Ref. Ares(2025)4365518 - 30/05/2025

Deliverable 2.18

Evaluation of up-dated dose-assessment at NORM involving situations

Work Package 2

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This project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 900009.

Document information

Project Acronym	**RadoNorm**
Project Title	**Towards effective radiation protection based on improved scientific evidence and social considerations - focus on radon and NORM**
Project Type	**RIA**
EC grant agreement No.	**900009**
Project starting / end date	**1st September 2020 – 31 August 2025**
Work Package No.	**2**
Work Package Title	**Exposure**
Deliverable No.	**2.18**
Deliverable Title	Original Title: Evaluation of up-dated dose-assessment at NORM industrial/legacy sites The title has been modified to “Evaluation of up-dated dose-assessment at NORM involving situations” to account for generic situations that have been evaluated and which do not refer to specific sites (locations).
Lead Beneficiary	**ISS**
Contractual Delivery Date	**28.02.2025**
Actual Delivery Date	**31.05.2025**
Type	**Report**
Dissemination level	**Public**
Authors	**Gennaro Venoso (ISS), Cristina Nuccetelli (ISS), Christian Di Carlo (ISS), Rosabianca Trevisi (INAIL), Federica Leonardi (INAIL), Raffaella Ugolini (ARPAV), Flavio Trotti (ARPAV), Danyl Pérez-Sánchez (CIEMAT), Almudena Real (CIEMAT), Jelena Mrdakovic Popic (DSA), Justin Brown (DSA), Amandine Katz (ASNR), Paul Wigniolle (ASNR), Edouard Veilly (ASNR), Helene Caplin (ASNR), Laureline Février (ASNR), Thuro Arnold (HZDR), F. Bok (HZDR), G. Montavon (SUBATECH), C. Bertin (BRGM), Laura Urso (BfS)**

To be cited as:

Venoso G., Nuccetelli C., Di Carlo C., Trevisi R., Leonardi F., Ugolini R., Trotti, F., Pérez-Sánchez D., Real A., Mrdakovic Popic J., Brown J., Katz A., Wigniolle P., Veilly E., Caplin H., Février L., Arnold T., Bok F., Montavon G., Bertin C., Urso L. (2025): Evaluation of up-dated dose-assessment at NORM involving situations. Deliverable D2.18 of project RadoNorm.

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Acknowledgement

This document is a deliverable of the RadoNorm project. This project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 900009.

Status of deliverable		
	By	Date
Delivered (Lead Beneficiary)	Gennaro Venoso (ISS)	26/05/2025
Verified (WP Leader)	Laureline Février (ANSR)	28/05/2025
Reviewed (Reviewers)	WP Leader, PC	30/05/2025
Approved (PC)	PC	30/05/2025
Submitted to EC (PC)	PC	30/05/2025

Executive Summary

This RadoNorm Deliverable 2.18 presents an evaluation of dose assessment methodologies relevant to various situations involving radiation exposure due to Naturally Occurring Radioactive Materials (NORM). Based on recent scientific advances and evolving regulatory frameworks, particularly the European Basic Safety Standards (Council Directive 2013/59/Euratom), this work critically reviews the underlying assumptions, parameter choices and mathematical models. The primary objective is to provide updated, scientifically robust approaches for estimating radiological doses to humans and the environment across four NORM-relevant topics, thereby supporting informed decision-making in radiation protection.

The first area of investigation focused on the development of methodologies for assessing dose due to managing NORM residues in two distinct, yet common, end-of-life or reuse scenarios: the disposal of cleared NORM residues in conventional landfills and the agricultural application of NORM-containing sludge. Both analyses shared a common strategic objective: to establish screening levels called Operational Levels (OLs). These OLs are defined as activity concentrations of radionuclides of natural origin that, under specific exposure scenarios, would ensure compliance with established dose criteria, serving as practical screening tools for operators and regulators. A unifying element in both studies was the consistent application of updated ICRP dose coefficients for intake and external radiation and the utilization of internationally recognized modelling software, primarily the RESRAD codes, to estimate effective doses to workers and members of the public.

