V. U. Mme Claire Stievenart
Av. A. Huysmans 206, bte 10
1050 Bruxelles-Brussel

ISSN - 0250 - 5010

ANNALEN VAN DE BELGISCHE VERENIGING VOOR STRALINGSBESCHERMING

VOL. 43, N° 3, 2018

4e trim. 2018

5th European IRPA Congress
June 4-8, 2018
Den Haag, Nederland

Driemaandelijkse periodiek
1050 Brussel 5

Périodique trimestriel
1050 Bruxelles 5

ANNALES DE L'ASSOCIATION BELGE DE RADIOPROTECTION

Hoofdredacteur | Mr C. Steinkuhler | Rédacteur en chef
Rue de la Station 39
B- 1325 Longueville

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

Le cinquième congrès européen de l'IRPA (International Radiation Protection Association) a eu lieu à Den Haag du 4 au 8 juin 2018.
Ce numéro 3 du Vol. 43 des Annales de l'Association belge de Radioprotection reprend des articles d'auteurs belges.

Van 4 tot 8 juni vond te Den Haag het vijfde congres van IRPA (International Radiation Protection Association) plaats.
Dit nummer 3 van Vol. 43 van de Annalen van de Belgische Vereniging voor Stralingsbescherming herneemt artikels met belgische auteurs.

Sommaire – Inhoud

Vol.43/3/2018

5th European IRPA Congress
BELGISCHE BIJDRAGEN
PARTICIPATION BELGE

Annales de l'Association belge de Radioprotection, Vol 43, n°3, 2018
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.43, nr 3, 2018

NATURAL RADIOACTIVITY OF BUILDING MATERIALS IN BELGIUM: CURRENT SITUATION AND REGULATORY APPROACH

S. Pepin, G. Biermans, B. Dehandschutter[a], M. Sonck[b]

[a]Federal Agency for Nuclear Control,
rue Ravenstein 36, B-1000 Brussels, Belgium.
[b]ETRO, Vrije Universiteit Brussel, Brussels, Belgium.

Abstract
As all other EU member states, Belgium had to implement the relevant requirements of the EU BSS regarding natural radioactivity of building material. The Belgian radiation protection authority (FANC) organized in 2016-2017 a measurement campaign of 70 building materials. These building materials were selected taking into account the indicative list of annex XIII of the EU BSS; for imported natural stones, the selection was guided by the portal monitor measurements performed by the Customs on all containers in Belgian harbours. Based on the results of this and previous campaigns performed by other institutions, no building material of concern could be identified. The implementation in Belgian regulations of building materials aspects of EU BSS will thus focus on regular surveys of the natural radioactivity in building materials used in Belgium rather than on systematic measurements of specific categories of building materials. For the control of imported building materials, it will be striven for an increased collaboration with the Customs in order to more closely follow portal monitor data on building material containers in the harbours.

KEYWORDS: NORM, building material, regulations.

1. Introduction

The 2013/59/euratom BSS directive [1] sets a reference level of 1 mSv/a for external exposure to the natural radioactivity of building materials. It

asks member states to identify building materials of concern from radiation protection point of view taking into account an indicative list published in Annex XIII of the directive. For that purpose, the activity index I may be used as a conservative screening tool in the identification of the building materials which could induce an external exposure exceeding the reference level:

$$I = C_{Ra\text{-}226}/300 + C_{Th\text{-}232}/200 + C_{K\text{-}40}/3000 \quad (1)$$

If this index is higher than 1, a specific dose-assessment must be performed in order to compare with the reference level. A stepwise method for calculating this external dose has been proposed by the European Centre for Normalization (CEN) in a recently published technical report [8].

Studies on the radioactivity of Belgian building materials have already been carried out in the 1980s [2]. More recently, the University of Hasselt made a survey of more than 120 building materials on the Belgian market; this survey included natural stones, tiles, cement, concrete, bricks and gypsum. No one of the samples exceeded the reference level of 1 mSv/a [3] [4]. To complement these results, the Federal Agency for Nuclear Control (FANC), the Belgian radiation protection authority, performed another survey in 2016-2017.

2. Natural radioactivity of building material produced or imported in belgium: results of FANC survey

2.1 Overview of the campaign

The FANC survey included 73 samples which were analysed by gamma spectrometry in the Belgian laboratories of SCK-CEN and IRE-Elit. The samples were collected in collaboration with professional associations and companies from the building sector. They covered the categories listed in Table 1. Cement and natural stones were the categories for which the most samples were analysed. Cement samples included Portland CEM I cement (made essentially purely of clinker), composite cement CEM II and CEM V, blast furnace cement CEM III.

Imported natural stones were selected on basis of the portal monitor measurements performed by the customs in the Belgian harbours of Antwerp and Zeebrugge.

Table 1 shows an overview of the results of the laboratory measurements for the activity index I.

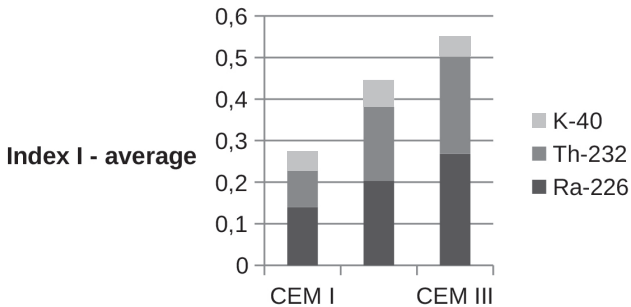
Table 1: *activity index I results for the different categories of building material*

Categories	# samples	I min	I max	I average
CEM I (Portland)	13	0.15	0.36	0.27
CEM II + V (composite)	11	0.31	0.61	0.45
CEM III (blast furnace)	14	0.41	0.69	0.55
aggregates	4	0.09	0.67	0.37
bricks	4	0.52	0.67	0.61
gypsum	6	0.05	0.46	0.24
Imported natural stones	21	0.59	1.47	1.08

2.2 Results for cement samples

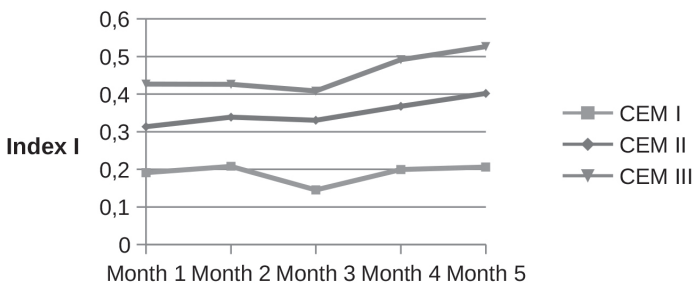
Annex XIII of the EU BSS lists materials containing fly ashes or blast furnace slag among the indicative list of building material potentially of concern. Fig. 1 shows the results for the average value of the activity index I for the three cement categories. It shows also the relative contribution of K-40, Th-232 and Ra-226. Cement incorporating fly ashes (CEM II and CEM V) or blast furnace slag (CEM III) have indeed a higher activity concentration in natural radioactive substances compared to CEM I cement but the activity index I stays lower than 1 for all cement categories.

Fig. 1: *average activity index for the three main categories of cement, CEM I, CEM II and V, and CEM III. The different colours correspond to the respective contribution of K-40, Th-232 and Ra-226*



In one factory, one sample of each cement category CEM I, CEM II and CEM III has been analysed for 5 consecutive months in order to check the temporal variation of the activity concentration. Results are displayed in Fig. 2. The index of CEM I is essentially constant on the 5 months period but there is some more variability (25 – 30%) in the activity index of CEM II and CEM III cement.

Fig. 2: *variation of activity index of CEM I, CEM II and CEM III on 5 consecutive months*



2.3 Bricks, aggregates, gypsum

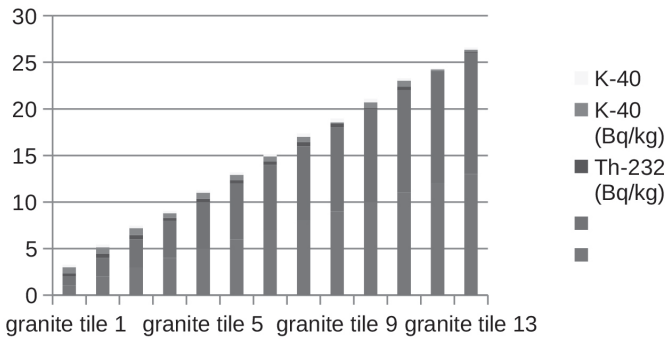
In Table 1, bricks have a higher activity index among the building materials used in bulk – essentially due to the contribution of K-40. The index I however does not exceed 0.67 - well below the reference level. The same is true for the aggregates, which included two samples of limestone and one of porphyry from Belgian quarries, as well as an expanded clay aggregate. Porphyry is cited in the indicative list of Annex XIII of the EU BSS but the Belgian sample has an activity index of 0.5, slightly higher than the limestones (I = 0.1 and 0.2) and slightly lower than the expanded clay (I = 0.67).

In Belgium, phosphogypsum from phosphate production is used as a building material. The activity concentration of this phosphogypsum is measured by the producer and is quite low due to the use of magmatic phosphate ore in the production process [5]. The results on the building material made of that phosphogypsum confirm these low values of radioactivity with an activity index which does not exceed 0.46.

2.4 Results for natural stones

The samples have been selected on basis of the level of gamma radiation measured on the portal monitor in the harbours of Antwerp and Zeebrugge [6]. Shipments of granite systematically presented the highest values on the portal monitor: 13 samples of granite tiles have been analysed. 2 slate tiles and one sandstone tile have also been selected. Fig. 3 shows the results for the activity index for the 13 granite samples. Although the majority of the samples have an activity index higher than 1, we will see in section 3 that the dose-impact stays largely below 1 mSv/a – these materials being devoted to superficial applications in buildings.

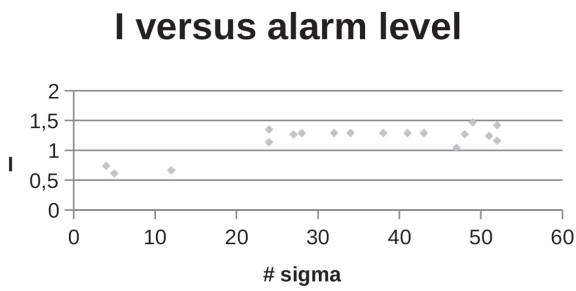
Fig.3: *activity index of the 13 samples granite tiles. The different colours correspond to the respective contribution of K-40, Th-232 and Ra-226.*



2.5 Correlation with portal monitor measurement

The alarm level on the portal monitor is expressed as a number of « sigma » (standard deviation of natural background). Fig. 4 shows the relation between the activity index of the material and the number of « sigma » on the portal monitor for the corresponding container. Materials with an activity index lower than 1 induce a lower signal on the portal monitor but the relation between the measurement on the portal monitor and the index I is not linear. For a same activity index (I ~ 1.3), the number of "sigma" may vary between 25 and 50. This is probably due to the numerous factors influencing the measure on the portal monitor: geometry of the shipment, density of the materials, speed of the truck, etc.

Fig.4: *activity index and corresponding measurement on the portal monitor*



2.6 “Inter-comparison” between laboratories

Some of the materials have also been analysed by the producer in another laboratory. Although it can not really be considered as an inter-comparison (the different laboratories analysed the same material but not necessarily the same sample), it still give an idea of the robustness of the results. The results are summarized in Table 2. The agreement between the measures performed by the different laboratories is satisfactory as the difference in the results didn’t exceed 10%.

Table 2: *activity index of some building materials as measured by three different laboratories*

Sample	Laboratory 1	Laboratory 2	Laboratory 3
Sample 1	0.65	0.62	0.65
Sample 2	0.67	0.66	0.66
Sample 3	0.59	0.54	0.55
Sample 4	0.52	0.51	0.52
Sample 5	0.72	0.7	-
Sample 6	0.41	0.46	-
Sample 7	0.41	0.39	-

3. Dose-assessment

Section 2 showed that only imported granite tiles have an activity index higher than 1. These granite tiles are superficial material and the activity index is obviously far too conservative to assess their effective dose-impact. The European Committee of Normalization (CEN) developed a formula [8] which takes into account the density and thickness of the material to assess the dose. Using this formula, the dose induced by the granite tile with the highest activity index (I=1.47) can already be shown as being lower than 1. This formula still is quite conservative as it assumes that the room is fully covered by the material. More realistic calculations can be performed e.g. using the tables of the CEN technical report and calculating the contribution of each building product and each structure of the room to the external dose. Alternatively, a specific calculation code such as RESRAD-BUILD may be applied. These calculations have been detailed in [7]. One assumes the standard CEN room with four walls made of bricks, a ceiling and a floor made of concrete; the floor is covered with

the granite tiles. The activity concentration for each of the building material is taken from the present measurements. The resulting external dose is 0.35 mSv/a (resp. 0.27 mSv/a) using the CEN method (resp. RESRAD-BUILD code) with inclusion of the granite floor tiles. Without granite tiles, the external dose will be 0.315 (resp. 0.21) mSv/a. The incremental dose due to the use of granite floor tiles is thus only of 30 – 60 µSv/a depending on the calculation method. In any case, the external dose in a room made of building materials typical for the Belgian market is only a fraction of the reference level.

