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A methodology for the systematic identification of naturally occurring radioactive materials (NORM)

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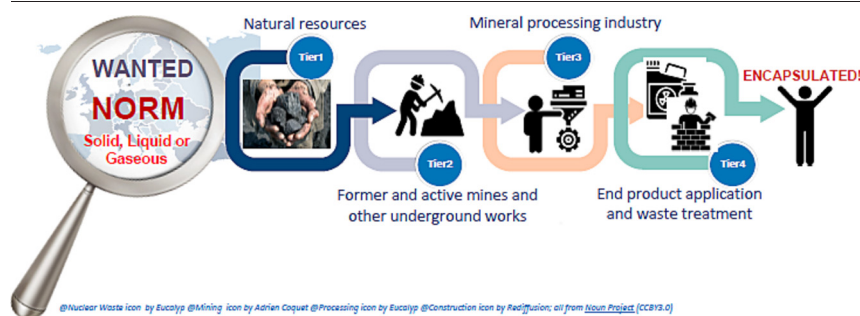
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HIGHLIGHTS

- Naturally occurring radioactive materials (NORM) can impact humans and biota.
- A systematic method for determination is needed.
- NORM affects many industrial sectors often not considered in terms of radioactivity.
- Case-by-case made inventories of NORM may not cover all situation of concern.
- Methodology facilitating initial or periodic review of NORM inventories is provided.

GRAPHICAL ABSTRACT



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ABSTRACT

Naturally occurring radioactive materials (NORM) are present worldwide and under certain circumstances (e.g., human activities) may give radiation exposure to workers, local public or occasional visitors and non-human biota (NHB) of the surrounding ecosystems. This may occur during planned or existing exposure situations which, under current radiation protection standards, require identification, management, and regulatory control as for other practices associated with man-made radionuclides that may result in the exposure of people and NHB. However, knowledge gaps exist with respect to the extent of global and European NORM exposure situations and their exposure scenario characteristics, including information on the presence of other physical hazards, such as chemical and biological ones. One of the main reasons for this is the wide variety of industries, practices and situations that may utilise NORM. Additionally, the lack of a comprehensive methodology for identification of NORM exposure situations and

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the absence of tools to support a systematic characterisation and data collection at identified sites may also lead to a gap in knowledge.

Within the EURATOM Horizon 2020 RadoNorm project, a methodology for systematic NORM exposure identification has been developed. The methodology, containing consecutive tiers, comprehensively covers situations where NORM may occur (i.e., minerals and raw materials deposits, industrial activities, industrial products and residues and their applications, waste, legacies), and thus, allows detailed investigation and complete identification of situations where NORM may present a radiation protection concern in a country. Details of the tiered methodology, with practical examples on harmonised data collection using a variety of existing sources of information to establish NORM inventories, are presented in this paper. This methodology is flexible and thus applicable to a diversity of situations. It is intended to be used to make NORM inventory starting from the scratch, however it can be used also to systematise and complete existing data.

1. Introduction

Naturally occurring radioactive material (NORM) is defined as radioactive material containing no significant amounts of radionuclides other than naturally occurring radionuclides (NOR). Many NOR occur in nature, nevertheless from radiation protection perspective uranium (^{238}U), thorium (^{232}Th), their decay products, and potassium (^{40}K) are considered the most significant contributors to radiation exposure and the presence of other NOR are usually neglected. The term NORM also covers materials in which activity concentrations of NOR are enhanced due to human activity, even unintentionally (IAEA, 2019). However, decisions on the meaning of 'significant amounts' vary, being made differently at national, and even local authority levels. Therefore, the classification of materials as NORM may vary from country to country, and even at wider global regional levels.

NORM may be associated with raw materials including ores, fossil fuels and associated rocks or formation water. Additionally, the mining and processing of raw materials (for example: extractive metallurgy, ore beneficiation and smelting, chemical and physical separation processes, water, and liquid effluent treatment, as well as flue gas cleaning) may lead to accumulation of NOR, even when exploited raw material or feedstock do not contain substantial amounts of them. Therefore, residues, waste, by-products, and final products that should be considered as NORM are expected to occur in a wide variety of industrial sectors not directly related to nuclear industry (Michalik, 2009; European Commission, 2003; Hofmann et al., 2000; Garcia-Tenorio et al., 2015; Chen, 2022). The variety of NORM issues are well depicted in proceedings from NORM symposia organized periodically since 1990s (IAEA, 2005, 2008, 2011, 2015, 2018).

According to Article 23 of Council Directive 2013/59/Euratom (European Commission, 2014), commonly called the European Basic Safety Standards Directive - EU BSSD, European Member States shall ensure the identification of practices involving NORM that may lead to exposure of workers or members of the public, which cannot be disregarded from a radiation protection point of view. Such identification shall be carried out by appropriate means, considering the Indicative List of industrial sectors given in the EU BSSD Annex VI. Similarly, the International Atomic Energy Agency (IAEA) provides a White list of activities that involve NORM that should be considered by countries worldwide with respect to radiological protection issues (IAEA, 2006a, 2006b). Based on these lists (Table 1) and elaborated international documents dealing with different aspects of NORM (European Commission, 1999, 2001; IAEA, 2006a, 2006b); some of the activities of concern are well identified (IAEA, 2003a, 2003b, 2003c, 2007, 2012, 2013a, 2013b; IAOGP, 2008).

However, the resulting NORM inventory, based on a case-by-case approach that is applicable to the local context and to specific issues, is limited to known industrial sectors that utilise NORM and there is a substantial likelihood that some important routes to exposure may not be considered (IAEA, 2022; Chang et al., 2007). Industries processing NORM as input materials are only some of the processes that may be of concern. Thus, even starting with raw materials with a low content of a specific radionuclide, an industrial process may at some point of the life-cycle of these materials be responsible for enhancing NOR concentration in discharges, solid waste, or final products. Certain examples are well known, such as combustion of fossil fuels (Smith et al., 2001) or technological processes applied in

inorganic chemistry (Timmermans & van der Steen, 1996; Burnett et al., 1996) with significant reduction of feedstock mass or volume. Other situations of concern include specific workplaces situated in spaces that may be confined or poorly ventilated or both (for example: underground mines). In such cases, regardless the properties of excavated material or conditions for ongoing activities, favourable circumstances for radon exhalation may be created (UNSCEAR, 2016; Skubacz et al., 2019; Bonczyk et al., 2022a, 2022b), formation water may be released (Wysocka et al., 2019; Haluszczak et al., 2013; Kharaka et al., 2013) or increased exposure to external radiation emitted by surrounding walls and rocks may result (Wysocka et al., 2019). Finally, a further obstacle to the identification of NORM exposure situations is the fact that the processing of excavated raw material, use of commodities, products, or merchandise as well as waste treatment often happens outside the industry where they were initially created. Thus, sequences of relevant processes and relationships among different industrial (or even agricultural) sectors must be considered (Trevisi et al., 2018; Blissett and Rowson, 2012; Flannery et al., 2018).

