ISSN - 0250 - 5010

ANNALEN VAN DE BELGISCHE VERENIGING VOOR STRALINGSBESCHERMING

==========

VOL. 21, N° 4 4e trim. 1996

IRPA 9

BELGISCHE BIJDRAGE

PARTICIPATION BELGE

Driemaandelijkse periodiek
2400 MOL 1

Périodique trimestriel
2400 MOL 1

==========

ANNALES DE L'ASSOCIATION BELGE DE RADIOPROTECTION

Hoofdredacteur Dr M.H. Faes Rédacteur en chef
Fazantendreef, 13
B- 3140 Keerbergen

Redactiesecretariaat Mme Cl. Stiévenart Secrétaire de Rédaction
Av. Armand Huysmans 206, bte 10
B- 1050 Bruxelles - Brussel

Van 14 tot 19 april 1996 vond te Wenen het negende congres van de IRPA (International Radiation Protection Association) plaats. Verschillende landgenoten hebben op dit congres een poster voorgesteld. Het Bureau van de Belgische Vereniging voor Stralingsbescherming heeft besloten om de uitgewerkte teksten van deze posters te bundelen in dit nummer van haar Annalen.

Du 14 au 19 avril 1996 a eu lieu à Vienne le neuvième congrès de l'IRPA (International Radiation Protection Association). Plusieurs Belges y ont présenté un poster. Le Bureau de l'Association belge de Radioprotection a décidé de grouper les textes élaborés de ces posters dans le présent numéro de ses Annales.

Inhoud **Sommaire**

Annales de l'Association belge de Radioprotection,
Annalen van de Belgische Vereniging voor Stralingsbescherming, vol.21, n°4, 1996
pp 317-321

EXHALATION OF RADON AND THORON FROM PHOSPHOGYPSUM USED AS BUILDING MATERIAL

Hans Vanmarcke

SCK-CEN, Nuclear Research Center,
Boeretang 200, 2400 Mol, Belgium

ABSTRACT

The radioactive properties of two types of phosphogypsum, were determined. Gypsum plates with different thickness were produced. The ^{226}Ra and ^{232}Th concentrations were measured by means of high resolution gamma spectrometry. The results are for type 1 ^{226}Ra : 75 Bq/kg and ^{232}Th : 230 Bq/kg and for type 2 ^{226}Ra : 155 Bq/kg and ^{232}Th : 160 Bq/kg. The radon (^{222}Rn) exhalation rate was evaluated by closing the plates in air-tight barrels and measuring the radon concentration. The exhalation rate of type 1 is 1.2 10^{-5} Bq/(kg s) and type 24.7 10^{-5} Bq/(kg s).
In combination with the ^{226}Ra concentration an emanating fraction of respectively 7.6% and 14% is obtained.
The ^{220}Rn (thoron) exhalation of the plates was determined by measuring the concentration of the decay products in a chamber of 1 m^3 with normal aerosol concentrations. The exhalation rate was found to be independent of the thickness of the plates, as expected from the short half life of ^{220}Rn Covering the entire surface of the plates with two layers of a common Latex paint decreased the thoron exhalation by a factor of 10 to 20.

The laboratory results for the radon and thoron exhalation were converted using realistic assumptions for a room. The contribution of phosphogypsum to the average radon concentration in a room is found to be about 1 Bq/m^3 for type 1 and 4 Bq/m^3 for type 2 resulting in an annual effective dose of the order of 0.1 mSv/year. The contribution to the effective dose from the thoron exhalation is much greater, namely, 1.8 mSv/year for type 1 and 0.9 mSv/year for type 2. Painting the gypsum lowers the thoron dose by a factor of 10 to 20 making the thoron dose comparable to that of radon.

INTRODUCTION

Gypsum is widely used in Belgium as a building material. The walls in most dwellings are covered with a layer of 1 to 2 cm of gypsum before they are painted or wallpapered. A considerable fraction of the gypsum applied in Belgian dwellings comes from the phosphate industry. The reaction of phosphate ore with sulfuric acid yields a variety of phosphate products and calcium-sulfate (gypsum). On average 4 kg of gypsum are produced for each kg of phosphoric acid (1). The phosphate ore has generally a high content of ^{238}U and/or ^{232}Th in radioactive equilibrium with its daughter products. Most of this radioactivity will be found in the gypsum waste which is

called phosphogypsum.

The paper consists of two parts. In the first part the radioactive properties of two types of phosphogypsum are determined. Results are given concerning the ^{226}Ra and ^{232}Th concentration, the radon (^{222}Rn) exhalation rate and the thoron (^{220}Rn) exhalation with and without painting. The radiological impact of using these types of phosphogypsum in home construction are discussed in the second part.

METHODS AND RESULTS

From each of the two types of phosphogypsum 60 plates with a surface of 0.295 x 0.210 m were made. Half of the plates was produced with a thickness of 0.5 cm and the other half with a thickness of 2 cm.

The ^{226}Ra and ^{232}Th concentrations of the two types of phosphogypsum were measured by means of high resolution gamma spectrometry. The detector of the measurement system is a 20% efficient germanium cristal with a resolution of 1.70 keV (fwhm). The detector is calibrated in the same geometry with a ^{134}Cs source traceable to NIST. The results for the two types are:
Type 1 : ^{226}Ra : 75 Bq/kg ^{232}Th : 230 Bq/kg
Type 2 : ^{226}Ra : 155 Bq/kg ^{232}Th : 160 Bq/kg
The radium concentrations are lower and the thorium concentrations are higher than those found in phosphogypsum from Moroccan origin (1).

The radon (^{222}Rn) exhalation rate was measured by closing some of the plates in an airtight barrel flushed previously with radon free air. One week or more later, the radon concentration in the barrel was detennined by means of the Lucas technique. From the dimensions of the barrel the weight of the sample and the radon concentration the exhalation rate was calculated:
Type 1 : 1.2 10^{-5} Bq/(kg s)
Type 2 : 4.7 10^{-5} Bq/(kg s)
In combination with the ^{226}Ra concentration an emanating fraction of 7.6 % for type 1 and 14% for type 2 is obtained.

The thoron exhalation (^{220}Rn) of the plates was evaluated by means of its decay products because of the short half life of thoron (55.6 s). The plates were brought into a 1 m^3 chamber which was ventilated continuously at a rate between 0.3 to 0.4 h^{-1} with outside air containing natural aerosol particles. The outside air was taken at a height of 10 m where the thoron and thoron decay product concentrations are very low. Each sample was measured 3 to 4 times. The reproducibility of the equilibrium equivalent thoron concentration was about 25% due to fluctuations in the aerosol concentration in the chamber. The equilibrium equivalent thoron concentration (EEC_{Th}) is calculated from the ^{212}Pb and ^{212}Bi concentrations in the following way :
EEC_{Th} = 0.91 C (^{212}Pb) + 0.09 C (^{212}Bi)
The results given in table 1 are normalised to an exhalation surface of 4 m^2 and corrected for

the influence of humidity on the exhalation of the freshly produced gypsum plates.

TABLE 1 . Equilibrium equivalent thoron concentration of the phosphogypsum plates with and without painting in the experimental chamber

Type	**Thickness**	**Not painted EEC_{Th} Bq/m^3**	**Painted EEC_{Th} Bq/m^3**	**Ratio Not painted / Painted**
1	0.5	35	2.2	16
1	2.0	25	2.3	11
2	0.5	14	0.7	20
2	2.0	12	1.0	12

The thoron exhalation is, as expected from the short half life of ^{220}Rn depended of the thickness of the samples. The exhalation of type 1 is about two times higher than that of type 2.
This is more or less in line with the rneasured ^{232}Th concentrations.

The effect of covering the entire surface of the plates with two layers of a common Latex paint was also investigated. The set-up was identical to the measurements of the un-painted plates. The measured equilibrium equivalent thoron concentrations are shown in table 1. The reduction in thoron exhalation by painting the gypsum plates is a factor of 10 to 20.

DISCUSSION

In order to evaluate the radon and thoron exhalation of the phosphogypsum plates, the laboratory results were converted using realistic assumptions for a room:

- size of the room : 4 x 5 x 3 m (60 m^3);
- ventilation rate : 0.5 h^{-1} ;
- walls and ceiling covered with type 1 or type 2 phosphogypsum (74 m^2);
- residence time : 80%.

Covering the walls and the ceiling of the room with 1 cm of phosphogypsum increases the radon concentration of the room with 1 Bq/m^3 for type 1 and 4 Bq/m^3 for type 2 resulting in a contribution to the effective dose of the order of 0.1 mSv per year.

In the calculation of the effective dose from the thoron exhalation the conversion factor between the equilibrium equivalent thoron concentration and the effective dose from ICRP publication 50 (2) is taken: 3.9 10^{-5} mSv/(Bq h/m^3). The effective dose from the thoron exhalation of the uncovered phosphogypsum is much greater than the one from the radon exhalation, namely, 1. 8 mSv/year for type 1 and 0.9 mSv/year for type 2. Painting the gypsum reduces the thoron exhalation by a factor of 10 to 20 making the thoron dose comparable to that of radon

REFERENCES

1 . L.H. Baetsle, Proceedings International Symposium on Remediation and Restoration of Radioactive-contaminated Sites in Europe, CEC, Doc. XI - 5027/94, Radiation Protection-74, 233 - 252 (1993).
2. ICRP Publication 50, Annals of the ICRP 17, N° 17 (1986).

Annales de l'Association belge de Radioprotection
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.21, n°4, 1996
pp 323-328

THE MEASUREMENT OF ^{241}Am IN THE BODY WITH A DOUBLE LOW ENERGY GERMANIUM DETECTOR SYSTEM

J.L. Genicot[1], J.P. Culot[2], F. Hardeman[1], J.P. Alzetta[1], H. Vanmarcke[1]

[1] SCK•CEN - Boeretang 200, 2400 Mol Belgium
[2] A.V. Nuclear - Avenue du Roi 157, 1190 Bruxelles Belgium

ABSTRACT

A system based on two low energy germanium detectors has been calibrated for the investigation of internal contamination with ^{241}Am. The two detectors have a total active area of 5775 mm^2 and a resolution of 600 eV at the energy of 59.5 keV. The measurement system was calibrated with the realistic torso phantom of Livermore which was obtained from the IAEA. The detection efficiency was determined in real measurement conditions with the two detectors above the right lung. Assuming a standard tissue thickness of 25 mm, a detection efficiency of 0.083 cpm/Bq was found. For a counting time of 50 minutes this corresponds to a detection limit of 6 Bq of ^{241}Am. Applying the new ICRP lung model and assuming a typical Pu (MOX) mixture with 1.2 % (w) ^{241}Am, an inhalation corresponding to an internal dose of 20 mSv is measurable 30 days after the intake for a class M compound. In case the americium of the Pu(MOX) mixture can be classified as slow (S), a single intake corresponding to a dose of 20 mSv is detectable over a period of about six months. The subject specific background is evaluated from a background prediction region above the peak region. This system allows a fast response in case of accident. The detectors are also calibrated for the measurement of americium and plutonium in the liver, for uranium and americium in the tracheo-bronchial lymph nodes and for plutonium in the lungs.

INTRODUCTION

The in-vivo measurement of lung contamination with actinides has always been a difficult task because the low energy X-rays emitted by most of the nuclides are strongly absorbed by the tissues. For up two years now, a system using two low energy HPGe detectors has been used in the SCK•CEN for this type of measurements. This system greatly improves the capabilities for quantitative assessment of actinides in the lungs, the liver and the tracheo-bronchial lymph nodes.

The two crystals having a total area of 5775 mm^2 are placed over the right lung and positioned with an angle between them of about 30 degrees. The technique is based on the quantification of ^{241}Am associated with plutonium in MOX fuel particles. With this system, the detection limit is greatly reduced to values

between 6 and 10 Bq ^{241}Am in the lungs for a 50 minute acquisition time. The counting room is made of steel plates 200 mm thick lined with thin plates of Pb (3.2 mm), Sn (1. mm) and Fe (0.5 mm).