4. Future regulations

The results of FANC survey and of previous studies didn't allow identifying any building material of concern in Belgium. Consequently, in its transposition of art. 75 of the EU BSS, Belgium didn't define any specific categories of building material and choose not to implement a general obligation of measurements for the producers or importers of building materials. However, FANC has defined the mechanisms needed to take action in case some building material of concern would nonetheless be identified. In particular, FANC developed mechanisms to insure a long-term follow-up of the issue of natural radioactivity in building material: it has integrated the radioactivity of building material in its radiological surveillance program. FANC intends to analyse around 40 samples of building materials each year in order to follow the evolution of natural radioactivity in building materials and of the corresponding public exposure. For the selection of these samples, FANC will collaborate with the ministry of Economy which is in charge of the control on the application of the Construction Products Regulations (CPR). FANC will also continue to work together with the customs which are in charge of the portal monitors installed in Belgian harbours. Anomalies on the radiation level of incoming shipments will be controlled.

5. Conclusions

Literature data and survey carried out by FANC didn't allow identifying any building material of concern in Belgium. Based on a standard room model, external exposure due to the investigated building material may

be estimated to approximately 0.3 mSv/a – well below the reference level of 1 mSv/a. Consequently, FANC will not impose a general obligation of measuring radioactivity for given categories of building materials. The follow-up of the exposure from building materials will rather be integrated in the national radiological surveillance program of FANC. These and additional controls and surveys will be undertaken in collaboration with other authorities, such as customs, which monitors the radioactivity of incoming sea shipments, and the ministry of Economy, which controls the application of the Construction Product Regulations.

6. References

[1] Council directive 2013/59/Euratom of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, and repealing Directives 89/618/Euratom, 90/641/Euratom, 96/29/Euratom, 97/43/Euratom and 2003/122/Euratom.

[2] Poffijn A., Bourgoignie, R., Marijins R., Uyttenhove J., Janssens A. & Jacobs R., "Laboratory measuments of radon exhalation and diffusion", Radiat. Prot. Dosim. vol.7, no.14, 7779, 1984;

[3] M. Stals, V. Pellens, B. Boeckx, P. Vannitsen, W. Schroeyers, S. Schreurs, "Eindrapport van het B-NORM project, Kennisverspreiding over de problematiek van natuurlijk voorkomende radioactiviteit in bouwmaterialen", 2013.

[4] M. Stals, S. Verhoeven, M. Bruggeman, V. Pellens,W. Schroeyers, S. Schreurs, "The use of portable equipment for the activity concentration index determination of building materials: method validation and survey of building materials on the Belgian market", J. Env. Rad. 127 (2014) 56.

[5] IAEA, Safety Report 78, ''Radiation Protection and management of NORM residues in the phosphate industry", 2013.

[6] V. Pellens, T. Clerckx, L. Hulshagen, W. Schroeyers, P. Fias, T. Peeters, F. Biermans, S. Schreurs, "Transport of NORM in the port of Antwerp: From megaports to a special purpose measurement methodology", Proceedings of NORM VI symposium, Marrakesh (Morocco), IAEA, 2010.

[7] S. Pepin, "Using RESRAD-BUILD to assess the external dose from the natural radioactivity of building materials", Construction and Building Materials 168 (2018) 1003–1007.

[8] Technical Report, CEN/TR 17113:2017 E, Radiation from Construction Products – Dose assessment of emitted gamma radiation, European Committee for Standardization (CEN), 2017.

Annales de l'Association belge de Radioprotection, Vol 43, n°3, 2018
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.43, nr 3, 2018

ASSESSING THE PUBLIC EXPOSURE RELATED TO THE USE OF NORM IN THE NEX TYPES OF BUILDING MATERALS

Wouter Schroeyers[a*], Tom Croymans[a], Zoltan Sas[a,b], Gergo Bator[c], Rosabianca Trevisi[d], Cristina Nuccetelli[e], Federica Leonardi[d], Sonja Schreurs[a], Tibor Kovacs[c]

[a]Hasselt University, CMK, NuTeC, Nuclear Technology - Faculty of Engineering Technology, Agoralaan building H, B-3590 Diepenbeek, Belgium
[b]School of Natural and Built Environment, Queen's University Belfast, David Keir Bldg., 39-123 Stranmillis Rd, Belfast BT9 5AG, United Kingdom
[c]University of Pannonia, Institute of Radiochemistry and Radioecology, H-8200, Egyetem St. 10., d Veszprém, Hungary
[d]INAIL (National Institute for Insurance against Accidents at Work)- Research Sector, DiMEILA, Via di Fontana Candida 1 00078 Monteporzio Catone (Rome), Italy
[e]ISS (National Institute of Health), Technology and Health Department, Viale Regina Elena, 299, Rome, Italy

**Wouter.schroeyers@uhasselt.be*

Abstract

For a safe reuse of Naturally Occurring Radioactive Materials (NORMs) in construction, it is of great importance to evaluate the radiological aspects of the reuse in addition to chemical, environmental, economic… aspects before the construction materials are introduced on the market. This is of particular importance for new types of construction materials, such as alkali activated materials, that allow the reuse of a large faction (wt%.) of residues. The Euratom BSS (basic safety standards) sets the requirement of the radiological evaluation of building materials that incorporate specific residues from NORM related industries. In the period 2014-2017, the COST Action Tu1301 NORM4Building initiated a lot of research on

the radiological evaluation of new types of construction material that are currently in the research state. In the course of the NORM4Building project a radiological database on NORM & building materials was developed. In addition, new dosimetrical tools were developed for a more realistic evaluate of the gamma dose related to the reuse of NORM in construction. These dosimetrical tools provide a more realistic radiological screening of the reuse of building materials in addition to the Activity Concentration Index (ACI) that is proposed by the EU-BSS as screening tool. In the current paper and linked presentation, the contents of the NORM4Building database will be presented next to the newly developed dosimetrical tools for the evaluation of the public exposure to gamma radiation from different types of building materials. The NORM4Building database is available via www.norm4building.org.

KEYWORDS Natural occurring radioactive materials, building materials, database, concrete, by-products, Euratom Basic Safety Standards

1. Introduction

Turning waste into resources is a key step on the Roadmap to a Resource Efficient Europe [1]. The recycled materials can however contain a measurable amount of natural occurring radionuclides such as ^{238}U, ^{232}Th and their decay products and ^{40}K and this aspect needs to be considered, particularly when the residues are included in building materials. Several industries that need to consider the precences of naturally occurring radioactive materials (NORMs) are listed in Annex VI of Council Directive 2013/59/Euratom [2]. An enhanced content of natural occuring radionuclides can be an issue for by-products such as fly ash from coal, peat and heavy oil fired power plants, phosphogypsum from phosphate industry, phosphorous slag from thermal phosphorus production, copper and tin slags from primary and secondary production, red mud from aluminium production and some residues from steel production. For the use of these by-products in building materials, the Council Directive 2013/59/Euratom (Euratom Basic Safety Standards; EU-BSS) [2] sets the requirement of the radiological evaluation of the produced building materials. In the EU-BSS, a screening parameter, the activity concentration index (ACI), is defined for the initial screening of the building materials incorporating NORM residues however the real criterion that determines if the use of

the considered residues in building materials is acceptable or not is the reference level of 1 mSv/year.

In the concrete industries, the considered residues are used in increased amounts as supplementary cementitious materials (SCMs) (as partial cement replacement or as mineral additions in concrete) and as aggregates [3]. In the ceramic industries metal smelting slags can be used as aggregates in clay-based ceramics [4]. In the bond system of clay ceramics residues, such as red mud, can be used [4]. Alternatives for cement and concrete using Alkali-Activated Materials (AAMs) are being developed. AAMs contain calcium silicate or a more aluminosilicate-rich precursor such as a fly or bottam ash, metallurgical slag or natural pozzolan, as solid aluminosilicate source [5].

In the period 2014-2017, the COST Action Tu1301 NORM4Building initiated a lot of research on the radiological evaluation of new types of construction material that are currently in the research state. In the course of the NORM4Building project a radiological database on NORM & building materials was developed. In addition, new dosimetrical tools were developed for the evaluation of the gamma dose related to the reuse of NORM in construction. These dosimetrical tools provide a more realistic radiological screening of the reuse of building materials in addition to the ACI that is proposed by the EU-BSS as screening tool. In the current paper (and linked presentation) the contents of the NORM4Building database will be presented with the scenarios used for the simulation of building materials incorporating NORM residues. In addition, newly developed more realistic dosimetrical tools for the evaluation of the public exposure to gamma radiation from different types of building materials are discussed.

2. Scenarios for incorporation of a by-product in concrete

Table 1 lists the compositions which were used to model the use of by-products in specific types of concrete.

Table 1 Description of concrete compositions used in the model compositions

Scenario ID	Construction Material	Composition (kg/m³)			
		Cement	By-product	Aggregates	Water
1	Reference concrete	400		1850	150
2	High volume fly ash (HVFA) concrete	160	220 (fly ash (FA))	1700	140
3	Concrete with FA as partial replacement of cement and sand'	320	130 (FA)	1750	150
4	Concrete with FA as partial replacement of sand	360	90 (FA)	1800	150
5	Concrete with slag as partial replacement of cement and aggregates'	80	720 (slag)	1850	150
6	Concrete with slag as partial replacement of cement	80	320 (slag)	1850	150
7	Concrete with slag as partial replacement of aggregates'	400	400 (slag)	1450	150
8	Alkali activated concrete containing red mud as partial replacement of cement and aggregates		1800 (red mud)	450	150

The activity concentration index (equation 1) [6] was calculated for several types of concrete using the compositions listed in table 1.

$$I-index = \frac{Ac_{226Ra}}{300Bq/kg} + \frac{Ac_{232Th}}{200Bq/kg} + \frac{Ac_{40K}}{3000Bq/kg} \qquad (1)$$

With Ac the activity concentration of the mentioned radionuclide expressed in Bq/kg. The average values of 0.38 and 0.45 were used in the calculations as I-indexes for respectively cement and soil/aggregates [7]. The results of the I-index calculation using the concentrations listed in Table 1 are described in detail in [8].

3. Databases to assess the use of NORM in construction

During the course of the COST Action NORM4Building several stategies to data collection and the verification of the collected were explored and

efforts were initiated to merge databases that contain data on NORM and the use of NORM in construction materials.

A lot of data on natural radioactivity in European building materials was collected by Trevisi et al. [7]. Recently, this dataset that mainly contained data on ^{226}Ra, ^{232}Th and ^{40}K activity concentrations of building materials in Europe was further enlarged and expanded with radon emanation and exhalation rates [9]. The data collection for the construction of this database and the verification of the collected results involved an extremely labour intensive process.

A new approach to data collection was developed in order to semi-automatically collect data from scientific publications. This appoach that relies on automated data mining via natural language processing and text link analysis is further described in [8]. This approach has important advantages: (1) hundreds of publications can be processed automatically at a monthly basis; (2) the approach allows a continuous (automated) search for newly published literature and is therefore very useful for keeping an inventory up to date; (3) the data search can be expanded or modified using different key-words which allows the construction of a more detailed and expanded database. In its current form, the approach has several limitations: (1) data from graphical images (eg.: histograms) is not collected; (2) the licence for datamining software is very expensive; (3) the reliability of the collected data is strongly dependent of the reliability of the included publications, an aspect that cannot be assessed by the automated datamining program, and therefore the validation of the results requires a labour intensive verification step; (4) the included publications enclose both papers that reported averaged results and papers that reported individual measurements in more detail. A drawback of this fact is that only a limited amount of statistical analysis can be applied on the collected data. On the basis of this approach the NORM4Building database was constructed and this database is available via www.norm4building.org. A large set of the data that is collected in this database is described in detail in [8] and [10].