A part of the RadoNorm project in the Horizon 2020 Framework Programme (www.radonorm.eu) is focused on the identification of NORM exposure situations throughout Europe, their detailed characterisation and evaluation of possible exposure scenarios. To do so, efforts have been focused on providing a systematic and structured mapping of NORM situations across all European countries. A methodology for the qualitative identification of NORM exposure situations has been developed as the first step of these activities. Furthermore, systematised tools, in the form of specific NORM questionnaires and registers for qualitative information and quantitative data collection, have been developed for industry, legacy sites, and non-disturbed areas with high natural background radiation (HBRA) (Mrđakovic Popic et al., 2023, submitted for publication).

The main objective of the current paper is to present and make publicly available the developed methodology for the systematic identification of NORM exposure situations and to provide clear and applicable examples on how the methodology can be applied in practice.

The main benefits that are foreseen for the developed methodology are:

- Facilitation of initial and periodic review of national NORM inventories (in European and worldwide countries), as requested by the EU BSSD and IAEA BSS, and,
- Identification of sites where it is necessary to gather the information and data of importance for an overall improved exposure characterisation, including the behaviour, mobilisation, and further transport of radionuclides.

Such improved exposure characterisation provides the necessary insight for the evaluation of existing exposure situation doses, as well as information on the presence of any additional non-radiological hazards.

2. Developing the methodology for identifying NORM exposure sites

2.1. Abbreviations in use

In common use, the NORM abbreviation is used towards materials containing natural radioactivity that may give rise to enhanced ionizing

Table 1

Lists of industrial sectors, activities and exposure sites involving NORM considered in the frame of the RadoNorm project and provided by the EU BSS and IAEA BSS.

RadoNorm Project	EU BSSD, Annex VI (European Commission, 2014)	IAEA SRS 49 (IAEA, 2006a, 2006b)
Extraction of rare earths elements from monazite	Extraction of rare earths elements from monazite	Extraction of rare earth elements
Mining of Th ore, production of thorium compounds as well as the manufacture and use of thorium-containing products	Production of thorium compounds and manufacture of thorium containing products	Production and use of thorium and its compounds
Processing of niobium and tantalum ore	Processing of niobium and tantalum ore	Production of niobium and ferro-niobium
Oil and gas production	Oil and gas production	Production of oil and gas
Geothermal energy production	Geothermal energy production	–
TiO ₂ pigment production	TiO ₂ pigment production	Manufacture of TiO ₂ pigments
Thermal phosphorus production	Thermal phosphorus production	–
Zircon and zirconium industry	Zircon and zirconium industry	The zircon and zirconia industries
Production of phosphate fertilisers	Production of phosphate fertilisers	Phosphate industry
Cement production, maintenance of clinker ovens	Cement production, maintenance of clinker ovens	–
Phosphoric acid production	Phosphoric acid production	–
Primary iron and steel production	Primary iron production	–
Tin, lead and copper smelting	Tin, lead and copper smelting	Production of tin, copper, aluminium, zinc, lead, iron
Coal-fired power plants including maintenance of boilers	Coal-fired power plants, maintenance of boilers	Combustion of coal
Heavy mineral sand ore processing, total heavy mineral concentrate (THM) production	–	–
Mining of ores other than uranium ore	Mining of ores other than uranium ore	Mining of ores other than uranium ore
Titanium metal smelting and refining	–	–
Other metal (not directly mentioned in the list) mining and processing	–	Mining and processing of uranium ore
Bauxite and aluminium industry	–	–
Refractories production and use	–	–
Abrasives production and use	–	–
Pulp and paper mills and primary paper production	–	–
Coal mining and processing	–	–
Ground water filtration facilities, including mineral waters	Ground water filtration facilities	Water treatment
Scrap recycling (cleaning, melting and recovery of mercury) and disposal of residues from recycling	–	–
Underground workplaces other than mines, such as tunnelling, touristic routes, caves	–	–
Radon spas	–	–
Use of geothermal waters or minerals (sediments) in health and cosmetic treatment	–	–
Building and construction industry	–	–
NORM affected legacy sites	–	–
High natural background radiation areas (HBRA)	–	–

radiation exposure to people and environment (IAEA, 2019). However, in literature many other abbreviations exist as first applied by Gesell and Pritchard (1975) Technologically Enhanced Natural Radiation – TENR, followed by others, e.g. Kathren (1998), Righi et al. (2000) or Papastefanou (2001). Baxter (1996) applied TERM (Technologically Enhanced Radioactive Material), Martin et al. (1997) in the first report completely dealing with European industry generating enhanced natural radioactivity, used the simple descriptive name ‘*materials containing natural radionuclides in enhanced concentrations*’ and Vandenhove (2002) used ‘*elevated levels of Naturally Occurring Radionuclides – NORs*’. Authors still use all the abbreviations interchangeably to describe situations when the presence of the natural radioactivity causes a not negligible exposure to radiation. Moreover, US EPA (US EPA, 2022) introduces the abbreviation TENORM ‘*Technologically Enhanced Naturally Occurring Radioactive Material*’ simultaneously providing the definition of NORM which is opposite to this one given by IAEA. To avoid problems with the context interpretation as well as semantic errors when planning an inventory of situations of concern, the recommendation in this paper is to focus first on a set of predefined naturally occurring radionuclides. Then, a material of concern can be classified as being or not being NORM following IAEA terminology as there is no official recommendation at the European level.

2.2. Natural background

Naturally occurring radionuclides are present around the world and the average derived effective dose to a member of the public is estimated at the level of 2.4 mSv y^{−1} including contributions from external radiation from radionuclides in the environment, intake by inhalation and ingestion, and exposure to cosmic radiation. Observed variations cover the range 1–13 mSv y^{−1}, mainly due to increment of cosmic radiation at high ground elevation and indoor accumulation of radon gas (UNSCEAR,

2008). Whatever the circumstances, this is considered as natural background. Thus, potential sites of concern with respect to NOR are those where practices and activities have resulted in the probability of NOR exposures at levels above the prevailing local background radiation dose that should be considered in terms of radiation protection. To characterise external circumstances influencing exposure situations useful terms can be defined, based on general terms provided by IAEA (2019), European Commission (2004), and Trivari et al., 2015, as follows:

- ‘*Natural environment*’ refers to the surroundings and conditions unaltered by human activity in which living and non-living things exist on the earth,
- ‘*Living environment*’ is defined as an assembly of the natural and built environment which is inhabited by a human population that perform various kinds of social, cultural, religious, economic, and political activities which induce peculiarities in the character of the living environment,
- ‘*Work environment*’ is used to describe the surrounding conditions in which human employees operate. The work environment can be composed of physical conditions determined by workplace features and equipment used. It can also be related to factors such as work processes or procedures.

Using these terms, it is easy to define the general conditions of ‘when’ and ‘where’ NOR should be scrutinised in terms of radiation protection, considering that radiation doses may be of concern. NOR present in the *natural environment* (in contrast to the living and working environment), undisturbed by any human activity, are appraised as a component of natural background and not subject to regulatory control regardless the activity concentration and so no actions are necessary from a radiation protection perspective (Article 2, EU BSSD). In contrast, NOR emerging in a *living* or *work environment* should be considered as a possible source of non-negligible exposure, and therefore, subject to regulatory control.