CALIBRATION

The efficiency of the counting system has been determined with the aid of a torso phantom of the Livermore type (lent by I.A.E.A.). The calibration is made for the assessment of ^{241}Am and of pure ^{239}Pu in lungs, in liver and in tracheo-bronchial lymph nodes. Different overlayers were used for the simulation of the different morphological types. These measurements show an important effect of the tissue thickness, tissue composition and distance detector-skin on the total efficiency of the system. An increase of 20 mm in the distance torso-detector results in a decrease of the efficiency of about 20%. The efficiency of our system with two detectors is 0.083 cpm/Bq. For a counting time of 50 minutes this corresponds to a detection limit [1] of 6 Bq of ^{241}Am for a man with a tissue thickness of 25 mm. The influence of the tissue thickness on the counting rate is shown in figure 1.

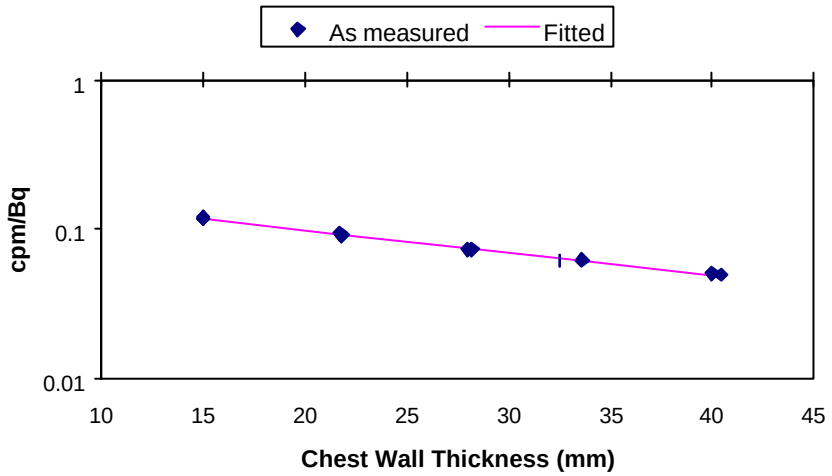

Figure 1: Decrease in counting rate with increasing chest wall thickness of a dual germanium system for the assessment of ^{241}Am in the lungs. The full line is obtained by a least square fitting including all observed values.

ANALYSIS PROCEDURE

A dispersion of 0.2 keV/channel is used for the counting. The examined zone between 59.2 and 60.0 keV, is compared to the zone between 61.4 and 64.2 keV used for the background calculation. The zone chosen for the background assessment is normally free of photopeaks in case of contamination by another radionuclide so that false negative results are minimised. Uranium, in case of contamination with mixed oxide fuel particles, because of the relative proportion of each element, will produce a negligible contribution in the 63 keV region as compared with ^{241}Am.

APPLICATION

A common bioassay tool for Pu(MOX) workers is chest counting for the ^{241}Am progeny of ^{241}Pu. An estimation of the intake requires working assumptions about the isotopic composition of the inhaled mixture and the lung's clearance dynamics.

Many factors have to be considered:

Particle size of the material: From a recent survey of the literature (Dorrian and Bailey, 1995) an AMAD of 5 µm and a median geometric standard deviation of 2.5 are realistic default values for occupational exposure when the particle size distribution is unknown.

Chemical form of the source material: The ICRP-30 and ICRP-68 designate all form of ^{241}Am as class W or class M material. However, a minor contaminant imbedded in a much more predominant matrix may exhibit behaviour characteristics of the matrix. It could be assumed that the ^{241}Am is of class Y or S such as the plutonium in the Pu(MOX) matrix.

Metabolic retention and dosimetric model: The new ICRP-66 lung model and ICRP-67 systemic model for Pu and Am provide an accurate basis for the retention and dose assessments. It will be noticed that ICRP-68 use a default value of 3. for the density.

In this work, a more realistic density of 10 is used, keeping all the other parameters of ICRP-68 (e.g. breathing rate of 1.2 m^3/h, resting for 31.3 % of the time and light work for 61.7%, size factor of 1.5).

Isotopic composition: In this work, a standard isotopic composition (in weight) is assumed: 1.2% ^{238}Pu, 61.1% ^{239}Pu, 23.5 % ^{240}Pu, 8.8% ^{241}Pu, 4.2% ^{242}Pu and 1.2 % ^{241}Am.

The doses due to all isotopes of the mixture vary with the solubility class when 1 Bq ^{241}Am is inhaled:

AMAD	1 µm	5µm	
Class M	0.23	0.13	mSv/Bq (Am)
Class S	0.20	0.11	mSv/Bq (Am)

Defining Q(20) as the measured activity in the lung that gives an effective dose of 20 mSv when we include the contribution of all the mixture:

Q(20)(Bq) = (20 mSv/Dose coefficient (mSv/Bqs Am))• Fraction remaining in the lung.

we found:

	^{241}Am class S (Bq)	^{241}Am class S (Bq)	^{241}Am class M (Bq)	^{241}Am class M (Bq)
Time (d)	1µm	5 µm	1 µm	5 µm
0.5	16.4	12.9	12.8	9.8
1	16.0	12.5	12.4	9.5
2	15.6	12.2	12.1	9.2
3	15.5	12.1	11.9	9.1
4	15.3	11.9	11.8	8.9
5	15.2	11.8	11.6	8.8
6	15.1	11.7	11.5	8.6
7	14.9	11.6	11.3	8.5
15	14.0	10.7	10.2	7.6
30	12.6	9.4	8.5	**6.2**
45	11.6	8.5	7.3	5.2
90	9.8	7.1	4.9	3.5
180	8.3	**6.0**	2.7	1.9
360	6.9	5.0	0.9	0.6

This table shows that, for a class S and 5 µm americium, an effective dose of 20 mSv could be assessed up to 180 days after an acute intake (inhalation), when the detection limit is of 6 Bq. The situation is less favourable for a class M compound but better for an older Pu(MOX) mixture.

REFERENCES

1) **Currie L.A.** *Limits for quantitative detection and quantitative Determination. Application to Radiochemistry.* Anal. Chem. 40, 586, 1968.

2) **Spitz HB, Bushbom RL, Rieksts GAA, Palmer HE.** *A New Method for Analysing High Resolution Spectra from Whole Body Counter in-vivo Meas.* Health Phys. V49, N°6. 1985.

3) **Summerling T.J., Quant S.P.** *Measurement of the human anterior chest wall by ultrasound and estimates of chest wall thickness for use in determination of transuranic nuclides in the lungs.* Radiation Protection Dosimetry, Vol.3, N° 4 pp. 203-210 (1982).

4**) Dorrian M.D. and Bailey M.R.** *Particle size distribution of radioactive aerosol measured in workplaces.* Radiation Protection Dosimetry Vol. 60, pp 119-133 (1995).

Annales de l'Association belge de Radioprotection
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.21, n°4, 1996
pp 329-337

FOLLOW UP OF THE CHERNOBYL ^{137}Cs CONCENTRATION IN BELGIUM BY MEANS OF IN SITU GAMMA SPECTROMETRY

J. Uyttenhove[1], S. Pommé[2], B. Van Waeyenberge[3], F. Hardeman[2], J. Buysse[3] and J.-P. Culot[4]

[1]University Gent, Physics Lab, Krijgslaan 281 (S-12), B-9000 GENT, Belgium
[2]Belgian Nuclear Research Centre, SCK-CEN, Boeretang 200, B-2400 MOL, Belgium
[3]University Gent, Nuclear Physics Lab, Proeftuinstraat 86 B-9000 GENT, Belgium
[4]A.V. NUCLEAR, Av. du Roi 157 B-l060 BRUSSELS, Belgium

Abstract

The residual radiocesium concentration, nearly 10 years after the Chernobyl accident, is measured at different sites on the Belgian territory by means of in-situ γ-spectrometry. A possible link with the rainfall at the beginning of May 1986 is investigated.

INTRODUCTION

On Friday, May 2, 1986 a cloud containing radioactive particles from the Chernobyl accident reached Belgium during the morning. At noon, an activity in the air of 58 Bq/m^3 was measured in Gent. Together with short living isotopes ^{131}I, ^{l32}Te, etc.) a concentration of 5 Bq/m^3 of the long living ^{137}Cs was registered (half live 30 y). One day later, on Saturday, May 3, the air activity was already reduced to a few percent of this peak value.
The aim of this research is to evaluate the situation concerning the residual radiocesium concentration in different parts of Belgium nearly 10 years after the Chernobyl accident by means of *in-situ g*-spectrometry.

EXPERIMENTAL PROCEDURE

The in-situ measurement technique for environmental radiation provides a means for the complete characterisation of the gamma radiation field at a given location. The *in-situ g*-spectrometry technique using efficient high resolution Ge-detectors allows a fast, accurate and sensitive determination of radionuclides in the soil. Also a better soil representativity can be achieved than with sample collection and subsequent laboratory analysis. Indeed, an unshielded detector placed at about one meter above the ground detects gamma rays from an area of at least a 10 meter radius, representing a huge volume of soil compared to the typical soil sample. The method is described by Beck et al. (1) and more recently by Miller et al. (2,3). The ICRU report 53 (4) gives a complete

review on the subject.

In this work, the measurements were performed with two different measuring chains, one of the University Gent (RUG) and the other of the SCK-CEN (Mol). The RUG detector is a 20.2% p-type (energy resolution 1.7 keV) with a normal 30 litre dewar. The SCK chain uses an n-type HPGe coaxial detector (resolution 1.83 keV and efficiency 10%) mounted on a small multi-attitude cryostat. Both spectrometers were calibrated independently and intercompared during two in-situ measurements at locations in Mol and Tihange. All results are in good agreement within the total experimental error of about 15%. Laboratory analyses at the SCK-CEN of soil samples confirm the in-situ data. Details about the calibration and the intercomparison are given in internal reports (5, 6).

Figure 1 gives a typical spectrum of a measurement with the RUG-detector in a high activity region; one can easily see the 662 keV ^{137}Cs line together with the 1461 keV ^{40}K line and the typical lines of the natural U and Th series. A measuring time between 30 and 60 minutes allows an accuracy better than 15% at the actual remaining ^{137}Cs concentrations from Chernobyl in Belgium (typical values from .5 to 5 kBq/m^2). Even a quick 10 minutes test measurement can give a good indication on the concentration.

After some preliminary measurements in Gent, Mol and the Ardennes, a systematic survey of the Belgian territory (30507 km^2) by in-situ gamma spectrometry has been carried out. About 55 measurements, equally distributed across the country were executed. The western part was covered by the RUG-team and the eastern part mainly by the SCK-CEN team. For all measurements, pastures that we know or consider as undisturbed during the last 10 years were selected. The penetration of fallout in the soil should therefore merely depend on natural processes.

A drawback of the method is its dependence on the assumption made on the depth distribution of fallout in the soil. In the calculations of the ^{137}Cs concentration we use an exponential depth profile with relaxation parameter α = 20 m^{-1} (equivalent with a relaxation lenght of 5 cm). This value was derived at the SCK-CEN from the comparison of the in-situ measured ^{137}Cs surface activity, S_A, with the mean activity S of soil samples (depth D=10 cm) taken at the same location :

$$S = \frac{1 - e^{-D\alpha}}{\rho . D} . S_A$$

In fact, the appropriate relaxation parameter is found by varying the α-value in the calculation of the surface activity, until the right hand side of the expression matches the left hand side. A typical value was deduced from measurements at 23 locations. Our actual choice of α = 20 m^{-1} complies with the data for aged fallout (table 3.5 in (4)). This relaxation parameter is rather critical for the calculations of the ^{137}Cs concentration; with α = 33.33 m^{-1} as used in earlier measurements (5,6) the values given in fig. 2 should be reduced by a factor 1.356. By using a fixed value, we ignore the existing variations on the different locations. This is however not critical on the interpretation of our data, in view of the strong ^{137}Cs concentration variations from one location to another.

RESULTS AND INTERPRETATION

Figure 2 shows a map of Belgium, indicating the ^{137}Cs surface activity (Bq/m^2) at the different measurement locations (assuming $l/\alpha = 5$ cm). Whereas the west side of the country shows very low concentrations, higher concentrations areas can be found on the east side. A peak value of 5600 Bq/m^2 was obtained near Sint-Hubert. The occurrence of regions with higher concentration coincides roughly with the zones of higher deposition as mentioned by Govaerts et al. (7).