In the context of conventional landfill disposal, the primary aim was to determine OLs for NORM residues that would ensure the annual effective dose to landfill workers and members of the public remains below 1 mSv/year. The methodological framework involved the definition of relevant exposure pathways (for each scenario), drawing upon relevant European guidelines such as *Radiation Protection 122 Part 2*, and the requirements of the European Directive 1999/31/EC for conventional landfill. A significant finding was that the landfill worker scenario often proved to be the dose-limiting case. The derived OLs for key decay chain segments, such as approximately 1.9 kBq/kg for Th-232 in secular equilibrium (Th-232sec) and around 2.5 kBq/kg for U-238 in secular equilibrium (U-238sec), were generally found to be higher than existing EU Clearance Levels and those reported in previous UK-based estimations. This discrepancy was primarily attributed to the incorporation of newer, and often lower, external radiation dose coefficients provided by recent ICRP publications. A particularly valuable outcome of this investigation was the demonstration that these derived OLs exhibited substantial independence from the specific physical dimensions (e.g., area, thickness) of the landfill, a characteristic that significantly enhances their generic applicability for preliminary screening and regulatory decision-making across a variety of landfill settings.

Similarly, the study addressing the agricultural use of NORM-containing sludge (e.g., from groundwater treatment facilities) focused on evaluating potential doses to resident farmers and deriving corresponding OLs. This assessment considered dose criteria of both 1 mSv/year and a more conservative 0.3 mSv/year. Scenarios of occasional (single) sludge application and, more critically, continuous annual application over a 50-year period, with dose projections extending to 100 years, were considered. Across various ICRP age classes, the infant age class was typically identified as the critical class due to higher radiological sensitivity and specific exposure parameters. Radium isotopes (Ra-226+, Ra-228+) showed to be the most radiologically significant, with OLs around 1 kBq/kg for a 0.3 mSv/year criterion considering sludge spreading on the same land over 50 years. These OLs offer conservative screening tools, aiding decisions that balance radiation protection with circular economy goals.

The second investigation explored the feasibility of complex water flow and solute transport models for NORM involving situations, using the Rophin legacy uranium mine site as a case study. Detailed site characterization informed conceptual models, which were then simulated using the 1D CIEMAT

model and the 3D MELODIE code. One-dimensional (1D) models simplify flow to a single direction, offering computational efficiency for assessing processes like vertical water movement or solute transport in stratified media. Conversely, three-dimensional (3D) models simulate flow in all spatial dimensions, providing a more realistic representation of complex aquifer systems, surface-groundwater interactions, and heterogeneous hydrogeology. Although 3D models offer greater accuracy, they demand significantly more data, computational resources, and calibration effort. The selection between 1D and 3D approaches is dictated by study objectives, data availability, system heterogeneity, and the scale of investigation.

Finally, a comprehensive review of approaches and tools for assessing the impact of NORM on non-human biota was conducted. It covered methodologies recommended by ICRP (Reference Animals and Plants, Derived Consideration Reference Levels), approach suggested in IAEA Standards, European initiatives (notably the ERICA Integrated Approach and Tool), and US frameworks (RESRAD-BIOTA). The ERICA Tool was identified as a possible robust starting point, with a tiered, graded assessment approach widely used. Key NORM-specific considerations include accounting for regional background radionuclide levels, adapting tools for diffuse sources, meticulously addressing natural decay chain disequilibrium, and specifically managing radon and thoron exposure pathways, for which ERICA provides tailored functionalities.

In summary, Deliverable 2.18 provides significant updates to methodologies for dose assessment in the context of NORM. It offers refined methodologies and screening values, underscores the impact of new dosimetric data, and emphasizes the necessity of scenario-specific considerations and tiered assessment strategies. The findings presented herein are also intended to provide a directly informative basis for the development of dose-assessment procedures to be included in the subsequent RadoNorm Deliverable 2.19.

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Glossary

Conservative dose assessment is an estimate of effective dose carried out by choosing parameters, assumptions and mathematical structures such that the calculated effective dose never underestimates the real effective dose.

Dose assessment tool: a software/numerical tool that provides as output the effective dose. Equations, parameters that model the environmental pathways (e.g. transfer factors, distribution coefficients) and exposure pathways - ingestion, inhalation, external irradiation - (e.g. dose coefficients). Also, radionuclide decay is included as well as ingrowth of progenies.

Dose assessment: exposure assessment for humans or non-human biota, in which the considered agent is a radionuclide and a (effective) dose is quantified.