2.3. Broadened list of NORM exposure situations

In order to improve the effectiveness of NORM involving industries and activities identification, a broadened list of *NORM exposure situations* of diverse characteristics has been first developed. A comparison of this new list with EU and IAEA recommendations shows a significant number of practices and situations that may be omitted when an inventory is made based entirely on the already existing recommendations (Table 1).

2.4. Four tiers methodology for NORM exposure sites identification

Bearing in mind such a broad range of NORM involving industries, different perspectives from which related hazard may be perceived (e.g. human or ecosystem, workers or public) and the ambiguous terminology used for its description, identification of all exposure situations of potential radiation protection concern needs a well-developed methodology. The methodology must be sufficiently comprehensive and include inter-related tiers that would cover, not only the identification of already confirmed or potential sources, but also account for the entire life cycle of these natural resources in the different value chains that may result in different types of commodities, products, by-products and wastes that may generate different potential NORM exposure situations.

A systematic tiered approach has been developed to identify situations when exposure to NOR cannot be neglected from a radiation protection perspective (Fig. 1). NOR characteristics, potential changes to NOR activity concentration implied either by the successive decay or influence of various industrial processes and knowledge gathered from an extensive international literature research and specific industrial processes understanding have been taken into consideration.

The developed approach, assumes an initial position of limited knowledge of the situation and proposes several tiers for determining the NORM inventory as follows:

- I. Analysis of the presence of natural mineral resources (the content of NOR is an inherent feature of all materials of mineral origin)
- II. Identification of ongoing and any former mining industries (including other underground workplaces)
- III. Identification of ongoing and former mineral and fossil fuel processing industries
- IV. Analysis of commodities and products life cycle, including their application, final disposal and secondary wastes generated

The first tier aims to provide general information to support identification of potential NORM exposure situations and consideration of potential radiation exposure related problems at the level of industry design and strategy for the exploitation of mineral resources in a country, prior to the start of any exploitation. Working through remaining tiers (II, III and IV, Fig. 1), at a theoretical level, can inform on possible exposures that could arise and support the development of any necessary prevention or mitigation methods in advance for all aspects of the life cycle of a planned commodity or product. It is important to note that, when natural resources have not yet been exploited, any existing exposure should be considered as natural background.

Tiers II, III and IV are then focused on existing or planned activities associated with the identified natural resources. Possible activities are limited, and their sequence is well defined as follows: mining of natural resources, resource (or mineral) processing (including transportation and storage), products and commodities industrial applications and final goods use. Such a sequence may seem obvious, leading to doubts whether tiers II, III and IV must be emphasised explicitly in this methodology. Therefore, it should be underlined that the content of NOR in processed material is important for possible exposure scenarios, but not crucial. Other circumstances defined by a technological process rather than an industry sector (for example: different exploitation methods applied in mining), the origin of processed raw material (imported materials also must be considered) as well as the different application and use of final goods could contribute to the possible exposure scenarios. The method proposed therefore assumes a two-way process of identification, following the subsequent relevant industries sectors (red arrows in Fig. 1) and the processed material fate at each level (yellow arrows in Fig. 1). Moreover, such an approach ensures that more attention is focused on each tier to analyse all possible relevant options where NORM may occur, with their increasing numbers from tier to tier, and with the potential for significant increase of possible uses of consumer goods are considered (as reflected by darkening shade of grey in Fig. 1). It must be underlined that waste management (including recovery, treatment, and disposal) is often a separate process and has not therefore, been directly included in the Fig. 1.

The proposed approach follows the sequence of subsequent processes or activities at each level and sequence of materials emerged is also tracked, therefore the likelihood that an important component is overlooked is minimised. Meticulous application of the methodology allows the development of complete national and regional inventories of NORM sites and situations. The proposed system of information collection is flexible and can

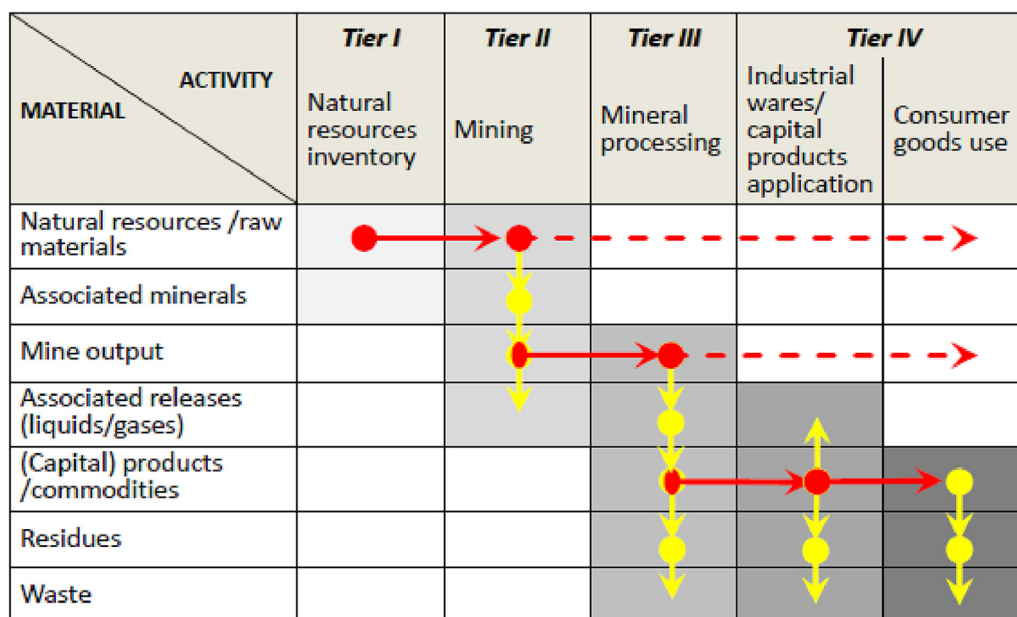


Fig. 1. Matrix of NORM identification tiers.

be launched at any tier (any point of the matrix in Fig. 1) and carried out even in reverse. However, if applied from tier I, there is a high probability that all cases and situations when NOR occurrence could be important from a radiation protection perspective will be identified and situations of concern will not be overlooked.

2.4.1. Tier I – Inventory of natural resources

Information and data from geological surveys and lists of raw materials of economic importance, regularly available in national registers. For example, the **Mineral Resources of Poland** database contains more than 14,000 mineral raw materials deposits (Polish Geological Institute, 2022). In Portugal, an extensive database also describes the main mineralogical occurrences throughout the country (Geoportal, 2022) and can be used to identify all relevant natural resources containing elevated NOR. Additionally, associated minerals, overburden or rock that is not sufficiently mineralised to be of commercial value, but which may contribute significantly to the generated waste stream (when exploitation is underway), must also be considered (IAEA, 2013b).

Features of minerals constituting identified raw materials can be checked regarding NOR content using literature data or widely available databases, (these include www.minerales.info, 2022; www.webmineral.com, 2022b; where about 400 minerals are directly classified as radioactive). The qualitative information concerning specific minerals of potential radiological protection concern should be completed with quantitative data on radionuclide activity concentrations, if available. Where no such data are available, screening measurements, including gamma background variation, or NOR activity concentration in randomly collected samples on site of concern are recommended to verify predicted situations. If screening measurements cannot be made, samples from former geological investigations can be used. Such material is often preserved in national archives. The sequence of activities included in tier I are depicted in the (Fig. 2).