In figure 3, a contour map was created from our database with the Kriging gridding method using SURFER for Windows (8). The numbers on the axes are the Lambert co-ordinates, a local system widely used in Belgium; the country is situated between 49.5 and 51.5 N and 2.5 and 6 E. We have also drawn contour maps of the rainfall in the critical days of May 1986, based on the rainfall data from 270 observation points obtained from the Royal Meteorological Institute (KMI, Ukkel). Only on May 3, a considerable rainfall occurred in (the eastern part of) Belgium, as is shown in figure 4.
By comparing the maps of fig. 3 and 4 some overall correlation can be found between the rainfall and the residual ^{137}Cs concentration, but the maximum values do not coincide. This can be explained by the fact that the rainfall data were integrated over the whole day, whereas a considerable variation in the air activity must have occurred.

Some discrepancies between neighbouring points can possibly be caused by the variations in the use of the grassland. In spite of our careful selection of the measuring sites, it can not be completely excluded that some pastures have been ploughed or modified in some way, disturbing the natural distribution of the Cs.

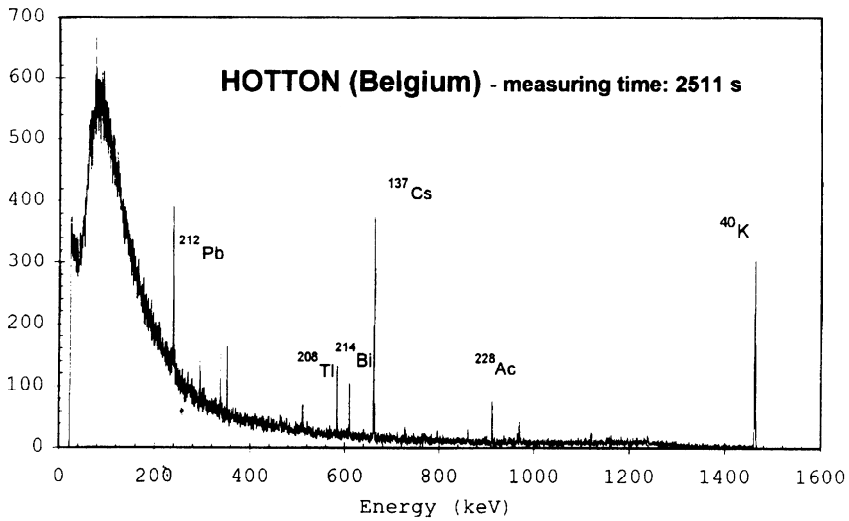

Figure 1 - A typical in-situ gamma spectrum measured in the Hotton region (Ardennes). The ^{137}Cs concentration is about 4600 Bq/m^2.

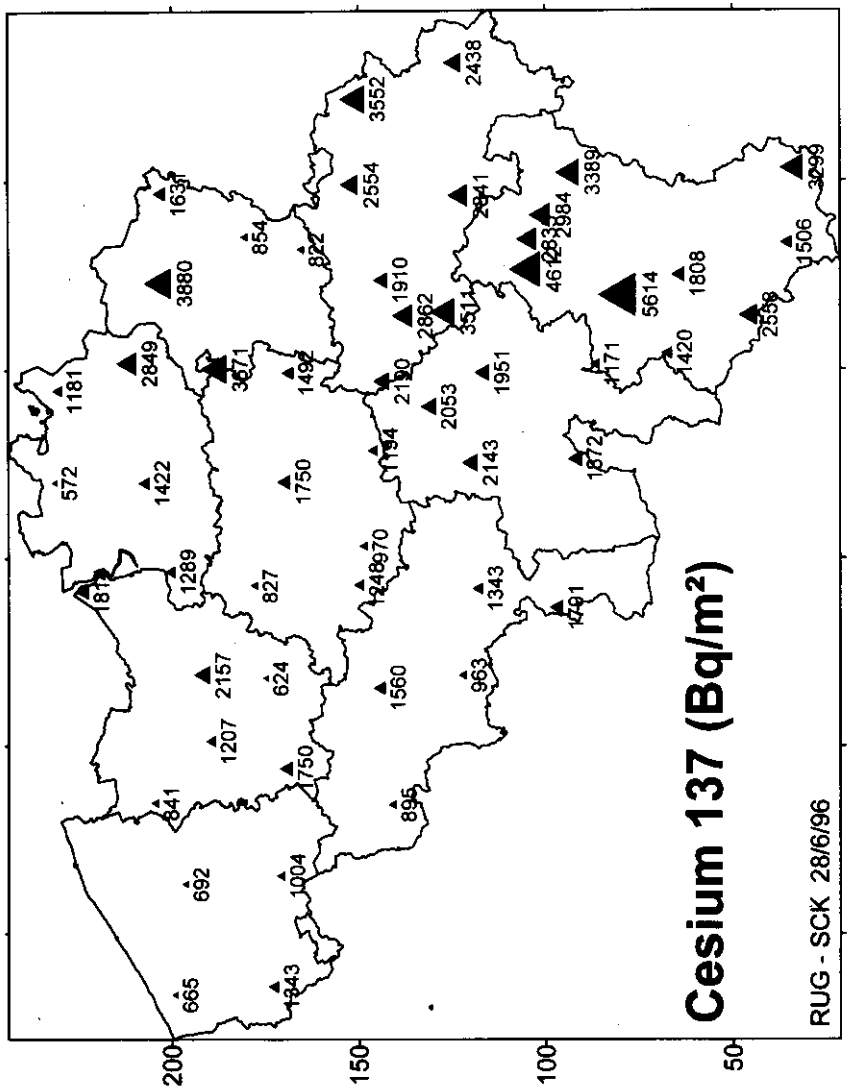

Fig.2 Map of Belgium with the ^{137}Cs concentrations (in Bq/m^2) at the measuring points

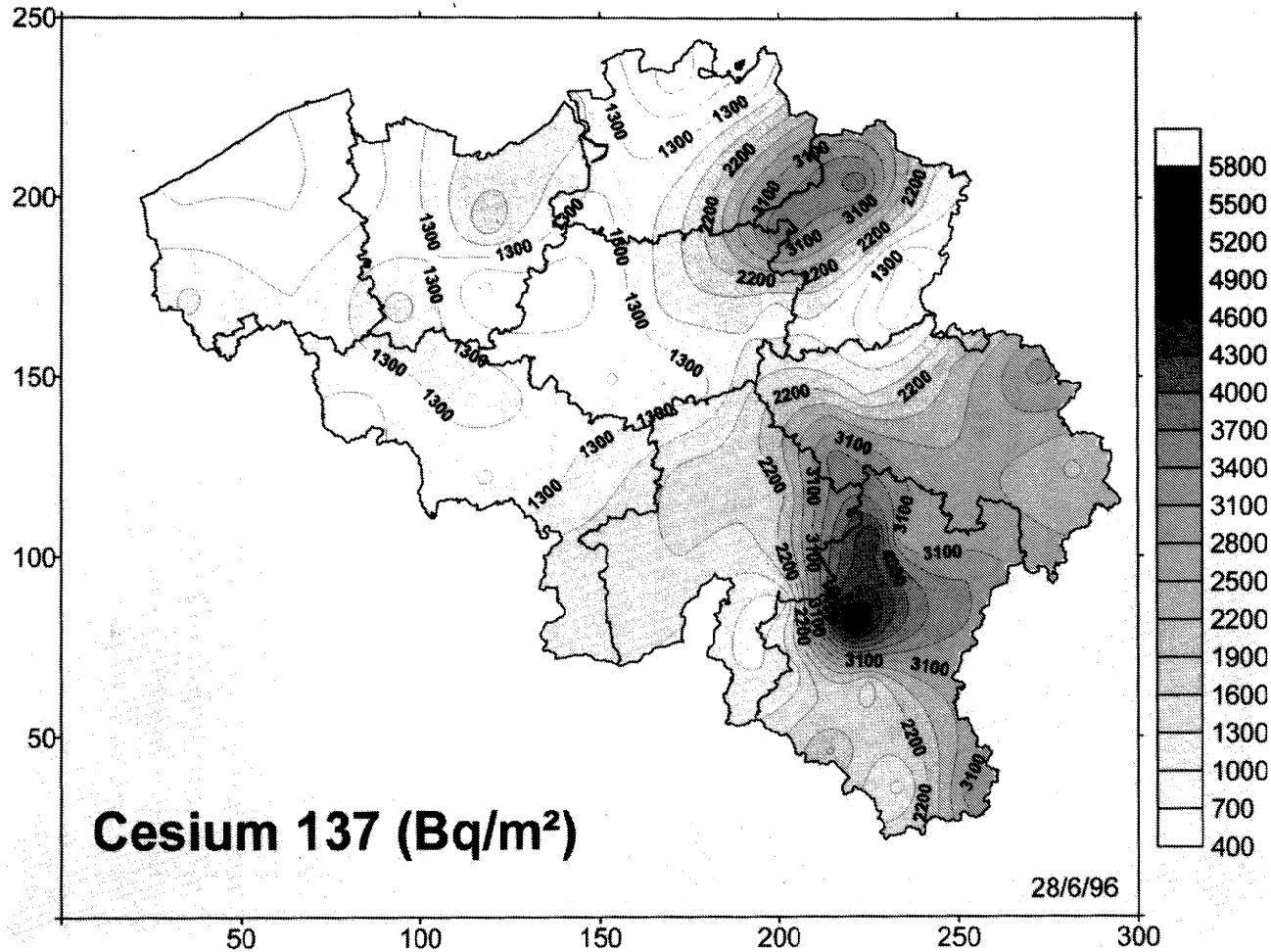

Fig.3 Contour map of Belgium with the ^{137}Cs concentrations (in Bq/m^2)