To allow a more in-depth statistical analysis, a database that is purely based on individual measurement entries and not on average results

reported in literature was constructed by Sas et al. [11] ('By-BM database'). This database allows many interesting analysis and visualisation options. A future aim is to step by step investigate the data reported in the previously mentioned databases and to track the underlying 'individual measurement results' (if they can be found) and to incorporated these, after a verification step, in the By-BM database. The combination of the described approaches and databases can provide important added value especially if automated data collection can be combined with more in-depth statistical analysis options. The By-BM database is accessible online via http://bybmproject.com/ and the data included was discussed in [11].

4. Expanded set of screening tools for gamma dose assessment

For the assessment of gamma ray exposure from building materials several methods have been developed ranging from simple indices to more sophisticated Monte Carlo simulations [2][12][13][14]. In the dose assessment calculations based on gamma ray attenuation and build-up factors the density and wall thickness were identified as very critical parameters [15][16][17]. The approach implemented by the EU-BSS uses an Activity Concentration Index (ACI) [2] that does not include the density and wall thickness as modifiable parameters. Technical guide Radiation Protection (RP)-112 [6] describes the index, originally developed by Markkanen [18], in more detail. The index described in RP-112 assumes a standard room with dimensions 400 cm x 500 cm x 280 cm, uses the density of concrete (2350 kg/m^3) and assumes a thickness of 20 cm for walls, floors and ceilings. A screening method that takes into consideration density an thickness via a density and thickness corrected index I(ρd) was proposed by Nuccetelli et al. [19] Complementary to the methodology proposed by the EU-BSS, the technical report CEN/TR 17113:2017, potentially a precursor for the development of a harmonized European Standard, also included a more elaborate index that allows modifying the density and the thickness [20].

A new study described by Croymans et al. [21] provided a dose calculation assessment using the original dose calculation of Markkanen with an expanded set of gamma lines and a higher total gamma intensity. The developed model by Croymans et al. [21] that uses an expanded set of gamma lines is complementary to the existing ACI model, proposed in

the EU-BSS and the density and thickness corrected assessments proposed by Nuccetelli et al. [19] and CEN/TR 17113:2017 [20]. An initial screening can be based on the ACI proposed by the EU-BSS, especially useful in case that the building materials are thinner than 20 cm or lighter than 2350 kg/m^3. For building materials thicker than 20 cm or heavier than 2350 kg/m^3 it is advisable to use a density corrected assessment tool, especially useful for standard room sizes. The expanded gamma dose assessment method, allowing the assessment of non-standard rooms, can be used in specific cases. This model also allows considering the presence of doors and windows in the considered model.

5. Conclussion & outlook

A semi-automated database for screening, identifying materials of concern from a radiological perspective was set-up by the COST-Action NORM4Building. More realistic scenarios are proposed for assessing the impact of the use of NORM in building materials. Complementary tools for the evaluation of the gamma dose related to the use of NORM in building materials were developed.

The NORM4Building network joint forces with the EAN-NORM & EU-NORM networks to form the European NORM Association (ENA). Future aims involve the further integration of the developed databases and implementing the developed tools for gamma dose assessment in the online database.

6. Acknowledgement

The authors acknowledge the networking support by the COST Action TU1301. www.norm4building.org. The authors wish to thank the University of Pannonia for the management and setting up of the database and datamining approach. In addition, the authors would like to thank all the colleagues who helped creating and evaluating the database and are very grateful to University of Hasselt for support on accessing the e-Journals. This work was supported by the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 701932.

7. References

[1] European Commision, An EU action plan for the circular economy, Com. 614 (2015) 21. doi:10.1017/CBO9781107415324.004.

[2] EU, Council Directive 2013/59/Euratom, laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, and repealing Directives 89/618, 90/641, 96/29, 97/43 and 2003/122/Euratom, Off J Eur Commun L13. (2014). doi:10.3000/19770677.L_2013.124.eng.

[3] R. Siddique, M.I. Khan, Supplementary Cementing Materials, Springer, 2011.

[4] Y. Pontikes, G.N. Angelopoulos, Effect of firing atmosphere and soaking time on heavy clay ceramics with addition of Bayer's process bauxite residue, Adv. Appl. Ceram. 108 (2009) 50–56.

[5] C. Shi, P.V. Krivenko, D.M. Roy, Alkali-Activated Cements and Concretes, Taylor & Francis, 2006.

[6] EC, Radiological protection principles concerning the natural radioactivity of building materials - Radiation Protection 112, Eur. Comm. (1999) 1–16.

[7] R. Trevisi, S. Risica, M. D'Alessandro, D. Paradiso, C. Nuccetelli, Natural radioactivity in building materials in the European Union: A database and an estimate of radiological significance, J. Environ. Radioact. 105 (2012) 11–20. doi:10.1016/j.jenvrad.2011.10.001.

[8] W. Schroeyers, Z. Sas, G. Bator, R. Trevisi, C. Nuccetelli, F. Leonardi, S. Schreurs, T. Kovacs, The NORM4Building database, a tool for radiological assessment when using by-products in building materials, Constr. Build. Mater. (2017). doi:10.1016/j.conbuildmat.2017.11.037.

[9] C. Nuccetelli, S. Risica, S. Onisei, F. Leonardi, R. Trevisi, Natural radioactivity in building materials in the Eurpean Union: a database of activity concentrations, radon emanations and radon exhalation rates, Rapporti Istisan, 2018.

[10] Cost network NORM4Building, Naturally Occurring Radioactive Materials in Construction, 2017. https://www.elsevier.com/books/naturally-occurring-radioactive-materials-in-construction/schroeyers/978-0-08-102009-8.

[11] Z. Sas, R. Doherty, T. Kovacs, M. Soutsos, W. Sha, W. Schroeyers, Radiological evaluation of by-products used in construction and alternative applications; Part I. Preparation of a natural radioactivity database, Constr. Build. Mater. 150 (2017) 227–237. doi:10.1016/j.conbuildmat.2017.05.167.

[12] L. Koblinger, Calculation of exposure rates from gamma sources in walls of dwelling rooms, Health Phys. 34 (1978) 459–463.

[13] M. Zeeshan Anjum, S.M. Mirza, M. Tufail, N.M. Mirza, Z. Yasin, Natural radioactivity in building materials: dose determination in dwellings using hybrid Monte Carlo-deterministic approach, in: Int. Conf. Nucl. Data Sci. Technol., 2007: pp. 1–4.
[14] C. Nuccetelli, S. Risica, M.D. Alessandro, R. Trevisi, Natural radioactivity in building material in the European Union: robustness of the activity concentration index I and comparison with a room model, J. Radiol. Prot. 32 (2012) 349–358. doi:https://doi.org/10.1088/0952-4746/32/3/349.
[15] E. Stranden, Radioactivity of building materials and the gamma radiation in dwellings, Phys. Med. Biol. 24 (1979) 921–930.
[16] R.. Mustonen, Methods for evaluation of doses from building materials, Radiat. Prot. Dosimetry 7 (1984) 235–238., Radiat. Prot. Dosim. 7 (1984) 235–238.
[17] S. Risica, C. Bolzan, C. Nuccetelli, Radioactivity in building materials: room model analysis and experimental methods, Sci. Total Env. 272 (2001) 119–126.
[18] M. Markkanen, Radiation dose assessments for materials with elevated natural radioactivity, 1995.
[19] C. Nuccetelli, F. Leonardi, R. Trevisi, A new accurate and flexible index to assess the contribution of building materials to indoor gamma exposure, J. Environ. Radioact. 143 (2015) 70–75. doi:10.1016/j.jenvrad.2015.02.011.
[20] CEN, Construction products. Assessment of release of dangerous substances. Radiation from construction products. Dose assessment of emitted gamma radiationt, PD CEN/TR 17113:2017, 2017.
[21] T. Croymans, F. Leonardi, R. Trevisi, C. Nuccetelli, S. Schreurs, W. Schroeyers, Gamma exposure from building materials - A dose model with expanded gamma lines from naturally occurring radionuclides applicable in non-standard rooms, Constr. Build. Mater. (2017). doi:10.1016/j.conbuildmat.2017.10.051.

Annales de l'Association belge de Radioprotection, Vol 43, n°3, 2018
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.43, nr 3, 2018

WHAT IS THE RADIOLOGICAL/ECOLOGICAL IMPACT OF NORM RESIDUES AND EFFLUENTS ON THE ENVIRONMENT?

Hildegarde Vandenhove, Talal Al Mahaini[a], Lieve Sweeck[a]

[a]Belgian Nuclear Research Centre, Institute Environment Health and Safety, Mol, Belgium

Abstract

Some industrial activities such as oil and gas extraction, phosphate fertiliser production, ceramic production, coal combustion in power plants or mining and ore processing for the production of metals (tin, aluminum, …), geothermal energy production, … involve the use or generation of materials, usually regarded as non-radioactive but which contain naturally occurring radionuclides (NORM). NORM industries may be of radiological concern for the general public and the environment as a result of their discharges and wastes. We here present a short overview of the waste production processes and the radiological content of the raw materials, residues and discharges for the NORM industries that may require regulatory control. The main sources and pathways by which technologically enhanced radioactive materials can impact on man and environment and the methodologies to assess radiation doses to humans and wildlife are described. Taking an example case from the Belgian phosphate industry, we perform a preliminary radiological impact assessment for man and environment for the Veldhoven phosphate sludge deposit and the inundation areas of the Grote Laak.

KEYWORDS: NORM, TeNORM, Environmental impact assessment, phosphate industry

Introduction

NORM (TeNORM) impacts an important number of industries but not all are recognised as having NORM issues associated with them. The production and processing of uranium is well known as an industry affected by NORM. Most operations have radiation monitoring programs for occupational, public and environmental exposures and waste disposal is a concern due to the large quantities produced. We are now well aware that the production of oil and gas results in sludge and scales which contain enhanced quantities of ^{226}Ra. The disposal of these wastes has been problematic in some operations. Mineral sands typically have a large amount of thorium present. The production of phosphates for fertiliser and other uses may have uranium and radium issues including scales and waste disposal issues (phosphogypsum waste dumps) and so forth.

Depending on the NORM industry and waste stream, there may be different radionuclides of major concern and a range in radionuclide concentrations exist: e.g. ^{226}Ra is present in fairly low levels of ~1 kBq/kg in phosphogypsum sludge from the phosphate industry up to huge concentrations (10^6 kBq/kg) in scales in tubing of the petroleum industry. NORM and TeNORM wastes come in different physical forms: as waste water from oil and gas production, as sludge from phosphate-fertiliser production, water treatment or metal processing, as scales in oil and gas and phosphate industry, as ashes and slag from metal processing, coal industry or as waste rock. Also non-radioactive hazardous components need to be considered when evaluating the potential impact from the NORM industries [1, 2]. Non- radiological parameters may also drive the dispersion of radioactive contaminations (e.g. pH, ground water head, sulphuric acid content).

During operation the most important release mechanisms are dust emission and release of ^{210}Pb and ^{210}Po from stacks from smelters or furnaces [3], release of waste streams to rivers and seas in case of sea dumping of radium scales from the oil and gas industry, sea or river dumping of $CaCl_2$ from the phosphate industry and routine releases of process water for example in case of coal mining and geothermal energy production. Releases of radioactivity from NORM disposal sites occurs via dissolution of radionuclides present in the leachate and discharge to

ground and surface water, release of radon and decay products from the waste heap, the emission of dust. Exposure from NORM sites occurs via atmospheric, terrestrial and aquatic pathways. The inhalation of radon and the subsequent deposition of radon decay products in the lungs and inhalation of dust is one of the major pathways by which occupational exposure can occur. Radioactivity in air can also lead to external exposure. Exposure via the terrestrial and aquatic pathway is possible by ingestion of contaminated foodstuffs and water and via external irradiation. There are only few studies that evaluate the exposure of the public from NORM sites and liabilities (e.g. [4, 5]).

Radiation exposures resulting from the mining and processing of raw materials containing NORM are required to be controlled through a system of radiation protection. The requirement of the IAEA Basic Safety Standards (BSS) for the optimisation of protection and safety [6] (§ 1.6) specifies: "…the system of protection and safety aims to assess, manage and control exposure to radiation so that radiation risks, including risks of health effects and risks to the environment, are reduced to the extent reasonably achievable." The IAEA BSS also state that (§ 1.35) "These Standards are designed to identify the protection of the environment as an issue necessitating assessment, while allowing for flexibility in incorporating into decision making processes the results of environmental assessments that are commensurate with the radiation risks."