Criteria used for data interpretation are mainly activity concentrations of radionuclides from the ^{238}U and ^{232}Th decay series, as well as ^{40}K . In the case of raw materials, secular equilibrium in decay series may assumed, but such an assumption will ultimately need verification. However, in addition to the parent radionuclides ^{238}U and ^{232}Th , it is commonly important to individually consider radium isotopes, especially ^{226}Ra , as it can migrate in the geological environment independently of its parent. This also applies, to some extent, to the radionuclide ^{230}Th . Separate evaluation of uranium isotopes ^{238}U and ^{234}U to consider their radiotoxicity may also be warranted. It should also be noted that radionuclides with relatively short half-lives (e.g., ^{228}Ra and ^{228}Th) are of importance when technological processes are considered, and are, thus, increasingly important in subsequent tiers (Michalik et al., 2013; Nucetelli and Risica, 2008; Nelson et al., 2015).

2.4.2. Tier II - Inventory of ongoing/former mining industry

In this tier, mining activities dealing with raw materials containing enhanced activity concentration of NOR should be considered first. However, independently of the natural materials mined, all mining activities should be the subject of analysis due to the radiation risk-favourable circumstances that may be created by this type of activity (IAEA, 2002; Wysocka et al., 2021; Skubacz et al., 2019; Skubacz et al., 2011; Santos et al., 2015). Based on national registries, an inventory of active and former mines, including other underground workplaces, should be made in this tier.

Mining sites should be considered in terms of the type of mined raw material. If raw material is identified in the frame of tier I as NORM, subsequent tiers are necessary. Notwithstanding with the formal classification undertaken in tier I, it is recommended to carefully check other circumstances, which may give rise to either NOR accumulation or release NOR not directly attached to the mined material, such as:

- local hydrogeological conditions, considering mainly formation water occurrence (Wysocka et al., 2019),

Tier I

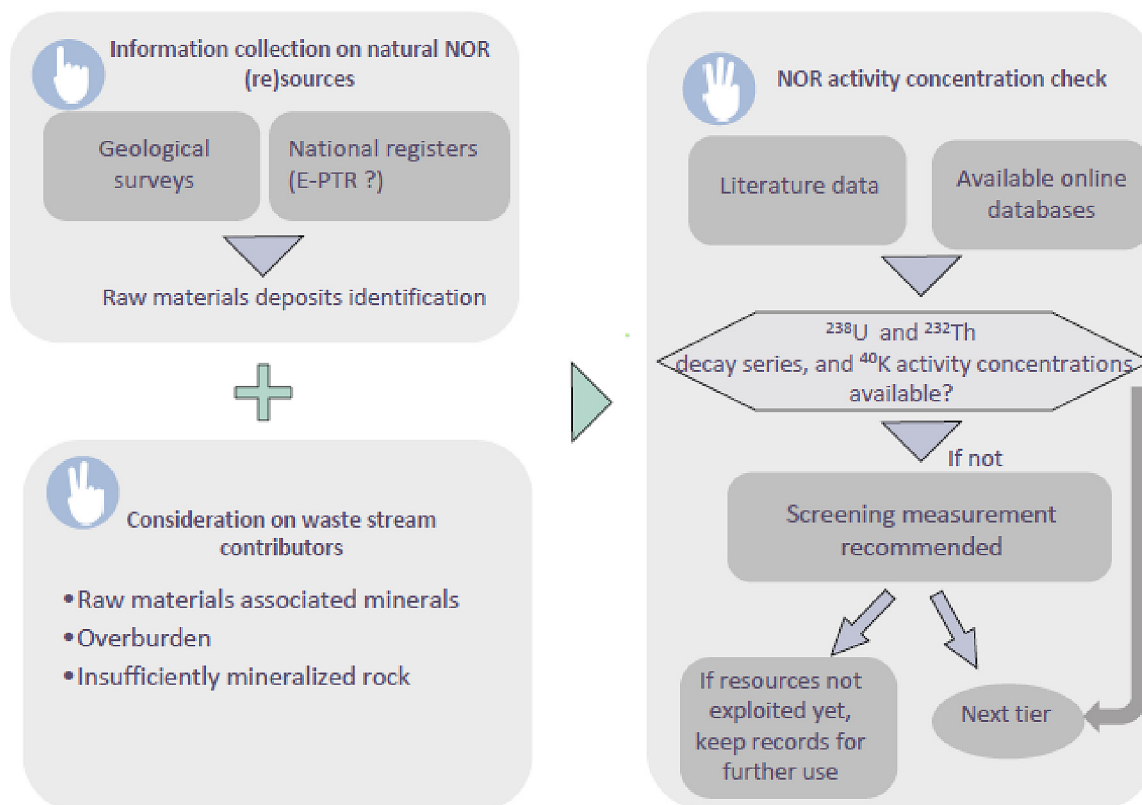


Fig. 2. Schematic representation of tier I.

- method of raw materials, ores, and fossil fuel mining, considering circumstances influencing radon exhalation as well as exposure to external radiation to some extent (Chen, 2022),
- type of applied ventilation system, considering radon and especially radon and thoron progeny ingrowth and accumulation/dilution (Skubacz et al., 2019),
- type of ore treatments, considering residues generated as well as dust, leachates and physical hazards related (Garcia-Tenorio et al., 2018; Bahari et al., 2007; Angadi et al., 2015),
- mine tailings deposition sites, considering possible impact on environment and public (Michalik, 2011; Michalik et al., 2020),
- mine drainages storage and treatments, considering possible impact on workers, environment and public as well as NOR accumulation in residues emerged after treatment (Michalik et al., 2022).

When verifying a mine, all substances involved in the mining process should be identified, based on the technological process description (Pires do Rio et al., 2002) and using a national waste catalogue since, due to the technological process applied, properties of excavated material, residues and waste can differ from the properties of minerals defined in the frame of tier I. If any material is classified as NORM, it is important to follow its fate during subsequent tiers. Nonetheless, irrespective of whether substances identified in association with mining processes include NORM, there are still potential sources of radiation exposure that may need require investigation, such as exhalation of radon and the presence of radon progeny in air. The sequence of activities included in tier II is depicted in the Fig. 3.

As different mining technologies may be in use (Ghose, 2009; IAEA, 2003a), the specific features of the applied excavation process must be considered to evaluate all details related to potential exposure to radiation. For example:

- *Surface mining (open pit and strip mining)* - the radiological hazards are mainly related to the type of deposit, and the exposure either to workers or environment is proportional to the increase in NOR in the mined material or residues. Sources of exposure include external gamma radiation and intake of radionuclides by inhalation and accidental ingestion (Pillai and Khan, 2003). If there is no enhanced content of NOR, existing circumstances will not give rise to enhanced exposures to people or the environment (IAEA, 2003a).
- *In situ mining (borehole mining)* - mainly related to oil and gas extraction - radionuclides (mainly radium isotopes, and uranium to some extent) are present in formation water that usually coexists with fossil fuels. Due to changes in physical parameters (pressure and temperature) as well as chemical properties of a mine environment, these radionuclides are further accumulated in scale, slurry, silt, and sludge. This phenomenon is

widely reported in scientific literature and well summarised in relevant documents from the IAEA and oil and gas producers' associations (IAEA, 2003a, 2003b, 2003c; IAOGP, 2008). Often, internal surfaces of the used installation and pipelines are covered with a layer of scale containing significantly increased concentrations of radium isotopes and ^{210}Pb and ^{210}Po (Khan and Al-Shehhi, 2015). In these situations, the main sources of occupational exposure relate to external gamma radiation and the intake of radionuclides by inhalation and accidental ingestion when in contact with scale and sediments. Special attention should be paid to unconventional shale gas exploitation as to date many reports are available about NOR-rich effluents created due to this activity (Kondash et al., 2017).