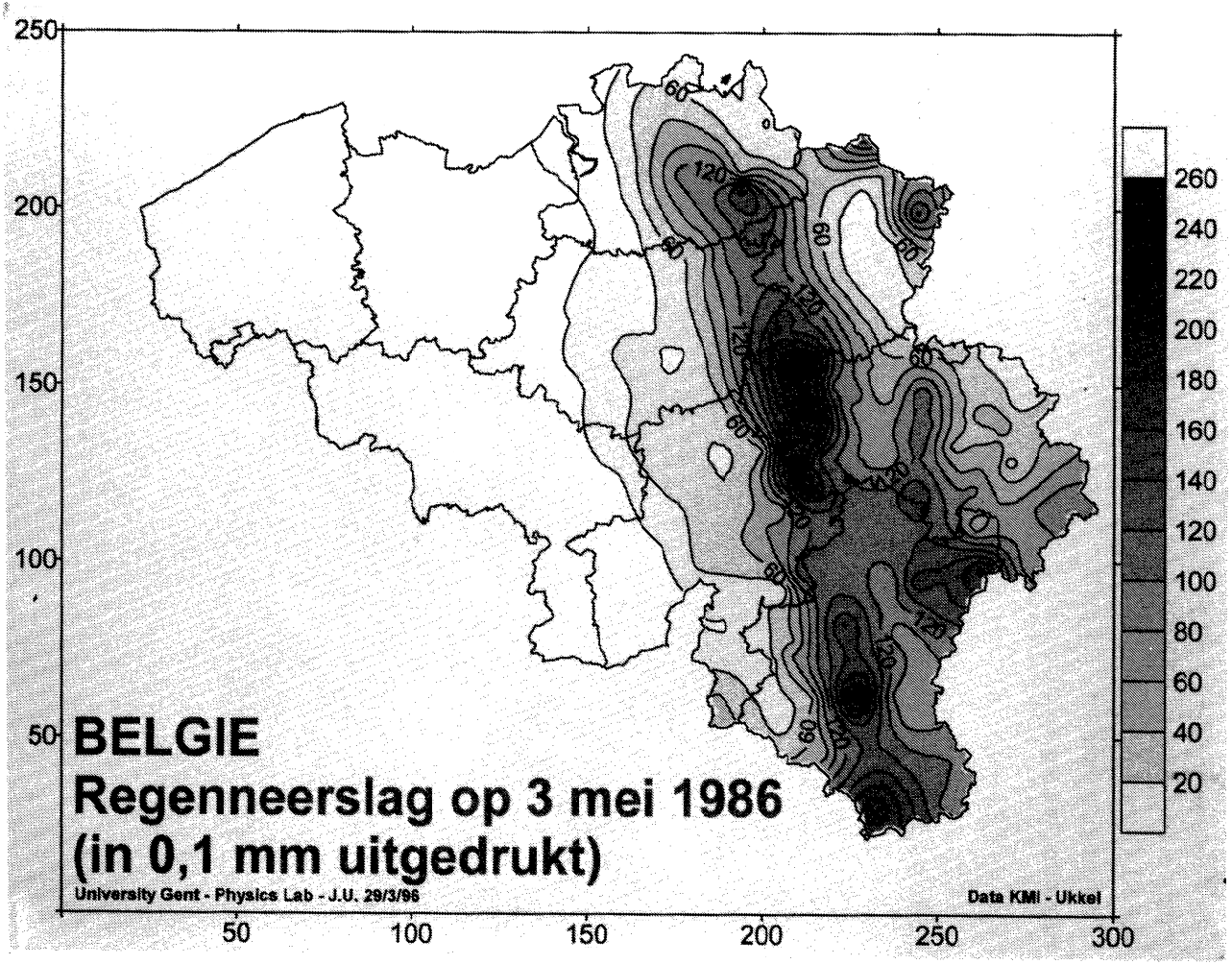

Fig.4 Map of Belgium with rainfall on May 3, 1986

ACKNOWLEDGEMENTS

The SCK-CEN authors wish to thank all their colleagues of the γ-spectrometry group, in particular F. Janssens, who participated in the measurement campaign. The RUG-team is indebted to Prof H. Dumont (Ecology Laboratory, RUG) for transportation facilities. The authors acknowledge the cooperation of the KMI by extracting the requested rainfall data from its databases.

REFERENCES

l. Beck, H., De Campo J. and Gogolak C. , USDOE Report HASL-258 (1972)
2. Helfer, 1. and Miller. K. , Health Physics, 55 No 1, 15-29 (1988}
3. Miller, K. And Shebell P. , EML - USDOE , ENL,-557 (1993)
4. ICRU report 53. ICRU, Bethesda, Maryland, USA (1994)
5. Van Waeyenberge B.. thesis Univ. Gent, (1995)
6. Pommé S., internal report SCK-CEN, BLG-695 (1995)
7. Govaerts P., Fieuw G., Deworm J.P. and Zeevaert Th., report SKV-H6-1013 SCK-CEN. Mol
8. SURFER for Windows, User Guide, Golden Software Inc., Golden, Colorado, USA

Annales de l'Association belge de Radioprotection
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.21, n°4, 1996
pp 339-344

ON THE USE OF THE NORMALIZED LEAST-SQUARES METHOD TO RECONSTRUCT THE SOURCE TERM FROM TRACER EXPERIMENTS

Carlos Rojas-Palma and Alain Sohier
Belgian Nuclear Research Center (SCK-CEN)
Boeretang 200, B-2400 Mol, Belgium

INTRODUCTION

Should an accident in a nuclear power plant (NPP) result in a major release of radioactivity into the atmosphere, a number of protective actions must be issued to avoid excessive exposure of the population to ionizing radiation. Early countermeasures will be based on the current status of the NPP such as burned-up fuel, cooling time, release pathways , etc. However, once the release has begun, off-site emergency actions will strongly depend upon one's ability to collect in a minimum of time enough data to characterize the accident, and to use these data to reconstruct the source term, i.e. the amount and nature of the material being released. New estimates of the source term are then fed into a dispersion-deposition model to obtain an updated view of the affected areas downwind, providing the decision makers with new information.
This paper concentrates on the use of the normalized least-squares method as a tool to reconstruct the source term used during several tracer experiments, as well as on a discussion of its applicability considering the time and data constraints of a real situation.

MATERIALS AND METHODS

Experimental and Modeling

The data set used in this work was generated during several (11) experiments conducted during the early eighties at the location of Mol, Belgium. These experiments consisted of releasing a known amount of a tracer gas (SF_6) from a height of about 2 m. Samples were collected at a number of stations situated downwind on arcs around the point of release, and covering a distance of a few kilometers. For details on the sampling technique as well as on the analytical procedures used to determine the air concentrations, the reader is referred to [1]. The release rate was kept constant throughout the experiment, and the time span between release and sampling was long enough to ensure that the plume had reached the observation point.
A data set of model estimates was computed for each tracer experiment using the segmented-puff dispersion deposition model ATSTEP [2]. This model has been chosen due to its degree of complexity which lies between a simple bi-gaussian model and a particle model, the reduced number of input parameters required, and overall speed, which are all elements of extreme importance in

real-time response to an emergency. Model predictions were done assuming a stationary meteorology, i.e. wind direction, speed and atmospheric stability did not change throughout the experiment, and a constant release rate of 1 g s^{-1} .

Normalized least-squares method (NLSM)

This approach does not differ significantly from the one used in [3] for the source term reconstruction using the deposited activity after a nuclear accident, and it is based upon the assumption that the modelled quantities, airborne concentrations in this case, satisfy the equation:

$$L_i = Q\, L_i\,(P)$$

where the subscript i represents a given point, Q is the source tenn, and P summarizes all the parameters such as release height, wind speed, atmospheric stability, etc. Bearing this in mind, one can define an objective function F such that,

$$F = \frac{\sum_i (QL_i - Z_i)^2}{\sum_i Z_i^2}$$

with Z_i being the observed quantities, e.g. airborne concentrations and L_i the modelled quantities. The normalization to the sum of the Z_i ensures that all possible values of F are between zero and one, which makes the interpretation of the results somewhat easier. The source tenn (Q) can be determined by minimizing F, which is a straightforward procedure.

RESULTS AND DISCUSSION

A summary ofthe results obtained for 11 tracer experiments, is shown in the Table 1 below:

Case	Qreal	Qnslm	Qinvgauss
1	2.2	1.1	1.2
2	3.2	6.7	6.5
3	3.2	8.0	10.4
4	3.2	3.6	4.1
5	3.4	3.5	4.4
6	3.4	2.9	2.8
7	1.0	0.8	1.1
8	3.3	3.1	4.7
9	3.3	12.0	13.1
10	3.5	1.6	1.4
11	4.2	2.7	3.4

Table 1. Comparison of the real source term (g s^{-1}) with the reconstructed using NLSM and that obtained by simply inverting a bi-gaussian model.

It can be seen from this table that the NLSM procedure yields (cases 4 through 8) an estimated source term that agrees fairly well with the real one, whereas the maximum difference (factor 4) was observed in case 9. In this particular case, the atmosphere was very stable and this resulted in a very narrow plume, as shown in Figure 1, causing that a significant number of receptors did not receive any signal at all. Even in this case (9) a difference of a factor of 4 seems quite acceptable, considering that the meteorological conditions were assumed constant throughout the experiments. Even though this method has proven to be a very useful tool for the estimation of the source term, its applicability to a real situation is very limited, and this is mainly due to the scarcity of observation points during the early phase of the accident. Furthermore, even if several observation points were available, they should be near the plume's center line to avoid situations similar to number 9 above, i.e. when the plume is very narrow.

The last column in Table 1 shows the source term resulting from the inversion of a bi-gaussian model and considering only one point, namely the one with the highest concentration. It is worthy to mention that this procedure also yields significant results and has the advantage of needing very few observation points. Therefore, the reconstruction of the source term during an accident could be possible if one follows a monitoring strategy that focuses mainly on locating the plume's axis. A number of monitoring guidelines are still under development to achieve this goal.

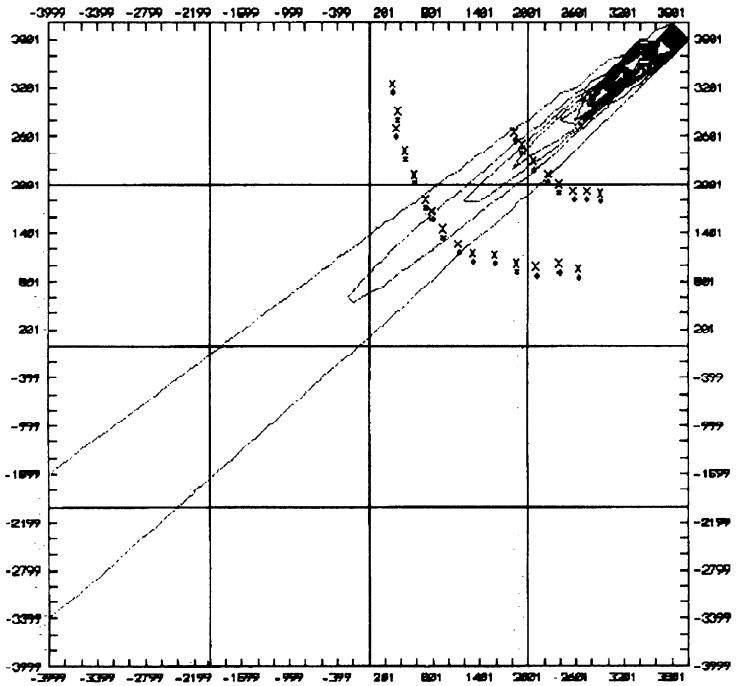

Figure 1. Plot of the resulting plume for case 9. The position of the source in this 41x41 grid has been chosen according to the prevailing wind direction (NE), and the resolution was set to 200m. The location of the receptor points is also shown.

REFERENCES

1 . J.G. Kretzschmar, I. Mertens and R. Vandenborght, *Final Report* Contract SR-028-B (1984)
2. J. Pasler-Sauer, *Radiation Protection Dosimetry*, Vol 50, pp 219-226 (1993)
3. B.L. Semenov, R.V. Arutyunjan et al. Jahrestagung Kerntechnik, pp 68-71 (1995)

Annales de l'Association belge de Radioprotection,
Annalen van de Belgische Vereniging voor Stralingsbescherming,Vol.21, n°4, 1996
pp 345-352

AFCF AS AN AGRICULTURAL COUNTERMEASURE: SIDE-EFFECTS OF APPLICATION OF MANURE FROM TREATED ANIMALS AND POTENTIAL EFFICIENCY WHEN APPLIED DIRECTLY ON SANDY SOIL.

C.M. Vandecasteele, M. Van Hees, S. De Brouwer and H. Vandenhove

Radioecology laboratory, SCK•CEN, B-2400 Mol

Abstract

AFCF is known to be a very effective countermeasure to limit the radiocaesium uptake by animals. Our results show that it could also be directly applied on recently contaminated sandy soil to reduce the radiocaesium availability for plant uptake: a 4-fold reduction in TF can be achieved under strictly controlled conditions with 10 $kg.ha^{-1}$. One ton $AFCF.ha^{-1}$ even resulted in a 225 fold reduction of the transfer factor. Up to application rates as high as 1 $t.ha^{-1}$, AFCF does not affect plant growth nor induces visual toxic symptoms. On another hand, the presence of AFCF in animal faeces returning to pastures and field as manure reduces also the availability for plant uptake of the already present caesium in soil; this effect lasts for at least one year after the application. Taking into account the absence of toxic effect of AFCF addition on plant growth when it is applied on sandy soils as a water solution in concentrations of up to 1 $t.ha^{-1}$, it is improbable that applying on field or pastures, manure from AFCF treated animals in appropriate doses would have negative side-effects on the vegetation biomass production.

INTRODUCTION

AFCF [Ammonium-ferric-hexacyano-ferrate] is a very effective caesium binder. Mixed with the animal feed, presented in the form of salt licks or introduced into the rumen as slow release *boli*, this compound is an efficient countermeasure to limit the gastro-intestinal uptake of radiocaesium by farm animals and wild ruminants [1-3]. Less than 1 % of the ingested AFCF is excreted in urine or secreted in milk [2], suggesting that it crosses the gastro-intestinal tract unabsorbed to be finally excreted in faeces together with the caesium bound in the gut. This means that AFCF from treated animals returns directly to pastures while animals are grazing or that it can be spread on fields fertilised with animal manure. Although no toxicological problems have been observed on animals given hexacyanoferrates in the recommended doses, the fate of this molecule in the environment after excretion is not well documented. Except for limited data obtained in Norway and in the CIS [4], practically no information is available regarding its action on the availability of Cs present in the soil, nor concerning potential side-effects of its possible degradation to cyanides and other materials with a concomitant release of bound Cs over long periods of time [2, 3].