In the next paragraphs we provide a preliminary assessment of the public exposure from a waste disposal site from the phosphate industry and of public and environmental exposure in inundation areas of rivers to which $CaCl_2$ from the phosphate industry was released.

2. Tier 1 impact study for the phosphate industry: example case Tessenderlo chemie.

From 1920 to 2013, phosphate ore processing was an important activity of the Tessenderlo group. Phosphate ores mostly from Moroccan origin, were converted into dicalcium phosphates via an hydrochloric acid leaching process. These industrial activities have generated a huge amount of NORM waste, predominantly CaF_2 sludge, disposed of in several

landfills. The Veldhoven landfill covers 55 ha and is still partly in operation. $CaCl_2$ is formed as an effluent and until 1995, 70% of the ^{226}Ra in the raw phosphate was released to the Grote Laak or Winterbeek rivers, leading to an accumulation of radium in the bed sediment. By introducing a $BaCl_2$ precipitation step, radium solubility was decreased and since then most radium is disposed on the CaF_2 dump. Due to flooding and dredging, the river banks of the Grote Laak became contaminated. Here, we discuss the dose impact of the 'Veldhoven' landfill and the contaminated river banks of the Grote Laak. The impact assessment focuses on the main exposure pathways. For the inundation areas of the Grote Laak we also assess the impact on fauna and flora.

2.1 Site description

2.1.1. Veldhoven landfill

The landfill 'Veldhoven' contains about 9 million m^3 sludge. The local water table is at a depth of 2 m below land surface. The average ^{226}Ra concentration is approximately 3.5 kBq/kg. Dose rates measured at surface are up to a few µSv/h. In 1993, the radon concentrations varied from 15 Bq/m^3 to 60 Bq/m^3 (background is about 10 Bq/m^3). Similar values are found for the period 2010 – 2014. Between 2004 and 2009, the total alpha activity in groundwater samples of the Veldhoven site were measured using two piezometers. These activities ranged from values below 15 mBq/l to a maximum value of 139 mBq/l and are a factor of 2 to 10 higher than the ^{226}Ra measurements carried out in March 1993 [7-9].

Figure 1: The Veldhoven landfill

2.1.2. River banks of the Grote Laak river

The Grote Laak is a small river with a catchment of about 125 km^2. About 14.5 km from the discharge point of the liquid waste, the river flows into the Grote Nete river. Until 1990, the average ^{226}Ra concentration of the waste water was 20 Bq/l. Following the introduction of a $BaSO_4$ step, the concentration was reduced to 3 Bq/l. Floodings and placing dredged bed sediment on the river banks have led to a contamination of banks and flood plains. Dose rates up to 1 μSv/h were measured on the river banks. ^{226}Ra concentrations on the right river bank (more contaminated) vary from 400 Bq/kg to 2000 Bq/kg (average 800 Bq/kg) [10]. The radon concentrations in dwellings less than 100 m from the river vary from 19 Bq/m^3 to 134 Bq/m^3.

2.2. Human radiological impact assessment

2.2.1. Scenarios for impact assessment

Veldhoven disposal site: Two possible future exposure scenarios were evaluated for the 'Veldhoven' landfill. The first radiological impact scenario is the 'well' scenario. It describes the normal evolution of the landfill. Radionuclides leach out into the groundwater and are transported into a well at 50 m from the disposal site. The representative person is a self-sustaining farmer who uses the well water for drinking, irrigation of crops and watering cattle. All his food is coming from the contaminated site and he spends 1800 h/y on the field. This is a conservative approach. The second scenario is the 'residence' scenario. It is assumed that a house will be built on the site and that there is no cover on the site. The representative person is an inhabitant with a kitchen garden on the contaminated soil who spends 100% of his time (20% outdoors, 80% indoors) on the site. This is the worst-case scenario.

Contamination of the surroundings of the Grote Laak: Two exposure scenarios are assumed for the contamination of river banks: a recreational scenario and a residence scenario. In the recreational scenario we assume that people spend 2 hours per day on the river banks (e.g. walking, fishing). In the residence scenario, we consider the houses built at less than 100 m from the river banks and assume, similar to the aforementioned residence

scenario, that the inhabitants have a kitchen garden on the contaminated area and spend 100% of their time on the contaminated area.

2.2.2. Approach to human dose assessment

We used the standard advection-dispersion transport module in the HYDRUS-1D code [11] to estimate the amount of radionuclides leached from the sludge to the underlying aquifer due to rain water infiltration. The SCK•CEN biosphere model [12] was used for the calculation of the contamination of the food chain and dose to humans. The biosphere assessment model is a representation of radionuclide transfer mechanisms in the biosphere, along with related assumptions and simplifications. The model is used for calculating the concentrations in the different biosphere compartments and the dose to representative persons living in the biosphere. A schematic presentation of the model is given in Fig. 2.

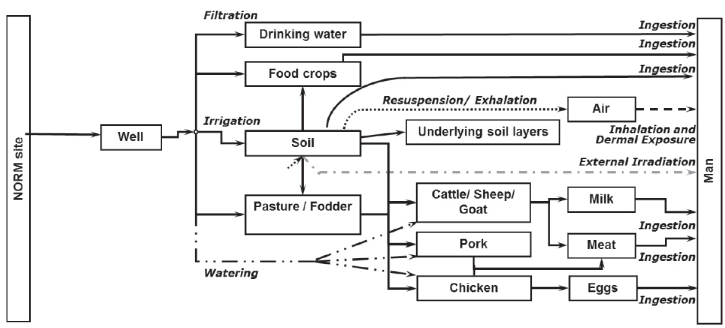


Figure 2: Conceptual representation of the Biosphere model

The HYDRUS-1D code was set up assuming the profile composition presented in Table Table 1. The physical and hydraulic properties of the profile materials were adapted from [13]. Water flux through the waste (i.e. the upper boundary) was set equal to the long-term average infiltration rate at the site (100 mm/y). Assuming an average bulk density of 1500 kg/m³, the height of the sludge layer is about 16 m above land surface. The activity concentration of ^{226}Ra has been previously determined in sludge samples (3.5 Bq/kg). We assume that the mother ^{238}U and daughters ^{210}Pb and ^{210}Po are in secular equilibrium. Sorption of radionuclides onto profile materials was quantified using the solid-liquid distribution

coefficient (K_d). K_d values of the radionuclides of interested are given in Table 2. After leaching to the groundwater, the radionuclides in the sludge were transported (through advection and dispersion) with the groundwater from the landfill site to a well located 50 m down-gradient. The hydraulic and chemical properties of the aquifer material were assumed identical to those of the sand in Tables 1 and 2. The maximum activity concentration in the leachate was used in the groundwater transport calculations. Dilution in of the leachate in the aquifer was accounted for by applying a dilution factor calculated as suggested by [14].
The biosphere parameters used to calculate the environmental transfer of radionuclides and human dose impact are derived from the database compiled for NIRAS/ONDRAF [15-17].

Table 1: Hydraulic parameters used in HYDRUS-1D to simulate infiltration of rain water through the sludge and leaching of radioηnuclides to the aquifer. ρb: bulk density, θr & θs residual and saturated moisture content, η: shape parameter, Ks: saturated hydraulic conductivity. Values adapted from Mallants [13].

	Thickness (m)	ρ_b kg m^{-3}	θ_r (m^3 m^{-3})	θ_s (m^3 m^{-3})	α (m^{-1})	η (-)	*Ks* (m d^{-1})
Sludge	16	1500	0.06	0.48	1.6	1.57	0.5
Sand	2	1300	0.03	0.33	7.44	2.69	285

Table 2: Kd values (m3/kg) used in the HYDRUS-1D simulation runs [15].

2.2.3. Results for the human dose assessment

^{238}U	^{226}Ra	^{210}Pb	^{210}Po
1.28	0.52	0.14	0.033

Results for the Veldhoven disposal site: The results for the well scenario and residence scenario are given in Tables 3 and 4. The dose results for the well scenario are conservatively estimated and are maximal doses. Realistic doses are about a factor of 10 lower. The results for ^{226}Ra in both tables also include the exposure to the daughters ^{220}Pb and ^{210}Po. The total dose of 1.05 mSv/y is mainly due to ^{238}U, because during the transport in the groundwater it decays much slower than ^{226}Ra, resulting in a higher maximum concentration in the well at 50 m from the waste disposal. The dose due to inhalation for ^{226}Ra is mainly due to the inhalation of radon

Table 3: Human dose impact (mSv/y) for the well scenario in vicinity of the Veldhoven sludge basin.

Radionuclide	External irradiation	Inhalation	Ingestion				Total dose
			water	crops	milk, meat	soil	
^{226}Ra	2.15E-08	1.34E-08	7.09E-05	1.31E-05	3.81E-06	3.90E-07	8.82E-05
^{238}U	2.01E-04	6.86E-05	2.65E-03	6.46E-03	1.10E-03	8.03E-05	1.04E-02
							1.05E-02

(> 97% of the dose). The results are conservative, since we assume that all food is coming from the contaminated area. It can be expected that a realistic predicted dose is at least 10 times lower.

The results for the residence scenario show that the dose is mainly due to the radon inhalation indoors. This scenario is a worst-case scenario, assuming no remediation actions and subsistence farming. The external dose outdoors is also higher than 1. It is clear from these results that the access to the site should remain restricted during at least the next hundreds of years and a cover on the site is recommended.

Impact from residing at the contaminated river banks of the Grote Laak: The results for the recreational and residence scenario are given in Tables 5 and 6. The results for ^{226}Ra in both tables also include the dose due to exposure to the daughters ^{220}Pb and ^{210}Po. The total dose for the recreational scenario is mainly due to the external radiation of ^{226}Ra (Table 5). For the dose calculation from indoor Rn for the residence scenario, the measured average Rn concentration of 38 Bq/m^3 in houses less than 100 m from the river banks is used. Other dose results are calculated using the average measured ^{226}Ra concentration of 800 Bq/kg soil. It is seen that also for this scenario, the Rn inhalation gives the highest dose, although the differences with the other exposure pathways are less pronounced.

Table 4: Human dose impact (mSv/y) for the residence scenario at the Veldhoven sludge basin.

Radionuclide	External irradiation		Inhalation		Rn Exhalation		Ingestion		Total dose
	outdoors	indoors	outdoors	indoors	outdoors	indoors	crops	soil	
^{226}Ra	1.55E+00	2.01E+00	1.52E-03	1.85E-03	3.87E-01	3.45E+01	1.95E+00	2.17E-01	4.06E+01
^{238}U	1.86E-02	2.42E-02	6.60E-04	8.02E-04	/	/	5.76E-01	6.18E-03	6.26E-01
									4.12E+01

Table 5: Human dose impact (mSv/y) for the recreational scenario for the Grote Laak river banks.

Radionuclide	External irradiation	Inhalation	Rn Exhalation	Total dose
^{226}Ra	1.46E-01	1.43E-04	3.63E-02	1.82E-01
^{238}U	1.75E-03	6.21E-05	/	1.81E-03
				1.84E-01

Table 6: Human dose impact (mSv/y) for the residence scenario at the Grote Laak river banks.

RN	External irradiation		Inhalation		Rn Exhalation		Ingestion		Total dose
	outdoors	indoors	outdoors	indoors	outdoors	Indoors	crops	soil	
^{226}Ra	3.59E-01	4.66E-01	3.53E-04	4.29E-04	8.96E-02	9.58E-01	5.02E-02	4.52E-01	2.37E+00
^{238}U	4.32E-03	5.60E-03	1.53E-04	1.86E-04	/	/	1.43E-03	1.33E-01	1.45E-01
									2.51E+00

2.2.4. Conclusions for the human impact assessment

Predicted dose rates for an expected evolution scenario (well scenario) in the vicinity of the disposal site are well below the 1 mSv/a dose criterion for the general public. Given the (extremely) long half-life of ^{238}U and ^{226}Ra, an intrusion scenario is likely to occur with time. Covers will not last the time span of the radionuclides' half-life and good record keeping is a requirement. But this also holds for toxic and hazardous conventional waste. Residence on the contaminated river borders may lead to dose rates slightly over 1 mSv/a but still within the 1-20 mSv/a range proposed by IAEA [6] as reference annual dose rate range in respect to exposure to a representative person in existing exposure situations. For the Grote Laak and Winterbeek, a decision has been made to decontaminate the river borders by soil removal.