- *In situ leaching mining (ISL) or in situ recovery (ISR) mining* – a standard uranium production method – is a special form of solution mining that is normally applied to ore deposits located in sedimentary formations dominated by highly permeable stone and saturated aquifers. It consists in the injection of a solution (or lixiviant) that can be acid or alkaline, depending on the geology of the extraction site, into the ore through injection wells previously drilled to mobilize and recover the uranium by pumping the uranium bearing solution (pregnant solution) to the surface for further processing. Groundwater and natural spring contamination and acidification are the main concerns, which is a consequence of the presence of mining solution that remains in the mined aquifer and of residual water that may migrate to groundwater resources depending on the connectedness to the mine aquifer (IAEA, 2016).
- *Heap leaching* – used mainly for copper, nickel, and uranium extraction of low-grade ore. Heaps of low-grade crushed ore, 4 to 10 m high, are formed over a collecting system underlain by a watertight membrane. Acid or alkaline leaching solutions (depending on the geological characteristics of the ore) are distributed over the top surface of the heap. As the solution percolates through the heap, they dissolve a significant (50–75 %) amount of the elements in the ore. The elements are then recovered from the heap leaching liquor by ion exchange or solvent extraction. As in surface mining, the radiological hazards will depend on the type of deposit that is being mined. The exposure of either workers or environment is also proportional to the increase of natural radionuclides concentration in the mined material or residues. Risk to workers is mainly associated to gamma radiation and long-lived radioactive dust exposure through inhalation and ingestion. Environmental hazards are mainly associated to soil and groundwater chemical and radiological contamination and acidification due to leaching liquor seepage and long-lived radioactive dust spreading to the surrounding areas (NEA, 2006; Hoummady et al., 2018)
- *Underground mining* – the main features of this mining type include confined spaces, poor ventilation, and enhanced radon exhalation from crushed rocks that together can lead to a significant increase in the

Tier II

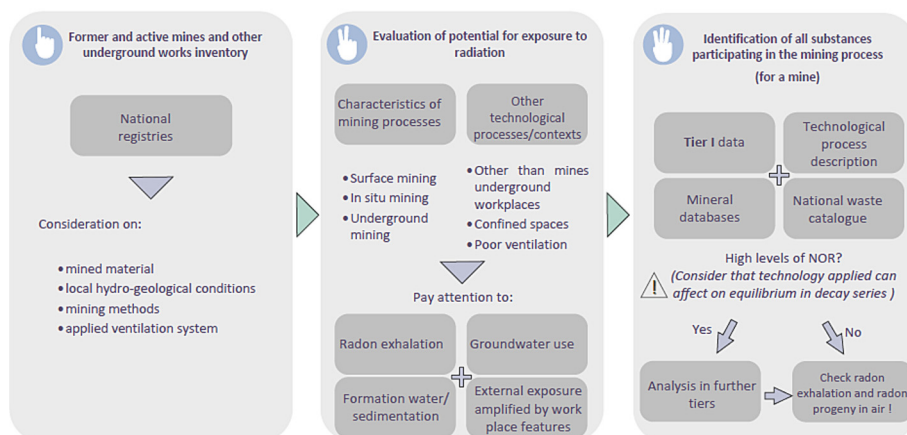


Fig. 3. Schematic representation of tier II.

concentration of radon decay products in the air, even if the concentration of parent radionuclides in the surrounding rocks and excavated minerals high enough to be classified as NORM (Trevisi et al., 2012). An additional factor increasing miner exposure can be formation waters enriched with natural radionuclides, which flow into mining excavations. However, the main source of exposure is radon, especially radon progeny without regard to excavated material besides that for uranium ore (Kavasi et al., 2011; Skubacz et al., 2019). Typical exposure pathways include inhalation of radionuclides, external gamma radiation and radionuclide ingestion, which depend, in this case, on mined minerals and local hydrogeological situation, such as the presence of radium enhanced formation water (Ashraf et al., 2004; Wysocka et al., 2021).

Conditions like those found in an underground mine can occur when using other technological processes or in other contexts. Therefore, a list of conditions and situations causing possible exposure to natural radiation independently from the processed or extracted material should be broadened to identify possible situations of concern with regards to radiation protection (IAEA, 2003b). Examples can be given as special cases of workplaces located in confined, underground spaces, such as (Fijałkowska-Lichwa and Przylibski, 2022; Margineanu Romul Mircea, 2019; Grygier et al., 2022):

- caves, especially those adapted for tourism purposes where regular staff are involved as guides or for facility maintenance,
- underground museums.

Radon and radon progeny are the first radiation exposure dose contributors that should be considered. As formation water is a common carrier of natural radionuclides, groundwater use (geothermal energy, spas, water treatment plants) should also be considered in a special category of relevance for radiation protection. Special attention must be paid when formation water is filtered, purified etc. as these processes may lead to the accumulation of radionuclides in filters (IAEA, 2006a, 2006b; STUK Report A169, 2000; Allas and Kiisk, 2019).

In addition to exposures inherently related to mining activities, waste streams and products generated should also be followed. This includes plants for enrichment and purification of excavated material and mineral-ore concentrate preparation that are an inherent part of a mine. However, processes applied there should rather be analysed in the frame of tier III, despite the ore or fossil fuel treatment plant often being part of the mine of concern.

2.4.3. Tier III – Inventory of mineral processing industry

Tier III of the methodology for NORM identification is focused on industries processing materials of mineral origin. It is assumed that, at this level, all raw materials, ore concentrates, minerals and fossil fuels locally produced are already identified. If there are no active mines in a country, imported raw materials, concentrates and by-products of mineral origin should be considered. Information will be available as the outcome of tiers I and II, or in case of imported materials, from a trade or custom national register and then from available national registers on waste, pollution, or discharge, the same as in case of domestic resources processing.

The main objective of any kind of mineral processing industry is to recover the element or mineral of interest enclosed in a matrix of inert material. For that, many processes may be applied in general as chemical reaction or physical treatment. A specific process to be considered is energy generation from fossil fuels. Regardless the underlying principles of a process, feedstock is eventually divided into streams of materials differentiated from other in mass, volume, chemical composition, or even aggregate state as well as the radionuclide activity concentration. That is why all plants operating on materials of mineral origin should be considered in terms of mass reduction processes leading to NOR activity concentration enrichment:

- inorganic chemistry processes, such as titanium dioxide (Gázquez et al., 2014), phosphoric acid production (Vioque et al., 2010),
- combustion, such as coal combustion (Papastefanou, 2010),

- smelting, such as iron manufacturing (Li et al., 2018),
- fractionation, any separation in general as: sieving, distillation, filtration or sedimentation, magnetic separation, etc. (Moustafa, 2010; Vassas et al., 2006).