On another hand, considering its high binding potential for caesium and the fact that, in the days after an accident, it can easily be applied as a colloidal, aqueous solution on agricultural soils with a conventional pulveriser for liquid fertilisers or pesticides, its potential use as immediate countermeasure to reduce the radiocaesium availability for plant uptake can also be considered [5,6].

MATERIAL AND METHODS

^{134}CsCl and ^{137}CsCl sources were purchased from Amersham and AFCF (Industrial Giese Salt, containing 60-65% AFCF and 35-40% NH_4Cl) was obtained from Riedel-deHaen.

Blank and ^{137}Cs contaminated sheep faeces, with and without AFCF, were successively obtained from the same animal housed in a metabolism cage which was fed 150 g beet pulp per day and mixed grass hay *ad libitum* (fig. 1).

After a period of adaptation to the diet (4 days), Blanca faeces was collected for one week. From the 11th day onwards, the sheep was orally dosed 100 MBq ^{137}Cs.d^{-1} every morning for 5 days, then the contamination was suspended during 4 days and resumed at a rate of 60 MBq ^{137}Cs.d^{-1} for another 5 days' period. Giese salt administration (2 g.d^{-1} given *per os* in the morning, 15 minutes after the administration of Cs) was started two days before the second Cs contamination phase, and carried on until two days after. Both morning and night faeces production was collected. After g-counting (minaxi, auto-gamma 5000 series, Packard Instrument), faeces with a high and rather constant contamination level was gathered, mixed and ground. Cs-AFCF faeces was diluted with Blank faeces to reach the same radioactivity level as in Cs faeces (i.e. 63 kBq.g^{-1} dw). The AFCF-level of the contaminated Cs-AFCF faeces was estimated from the amount dosed and the faeces dry weight production, assuming that an equilibrium between AFCF ingestion and excretion had been reached when the Cs-AFCF faeces collection was started. After dilution with Blank faeces the concentration was about 0.10 g AFCF.g^{-1} dw faeces. These faeces were mixed with blank and ^{134}Cs contaminated soil. Since 10 g faeces were applied (3.3 t.ha^{-1}), this resulted in a ^{137}Cs contamination level of 630 kBq per container, mixed in the top 0.4 cm layer. The AFCF amount applied with Cs-AFCF faeces was equivalent to 10 kg AFCF.ha^{-1}.

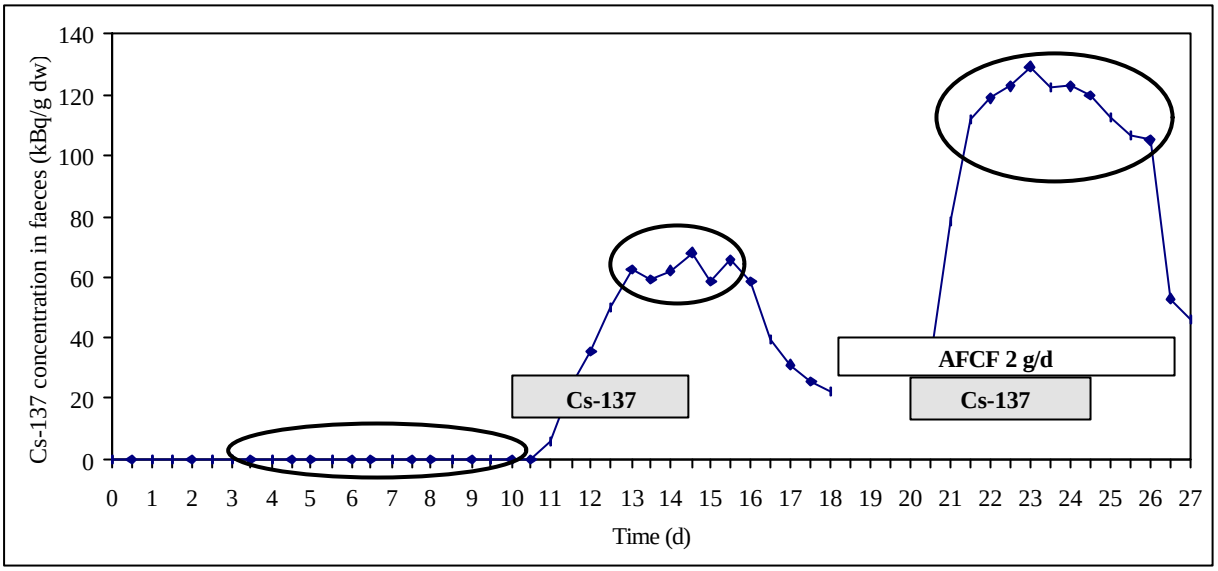

Figure 1: Blank, ^{137}Cs and ^{137}Cs-AFCF faeces production schema. The samples surrounded by an ellipse were pooled to give the different faeces types.

The transfer to ryegrass was studied in greenhouse conditions. Rectangular shaped darkened containers (20x15x11 cm^3) were filled with 3200 g of dry soil (Orthic Podzol). 2 g of Italian ryegrass (*Lolium multiflorum*) was spread evenly over the soil. The seeds were covered with a 0.4 cm moist soil layer homogeneously contaminated with ^{134}Cs (1.0 Mbq per container), mixed with different additives, according to treatment applied.

To investigate the potential side-effects of the application on fields of manure from AFCF treated animals, four treatments (in triplicates) were considered :

1. no additives other than ^{134}Cs in the top soil layer,
2. a soil amendment with ^{137}Cs contaminated sheep faeces (3.3 t dw.ha^{-1}) from an AFCF treated animal,
3. a soil amendment with ^{137}Cs contaminated sheep faeces at the same rate but from an untreated animal,
4. the application of an AFCF water solution (30 m^3.ha^{-1}) at a rate of 10 kg Giese salt per ha.

Its efficiency to reduce the radiocaesium bio-availability in soil and potential toxic effects on plant growth were tested by surface application of an AFCF aqueous solution onto the superficially contaminated soils. The AFCF application rates ranging from 100 g to 10 t.ha^{-1} were investigated in two experiments respectively considering ranges from 100 g to 10 kg AFCF.ha^{-1} and 10 kg to 10 t AFCF.ha^{-1}.

Pots were watered every two days. Ryegrass was harvested every two or three weeks, depending on growth. Dry weight was determined and activity measured by g-counting. The ^{137}Cs and ^{134}Cs transfer factors are defined as (Bq.g^{-1} dry plant material)/(Bq.g^{-1} dry soil). After every harvest, plants were given $N_{13}P_{13}K_{21}$-fertiliser (30 ml.dm^{-2} of a solution containing 17 g fertiliser.L^{-1}). In total 17 (low range AFCF application) or 19 (high range AFCF application and experiment with manure) cuts were harvested over one year.

The data were analysed using "Statistica®" as statistical software [7].

RESULTS AND DISCUSSION

Application to soil surface of aqueous solutions of AFCF

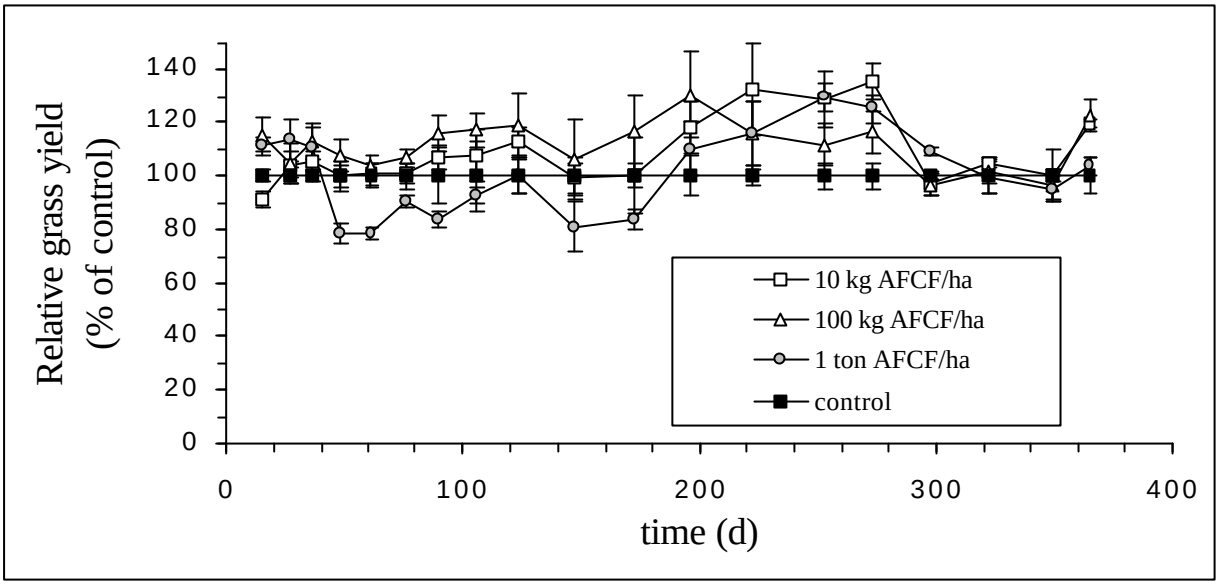

Fig. 2: Relative ryegrass yield (in % of the control) for AFCF applications on the soil surface ranging from 10 kg to 10 t AFCF.ha^{-1}. Error bars represent one standard deviation.

The yields of the successive ryegrass harvests in each experiment was compared to their respective control. The results from the one dealing with the highest range (10 kg to 10 t AFCF.ha^{-1}) are presented in fig. 2. Neither growth reduction nor visual toxic symptoms were observed with increasing surface additions of AFCF from 10 kg to 1 t.ha^{-1}. Lower application rates also had no effect on the ryegrass productivity. Only the very unrealistic application of 10 t.ha^{-1} prevented seed emergence through the soil surface clogged by AFCF.

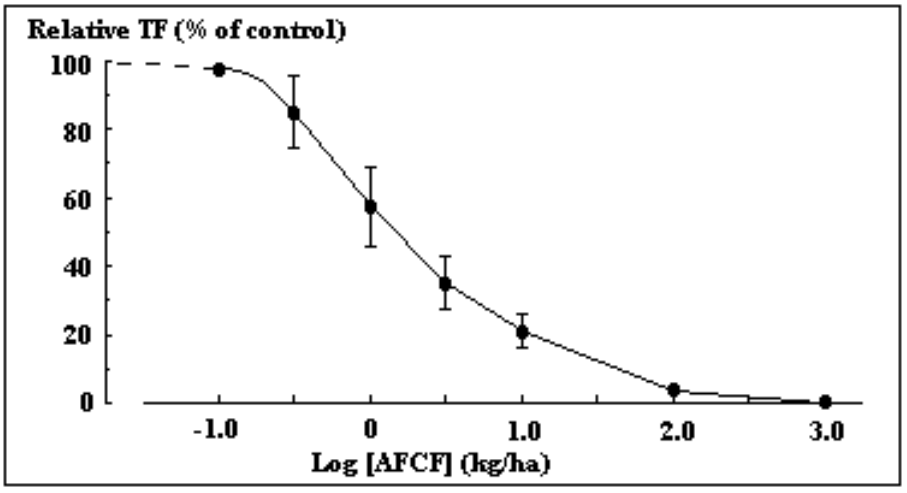

Fig. 3: Relative transfer factor (in % of the control) for AFCF applications on soil surface ranging from 0.1 kg to 1 t AFCF.ha^{-1}. Error bars represent one standard deviation.

The TF ($Bg.g^{-1}$ plant dw/$Bq.g^{-1}$ soil dw) for the control (1.869 ± 0.946) (Fig. 3) is in agreement with literature data [8,9]. Levels of 1 kg AFCF.ha^{-1} and higher significantly reduce the soil-to-plant transfer of ^{134}Cs: by a factor of 4 when 10 kg AFCF.ha^{-1} is applied and by a factor of 25 and 225, with 100 and 1000 kg AFCF.ha^{-1}, respectively, resulting in transfer factors of 0.069 ± 0.036 and 0.008 ± 0.007. AFCF additions of less than 1 $kg.ha^{-1}$ were however hardly effective in reducing the TF to ryegrass.