Dose coefficients (DCs) are quantities that, when multiplied by a measurement of radionuclide intake, air (kerma), particle (fluence), or environmental radioactivity concentration, will yield an organ equivalent dose or the effective dose to the exposed individual. The units of the dose coefficients are (Sv Bq^{-1}) for inhalation and ingestion and (Sv g Bq^{-1} s^{-1}) for external exposure, respectively.

Non-human-biota: Flora and fauna, in general living organisms other than human ones.

NORM-involving legacy sites are areas affected by past industrial activities involving NORM. They are abandoned or ownership is uncertain.

Radioecological model: combination of mathematical structure of the model, discrete values or probability distributions of model parameters, assumptions (e.g. conventions about human habits, exposure times).

Realistic dose assessment is an estimate of effective dose carried out by choosing parameters, assumptions and mathematical structures such that the estimated dose is of the order of the real dose.

Reference person: An idealised person for whom the equivalent doses to organs and tissues are calculated by averaging the corresponding organ doses in the Reference Male and Reference Female. The equivalent doses of Reference Person are used for the calculation of effective dose.

Representative person: an individual receiving a dose that is representative of the more highly exposed individuals in the population, excluding those individuals having extreme or rare habits.

Acronyms and abbreviations

ADR, formally the Agreement of 30 September 1957 concerning the International Carriage of Dangerous Goods by Road is a 1957 United Nations treaty that governs transnational transport of hazardous materials. "ADR" is derived from the French name for the treaty: ***A**ccord relatif au transport international des marchandises **D**angereuses par **R**oute*).

AOL: Annual Operational Level

AMAD: activity median aerodynamic diameter

ARPAV: Agenzia Regionale per la Prevenzione e Protezione Ambientale del Veneto (Environmental Protection Agency of Veneto, Italy)

ASNR: Autorité de Sûreté Nucléaire et de Radioprotection (French Nuclear Safety and Radioprotection Authority

BCG: Biota Concentration Guides

BLM: Biotic Ligand Model

BRGM: Bureau de Recherches Geologiques et Minières (French Geological Survey)

CEA: French Alternative Energies and Atomic Energy Commission

CIEMAT: Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Research Centre for Energy, Environment and Technology, Spain)

CL: Clearance Level

DCs: Dose Coefficients

DCLR: Derived Consideration Reference Level

DOC: Dissolved Organic Carbon

DSA: Direktoratet for strålevern og atomsikkerhet (Norwegian Radiation and Nuclear Safety Authority)

ECOMOD: Ecological MODel (for radionuclides in aquatic biota)

EDEN: Elementary Dose Evaluation for Natural environment

EMRAS: Environmental Modelling for RAdiation Safety

ENA: European NORM Association

ENVIRONET: IAEA Network of Environmental Management and Remediation

EPA: Environmental Protection Agency (USA)

ERICA: Environmental Risk from Ionising Contaminants: Assessment and Management Tool

EU-BSS indicates the European Basic Safety Standard for protection against dangers arising from exposure to ionising radiation - Council Directive 2013/59/Euratom of 5 December 2013

FASSET: Framework for ASSessment of Environmental impact

FREDERICA: Font for the REassessment of Doses from Environmental RAdioactivity (database)

FVFE: Finite Volume Finite Element

GCLs: General Clearance Levels

HYDRUS: Software Package for Simulating the One-Dimensional Movement of Water, Heat, and Multiple Solutes in Variably-Saturated Media

HYTEC: Module-oriented modeling of reactive transport

HZDR: Helmholtz-Zentrum Dresden-Rossendorf

IAEA: International Atomic Energy Agency

ICRP: International Commission on Radiological Protection

IRSN: Institut de Radioprotection et de Sûreté Nucléaire (Institute for Radiological Protection and Nuclear Safety, France)

ISS: Istituto Superiore di Sanità (National Institute of Health, Italy)

INAIL: Istituto Nazionale Assicurazione contro gli Infortuni sul Lavoro (National Institute for Insurance against Accidents at Work, Italy)

K_d: Distribution Coefficient

LIETDOS-BIO: LIThuanian DOsimetry system for BIOta

MELODIE: Model for long-term assessment of radioactive waste repositories *(Modèle d'Evaluation à LOng terme des Déchets Irradiants Enterrés)*