Industries processing feedstock already classified as NORM are immediately identified based on the results of tier I or II. For the remaining processing industries, non-radiological information including type of material, chemical properties or reactions, quantity or volume and process temperature, may give important information on which NOR accumulation related issues can be expected (e.g. coal combustion). This type of information helps to narrow down the list of materials and industries that need further attention. Then, for each case, and consistent with the approach in tier II, the inventory of all materials associated with the process should be developed and checked with respect to possible NOR content. Contrary to tier I, at this level of analysis, attention should be paid not only to the parent radionuclides of a decay series, but also the decay products including the ^{228}Ra , ^{228}Th or ^{210}Pb and ^{210}Po as well as radon isotopes (^{222}Rn and ^{220}Rn). Even if no NORM is identified, workplaces should be checked for radon and radon progeny, especially in confined poorly ventilated spaces. The sequence of activities covered by tier III are depicted on Fig. 4.

NOR often accumulate in residues, and residues or waste currently considered as NORM, such as red mud (Wang and Liu, 2012), coal combustion products (Papastefanou, 2010) or slags from metal smelting (Croymans et al., 2017), may have been generated over many years and already be subject to regulation and treatment. All active industries managing enhanced concentrations of NOR are subject to European and national regulations, and, eventually, all residues must be categorised and treated following the methodologies defined in these regulations (European Commission 2008; European Commission, 2006, 2000). Therefore, when NORM situations are identified a recommended step is to check if treatment arrangements already applied meet additional requirements related to the presence of radioactivity.

Whilst NOR commonly accumulates in residues, sometimes accumulation can occur in products and by-products. Information on NORM in products and by-products can be obtained by conducting the analysis detailed in tier IV.

2.4.4. Tier IV Inventory of end product application and waste utilisation

The last tier for identification of possible NORM exposure situations is focused on the identification of industrial materials, by-products, and tools as well as commodities which are present directly in the working or living environment and potentially containing NORM.

Objects of everyday use containing high levels of NOR are exceptionally rare in Europe because of legal restrictions set by the EU BSSD. The use of luminescent paints based on radium, historically applied to watch dials, indicators, warning signs, are currently banned and have been replaced by non-radioactive alternatives (Martinez et al., 2022). However, it is foreseen that the application of the presented tiered methodology may result in the identification of new cases of radiation protection concern. For example, some alternative medicine products, such as scalar energy products (Bonczyk et al., 2022a) are stated by manufacturers to be made of volcanic materials or other minerals that are claimed to have health benefits such as emanating positive energy. Products advertised as having similar properties are becoming more widely, especially in cross border trade via the internet (Abu et al., 2022). These products contain natural radionuclides, mainly ^{232}Th and its decay products, and may be an additional source of exposure when used as recommended (Bonczyk et al., 2022a) and currently not being subject to any regulatory controls.

The probability of finding products containing NORM is higher in:

- industrial materials and tools (such as welding rods, refractories, abrasive materials, ceramics),
- construction and building materials (such as tiles, glaze, natural rocks, products with waste origin),
- fertilisers.

Tier III

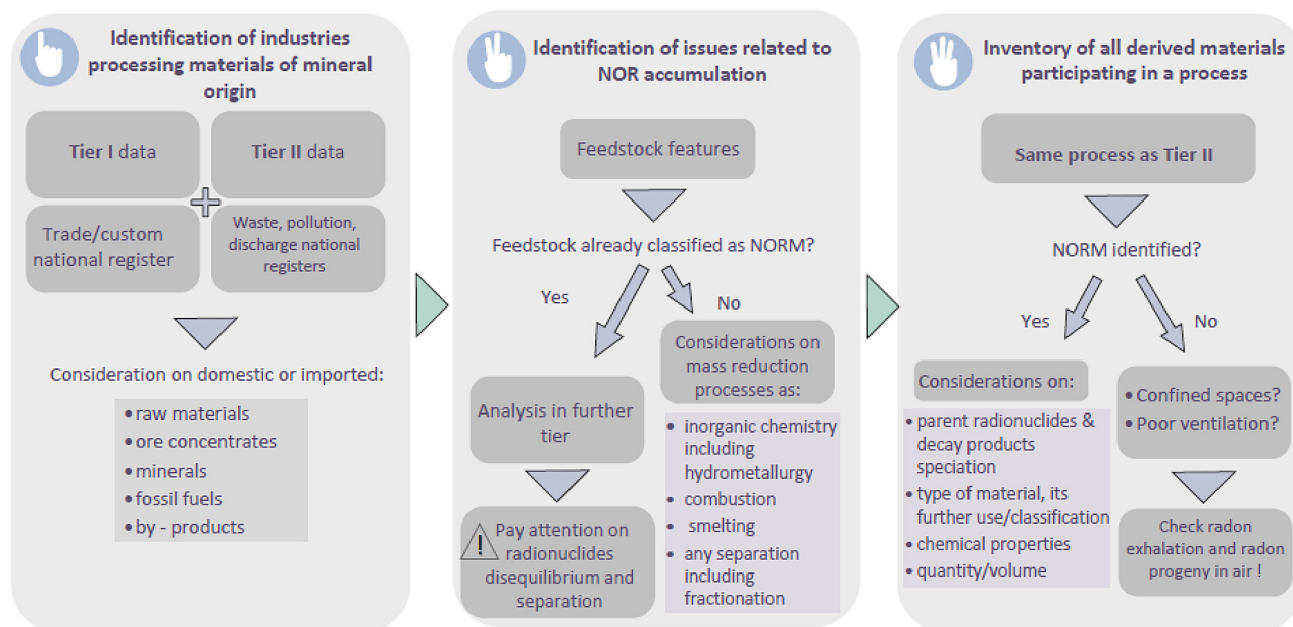


Fig. 4. Schematic representation of Tier III.

The list of possible items is not expected to be exhaustive, and all should be identified through application of the proposed four-tiered methodology. If there are no data resulting from tiers I-III, the main criterion to select an item for further investigation is the origin of material that the given item is made of. Items made from wood, plastic, pure metals, and fabrics are largely free from measurable NOR content, while items made from mineral substances should be investigated. Items entirely made of rock or uncommon minerals should be prioritised for consideration. Currently, there is no obligation for manufacturers to provide information about the NOR content of manufactured products (besides building materials). If there is a serious suspicion of enhanced NOR activity in a product, the proposed methodology can be applied in a reverse direction; starting with the product, and ending with the feedstock used for its production. However, meticulous application of the proposed methodology allows one to identify all cases of concern and avoid investigation of multiple items.

Among the products present in the living environment that give rise to human exposure, building materials are by far the most common. The recommendations given by the EU BSSD are applicable to categorise them in terms of radiation protection. In other cases, individual items containing NORM are small; the total activity of NORM is not so high and related activities do not contribute significantly to exposure (for example: TIG welding rods, refractories, or specific ceramics (Mcelearney and Irvine, 1993; Saito et al., 2003; Caridi et al., 2021; Hobbs, 2000)). In case of products identified as NORM, in addition to technological process features, special attention should be paid to situations related to wholesale trading, transport, and storage. Schematic representation of tier IV activities is shown in Fig. 5.