Application to soil of manure from AFCF treated animals

Compared to the control, no statistically significant effect on the biomass production was observed due to the addition of animal manure (3.3 t $dw.ha^{-1}$) (data not shown). The analysis of the variance (two-way anova) underlines a highly significant effect of the time (with an increased yield in spring and early summer), but no difference between the four treatments. The absence of toxic effects on plant growth due to the AFCF applied with faeces is not surprising considering that no effect (visual toxicity symptoms or biomass production decrease) of AFCF has been observed on the same soil type (sandy podzol), after application of up to 1 t AFCF.ha^{-1} in a water solution. Such a high dose would only be achieved after more than 270 y on pastures supporting a grazing pressure of 5 $cows.ha^{-1}$, in so far as the cows are constantly dosed 2 $g.d^{-1}$, the farm manure produced in the winter period is spread on the grazed pastures and the AFCF molecule is not degraded and its application is thus cumulative.

The availability for plant uptake of radiocaesium excreted with faeces (^{137}Cs) was significantly lower, by a factor 5, in Cs-AFCF-faeces compared to that in Cs-faeces from untreated animal. Moreover, the AFCF brought to the soil (10 $kg.ha^{-1}$) with Cs-AFCF-faeces (3.3 t $dw.ha^{-1}$) decreased in the same proportion the bio-availability of ^{134}Cs present in the soil surface layer than that of ^{137}Cs in manure (fig. 4). A comparable effect was obtained by the application at the same rate of AFCF in water.

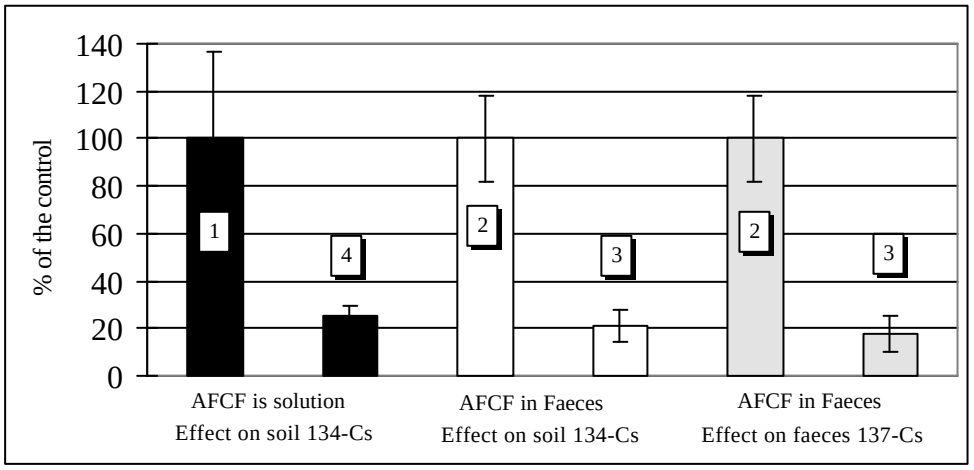

Fig. 4: Effect of AFCF applied at a rate of 10 $kg.ha^{-1}$ on the transfer to plants of caesium from faeces or already present in the soil top layer. Average over one year observation (19 successive cuts) for the treatments and their respective control (± s.dev. between 4 replicates)

From the comparison of treatments 1 and 3, regarding the soil ^{134}Cs transfer to plants, we notice that the addition of organic matter can also contribute to decrease the availability for plant uptake of the ^{134}Cs in soil by increasing the soil exchange capacity. In our case, the application of 3.3 $t.ha^{-1}$ of animal manure to a soil containing some 4 % organic matter induces a 20 % reduction of the transfer to plants compared to the non amended control.

CONCLUSIONS

From the results presented here, some conclusions can already be drawn :

- AFCF application to sandy soil surface does not affect plant growth nor induces visual toxic symptoms.
- AFCF is effective in reducing radiocaesium transfer from soil to ryegrass: a 4-fold reduction in TF can be achieved under strictly controlled conditions with 10 $kg.ha^{-1}$. One ton $AFCF.ha^{-1}$ even resulted in a 225 fold reduction of the transfer factor. The relatively low cost ($\approx$ 650 $US\$.kg^{-1}$ which could be reduce by an upscaled production), its easy application (an AFCF solution can be sprayed making use of the machinery for applying pesticides), its non-toxicity for plants and its long-term effectiveness (even after 1 year AFCF was still effective in reducing the TF) make AFCF a seemingly ideal countermeasure on soils with a low affinity for Cs.
- AFCF in faeces from sheep treated with a 2 $g.d^{-1}$ dose limits the availability for plants of the caesium excreted in the faeces.
- The presence of AFCF in the faeces acts also on the availability of the already present caesium in soil, and reduces its bio-availability for plant uptake. This effect lasts for at least one year after the application.
- Taking into account the absence of toxic effect of AFCF addition on plant growth when it is applied on sandy soils as a water solution in concentrations of up to 1 $t.ha^{-1}$, it is improbable that applying on field or pastures, manure from AFCF treated animals in appropriate doses would negatively affect the biomass production.

However, some attention should be given to the influence of AFCF on the Cs mobility in soil and the risk linked with a potential contamination of the (under)ground waters.

REFERENCES

1. Giese W.W. (1989), Countermeasures for reducing the transfer of radiocaesium to animal derived food, Sci. Tot. Environ., 85: 317-327.
2. P.J. Coughtrey (1993), Effects of countermeasures on radionuclide transfer to animal products, IUR Pub R-9301-03, 52pp.
3. G. Voigt (1993), Chemical methods to reduce the radioactive contamination of animals and their products in agricultural ecosystems, Sci. Tot. Environ., 137: 205-225.
4. Hove, P. Strand, B. Salbu, D. Oughton, N. Astasheva, A. Vasiliev, A. Ratnikov, T. Jigareva, V. Averin, S. Firsakova, M.J. Crick, and J.I. Richards, *IAEA-SM-339/153*, 75-76 (1995).

5. Segal M.G. (1993), Agricultural countermeasures following a nuclear accident, Sci. Tot. Environ., 137: 31-48.
6. Jones D.R., Paul L. & Mitchell N.G. (1994), Field investigation of the effects of ameliorative treatments applied to improved upland vegetation in the UK contaminated by radiocaesium, Joint XXIVth ESNA/UIR meeting, Varna Bulgaria, September 12-16, 1994.
7. StatSoft, Inc. (1996). STATISTICA for Windows [Computer program manual]. Tulsa, OK: StatSoft, Inc., 2300 East 14th Street, Tulsa, OK 74104.
8. Alexakhin R.M. (1993), Countermeasures in agricultural production as an effective means of mitigating the radiological consequences of the Chernobyl accident, Sci. Tot. Environ. 137: 9-20
9. International Union of Radioecology (1989), Vth Report of the working group soil-to-plant transfer factors. Report of the working group meeting in Guttannen, Grimpelpass, Switzerland, May 24-26, RIVM, Bilthoven, The Netherlands, pp. 215.

Résumé.
L'AFCF est connu pour son efficacité en tant que contre-mesure visant à limiter l'absorption du radiocésium par les animaux. Nos résultats démontrent qu'il pourrait également être appliqué directement sur des sableux sols récemment contaminés afin d'en réduire la disponibilité du radiocésium pour le végétaux: en conditions strictement contrôlées, une diminution d'un facteur 4 du transfert du sol à la plante est obtenue par l'application de 10 $kg.ha^{-1}$. L'application d'1 $t.ha^{-1}$ aboutit à une réduction du transfert par un facteur 225. Jusqu'à des doses aussi élevées que 1 $t.ha^{-1}$, l'AFCF n'affecte pas la croissance des végétaux, ni n'induit de symptôme de toxicité visible. D'autre part, l'AFCF dans les excréments animaux retournant à la prairie ou répandu comme engrais réduit également la disponibilité pour les végétaux du radiocésium présent dans le sol; cet effet persiste au moins un an après l'application de l'amendement organique. Considérant l'absence d'effet toxique sur la croissance des végétaux liée à l'application sur sols sableux d'AFCF en solution aqueuse jusqu'à des doses d'1 $t.ha^{-1}$, il est improbable que l'épandage sur des champs de fumier provenant d'animaux traités avec de l'AFCF aux doses recommandées ait des effets secondaires négatifs sur la productivité végétale.

Samenvatting.
AFCF is bekend als een efficiënte tegenmaatregel om de absorptie van radiocesium bij dieren te beperken. Onze resultaten tonen aan dat AFCF ook op recent besmette zandgronden zou kunnen gebruikt worden om de beschikbaarheid van radiocesium voor planten te reduceren: onder strikt gecontroleerde omstandigheden wordt een vermindering van de transfer van de grond naar de plant met een faktor 4 bereikt met de toevoeging van 10 $kg.ha^{-1}$. De toevoeging van 1 $t.ha^{-1}$ leidt tot een reductie van de transfer met een faktor 225. Dosissen tot 1 $t.ha^{-1}$ hebben geen effekt op de groei van planten en veroorzaken geen zichtbare toxische verschijnselen. Anderzijds heeft AFCF aanwezig in dierlijke excreta die op de weiden terugkeren of in dierlijke mest die als organische meststof op velden verspreid wordt een vermindering van de beschikbaarheid voor planten van radiocesium die zich in de grond bevindt tot gevolg; dit effekt duurt ten minste gedurende één jaar voort. Rekening houdende met het feit dat er geen toxische effekten op de plantegroei voorkomen bij de verspreiding van waterige oplossingen van AFCF tot dosissen van 1 $t.ha^{-1}$ op zandgronden, wordt het onwaarschijnlijk geacht dat het verspreiden van mest van met AFCF behandelde dieren op velden secundaire negatieve effekten op de planteproduktiviteit zou hebben.

Annales de l'Association belge de Radioprotection,
Annalen van de Belgische Vereniging voor Stralingsbescherming,Vol.21, n°4, 1996
pp 353-358

RADIOLOGICAL CRITERIA FOR THE SELECTION OF A NEAR SURFACE DISPOSAL SITE

L. Baekelandt and J.-P. Minon, NIRAS/ONDRAF, Brussels
Th. Zeevaert and P. Govaerts, SCK/CEN, Mol

Introduction

The selection of a near surface disposal site requires a multi-attribute analysis. Some of these attributes deal with the radiological impact.
The most important radiological attributes are :

- for routine and normal evolution exposure :
  - individual doses for members of the critical group;
  - collective doses;
- for potential exposures, including accident situations :
  - individual risks for members of the critical group;
  - collective risks.

In case of potential exposure, the dose impact of the accident, as well as the probability of occurrence has to be considered.

The radiological impact has to be assessed during three distinct time periods :

- the operational phase;
- the period of institutional control;
- the period following institutional control.

During the operational phase, in routine situations, no radionuclides will be released into the biosphere. The same is true for a large part of the period of institutional control. In such cases, one has to consider only the exposure of workers (during transport, disposal and surveillance of the site); public exposure may be disregarded. During this same period, in accident situations, the individual risk to members of the public is likely to be insignificant. On the other hand, in regions with a high population density (continuously or temporary), the collective risk for the population cannot be disregarded.

During the post operational phase, the normal evolution will give rise to releases of radionuclides into the biosphere. During this period, routine and accidental exposure of workers during the surveillance of the disposal site will most likely be negligible.
For exposure of the general public, one has to consider only collective doses and collective risks, the individual dose being taken equal to the dose constraint of 0.3 mSv/year by accommodating the inventory of the facility to its full capacity.

The operational Phase

During the operational phase, one has to consider the transport and disposal operations.

Transport

Routine exposure is only relevant for workers. The distance between the disposal site and the waste

generator or the storage facility may in certain cases, have an impact on the individual dose of the workers, but will always have an impact on the collective dose to the workers.

For accidental exposure, the collective dose impact will depend on the population density along the transport route. Also the probability of occurrence of a transport accident will depend on the transport distance.