2.3. Environmental impact assessment

2.3.1. Approach

Impact assessment to wildlife was performed for the terrestrial and aquatic ecosystem. For the terrestrial environment, an assessment was done considering the average ^{226}Ra concentration at the right bank of 800 Bq/kg and the average ^{226}Ra concentration of the hot spots (5800 Bq/kg). For the aquatic ecosystem, the sediment and water concentrations of 1999 were applied (averages: 0.18 Bq/l and 528 Bq/kg ^{226}Ra and maxima:

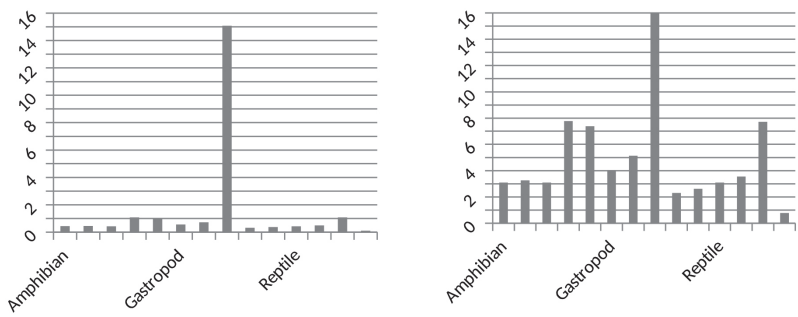
0.43 Bq/l and 902 Bq/kg ^{226}Ra). For ^{210}Po and ^{210}Pb, concentrations in soil or sediment are assumed to be at 80 % of the ^{226}Ra concentration.

2.3.2. Results

Terrestrial ecosystems: In Fig. 3, RQs are presented for the terrestrial wildlife exposed to activity concentrations monitored on the river banks of the Grote Laak. RQs were derived for average concentrations observed and averages of hot spots (highest dose rate locations). Fig. 3 shows that the average soil concentrations are unlikely to impact the terrestrial fauna and flora living on the river bank of the Grote Laak. For detritivorous invertebrates and soil invertebrates, RQs are slightly above one but effects data provided within the ERICA tool (which actually contains a link to a database of radiation dose effects [20, 21] show no effects for these organisms at the associated dose rates.

Dose rates to lichens and bryophytes are predicted to be 150 µGy/h for average soil activity concentrations. They exceed the screening dose rate applicable for organisms in a terrestrial ecosystem by a factor of 15. There are no effects data for lichens and bryophytes available in the FREDERICA database for these or higher dose rates. ^{226}Ra is the highest contributor to the total dose rate, except for lichens and bryophytes. The high dose rate for the latter species are due to the high concentration ratio of ^{210}Po (CR = 6 kg/kg), reflecting the high bioavailability of ^{210}Po for these organisms. For all scenarios analysed, dose rates were almost entirely due to internal exposure with ^{226}Ra and ^{210}Po contributing most to the dose (not shown).

Figure 3: Best estimate RQs for the terrestrial wildlife exposed to activity concentrations monitored on the river banks of the Grote Laak. RQs were derived for average concentrations observed (left) and averages of hot spots (highest dose rate locations) (right – RQ for Lichens and Bryophytes is 108).



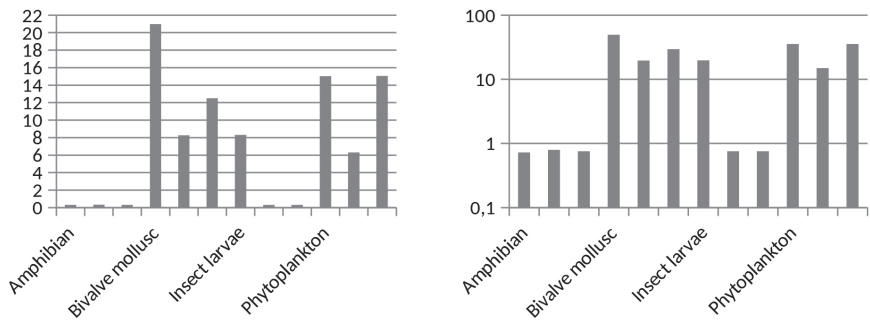
When considering average hot-spot concentrations, however, all RQs exceed 1, except for trees. At the related dose rates, there were either no FREDERICA effects data available or no statistically significant effects reported. Only some minor effects (generally related to mutations) are reported by FREDERICA at the associated dose rates for grasses and herbs, shrubs and mammals. For Bird, Rat, Deer, Grass and Tree, the predicted dose rates fall within the respective Derived Consideration Reference Level (DCRL) range proposed by ICRP [22] (a DRCL is considered by ICRP as a band of dose within which certain effects have been noted or might be expected).

Aquatic ecosystems: The situation is very different for the aquatic environment of the Grote Laak. High RQs are obtained for the 4 years monitored even for the reported average concentrations (Fig. 4). For 7 groups of reference organisms, the predicted dose rate exceeds the screening dose rate of 10 μGy/h. For all scenarios analysed, dose rates were almost entirely due to internal exposure and ^{226}Ra and ^{210}Po were contributing most to the dose (results not shown). Within a factor of 2 to 3 of the dose rates predicted for bivalve molluscs and gastropods (same effects data in FREDERICA database), some deleterious effects have been observed as reported in the FREDERICA database. For example, for oyster, a two-fold increase in frequency of abnormal larvae was reported at a dose rate of 125 μGy/h. For insect larvae, no effects were observed up to a dose rate of 200 μGy/h. For snails exposed to 270 μGy/h there are observations of either hormetic effects or decrease in capsules per snail. For all other organisms for which RQ>1, either no effects are reported for the dose rates obtained or no effects data are provided by FREDERICA.

Under the EU Euratom PROTECT project, organism group specific radiological benchmark values were derived for screening purposes [19, 23]. For plants, a screening value of 70 μGy/h was derived, and for invertebrates a screening value of 200 μGy/h was proposed. As these authors state, there are insufficient data to recommend values for specific organisms rather than groups of organisms. Even if these higher screening values are applied, RQs would still be higher than one. If we would consider the organism specific screening value for vertebrates of 2 μGy/h derived by Andersson [19, 23], the RQ would exceed 1 for birds, fish and

mammals. Dose rates predicted for these organisms when considering maximal concentrations would also be slightly higher than the lowest DCRL level proposed by ICRP [22]. As for terrestrial ecosystems, internal exposure appears dominant. ^{210}Po followed by ^{226}Ra were the highest dose contributors (results not shown). As expected, RQs are much higher for the maximum concentrations.

Figure 4: Best estimate RQs for aquatic wildlife exposed to activity concentrations monitored in river water and sediments of the Grote Laak (1999). RQs were derived for average concentrations (left) and for maxima (right).



2.3.3. Conclusions

This screening environmental risk assessment for presented phosphate industry case study shows that ^{226}Ra and ^{210}Po are the most important contributors to the wildlife dose and that the dose rate is almost entirely determined by internal dose rate. Since the best estimate RQs are higher than 1 for the Grote Laak river system impacted by the releases from the phosphate-fertiliser plant of Tessenderlo, it is advisable to assess the potential impact on the environment further, following a more detailed assessment with site-specific data including representative organism selection from a survey of the local biosphere and transfer parameters for local fauna and flora, as advised by the ERICA methodology.

3. General conclusions

As general conclusion it may be stated that the human and environmental impact assessment for NORM liabilities and legacy sites requires a site specific approach. The number of dedicated studies on public exposure

is rather limited, though it is clear that some exposure situations need a critical risk assessment. NORM is very long lived and impacts cannot only be considered in the short term but must include the potential impact for future generations. Long-term impact assessment, stewardship, long-term memory and long-term efficacy of remedial options are key for a robust management of NORM legacy sites.

4. References

[1] Wiegers R., Roelofs L., Kugeler E.A., 2000. Feasibility study on the technical and economic aspects of treating naturally occurring waste materials (NORM). Rep. 19130EN. Commission of the European Communities, Luxemburg.

[2] Schmidt G., Kuppens C., Robinson P., 1995. Handling of radium and uranium contaminated waste piles and other wastes from phosphate ore processing. ETNU-CT92-0084.

[3] Vandenhove H., Zeevaert T, Bousher A., et al., 2000. Investigation of a possible basis for a common approach with regard to the restoration of areas affected by lasting radiation exposure as a result of past or old practice or work activity – CARE. Final report for EC-DG XI-project 96-ET-006, Radiation Protection 115. Office for Official Publication by the European Communities, Luxemburg.

[4] Harvey M.P., Hipkin J., Simmonds J.R., et al., 1994. Radiological consequences of waste arising with enhanced natural radioactivity content from special metal and ceramic processes. Report EUR 15613. European Commission: Environment, Nuclear Safety and Civil Protection.

[5] Mead S., Wade B.O., 1996. Materials containing natural radionuclides in enhanced concentrations. B4-370/95/000387/mar/c3.

[6] IAEA, 2014. Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards. IAEA Safety Standards Series No. GSR Part 3. International Atomic Energy Agency, Vienna.

[7] Vanmarcke H., Zeevaert T., Govaerts P., 1993. Dosisevaluatie van het slibbekken Veldhoven. Studie uitgevoerd in opdracht van Tessenderlo Chemie.

[8] FANC/AFCN, 2011. Radiological monitoring in Belgium. Summary report 2011.

[9] FANC/AFCN, 2012. Radiological monitoring in Belgium. Summary report 2012.

[10] Vanmarcke H., Paridaens J., 1999. Onderzoek naar de radiumbesmetting van de Grote Laak. Studie uitgevoerd in de opdracht van het Ministrerie

van de Vlaamse Gemeenschap, department Leefmilieu en Infrastructuur AMINAKL afdeling Water.
[11] Šimůnek J., Šejna M., Saito H., et al., 2013. The HYDRUS-1D Software Package for Simulating the One-Dimensional Movement of Water, Heat and Multiple Solutes in Variably-Saturated Media, Version 4.17. Department of Environmental Sciences, University of California Riverside, Riverside, California, USA.
[12] NIRAS/ONDRAF, 2011. Biosphere model report - Project near surface disposal of category A waste at Dessel.NIROND-TR 2008–19E V2.
[13] Mallants D., Jacques D., Zeevaert T., 2004. Evaluatie van de mogelijke impact van de berging van zeer laagactief afval aan de oppervlakte in de nabijheid van de D1-stortplaats van UMICORE-OLEN. SCK•CEN Restricted report No. R-3772.
[14] Wiedemeier, T.H., Rifai, H.S., Newell, C.J., et al., 1999. Attenuation of sources and formation of plumes, in: Natural Attenuation of Fuels and Chlorinated Solvents in the Subsurface. John Wiley & Sons, Inc.
[15] NIRAS/ONDRAF, 2010a. Element dependent environmental input parameters for the biosphere model. NIROND-TR 2008-26E V2.
[16] NIRAS/ONDRAF, 2010b. Compendium of dose coefficients and related quantities for assessing human exposure. NIROND-TR 2008-27E V2.
[17] NIRAS/ONDRAF, 2012. Element independent biosphere parameters. Project near surface disposal of category A waste at Dessel. NIROND-TR 2008–28E V2
[18] Brown J.E., Alfonso B., Avila R., et al., 2008. The ERICA Tool. J Environ Radioactiv 99, 1371-1383.
[19] Andersson P., Garnier-Laplace J., Beresford N.A., et al., 2009. Protection of the environment from ionising radiation in a regulatory context (protect): proposed numerical benchmark values.
[20] FREDERICA, 2006. FREDERICA Radiation Effects Database. 2012. http://www.frederica-online.org/mainpage.asp.
[21] Copplestone D., Hingston J.L., Real A., 2008. The development and purpose of the FREDERICA radiation effects database. J Environ Radioact 99, 1456-1463.
[22] ICRP, 2008. Environmental protection: the concept and use of reference animals and plants. ICRP Publication 108. Ann. ICRP 38 (4–6).
[23] Andersson P., Beaugelin-Seiller K., Beresford N.A., et al., 2008. Numerical benchmarks for protecting biota against radiation in the environment: proposed levels and underlying reasoning. EC project N° FI6R-03642 Deliverable 5b. European Commission, Brussels.

Annales de l'Association belge de Radioprotection, Vol 43, n°3, 2018
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.43, nr 3, 2018

MONITORING OF NORM IN SECONDARY RAW MATERIALS FROM THE NON-FERROUS METALLURGY IN BELGIUM

S. Pepin, G. Biermans, B. Dehandschutter[a], M. Sonck[b]

[a]Federal Agency for Nuclear Control, rue Ravenstein 36,
B-1000 Brussels, Belgium.
[b]ETRO, Vrije Universiteit Brussel, Brussels, Belgium.