3. Life Cycle Analysis used to assess radiation exposure at all stages of a product life

The proposed four-tiered approach can, when used to assess the potential exposure to radiation associated with all stages of product life, from raw material extraction through materials processing, manufacture, distribution, use and possible re-use, and waste production and disposal, be described as a Life Cycle Assessment (LCA) oriented to the presence of NOR. In Fig. 6, an example focused on production and application of abrasives, including the final waste stream generated, is outlined. In each tier of analysis specific exposure situations can be defined resulting in recommendations for detailed quantitative exposure evaluation.

In practice, in industrial sectors as well as technological processes, enhanced NOR is not the only hazard. Such industries commonly give rise to multiple hazards, among which the radiological hazard may not be necessarily the dominant one. Therefore, to be able to completely characterise exposure situations in the different NORM involving industries, the identification of non-radiological hazards is also necessary. However, as overall circumstances are very similar when both radiological and non-radiological hazards are present, use of the LCA methodology and tools already developed in the context of non-radiological hazards, with adaptation to NOR related effects, is an optimal choice. Some efforts have already been made on including radioactivity in on-going research based LCA (Meijer et al., 2005; Schreiber et al., 2021; Koltun and Klymenko, 2020). However, to achieve reliable results, it would be beneficial to develop a NOR oriented LCA method either as a separate, supporting tool or as an inherent part of an IT tool facilitating the use of the methodology we propose.

4. An example of application of Tier II and III by the combination with the European platforms of polluters

Besides sources of information previously listed in the descriptions of tiers, for the identification of NORM exposure situations related to tiers II and III, information about known facilities listed in the European Pollutant Release and Transfer Register (E-PRTR The European Pollutant Release and Transfer Register, 2007) can be used. The E-PRTR contains compiled information on key environmental data of industrial facilities from different European countries, although without any information about possible radioactivity. Recently, after revisions proposed by the European Commission, a new website of the European Industrial Emissions Portal (EIEP The European Industrial Emissions Portal, 2020) was launched that includes information on the largest industrial complexes in Europe, including data on releases and transfers of regulated substances to environmental media and waste transfers. The portal also provides detailed data on energy input and emissions for large combustion plants in EU Member States and Iceland, Liechtenstein, Norway, Serbia, Switzerland, and the United Kingdom. This portal gives public access to detailed information on the emissions and the off-site transfers of pollutants and waste from around 60,000 industrial sites from 65 economic activities across Europe. Such a source of data provides additional

Tier IV

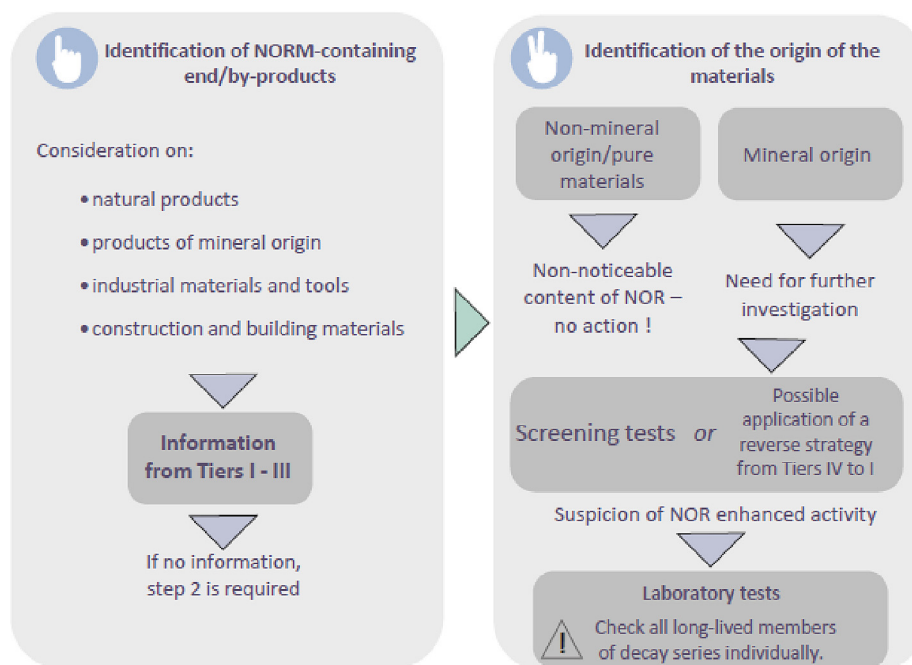


Fig. 5. Schematic representation of tier IV.

possibilities for identifying NORM exposure situations complementary to the options implied by the abovementioned descriptions of tiers, where it is assumed to start with a singular raw material and identify all related industries. By using E-PRTR at the outset, an almost complete set of options is available, from which NORM situations can be identified by elimination. The optimal approach will depend on the accuracy of the natural raw materials register and E-PRTR.

The activities covered by E-PRTR fall within the following sectors:

- energy,
- production and processing of metals,
- mineral industry,
- chemical industry,
- waste and wastewater management,

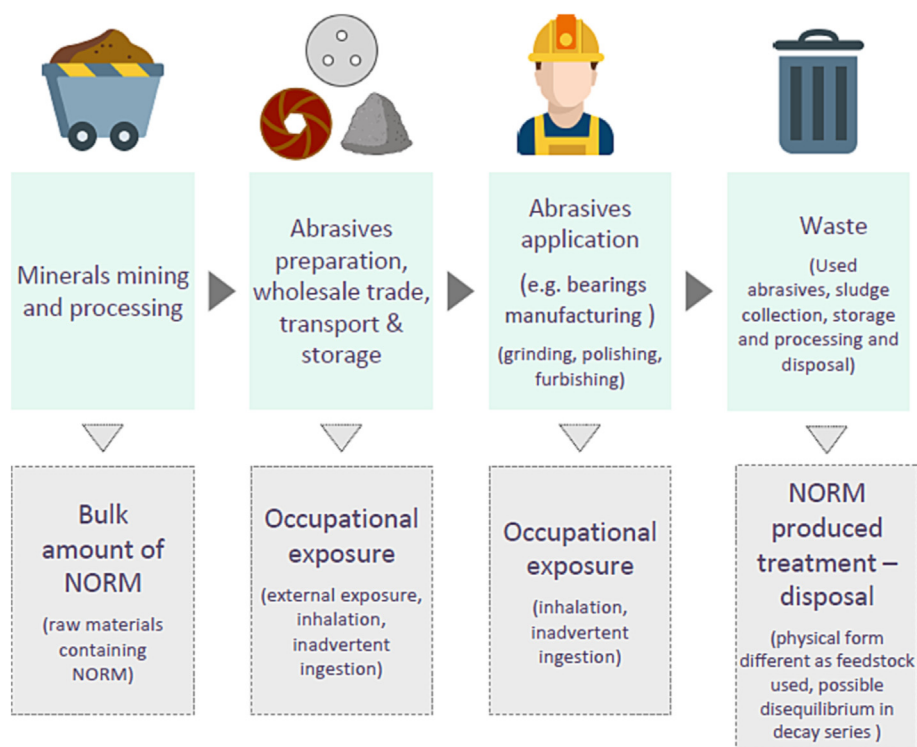


Fig. 6. Life Cycle Analysis (LCA) of abrasives applied in bearings manufacturing.