The impact of transport by road (dose impact as well as accident probability of occurrence) may be different from the impact of transport by rail.

Disposal

Routine exposure does not depend on site-specific parameters.

In accident situations, the individual doses to workers are not site-specific. The collective impact for the general public depends on the population density in the neighbourhood of the disposal facility.

As far as the probability of occurrence of accidents is concerned, one has to consider the following types of accident :

- accidents while handling waste packages;
- accidents of external origin : air plane crash;
- accidents of natural origin : earth quake and flooding.

Given the site selection exclusion criteria, earth quakes and flooding must not be considered. Therefore, only the probability of occurrence of an air plane crash is to be considered as a site-specific parameter. This probability depends on the presence/absence of air corridors and airports (military and general).

The period of Institutional Control

With respect to routine exposure of workers, individual as well as collective doses are not site-specific.

With respect to accidental exposures, one has to consider the same accidents as during the operational phase. However, since the disposal facility will be closed and completely covered, the risks for workers, as well as for the general public, will be negligible.

Concerning the normal evolution during this period, only very mobile radionuclides (tritium, carbon, iodine, technetium) may be released into the biosphere. For the radiological impact to the general public, a dose constraint of 0.3 mSv/year is assumed to be reached when the full capacity of the disposal facility is used. Therefore, given a fixed individual dose, only the collective dose must be considered. In that case, the most relevant site-specific parameters are :

- the longitudinal dispersivity for groundwater transport;
- the groundwater transport distance to the receptor (well or surface water).

Other site-specific parameters are : the population density in the neighbourhood of the receptor and the use of the surface water, in particular for irrigation purposes; for such purposes, texture and drainage class of the soil are of some importance.

The Period following Institutional Control

The normal evolution scenarios during this period and during the period of institutional control are identical.

For accident scenarios, one has to consider different types of human intrusion :

- construction of buildings, roads and canals;
- construction of water wells;
- construction of buildings for human residence.

In the Belgian context, the construction of canals appears to be the most site-specific parameter with respect to the probability of occurrence.
With respect to the dose impact of the intrusions, one has to make a distinction between the direct exposure to the individuals responsible for the intrusion, and the future exposure of individuals because of the (earlier) release of the radionuclides into the biosphere. The site-specific parameters for human intrusion are the same as those for the normal evolution.

Evaluation of the radiological attributes

Individual and collective doses have been evaluated for three representative radionuclides and for two extreme situations : a very favourable one and a very conservative one.
The calculated ranges are given in the following tables.

Table 1 - Individual Doses (mSv/year)

	Transport		Disposal		Post-operational phase	
	Routine	Accident	Routine	Accident	Normal evolution	Intrusion
Nb-94	2.5 - 3.7	0 - 0.4	5.2 - 20	$< 10^{-3}$	$3.2\ 10^{-3}$ - 0.3	$1.5\ 10^{-3}$ - $6\ 10^{-3}$
I-129	2.5 - 3.7	0 - 0.4	$8\ 10^{-3}$ - $1.4\ 10^{-2}$	$< 10^{-3}$	0.3	$< 10^{-3}$
Pu-239	2.5 - 3.7	0 - 0.4	0	$< 10^{-3}$	0.2 - 0.3	0.1 - 0.3

Table 2 - Collective Doses (man.Sv)

	Transport		Disposal		Post-operational phase	
	Routine	Accident	Routine	Accident	Normal evolution	Intrusion
Nb-94	0.3 - 3.7	0 - 0.3	1.5 - 6	0 - $5\ 10^{-8}$	0.6 - 20	$7\ 10^{-2}$ - 0.3
I-129	0.3 - 3.7	0 - 0.3	$2\ 10^{-3}$ - $4\ 10^{-3}$	0 - $4.6\ 10^{-4}$	290 - 6000	$1.4\ 10^{-4}$ - $2.6\ 10^{-4}$
Pu-239	0.3 - 3.7	0 - 0.3	0	0 - $5.2\ 10^{-3}$	140 - 3200	5.3 - 15

Conclusions

In general, the most relevant radiological attributes are associated with the routine transport and disposal operations and with the collective dose as a result of the release of radionuclides into the biosphere during normal evolution. It must be recognized however, that :

- doses due to routine operations can be reduced in applying normal radiological protection procedures;
- some of the differences between sites are a direct result from the fact that the capacity of the sites is coupled to the dose constraint of 0.3 mSv/year. As a consequence, the capacity of the "worst" sites is reduced, to such extent that some of the attributes (collective doses or risks) at these sites become more favourable than those for the "better" sites;
- the collective doses are integrated over a long time period.

REFERENCES

[1] Th. Zeevaert and P. Govaerts, "Radiologische criteria in site selectie voor oppervlakteberging", SCK/CEN E23-311.94 (September 1994)

[2] Th. Zeevaert, P. Govaerts, H.-A. Pekka and G. Volckaert, "Site-specifieke factoren in radiologische criteria voor oppervlakteberging", SCK/CEN R-3062 (April 1996)

Annales de l'Association belge de Radioprotection
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.21, n°4, 1996
pp 359-365

DECOMMISSIONING ANALYSIS OF A UNIVERSITY CYCLOTRON

Gilbert X. Eggermont[1], N. Buls[2] and A. Hermanne[3]
University of Brussels (VUB), Radiation Protection Office[1],
Cyclotron Department[3], Brussels 1090, Belgium
Hogeschool Limburg[2], Hasselt 3500, Belgium.

INTRODUCTION

In the widespread use of some medical nuclear facilities, such as cyclotrons for isotope production, life cycle analysis, including decommissioning, was not taken into account. The structural materials of an accelerator and the concrete shielding of the bunker are activated by neutrons. This could yield a considerable volume of nuclear waste and needs radiation protection concern for occupational workers and the environment during some decennia (1).
At the university of Brussels (VUB) a prospective radiation protection and waste analysis is being made for the later decommissioning of their cyclotron (2). Only few similar studies have been published (1,3).
In Belgium future nuclear dismantling operations will be submitted to a radiation protection authorisation procedure. Meanwhile the nuclear waste authorities insist on dismantling planing, including financial provisioning.
An optimisation exercise was made at the VUB-cyclotron, taking into account international trends to clearance levels for low level nuclear waste.
Conceptual prevention opportunities e.g. selective material choice could be identified for future accelerator constructions.

ACTIVATION AROUND A CYCLOTRON

At the VUB a variable energy, multiparticle (protons, deuterons, α - and ^{3}He- particles) cyclotron (CGR-560) is in operation since 1985. The maximal current on target is 120 μA for 30 MeV protons. This machine is used for physics research and for radionuclide production in a shielded vault complex with 4 irradiation rooms (bunkers). The most important contribution to activation is due to ^{201}Tl isotope production at 27.3 MeV protons for radiopharmaceutical companies. The bunker shielding has a thickness of 2.5 m totalling a volume of 2700 m^3 of concrete.

The spectral characteristics of fast neutron beams were determined in different directions, showing an average energy of 12 Mev at 0°. Through multiple reflection on walls and elastic scattering in concrete, thermalisation of neutrons occurs in depth. The activation of trace elements in sand e.g. europium and of particular metals yields medium living radioactive products. They are created by capture reactions with high cross sections and by some threshold reactions with lower yield, as indicated below.

^{151}Eu(n,γ)^{152}Eu	σ_{th} = 5900 barn	$t_{1/2}$ = 13.33 year
^{153}Eu(n, γ)^{154}Eu	σ_{th} = 390 barn	$t_{1/2}$ = 8.8 year
^{133}Cs(n, γ)^{134}Cs	σ_{th} = 29 barn	$t_{1/2}$ = 2.06 year
^{59}Co(n, γ)^{60}Co	σ_{th} = 37 barn	$t_{1/2}$ = 5.3 year
^{45}Sc(n, γ)^{46}Sc	σ_{th} = 26.5 barn	$t_{1/2}$ = 83 days
^{58}Fe(n, γ)^{59}Fe	σ_{th} = 1.15 barn	$t_{1/2}$ = 44 days
^{64}Zn(n, γ)^{65}Zn	σ_{th} = 0.78 barn	$t_{1/2}$ = 244 days
^{55}Mn(n,2n)^{54}Mn	σ_{max} = 910 mbarn (18MeV)	$t_{1/2}$ = 312 days
^{54}Fe(n,p)^{54}Mn	σ_{max} = 590 mbarn (10MeV)	$t_{1/2}$ = 312 days

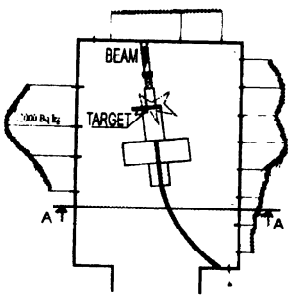

Fig 1 : Spatial distribution of maximal ^{152}Eu in concrete shielding.

The activation distribution was assessed by γ -spectrometry on bore samples of 5 cm diameter. They were systematically taken up to a depth of 50 cm throughout the shielding structure presented in fig. 1 .

RESULTS OF ACTIVATION MEASUREMENTS

The highest specific activation in concrete occurs laterally near the target at 15 cm depth (fig 2). The iron reinforcement bars in the concrete shielding show maximum activation levels of 32 Bq/g. The activity decrease is exponential for both cases. Prediction of depth profiles could be derived from the measurements (fig 3). The irradiation room infrastructure consists for 70 % of steel. Specific activities up to 360 Bq/g ^{54}Mn and ^{60}Co are measured.
The huge steel accelerator yoke of 80 ton however is showing much lower level activation due to its low ^{59}Co content. These values can be compared with clearance levels of about 0,3 Bq/g (4).

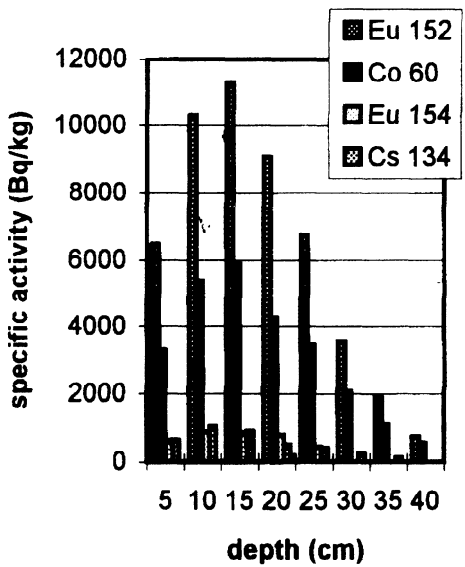

Fig 2 : Depth distribution in a concrete bore sample of specific activity for ^{152}Eu, ^{60}Co, ^{154}Eu, ^{134}Cs

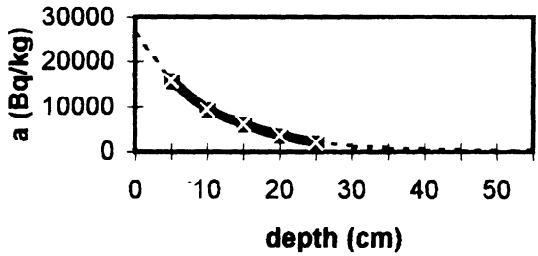

Fig 3 : Measured ^{60}Co activation in iron reinforcement bars

OPTIMISATION OF DISMANTLING SCENARIOS

Dismantling of a cyclotron will be submitted to an authorisation procedure in future Belgian regulations. Waste quantities have to be estimated with specification of their destination. The protection of man and the environment against radiation has to be guaranteed during preparatory work and dismantling operations.
Site destination and future use should be specified.
In this preliminary study, the following scenarios were considered :

- Early dismantling of a cyclotron infrastructure, directly after shut-down is confronted with an internal contamination risk by ^{65}Zn. This copper activation product is easily dispersed. ^{65}Zn is detected regularly at acceptable levels in whole body monitoring of cyclotron maintenance workers.
- The waste from metal infrastructures having activities up to 1000 times clearance level could be stored for decay during 35 years in the most activated cyclotron vault. This should be done after the management of the contamination problems.
- Early decommissioning of the concrete walls to 30-50 cm depths could yield up to 100 m^3 of nuclear waste, applying IAEA proposed clearance levels (4). The dismantling techniques available in nuclear fuel cycle industries allow to remove the activated depths. Since no provisions are made, the cost of this option is too high for an university.
  Dilution techniques, mixing active and inactive crushed concrete and melting iron bars, could be applied in order to arrive at a reasonable cost. The authorisation of such an approach has been given in the UK (5). It is not evident in Belgium, where the obligation of an environmental assessment report could need to take alternative options into account.
- A decay on site of the activity of the concrete rooms and of the metal

infrastructures till clearance levels could be performed. Fig 4 illustrates that cooling times of maximum 70 y are necessary for this option.