Abstract

Non-ferrous metallurgy in Belgium uses a large range of secondary raw materials, some of which may show an enhanced concentration in natural nuclides. Identifying and characterizing the raw materials which may be of concern from a radiation protection point of view is often a challenge and requires in most cases the development of a cost-efficient screening approach.

Non-ferrous metallurgy has been included since 2012 in the "positive list" of NORM sectors subject to notification to the Belgian radiation protection authority (FANC): a short overview is given of the different types of non-ferrous metallurgy in Belgium (production of Sn, Pb, Co, Zn, noble metals,… through both pyro- and hydrometallurgical processes), of the type of secondary raw materials involved and of their radiological characteristics. Challenges regarding radiological screening and analysis (such as detection and characterization of enhanced Pb-210 and Po-210 activity concentration) will be discussed.

From a regulatory perspective, this large range of exposure circumstances exemplifies the need of a graded-approach to the issue.

KEYWORDS: NORM industry, non-ferrous metal, secondary raw materials, screening measurement.

1. Introduction

Extraction of the non-ferrous metals tin, lead and copper is cited as a potential NORM industry in the indicative list of annex VI of the 2013/59/euratom directive (EU BSS) [1] as well as in IAEA Safety Report N°49 [2] (which also explicitly includes zinc and aluminium). These activities however cover a wide range of practices ranging from direct metal extraction from ores or concentrates to recycling or metal extraction from a large set of secondary raw materials. An overview of the non-ferrous metallurgy in Europe, its processes, its raw materials, etc. has recently been published by the Joint Research Centre of the European Commission [3].
This variety of processes and raw materials constitutes challenges for the identification of NORM issues in these sectors, as it is often difficult to identify beforehand which processes or raw material may or may not be of concern. In the processing of secondary raw materials, an additional challenge is due to the general absence of secular equilibrium in the materials, making also the identification of the most relevant nuclides challenging.

2. Overview of non-ferrous metal industry in Belgium

Since 2012, FANC has included all non-ferrous metal production activities in its "positive" list of NORM industries. These industries must introduce a declaration to FANC which allows identifying potential NORM issue and assessing the risk of exposure for the workers and the public. In case risks of exposure to NORM are identified, FANC may impose "corrective measures" in order to make sure that the exposure stays below the level of 1 mSv/a. Details on the Belgian regulatory framework for NORM activities and residues may be found e.g. in [4].

Nine non-ferrous metal companies have introduced a declaration to FANC. The range of metals produced is large; when some of them only produces a few main metals, some other produces a large spectrum of metals: next to the most common, such as zinc, lead, copper or tin, other substances like chrome, bismuth, antimony, molybdenum, cobalt, germanium or noble metals are also produced. Different processes are being used, both pyro-metallurgical and hydrometallurgical. Several companies combine pyro- and hydro-metallurgical processes depending on the type of metal produced. The raw material may be processed thermally in different steps,

including e.g. roasting or smelting, followed by chemical or electrolytic separation and purification. In hydrometallurgical processes, the materials are essentially chemically processed through reaction with acid and a set of other chemical reactions.

After review of their raw materials, processes and residues, NORM issues were deemed to be significant in three of these companies and FANC imposed them some corrective measures. These measures consisted essentially in the obligation of implementing a measurement procedure for their raw materials and relevant residues and to report the results to FANC. How these companies tackle the measurements procedure will be described in the next section.

Table 1 gives an overview of some raw materials presenting an enhanced concentration of natural nuclides. The main nuclides of concern in these materials and the range of their activity concentration are also indicated. It must be noted that these examples are not necessarily representative for the majority of raw materials used in the process. Generally, these specific raw materials are mixed with other materials and the impact of their processing on the end-products or on the residues is in most cases limited.

Table 1: some raw materials used in non-ferrous metal extraction and their activity concentration

	Raw material	**Main nuclide of concern**	**Range of activity concentration (kBq/kg)**
Sn production	Cassiterite	U-238sec Th-232sec	Up to 50
Sn/Pb production	Sn/Pb ingot from primay tin extraction	Pb-210 Po-210	Up to 600
Co production	Co concentrate	U-238 (without progenies)	0.1 up to 10
Cu production	Copper cement[1] (from Zn or Co production)	U-238 (without progenies)	1 up to 50
Zn and Pb production	Flue dust from primary Zn production	(Pb-210)	Up to 0.6
		Po-210	Up to 4
	Residues from flue dust leaching	Pb-210	15
		Po-210	80

1 Cobalt cement may contain similar or higher activity concentration

In many cases, the residues of one process will be the raw material of another: e.g. copper cement may be produced by zinc or cobalt production factories as a result of the cementation of a leaching solution and will be sold as a raw material for copper production.

In the case of cobalt production, uranium is present in the raw material coming from mining operation. Due to the pretreatment of the ores, the progenies of uranium are not present anymore. Moreover, the concentration of uranium displays a significant variability, from less than 10 ppm up to 600 ppm. In the production process, the uranium precipitates into an iron residue with an activity concentration which in some cases may reach 25 Bq/g U-238. This residue needs then to be disposed of on a landfill authorized by FANC for the acceptance of NORM residues.

Some of the non-ferrous metal production companies also face issues regarding orphan sources in their raw materials. Scrap non-ferrous metals may constitute a significant part of their raw materials or they may be confronted to the consequences of the melting of an orphan source by one of their suppliers of secondary raw materials (contaminating the resulting flue dust). Several cases of contamination of secondary raw material, such as flue dust, with Cs-137 have been reported.

3. Legacies related to non-ferrous metal extraction

Metal extraction in the past also led to NORM legacies. Radium extraction is an obvious example but is generally not considered as a NORM activity. The issue of legacies from radium industry has been thoroughly described elsewhere (e.g. in [6]). Ferro-niobium extraction can also lead to an enhanced concentration of Th-232 and to a lesser extent of U-238 in the resulting slags [7]. Slags from zinc production or so-called zinc ashes have been landfilled or used as backfilling material in road construction. In one sample analysed by FANC, the activity concentration amounted to 120 Bq/kg of U-238sec. In the Netherlands, radioactivity had been used as a proxy

to identify the presence of zinc slags in the basement of some roads through on-field gamma radiation measurement [8].

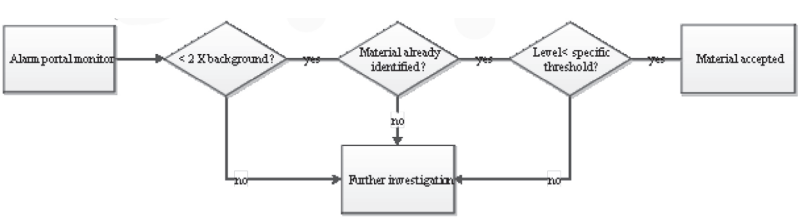
4. Monitoring of raw materials

4.1 Use of portal monitors

Most of non-ferrous metal production companies also recycle scrap metal. They are generally well aware of the risks related to orphan sources and have installed portal monitors to screen their raw materials. The installation of a portal monitor is compulsory in Belgium for all companies recycling at least 25 000 tons of scrap metal. Procedures have been published by FANC regarding the actions which have to be undertaken in case of detection of orphan sources.

Although the primary goal of a portal monitor is the detection of sources, they may also be used as a screening instrument for the monitoring of NORM in raw materials. The alarm level of most portal monitors is generally set to a few times the standard deviation of background level. This alarm level is exceeded for most common NORM material, including many materials which are out of regulatory concern (i.e. materials for which the activity concentration, although higher than background levels in Belgian soil, are below exemption levels: for instance alumina refractories or potassium compounds). For raw materials unlikely to contain any orphan source, an action level is defined which is generally equal to two times the normal background. A sample will be taken and analysed only if this action level is exceeded. Some companies also defined material-specific thresholds derived from experience. Exceeding this threshold for the material in question is considered as an anomaly; sampling and further characterization will be undertaken before processing the material. This procedure of routine control is summarized in Fig. 1.

Fig. 1: flowchart for management of alarm on a portal monitor



4.2 Screening measurement in the prospection phase

Portal monitors give an indication of the radiation level on materials which have already been accepted for production by the metal processing company. That information is not available when the company makes the initial characterization in the phase of prospection and selection of their raw materials. The procedures for controlling radioactivity in samples during the prospective phase vary from one operator to another. In one of the company, all samples are measured with hand-held instruments (contamination monitor). When the counts in beta/gamma or alpha signal exceed two times the background, the sample is often analysed in laboratory with gamma or/and alpha spectrometry. A simple gamma counter is generally not sufficient to identify materials possibly of concern as in several cases Pb-210 and Po-210 are the most relevant contaminants. For such material where Pb-210 and Po-210 are the dominant nuclides, a gamma spectrometry will not provide a complete characterization of the material: as can be seen from Table 1, Po-210 may sometimes be one order of magnitude higher than Pb-210 activity concentration.

4.3 Additional issues

The significant time necessary to carry on radiometric analysis often conflicts with commercial imperatives where the decision to buy a given material generally needs to be taken quite quickly.

The world-wide character of the trade of (secondary) raw materials is an additional challenge: suppliers of raw materials are located all over the world, from Mexico to Europe, from Africa to Australia. Standards, regulations, measurement protocols, awareness of NORM issues greatly vary and communication and transfer of information between processing company and supplier may be challenging.

5. Conclusions

Most of non-ferrous metal extraction factories process a large range of secondary raw materials, the residue of one being often the raw material of the other. Due to the variety of processes and raw materials involved,

it is generally difficult to predict beforehand which specific raw material will be of concern from radiation protection point of view and which nuclide from the natural decay chains will be the most prominent. As a complete and systematic radiometric characterization of each raw material is not cost-efficient in the current circumstances, screening methods are generally applied to identify potential material of concern. They generally involve a combination of portal monitor, hand-held and laboratory measurement. Although progress has been made e.g. in the context of the recent MetroNORM project [9], screening identification of enhanced Pb-210 and Po-210 activity concentration in raw materials remains in many cases a challenge.

6. References

[1] Council directive 2013/59/Euratom of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, and repealing Directives 89/618/Euratom, 90/641/Euratom, 96/29/Euratom, 97/43/Euratom and 2003/122/Euratom.

[2] IAEA, « Assessing the need for radiation protection measures in work involving minerals and raw materials", IAEA Safety Reports Series N°49, 2006.

[3] European Commission Joint Research Centre, Gianluca Cusano, Miguel Rodrigo Gonzalo, Frank Farrell, Rainer Remus, Serge Roudier, Luis Delgado Sancho; « Best Available Techniques (BAT) Reference Document for the Non-Ferrous Metals Industries"; EUR28648,doi:10.2760/8224

[4] S. Pepin, B. Dehandschutter, A. Poffijn, M. Sonck, "Regulatory approach to the issue of NORM residues in Belgium", Proceedings of the International Symposium 50 Years BVS – ABR (2013).

[5] A. Poffijn, "Experience concerning NORM in the non-ferrous industry (in Belgium)", 1st EAN-NORM workshop, Dresden (2007).

[6] IAEA, « The environmental behaviour of radium : revised edition », IAEA Technical Report Series 476, 2014.

[7] A. Poffijn et al., « Residual environmental contamination at a former ferroniobium production site in Belgium", Proceedings of the NORM VII symposium (Beijing), IAEA, 2015.

[8] R.L. Koomans, P. van der Wal, T.P.J. Kamsma, "A Geophysical Technique for the Verification and Quantification of Zinc Slag in Roads", Near Surface 2009 conference - 15th EAGE European Meeting of Environmental and Engineering Geophysics.

[9] MetroNORM, “Metrology for processing materials with high natural radioactivity”, JRP report IND57, 2017.

Annales de l'Association belge de Radioprotection, Vol 43, n°3, 2018
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.43, nr 3, 2018

IMPROVING PERSONAL DOSIMETRY OF MEDICAL STAFF WEARING RADIOPROTECTIVE GARMENTS: DESIGN OF A NEW WHOLE-BODY DOSIMETER USING MONTE CARLO SIMULATIONS

Clarita Saldarriaga Vargas[(a)], Corinne Amalberto[(a)], Lara Struelens[(a)], Filip Vanhavere[(a)].

[(a)] Belgian Nuclear Research Centre (SCK•CEN), Radiation Protection Dosimetry and Calibrations, Mol, Belgium.

Abstract

Introduction

Medical staff working in interventional radiology and cardiology (IR/IC) are exposed to scattered ionizing radiation coming from the patient. Radiation protection garments (RPG) like a lead apron and a thyroid collar are therefore worn by this population, together with personal dosimeter(s) used to monitor the occupational dose.