MODARIA: MOdelling and DAta for Radiological Impact Assessments

NOR: Naturally Occurring Radionuclides

NORM: Natural Occurring Radioactive Materials

NORMALYSA: Model for NORM And LegacY Site Assessment

OIR: Occupational Intake of Radionuclides

OL: Operational Level

ORANO: a multinational nuclear fuel cycle company headquartered in France

RAP: Reference Animals and Plants

RESRAD: RESidual RADioactive materials Tool

RP: Radiation Protection

SADA: Spatial Analysis and Decision Assistance

SUBATECH: Laboratoire de SUBatomique et des TECHnologies associées (Laboratory of Subatomic physics and associated Technologies, France)

TSM: Thermodynamic Speciation Model

UNSCEAR: Scientific Committee on the Effects of Atomic Radiation

USDOE: US Department of Energy

ZATU: Zone-Atelier Territoires Uranifères

1. Introduction

Residues and effluents from the various types of industries that involve Naturally Occurring Radioactive Materials (NORM) can cause an increase of dose both to human and to non-human-biota. Exposure due to inhalation of dust, external radiation, ingestion including use of contaminated groundwater may be the reason for such increase. Situations can occur in which members of the public are exposed to such doses e.g. in legacy sites not properly managed (this can be avoided with a proper knowledge of the contaminated sites and providing adequate mitigation measures); if leachate occur, natural radionuclides might contaminate the ground water or rivers and basins which can later be used for irrigation or water consumption; if control on areas with NORM residues was not properly established or will be lost in the future, free access to members of public is possible or human intrusion e.g. creation of dwells may be possible; if NORM are managed into conventional landfills with a clean soil cover, which might erode with time, wind or human intrusion will cause resuspension of the dust contaminated with radionuclides which can be inhaled by the public. In addition, flora growing at the site and/or fauna living in the area may be exposed.

Dose assessment is not only necessary to understand the NOR impact at specific sites and for real situations, for example to decide whether mitigation activities are needed or not, but also in a regulatory perspective to check compliance with regulations. In order to carry out such dose assessments radioecological models are needed, which quantify the radionuclide concentration in different compartments of the system considered, including estimations of future evolution of contamination, and deliver effective dose to human and/or absorbed dose to non-human-biota.

In light of the European Basic Safety Standard for protection against dangers arising from exposure to ionising radiation (abbreviated here with EU BSS) (European Commission 2014), for which a distinction between natural and artificial sources is not retained anymore and a longer list of NORM industrial sector is considered of radiological concern, scientific and regulatory advances are available (e.g. new dose coefficients, updated dose assessment tools, increased consideration of long-term assessments, more focus on quantifying level of conservatism of models' predictions) and need to be considered to provide updated guidance to stakeholders that have to implement new regulations.

The approach of the EU BSS considering NORM-involving activities as planned exposure situations makes necessary the elaboration of updated guidelines and tools, shared by scientific community and endorsed by EU, for the dose assessment to comply with legal requirements more compelling than in the past.

In this respect, in the past years, activities at international level have been promoted with the intent to harmonise approaches for dose assessments at sites characterised by NORM-related contamination. Examples of such activities can be found within the projects from IAEA like EMRAS I and II, MODARIA and MODARIA II. These projects mainly dealt with development and testing of dose assessment procedures based on tiered-approach at various NORM-involving sites all over the world and on exchange between scientists, focusing on key questions related to physical, chemical and biological processes taking place at these sites, and stakeholders and operators, focusing on application of the new regulations in most pragmatic way. Another IAEA initiative, the ENVIRONET-NORM project, and the activities of the European NORM Association (ENA) deal with practical and regulatory aspects of dose control and mitigation activities.

In line with these activities and goals, RadoNorm Task 2.8 "Updating approaches for modelling long-term prediction of NORM transfer in the environment" focuses on critically reviewing assumptions, choice of parameters and mathematical models underlying dose assessments for NORM-involving situations and on the application of the most suitable models to specific NORM-involving situations. In addition, conceptual thinking of how scientific and regulatory advances may affect the revision of guidance documents (e.g. Radiation Protection 122 Part 2 or Radiation Protection 135) for specific topics is carried out, which will be the basis for the Deliverable 2.19: “Dose-assessment procedure for NORM involving situations. Recommendations".

Such type of analysis is carried out on several topics, which have been prioritised among others based on the relevance for the NORM community and on the experience of colleagues involved in the task.