Restricted use of cyclotron rooms as controlled areas in the intermediate period was evaluated. Doserates in the documented bunker were calculated from the activation measurements using conservative assumptions.

A maximal doserate of 60 μSv/h was derived. Measurements indicate actual doserates up to 15 μSv/h.

Occupational use of such rooms yield calculated doses between 6 and 120 mSv/y.

Surface doses were measured with TLD on concrete samples with a maximum of 2 mGy/y.

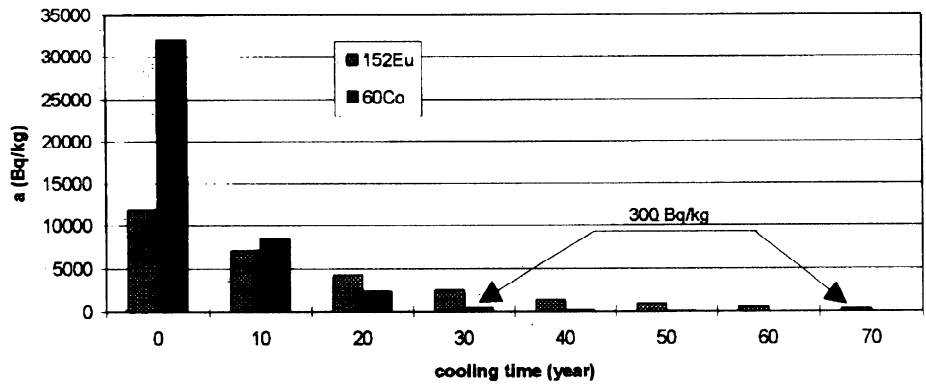

Fig 4 : Decay of Activation of Concrete Bunker

PREVENTION OPPORTUNITIES

The use of iron and scrap metal in the concrete shielding is not recommended for cyclotrons. The use of removable blocks for the inner part of the shielding walls could reduce dismantling costs. Local neutron absorption measures could be taken to reduce the source term. A selection of sand with low europium content for concrete preparation and a preference for low ^{59}Co steel or Al for the infrastructure could be taken into account in cyclotron complex conception.

CONCLUSION

Regarding the high cost of nuclear waste, decay during about 50 years of the infrastructure after shutdown is necessary to allow cooling to proposed clearance levels. Decontamination for ^{65}Zn could lbe performed after some years.
Considering dose limits for workers and the public, a restricted use of cyclotron rooms as controlled area, seems the most reasonable option. Life cycle analysis techniques should be integrated in the planning of isotope production facilities. It could contribute to reduction of later nuclear waste and to sustainable nuclear development.

REFERENCES

1 M.Braeckeveldt and M. Schrauben, *IMechE Conf Transactions 95-7*, 141-148,London.
2 A. Hermanne et al, presented at DECOM95, *IMechE Conf 95-7*, London
3 Ken-ichi Kimura et al, *Health Physics*, 67, 6, (1994).
4 IAEA, *Safely Series No. 89*, Vienna (1988).
5 D. Loughborough et al, *IMechE Conf. Transactions 95-7*, 343-354, London

Annales de l'Association belge de Radioprotection
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.21, n°4, 1996
pp 367-372

RADIOSENSITIVITY OF THE GUINEA-PIG OOCYTE AT DIFFERENT STAGES OF FOLLICULAR DEVELOPMENT

P. Jacquet[1], L. de Saint-Georges[1], J. Buset[1], J. Vankerkom[2], and L. Baugnet-Mahieu[1]

[1] CEN/SCK, Laboratory of Radiobiology, B-2400 Mol, Belgium,
[2] VITO, B-2400 Mol, Belgium

INTRODUCTION

Recently, we have shown that the guinea-pig represents an excellent model for studies on genetic hazard of radiation in man (1). We also developed techniques for culturing oocytes of this species and preparing their meiotic chromosomes for cytogenetic examination (2). In the experiments reported here, we examined the radiosensitivity of oocytes at two different stages of maturation, separated by only one week.

MATERIAL AND METHODS

In a first series of experiments, ovaries of adult females were X-irradiated with 1 or 2 Gy on day 3 of the 17-day oestrous cycle. Their meiotically competent oocytes were punctured from the growing follicles on day 10. They were cultured for 6 h to the MI stage, and fixed for cytogenetic analysis.
In a second series of experiments, ovaries were irradiated with the same doses on day 10 of the oestrous cycle. In this case, oocytes were collected and cultured either immediately after irradiation, or at various time intervals thereafter.

RESULTS

As shown in Table 1, important differences were found in the yields of chromosome aberrations, according to the time of the oestrous cycle at which irradiation had occurred.
Irradiation at the beginning of the cycle induced low numbers of chromatid interchanges and slightly higher numbers of other aberrations (chromatid and isochromatid breaks and fragments).
These effects were dose-dependent. The frequency of oocytes carrying interchanges was about 1.5 Gy^{-1}, a value rather comparable to that obtained after irradiation of the immature oocyte (3).
When irradiation was performed one week later, and oocytes were collected and cultured immediately thereafter, the frequencies of oocytes showing chromosome anomalies revealed much higher. This was true for interchanges as well as for other types of chromosome aberrations. Interchanges involved variable numbers of chromosomes and in many cases the number of chromosomes involved could not be determined. These highly damaged metaphases were frequently associated with chromatid breaks. The frequency of oocytes carrying interchanges was 30-35 % Gy^{-1}.
In some experiments, oocytes which had been irradiated on day 10 were collected and cultured one day later. Surprisingly, MI preparations revealed impossible to obtain : all fixed oocytes were

already in MII, whether they had been irradiated with 1 or 2 Gy. This phenemenon was studied more in details, using various doses of X-rays and different times of cultures.
It appeared that most if not all oocytes which were in MII on day 11 had been stimulated to divide *in vivo*, before removal from the ovaries. The lowest dose of radiation which was able to induce such a rapid stimulation of the first meiotic division in meiotically competent oocytes was apparently in the range of 0.25-0.50 Gy. Further experiments showed that all MII oocytes had been eliminated from the ovaries by day 15, i.e. 2 days before ovulation, and been replaced by others that were contained in smaller follicles at the time of irradiation. Those were in diplotene and were able to evolute normally to the MI stage, when cultured for 6 h.

TABLE 1
Chromosome aberrations detected in metaphase I oocytes irradiated on days 3 or 10 of the oestrous cycle (examination 1 week or directly after irradiation).

Dose (Gy)	Day of irradiation	Number of oocytes examined	Number of abnormal oocytes	Number of oocytes with interchanges	Number of oocytes with break/fragm.
0	-	78	0 (0)[a]	0 (0)	0 (0)
1	3	132	5 (3.78)	2 (1.51)	4 (3.03)
2	3	147	15 (10.20)	5 (3.40)	11 (7.48)
1	10	122	50 (40.98)	44 (36.06)	27 (22.13)
2	10	120	89 (74.16)	76 (63.33)	53 (44.16)

[a] values in parentheses are percentages

DISCUSSION

Our data evidenced a dramatic increase in the radiosensitivity of the guinea-pig oocyte during a time interval of only 1 week : oocytes irradiated at the beginning of the oestrous cycle (day 3) had a low frequency of chromosome aberrations, while those irradiated at the middle of the oestrous cycle (day 10, when growing Graafian follicles are clearly visible at the surface of the ovaries) exhibited heavy chromosome damage.
Earlier, Oakberg and Clark (4) noted a marked decrease in the number of large oocytes, during the first ten days following irradiation of guinea-pig ovaries by 2 Gy. In agreement with this, we found that oocytes irradiated at the middle of the oestrous cycle were eliminated from the ovaries in a few days, after their evolution to the MII stage. The stimulation of the first meiotic division by radiation required less than 24 h after doses of 1 or 2 Gy, and was probably due to a very rapid killing or inactivation of follicular cells which normally exert an inhibitory effect on this process. When examined on day 15 of the oestrous cycle, i.e. 2 days before ovulation, ovaries of animals irradiated on day 10 again showed high numbers of growing Graafian follicles. Oocytes contained in those were in diplotene, and behaved normally in culture, reaching the MI stage after 6 h. Thus, the ovaries had apparently compensated for the loss of all large follicles by an acceleration of the maturation and growth of smaller oocytes.
An increase in the number of corpora lutea per female was noted by Cox and Lyon (5), in guinea-pigs irradiated with 4 Gy (the only dose tested) between days 6-12 of the cycle and mated at the first oestrous post-treatment. This effect was attributed to the ovulation of abnormally high numbers

of oocytes in irradiated animals ("superovulated effect"). Concomitantly, there was an apparent increase of the preimplantation loss in these animals, an effect possibly resulting from the induction of dominant lethals by radiation. It will be important, therefore, to have a precise idea of the level of chromosome damage present in the oocytes surviving an irradiation near the middle of the oestrous cycle, and able to reach ovulation after an accelerated growth. Such study is in progress in our laboratory.

Data on the mouse indicated that, in this species,the radiosensitivity of the oocytes is low during weeks 1-2 before ovulation, if one excepts the 12 hours immediately preceding ovulation (6,7). On the basis of our results, it can be concluded that the radiosensitivity of the growing guinea-pig oocyte (1 week before presumed ovulation) is clearly much higher than that of the corresponding stage in the mouse, both in terms of sensitivity to killing and to induction of chromosome aberrations.

Recent results have also shown that the mouse immature oocyte was sensitive to the induction of interchanges and other chromosome aberrations by radiation (8-10), and data obtained in our laboratory allowed to extend this conclusion to the immature oocyte of the guinea-pig (3). However, the results of Straume et al. (10) suggested that the radiosensitivity of the mouse immature oocyte should be rather high, while our data indicated that the radiosensitivity of the guinea-pig immature oocyte is rather low.

All together, our results suggest that important differences exist between the mouse and the guinea-pig, with regard to the sensitivity of their female germ cells. This underlines the necessity of performing studies in other mammalian species, in order to better define the genetic risks associated with an exposure of women to radiation.

ACKNOWLEDGEMENTS

This work was supported by Research Contracts from the European Communities (Contract No. FI3P-CT920005) and from the Belgian Government (Contract No. ST/52/030)

REFERENCES

1. P. Jacquet, J. Vankerkom and M. Lambiet-Collier, *Int. J. Radiat. Biol.*, 65, 357-367 (1994).
2. P. Jacquet, L.de Saint-Georges, J. Vankerkom and L. Baugnet-Mahieu, *Mutation Res.*, 334, 309-316 (1995).
3. P. Jacquet, L. de Saint-Georges, J. Buset, J. Vankerkom and L. Baugnet-Mahieu, *Proc. 10th Int. Cong. Radiat. Res., Würzburg*, 1995, abstract P26-7.
4. E.F. Oakberg and E. Clark, in "*Effects of Radiation on the Reproductive System*", Carlson & Gassner eds., Pergamon Press, 1964, pp. 11-24.
5. B. Cox and M.F. Lyon, *Mutation Res.*, 28, 421-436 (1975).
6. A. Caine and M.F. Lyon, *Mutation Res.*, 45, 325-331 (1977).
7. W. Reichert, I. Hansmann and G. Röhrborn, *Humangenetik*, 28, 25-38 (1975).
8. C.S. Griffin and C. Tease, *Mutation Res.*, 202, 209-213 (1988).
9. C.S. Griffin, C. Tease and G. Fisher, *Mutation Res.*, 231, 137-142 (1990).
10. T. Straume, T.C. Kwan, L.S. Goldstein and R.L. Dobson, *Mutation Res.*, 248, 123-133 (1991).