Whole-body personal dosimetry when wearing RPG is not straightforward mainly because such garments protect the body partially and are made of highly attenuating materials with a high atomic number (Z). These conditions make the exposure of the body highly inhomogeneous and affect the reading of conventional whole-body personal dosimeters in different ways depending on where the dosimeter is worn [1]. Some personal dosimetry methodologies based on $H_p(10)$ measurements have been proposed when wearing RPG and are currently applied for the dose monitoring of medical staff, namely single dosimetry (SD) and double dosimetry (DD) methods. However, several studies have shown that both methodologies can lead to unacceptable underestimations and excessive overestimations of the effective dose with RPG depending on the specific exposure conditions [1-3]. DD methods, which are in principle more accurate than SD, are moreover unpractical and not cost-effective

since they require the use of two dosimeters instead of one. Currently there is still a lack of personal dosimetry methods that can provide a good estimate of the effective dose while wearing RPG (*i.e.* a conservative estimate with minimized overestimation) under a wide range of relevant exposure conditions. This study aimed at designing a personal whole-body dosimeter worn over RPG capable of estimating the effective dose directly, without the intermediate step of $H_p(10)$, for photon exposures relevant for occupationally exposed IR/IC medical staff.

Materials and Methods

The design optimization of the dosimeter and virtual testing of its dose response were done by means of Monte Carlo simulations, taking as a reference the energy and angular dependence of the effective dose E_{RPG} calculated using an ICRP 110 reference male phantom equipped with a 0.5 mm-thick lead apron and thyroid collar [1]. Photon irradiations of the dosimeter and the ICRP phantom were simulated using Monte Carlo code MCNPX 2.7.0. The irradiation beams considered include monoenergetic photons and selected X-ray spectra of the Narrow (N) series of ISO standard 4037-1 with maximum photon energy up to 120 keV, and with positive angles of incidence (ϕ) parallel to the transverse plane of the body (with ϕ equal to 0° for an anteroposterior irradiation and 60° for a right frontal irradiation).

The new dosimeter stands as a single dosimeter and is conceived to be placed over the RPG worn by medical staff. A realistic virtual model was designed consisting of two silver-doped glass radiophotoluminescent (RPL) detectors, two sets of radiation filters and a housing plastic case. The dimensions and material characteristics of the RPL glass element and the housing case are practically the same as those of the commercial badge of Chiyoda Technol Corporation (Ibaraki, Japan) described by Maki *et al.* [4]. Two sets of filters (one for each RPL detection volume) were designed, and were optimized to fit within the space available in the commercial badge (none of the original filter inserts of the commercial badge were modelled). First detector (det1) has relatively light (low Z) frontal filtration, whereas the second detector (det2) is surrounded by high-Z filters.

The dosimeter dose (D_{dos}) is calculated from the linear combination of the dose received by the detectors (D_{det1} and D_{det2}) as follows (Equation 1):

$$D_{dos} = \alpha\,(D_{det2} + \beta\, D_{det1}) \quad (1)$$

where α and β are constants whose values are optimized to limit the over- and underestimation of E_{RPG} by D_{dos} for as many exposure conditions (photon energies and angles of incidence) as possible.

Results

Main design characteristics of the dosimeter model and its radiation filters are presented in Figure 1. The design of the two sets of filters is optimized so that the shape of the energy and angular dependence of D_{dos} is as similar as possible to that of the E_{RPG}. The photon energy and angular dependence of the dose to the detectors is shown in Figure 2. The overall energy and angular dependence of D_{det2} is very similar to that of E_{RPG}. A dose decrease is observed at 90 keV and higher energies due to the increased attenuation (*k*-edge) of Pb (88 keV) and Bi (90.5 keV) front filters at these energies. Given the low dose sensitivity of det2 at energies below 40 keV, the use of a detector with lighter filtration (det1) is necessary for estimating the dose contribution of low energy photons to the effective dose.

For this dosimeter design D_{dos} is estimated using Equation 1 with α and β equal to respectively 0.48 and 0.01. As can be seen in Figure 3, for monoenergetic beams the new dosimeter is capable of estimating E_{RPG} with a maximum deviation of ±25% for most beam energies and angles of incidence, except at 90 and 20 keV where respectively largest overestimation (+29%) and underestimation (-48%) are found. For more realistic beams the achieved performance is better: for spectra of the N series D_{dos} is always (*i.e.* for all the spectra and beam directions considered) within 20% of E_{RPG}. These deviations are significantly lower than the level of accuracy achieved with single and double dosimetry methods [1-3].

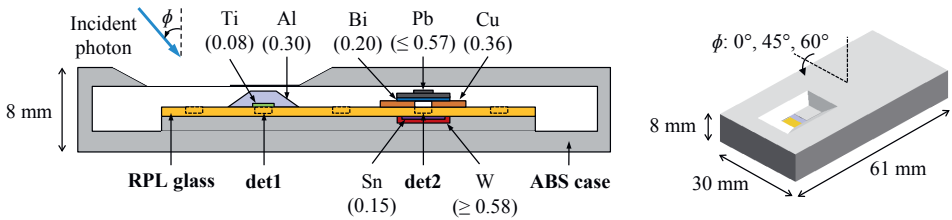


Fig 1. Schematic diagram of the cross section of the new dosimeter (left) and 3-dimensional view (right). Filter mass thickness ([g.cm^{-2}]) is indicated in parenthesis.

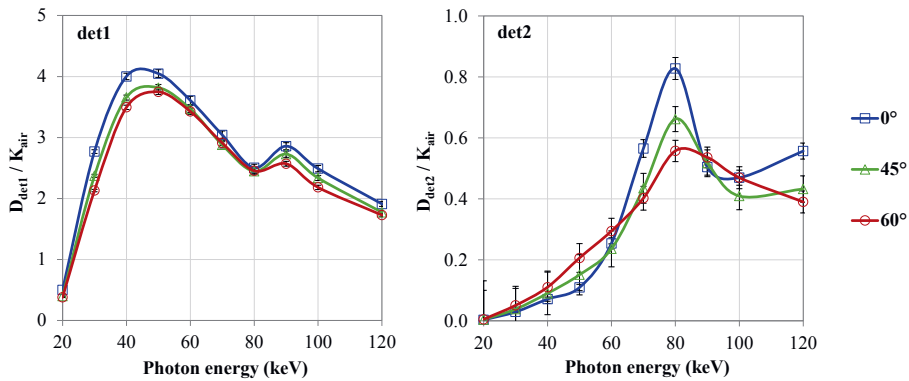


Fig 2. Dose response of det1 (left) and det2 (right) per incident air kerma as a function of energy and angle of incidence of the photon beam. Error bars indicate (±) the simulation statistical error.

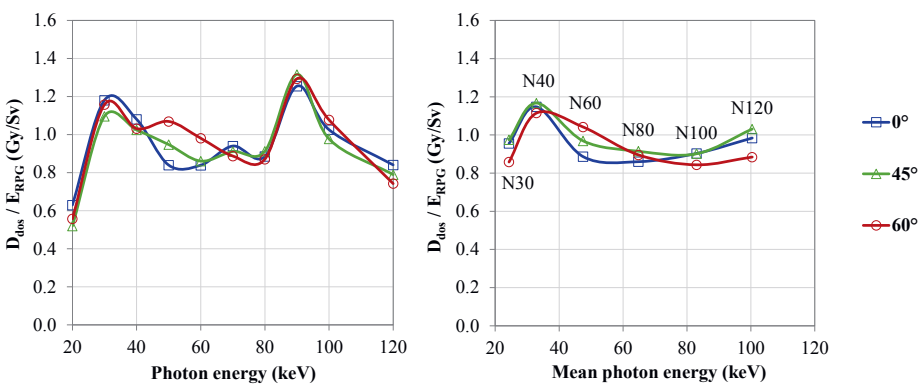


Fig 3. Ratio of the new dosimeter dose D_{dos} and the reference effective dose with RPG E_{RPG}, for different photon energies and angles of incidence. Results shown for monoenergetic photons (left) and X-ray spectra (right).

Conclusion

The concept of a stand-alone dosimeter suitable for estimating the effective dose of medical staff wearing RPG made of lead was demonstrated theoretically by means of radiation transport simulations. A virtual dosimeter design is presented based on two radiophotoluminescent detectors with different radiation filtration. The simulated dosimeter dose response indicates that it can estimate the reference effective dose with RPG with a maximum deviation of only ±20% for photon beams relevant in fluoroscopically-guided procedures. Although its dose response can still be improved and has to be validated experimentally, the *in silico* results presented here show the potential of such dosimeter to improve personal dosimetry of IR/IC personnel, by reducing the uncertainty in the estimation of the effective dose and offering a more practical and reliable solution than the dosimetry methods currently applied for this medical staff.

References

[1] C. Saldarriaga Vargas, L. Struelens, F. Vanhavere. The challenges in the estimation of the effective dose when wearing radioprotective garments. Radiat Prot Dosim 2018;178(1):101–111.

[2] T. Siiskonen, M. Tapiovaara, A. Kosunen, M. Lehtinen, E. Vartiainen. Monte Carlo simulations of occupational radiation doses in interventional radiology. Br J Radiol 2007;80:460–468.

[3] H. Järvinen, N. Buls, P. Clerinx, *et al.* Overview of double dosimetry procedures for the determination of the effective dose to the interventional radiology staff. Radiat Prot Dosim 2008;129(1-3):333–339.

[4] D. Maki, M. Ube, Y. Nagashima, *et al*. Development of the new glass badge. Radiat Prot Dosim 2016;171(3):337–345.

Email: csvargas@sckcen.be

Annales de l'Association belge de Radioprotection, Vol 43, n°3, 2018
Annalen van de Belgische Vereniging voor Stralingsbescherming, Vol.43, nr 3, 2018

CHEMICAL AND RADIOLOGICAL RISK-ASSESSMENT METHODOLOGY FOR SOIL CONTAMINATION IN BELGIUM: A COMPARISON

S. Pepin, G. Biermans, B. Dehandschutter (a), Sarah Radulovic (b)

(a) Federal Agency for Nuclear Control, rue Ravenstein 36,
B-1000 Brussels, Belgium.
(b) Ecole nationale supérieure de Chimie de Montpellier, France.

Abstract

In Belgium, environmental authorities have published detailed guidance on the chemical risk-assessment methodology for contaminated sites. These methodologies address both the risk-assessment to human health as to ecosystems and to groundwater and allow deriving generic as well as site-specific clean-up levels. For assessing the impact on human health of carcinogenic contaminants, a reference value of excess lifetime cancer risk of 10^{-5} is used; if the risk induced by the exposure to the contaminants exceeds this value, the soil contamination is considered to form a substantial threat. Moreover, the measured or predicted concentration of the contaminant in the environment has to be compared with relevant regulatory values, such as drinking water standards.

The clean-up levels are derived on basis of standard exposure scenarios defined for the five following ground-use: natural, agricultural, residential, recreational and industrial. The evaluation of human health risk from soil contaminants is made using the S-Risk model developed by the Flemish environmental institute VITO and used as reference model in all regions of Belgium. A former version of the S-Risk model was used a few years ago by soil contamination experts to assess the chemical risk of a Belgian site contaminated with uranium.

This methodology for assessing chemical risk to human-health is very similar to the methodologies used for assessing radiological risk for contaminated sites and could be used to derive clean-up levels for radioactive contaminants. A comparison between the methodologies for chemical and radiological human risk-assessment is presented. The present study confirms the conclusion of a recent US EPA paper where the consistency of US EPA and UK Environmental Agency methodologies for chemical and radiological risk-assessment of contaminated sites was demonstrated.

KEYWORDS: risk-assessment, contaminated site, mixed contamination, slope-factors.

1. introduction

Most, if not all, sites contaminated with radionuclides are also contaminated with non-radioactive pollutants, such as heavy metals. This is in particular true for NORM contaminated sites where in many cases the non-radioactive part of the contamination constitutes the main health risk and impacting factor on the environment. Some substances, such as uranium, present both a radiological and a non-radiological hazard. This entanglement between radiological and non-radiological hazards appeals for a consistent approach both in the risk-assessment as in the decision making process regarding a contaminated site.

