 were worked out in detail, taking into account the storage of the radioactive waste of the Bankloop and roads. The proposed multilayer cover consists of a 1 m thick layer of clay to prevent water infiltration and radon exhalation, and several layers of sand, gravel and porphyry to protect the clay layer. The multilayer cover has a total thickness of 2.25 m.

## EVALUATION OF THE RESTORATION OPTIONS

The future population exposure and the exposure of the workers involved in the restoration of the D1 dump were calculated for options 1 through 4. The calculation of the population exposure is not possible in option 5 because this depends on the final destination of the radioactive waste. The maximum dose received by a member

of the critical group is calculated for the normal exposure scenario and for some human intrusion scenarios after an institutional control period of 200 years. The parameters for the calculation of the different scenarios are given in (7). The results for the population exposure are summarised in table 3.

**Table 3.** Maximum doses to members of the critical group for the normal and human intrusion scenarios. Only the 1 m thick clay layer of the multilayer cover was assumed to be present in options 2 through 4.

Option	Normal evolution scenario		Construction scenarios			Residential scenario mSv/year
	Well mSv/year	Radon mSv/year	Dwelling mSv	Apartment mSv	Road * mSv	
<b>1</b>	0.00077	2.0	1.8	2.2	1.8	56
<b>2a</b>	0.00013	0.2 **	0.6	1.8	1.5	50
<b>2b</b>	0.00013	0.2 **	0.6	1.8	1.5	50
<b>3a</b>	0.00063	0.3 **	1.1	2.9	2.3	79
<b>3b</b>	0.00051	0.2 **	0.6	1.8	1.5	51
<b>4</b>	0.00003	0.0	0.02	0.8	1.3	< 14 **

\* construction of a road sunk halfway down into the tumulus

\*\* conservative value

The population exposure for the normal evolution scenario for a well at 100 m from the storage facility is given in table 3. They are orders of magnitude lower than the exemption level for practices of 0.01 mSv/year. After 500 years, a certain degradation of the clay cover is assumed and radon gas can escape the storage facility, except in option 4 where the chemical waste prevents the radon exhalation. The radon doses to people living at 50 m from the D1 dump are lower than 1 mSv/year, apart from option 1 (the present situation).

The dose impact of the intrusion scenarios is important, especially the radon exposure in the residential scenario. The calculated doses to the people living above the tumulus are higher than the current dose limit for radiation workers because the foundation of the dwellings penetrates the clay layer and reaches the radioactive waste. The radioactive waste in option 4 is located under the chemical waste so that the radon exhalation depends on the unknown radon transport properties of the chemical waste.

The collective exposure of the intervention workers is presented in column 4 of table 4. The collective dose in option 5 is an order of magnitude higher because of the need to pack the radioactive waste according to the specifications of the national agency for radioactive waste.

The criteria considered for the selection of the restoration option for the D1 dump are summarised in table 4. Options 1 and 5 can be eliminated. Option 1, leaving the situation as it is, is difficult to accept on the basis of the current radon exposure of the people living close to the D1 dump and the results of the intrusion scenarios. Option 5, packing the radioactive waste and sending it to the national agency for radioactive waste, is rejected for radiological reasons because of the high collective exposure of the intervention workers and for economical reasons (excessive costs).

The evaluation of the relevant criteria for the remaining options (2a, 2b, 3a, 3b and 4) gives similar results with regard to the cost, the surface area of the storage facility and the collective dose of the intervention workers, but very different results for the residential scenario. Only option 4 seems to be satisfactory for this criterion because of the reduced radon exhalation. Option 4 has some further advantages. The displacement of the waste to an area adjacent to the municipal and industrial disposal sites allows for a reduction of the surface area of the storage facility by increasing its height. The probability for human intrusion is in fact proportional to the surface area. The whole disposal area, consisting of the municipal, the industrial and the radium repositories will be a striking element in the landscape, limiting the probability for inadvertent human intrusion because of its magnitude. For these reasons, option 4 was selected as the best choice for the restoration of the D1 dump.

**Table 4.** Comparison of the restoration options for the D1 dump (in 1995 Euros).

Option	Cost of the facility	Surface area of the facility	Intervention workers Collective dose	Residential scenario Individual dose
--------	----------------------	------------------------------	---	---

	10 <sup>6</sup> Euro	m <sub>-</sub>	man.mSv	mSv/year
<b>1</b>	0	91 200	0	56
<b>2a</b>	** 6.77	46 500	63	50
<b>2b</b>	** 5.83	31 000	83	50
<b>3a</b>	** 6.92	24 000	150	79
<b>3b</b>	** 8.58	46 400	138	51
<b>4</b>	** 6.79	28 800	151	< 14
<b>5</b>	> 25	*	1 096	*

\* dependent on the final destination of the radioactive waste

\*\* radiological protection costs and chemical waste treatment costs are not included

Umicore has made a scenario in 2001, called project BREAM, in consultation with the competent authorities to implement the global restoration plan. The complete restoration and the transfer of the storage facility to the national agency for radioactive waste (NIRAS•ONDRAF) is planned for 2010.

## REFERENCES

1. IAEA. Nature and magnitude of the problem of spent radiation sources. IAEA-TECDOC-620 (1991).
2. E. Cottens, H. Vanmarcke, et al. Onderzoek naar de verspreiding van Ra-226 in het leefmilieu te Sint-Jozef-Olen en omgeving en de daaruit voortvloeiende dosisbelasting voor de bevolking. SCK-IHE report on behalf of DBIS•SPRI, Brussels (1993) (in Dutch).
3. E. Cottens, H. Vanmarcke, et al. Remediation and restoration of a thorium and radium contaminated site. p. 263-279 in: Remediation and restoration of radioactive-contaminated sites in Europe. Proc. Int. Symposium, Antwerp, 1993. European Commission publication N° XI-5027/94 (1994).
4. M. Loos, H. Vanmarcke, et al. Inventarisatie van de radiumbesmetting van de D1-stortplaats. SCK report on behalf of Umicore (Union Minière), Mol (1994) (in Dutch).
5. H. Vanmarcke, P. Geuzens and E. Vangelder. Chemische en radiologische karakterisatie van het diepteprofiel van de Bankloop te Sint-Jozef-Olen. SCK-VITO report on behalf of DBIS and OVAM, Mol, part 1 (1994); part 2 (1995) (in Dutch).
6. HEACON. Harbour and engineering consultants. Sanering van stortplaats V (D1). Report on behalf of Umicore (Union Minière), N° OUS1461 00008, (1995) (in Dutch).
7. Th. Zeevaert, L. Sweeck, et al. Radiologische impactstudie van de D1-stortplaats. SCK report on behalf of Umicore (Union Minière), R-3098, Mol (1996) (in Dutch).
8. H. Vanmarcke. Sanering van de omgevingsbesmetting met radium-226 te Olen en Geel. SCK report on behalf of DBIS, R-3179, Mol (1997) (in Dutch).
9. H. Vanmarcke and Th. Zeevaert. Restoration of the areas environmentally contaminated by the Olen radium facility. p. 517-539 in: Restoration of Environments with Radioactive Residues. Proc. STI/PUB/1092. IAEA, Vienna (2000).
10. J. Paridaens and H. Vanmarcke. Evaluatie van de radiologische risico's verbonden aan het gebruik van de met radium-226 besmette gronden langs de Bankloop gelegen ten noorden van de Roerdompstraat en ten zuiden van de Kleine Nete in Olen en Geel. SCK report on behalf of VLM, R-3637, Mol (2002) (in Dutch).