- paper and wood production and processing,
- intensive livestock production and aquaculture,
- animal and vegetable products from the food and beverage industry,
- other activities.

To perform an analysis of facilities and sites that may be classified as NORM exposure situations, the structure of the data in the E-PRTR should firstly be analysed and information screened against industries involving NORM listed in the EU BSSD and the RadoNorm project list of industries, activities, and sites presented in Table 1. The first five sectors, energy, and combustion industry; production and processing of metals; mineral industries; chemical industry; waste and wastewater management, should be analysed initially for the presence of NORM as these are more in line with NORM involving industries. Additionally, according to the list detailed in Table 1, a derivative wood industry (paper and cardboard) should also be included in the analysed industries since studies have shown that NORM can be present in precipitates and scale formed during the kraft process at pulp and paper mills (Fisher and Easty, 2003).

A preliminary inventory of NORM waste generating industries in Spain (Pastor et al., 2016) has already been developed using a national PRTR platform (PRTR-España, 2007), as illustrated in Fig. 7.

This strategy for the creation of an inventory of potential NORM exposure situations begins with the identification of NORM sites/facilities by discriminating them from the broader list of industries and facilities as described above. In general, it could be considered as a preliminary to the tier I or II activity aimed at preselecting industries of concern instead of focusing only on mining and mineral processing industries. The second step then consists of collecting information on radionuclides which may be associated with, on one hand, the production process involving different types of materials and the main products and residues, and, on the other hand, with releases to air, water bodies or soil from the facility. Residues can also be considered as a release and may contaminate all environmental compartments such that concern around NORM is commonly focused on their appropriate management including valorisation. Whilst different options for the management of NORM residues exist, optimisation of current management options or development of more appropriate strategies continue to be sought.

The approach for NORM sites identification using PRTR registers is beneficial for different end-users as the data in PRTR are widely available for different European countries. Information is structured and organized in a

proper way, which made the process of NORM exposure situations identification faster. Data from the register can be retrieved in common and easily used Excel spreadsheet format. Some information on chemical pollutants (for example: heavy metals) is available in the register, which may be useful for investigating multiple hazards at NORM exposure situations and for developing an integrated hazards assessment.

5. Conclusions

A methodology for the identification of NORM exposure situations through relevant information and data collection in a systematic and harmonised way, developed within the RadoNorm project, is presented in this paper. The proposed approach, consisting of four tiers for the identification of all cases where NOR may be a possible source of radiation exposure of concern, is based on nationally available data. Based on geological archives, mining, environmental licenses, import and export statistics and trade registers, qualitative information can be obtained on relevant natural resources, raw materials and industrial activities with potentially enhanced concentrations of NOR. To cover all possibilities and avoid gaps, it is recommended to start with the consideration of the natural resources inventory in the country (tier I), progress through the mining stage complemented with a study of the imported raw materials (tier II), and then consider the mineral processing industry (tier III) and products and waste and residues (tier IV).

For consideration of NORM in terms of waste management, it must be underlined that most NORM waste can be hazardous both from a chemical and radiological point of view. From a chemical perspective, NOR behave similarly to other non-radioactive elements. Hence, even when an industry was not characterised in terms of radiation protection to date, qualitative information can be extracted from already existing documents, such as environmental licenses, Environmental Impact Assessments (EIA), waste catalogues or the European Pollutant Release and Transfer Register (E-PRTR), as discussed in the Section 4.

When situations of concern with respect to radiation protection are identified, quantitative data should be compiled, including characterisation of technological processes, information on radionuclide behaviour and final fate. These data present the basis for radiation risk evaluation for workers, members of the public, non-human biota and the environment in general. NORM tools for harmonised and systematic data collection at identified NORM exposure situations are developed as a subsequent

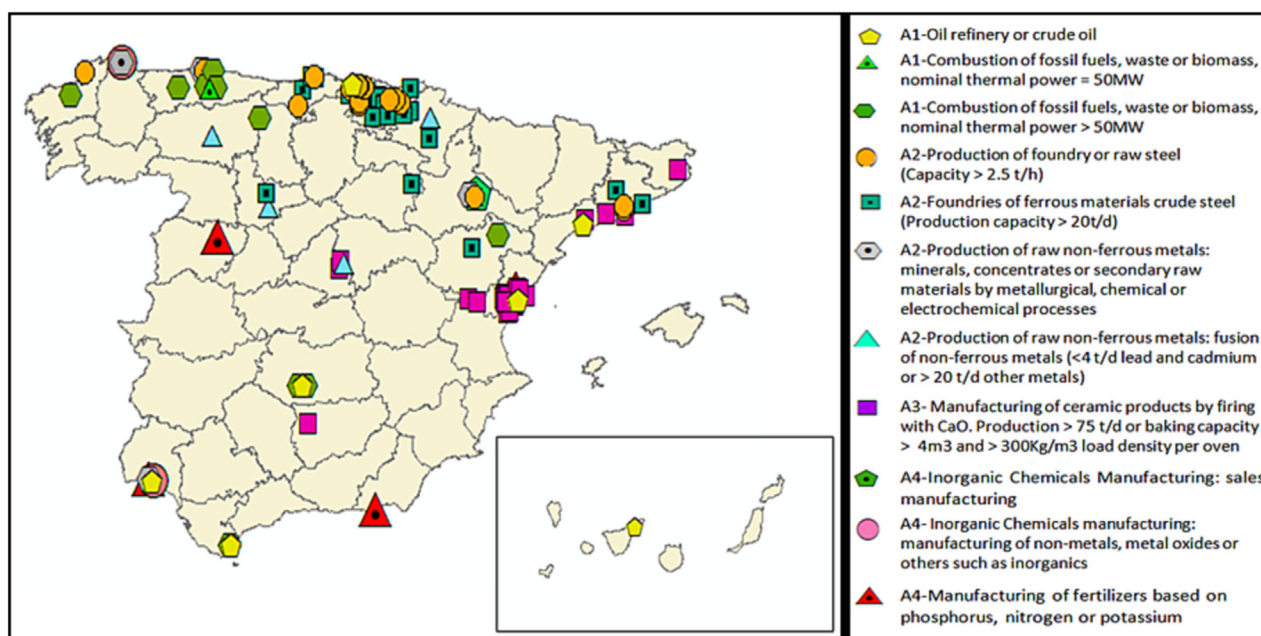


Fig. 7. The example of the companies corresponding to the different NORM involving industries mapping in ArcGIS elaborated in Spain.

activity in frame of the RadoNorm project and described in Mrdakovic Popic et al., 2023 (submitted for publication).

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Laureline Fevrier (IRSN, France), methodology, funding acquisition, project administration

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Lindis Skipperud, (NMBU, Norway), literature review, methodology,
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Antti Kallio (STUK, Finland) – literature review, methodology,
Jelena Mrdakovic Popic (DSA, Norway) - literature review, methodology, writing, review, supervision, activity coordination

Data availability

The data that has been used is confidential.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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