Such a consistent approach has already been derived and applied in different countries: in particular the US EPA has made the policy decision that risks from radionuclides exposures at remedial site should be estimated in the same manner as chemical contaminants; in the context of its Superfund program, EPA has defined slope-factors for radionuclides allowing to sum the excess cancer risk of both radioactive and non-radioactive contaminants so as to provide an estimate of the combined risks [1][2]. In the UK as well, the Radioactively Contaminated Land Exposure Assessment Methodology (RCLEA) [4] is based on the original Contaminated Land Exposure Assessment (CLEA) [5] approach that was developed for the assessment of non-radioactive contamination. Other studies (e.g. [6] in Germany)

have used the assumptions of non-radiological soil protection regulations to derive reference values for radionuclides in soil.

In Belgium, a standard model has been developed to assess the risk to human health of land contamination. Assumptions and parameters used in this model are comparable to the ones commonly used for radiological dose-assessment, confirming the consistency demonstrated in e.g. UK or US EPA approaches [20].

2. Risk-assessment versus dose-assessment: slope factors and dose conversion factors

In the derivation of clean-up levels for non-radioactive contaminants, a distinction is generally made between threshold effect and non-threshold effect. The latter essentially corresponds to carcinogenic, mutagenic or teratogenic effects, which obviously also applies to radioactive substances. In Belgium, the environmental authorities use for carcinogenic substances a criteria of 10^{-5} for individual excess life-time cancer risk[1]. The life-time cancer risk is calculated by multiplying the dose (intake of the contaminant expressed as a concentration per body weight) or the concentration with respectively a slope factor or a unit risk. Next to the criteria based on excess cancer risk, a second criteria based on compliance with legal limits for concentration of contaminant in the environment is also used in the assessment [9][10]. This is similar to the approach of the US EPA which considers that the remediation of a contaminated site (including radioactive contaminated sites) should achieve a level of risk within the 10^{-4} to 10^{-6} carcinogenic risk range based on the reasonable maximum exposure for an individual [2]. EPA also requests that compliance with so-called ARARs (*Applicable or Relevant and Appropriate Requirements*), such as drinking water standards or radon indoor level is taken into account in the risk-assessment.

In radiation protection, effective dose is generally used to characterize the risk and ICRP has attributed a nominal risk coefficient for cancer of 5.5 10^{-5} per mSv for the whole population [11]. In most cases, assessment of the

1 One risk of additional cancer for 100 000 exposed persons on their life-time.

impact of a site contaminated with radioactive substances will be done on basis of dose-conversion factors for the various nuclides and comparison with a reference level expressed as an effective dose. US EPA on the other hand has derived slope-factors for estimating incremental cancer risks resulting from exposure to radionuclides though inhalation, ingestion and external exposure pathways [3]. Slope factors for radionuclide represent the probability of cancer incidence as a result of a unit exposure to a given radionuclide averaged over a life-time using the linear non-threshold model [2]. This approach allows to assess in a consistent manner the impact of both radioactive and non-radioactive contaminants and to sum the risks of both radionuclide and non-radioactive contaminants with non-threshold effects. However, EPA studies have shown that there is not a simple one-to-one relationship between risks calculated with the effective dose and risks calculated using slope factors for each nuclide. The risks estimated directly from dose tend to be greater than those estimated with slope factors [1].

3. Comparison between exposure scenarios and default parameters

Various international and national guidance and models have been developed to assess the impact of radioactively contaminated land: e.g. in UK [4], Germany [12] or France [13]. As mentioned above, in Belgium, the model S-Risk is used as the standard model for assessing exposure and human health risks from (non-radioactive) contaminants present in soil [8]. This model has been developed by the Flemish environmental institute VITO and incorporates state-of-the-art values for assessment parameters taking into account specific circumstances for Belgium or its different regions (regarding e.g. diet or standard soil profile). Belgian soil protection regulations defines 5 standard soil use and S-Risk has implemented different standard scenarios corresponding to these standard uses. They are summarized in Table 1.

Table 1: Standard soil use and corresponding exposure scenario

Type of soil use		S-Risk default scenario
Type I	Nature area	REC-dayout (day recreation – incl. sport)
Type II	Agriculture	AGR (residence with vegetable garden in agricultural area)
Type III	Residential	RES-veg (residential with vegetable garden) RES (residential with garden) RES-ng (residential without garden)
Type IV	recreational	REC-dayout (day recreation – incl. sport)
		REC-dayin (day recreation indoor sport scenario)
Type V	industrial	IND-l (light industry)
		IND-h (heavy industry with outside activity)

S-Risk incorporates following exposure pathways:

a) Oral

- **Ingestion of soil and indoor settled dust;**
- **Intake of vegetables from local production (home-grown);**
- **Intake of meat and milk from local production;**
- **Intake of water (drinking-water or groundwater);**

b) Dermal

- Absorption from soil and indoor settled dust;
- Absorption from water during showering and bathing;

c) Inhalation

- **Inhalation of outdoor air (gas-phase + particles);**
- **Inhalation of indoor air (gas-phase + particles);**
- Inhalation during showering (gas-phase).

In bold, we have marked the pathways which are also relevant for radiological impact. External exposure pathway is obviously a missing exposure pathway for non-radioactive contaminants but needs to be taken properly into account in radiological impact assessment.

S-Risk incorporates also the Volasoil model [14] which allows calculating indoor air concentration of volatile compound due to vapour intrusion from soil or groundwater into the building. The model considers three different building types: with basement, with crawl space and with slab-on-grade; it takes into account both diffusive and convective transport. It also allows two options for the floor of the building: intact or with gaps and holes. Further work will focus on the applicability of this model to

estimate indoor radon concentration from radon concentration in soil-gas or radium activity concentration in soil.

S-Risk, like the models for radiological dose-assessment mentioned above, proposes a set of default values for the parameters used in the assessment. These default values differ from one model to another depending on national circumstances or different expert judgment. A comparison between default values used in [4], [13] , S-Risk and in the RESRAD-OFFSITE model had been performed in [16] : this report showed some discrepancies between parameters such as soil ingestion values, dust concentration or exposure time in different scenario. Table 2 gives an example of this comparison for inadvertent soil ingestion by an adult in a residential scenario with vegetable garden.

Table 2: soil ingestion values for an adult in a residential scenario with vegetable garden

	EDTTL, ASN, IRSN, "Gestion des sites potentiellement pollués par des substances radioactives – guide méthodologique ", 2011.	Environment Agency, "Radioactively Contaminated Land Exposure Assessment (RCLEA) Methodology: Technical Report	**RESRAD**	**S-Risk** C. Cornelis, A. Standaert, H. Willems, "S-Risk: Technical Guidance document", 2013/MRG/R/76 – revision, March 2017.
Quantity of ingested soil (g/a)	14.6	22	36.5	28

A similar conclusion has been reached in [7] where default parameters and assumptions of, e.g. EPA PRG calculator, RCLEA and RESRAD-ONSITE have been compared. This underlines the need for carefulness in the choice of any default parameters: it needs to take into account site-specific circumstances (e.g. the diet must correspond to the one of the affected population) and an appropriate degree of conservativeness. Default values of a specific model should only be used in the assessment context for which this model had been developed. Site-specific values, when available, should be preferred to generic assumptions.

4. The case of uranium

It is well known that the chemical toxicity of soluble uranium is higher than its radiotoxicity. This is why the World Health Organisation recommendation of 30 µg/liter for uranium concentration in drinking water [17] is more restrictive than the derived concentration for U-238 and U-234 as defined in the European directive 2013/51/euratom (respectively 3 and 2.8 Bq/l) [18].

In 2009, a plume of uranium contamination in the groundwater of a Brussels neighbourhood had been fortuitously discovered during a routine control of groundwater parameters. The contamination reached a maximum level of 660 µg/liter. Consequently, a characterization study allowed defining the vertical and horizontal contour of the groundwater contamination plume; limited uranium contamination was also found in some backfilling material. A risk-assessment study [19] was performed according to the rules of regional soil protection regulations; the latter rules do not take into account radiological aspects but the chemical toxicity of uranium had to be addressed in the assessment.

That study used a former version of the S-Risk model (Vlier-Humaan) and calculated the daily intake of uranium due to e.g. ingestion of vegetables. As decision criteria, the study followed the WHO recommendations of that time and used a Tolerable Daily Intake (TDI) for uranium of 0.6 µg/kg of body weight per day[2]. Chemical toxicity of uranium is thus considered as a threshold effect. The way chemical and radiological risks of uranium are assessed is therefore quite different: threshold effect and comparison with a TDI for the chemical toxicity, non-threshold effect and comparison with an excess cancer risk or an effective dose for the radiological toxicity.

The model calculated for the oral exposure pathway (ingestion of vegetable and of soil) a value of 1.27 µg/kg per day for adult and 2.9 µg/kg per day for a child, essentially due to ingestion of vegetables. As the TDI for uranium was exceeded, it concluded to the existence of a significant risk.

2 This TDI was significantly increased in the 2011 version of WHO recommendations.

The risk can however simply be mitigated by preventing consumption of vegetables grown in the contaminated area.

5. Conclusions

Models used to assess chemical and radiological risks show essential similarities. With the exception of external exposure, the exposure pathways will be the same and some regulators, such as US EPA, already made the policy decision to assess chemical and radiological risks in an integrated scheme and to sum both risks. Models for chemical risk-assessment, such as S-Risk, could be used to perform radiological assessment, either by applying the dose-conversion factors to the calculated values in terms of intake of Becquerel, either – following the EPA approach – in applying nuclide-specific slope factor to the calculation of excess cancer risk.

6. References

[1] Interagency Steering Committee on Radiation Standards, "A method for estimating radiation risk from total effective dose equivalent", ISCORS Technical Report 2002-02;

[2] US EPA, "Radiation Risk Assessment at CERCLA sites: Q&A", Directive 9200.4-40, EPA 540-R-012-13, May 2014.

[3] US EPA, "Cancer risk coefficients for environmental exposure to radionuclides: Federal Guidance Report N°13", EPA 402-RR-99-001. Office of Air and Radiation. 1999.

[4] Environment Agency, "Radioactively Contaminated Land Exposure Assessment (RCLEA) Methodology: Technical Report", CLR-14, May 2011.

[5] Environment Agency, "CLEA Software (Version 1.05) Handbook", SC050021/SR4, September 2009.

[6] R. Gellermann, P. Günther, B. Evers, "Beurteilung von Bodemkontaminationen mit Radioaktivität im Gebiet Hannover-List nach Masstäben und Ansätzen der BBodSch V", Altlastenspektrum Helft 1/2012.

[7] N. Shubayr, "Overview of radiation risk and dose assessment models for radioactively contaminated sites and selected default input parameters", October 2017.

[8] C. Cornelis, A. Standaert, H. Willems, "S-Risk: Technical Guidance document", 2013/MRG/R/76 – revision, March 2017.

[9] OVAM, « Basisinformatie voor risico-evaluaties : werkwijze voor het opstellen van bodemsaneringsnormen en toetsingswaarden, richtwaarden en streefwaarden", October 2016.
[10] DGO Agriculture, Ressources Naturelles et Environnement, "Code Wallon de Bonnes pratiques: guide de référence pour l'étude de risque – évaluation des risques pour la santé humaine », version 03.
[11] ICRP 103, "The 2007 Recommendations of the International Commission on Radiological Protection", Elsevier, 2007.
[12] Bundesamt für Strahlenschutz (BfS), "Berechnungsgrundlagen zur Ermittlung der Strahlenexposition infolge bergbaubedingter Umweltradioaktivität", BfS-SW-07/10, 2010.
[13] EDTTL, ASN, IRSN, "Gestion des sites potentiellement pollués par des substances radioactives – guide méthodologique", 2011.
[14] J. Bakker, J.P.A. Lijzen & H.J. van Wijnen, « Site-specific human risk-assessment of soil contamination with volatile compounds", report 711701049/2008, 2008.
[15] C. Yu et al., "Data Collection Handbook to Support Modeling impacts of Radioactive Material in Soil and Building Structures", ANL/EVS/TM-14/4, Argonne National Laboratory, 2015.
[16] S. Radulovic, "Etude comparative de scenarios génériques d'exposition dans le contexte d'une contamination radioactive des sols", FANC internship report 2017-08-31-SRA-7-3-1(2), 2017.
[17] WHO, "Guidelines for drinking-water quality", World Health Organisation, 2011.
[18] European Commission, "Council directive 2013/51/euratom laying down requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption", 2013.
[19] GEOSAN sa, "Etude détaillée et étude de risque GRES10.11048/02", 2013.
[20] N. Shubayr, "Study of Chemical and Radiation Risk Assessment Methods for the United States Environmental Protection Agency and the United Kingdom Environment Agency", October 2017.