The prioritised topics, which each is addressed separately, are:

1. Development of a methodology for dose assessment and estimate of amount of NORM residues disposable in conventional landfill.
2. Development of a methodology for dose assessment for use of sludges from liquid effluent depuration from NORM-involving industry in agriculture.
3. Feasibility study for use of complex water flow models for NORM situations.
4. **Non-human-biota.** Review of approaches and tools for assessing impact of NORM on non-human-biota.

The present document provides for each of these topics an overview of the work carried out, especially focussing on the radioecological models and dose assessment procedures under consideration.

2. Development of methodologies to obtain screening levels for assessing exposures related to NORM involving industries

Industries processing NORM generate residues and wastes that, while often containing low concentrations of natural radionuclides, necessitate careful assessment to ensure the protection of workers, the public, and the broader environment (ICRP 2019). This challenge is combined by the increasing societal and regulatory drive towards a circular economy, which encourages the reuse and recycling of materials where possible, and demands safe, sustainable disposal routes for those that cannot be valorised.

This section details two complementary studies sharing a primary objective: to develop robust, scientifically based methodologies for assessing the radiological doses associated with specific NORM management practices and to derive practical screening tools that can help in informed decision-making.

A central, unifying aim is establishing so called *Operational Levels (OLs)*: activity concentration (kBq/kg) for natural radionuclides in NORM residues calculated to ensure annual effective doses to the most exposed individuals (workers and public) remain below established dose criteria (typically 1 mSv y^{-1}). Practically, OLs serve as straightforward, conservative screening tools, usable even by non-experts in. radiation protection, for a preliminary assessment of whether NORM residues can be managed (via reuse or disposal) without significant radiological concern.

To achieve this common goal, both studies employ a consistent methodological framework, which includes: a detailed exposure scenarios (and exposure pathways) definition; a dose assessment modelling using internationally recognized dose assessment software; application of updated dose coefficients published by the ICRP; and generic conservative assumptions regarding the model parameters aiming for broad applicability. Both methodologies are designed to be inherently conservative, thus providing a protective baseline for assessment. This approach allows for subsequent refinement with site-specific data if initial screening using the generic OLs indicates a potential for exceeding given dose criteria.

The first study addresses the end-of-life management pathway, examining the radiological impact of disposing of NORM residues in conventional landfills. This is critical for NORM-containing materials that are not suitable for reuse or for which disposal is the most appropriate management option.

The second study focuses on the valorisation pathway, specifically investigating the radiological implications of using NORM-containing sludge (mainly from groundwater filtration facilities) as an agricultural fertilizer. This directly aligns with circular economy objectives by exploring a beneficial reuse for an industrial by-product.

The outcomes are intended to be valuable for regulators, industry operators, and other stakeholders involved in NORM management and may contribute to the ongoing refinement of relevant national and international guidance.

2.1 Development of a methodology for dose assessment and estimate of amount of NORM residues disposable in conventional landfill

2.1.1 Introduction

The goal of this study is to provide a methodology at European level to estimate disposable annual mass of NORM residues in a conventional landfill, for which dose criterion of 1 mSv y^{-1} for the members of the public and workers at the landfill is fulfilled.

The motivation underlying the study is at least twofold: firstly, the total disposable amount of NORM residues (mass) is not foreseen in the national legislations for final disposal; however, from a practical point of view, it is beneficial to have an idea of what amount of NORM residues in tonnes can be disposed of in a landfill for non-hazardous/hazardous material, complying at present but also in the future with the dose criterion of 1 mSv y^{-1}. Secondly, landfill owners' willingness to accept cleared NORM is often very limited due to misperception of the radiological risks and reluctancy of the local community to accept material that, despite being cleared, is still labelled as 'radioactive' (Gellermann *et al.* 2023). This is even more relevant when material is transported to the landfill as ADR class 7 material (Bergesen *et al.* 2018). Therefore, the study not only aims at providing scientific basis for the calculation of the amount of NORM but also aims at informing the general public about good practices in Radiation Protection, and in a broader context to spread the knowledge of Radiation Protection in society.

Central to the methodology is the derivation of the so-called 'Operational Levels' (OLs) related to the estimated maximum annual dose per unit activity concentration and dose criterion of 1 mSv y^{-1}. The methodology to derive the OLs is the same applied for example in IAEA (2005) and European Commission (2002) to obtain Clearance Levels. Additionally, the link between the OLs and the annual amount of residues (in mass) is provided.

The dose assessment procedure requires the application of radioecological models (including the use of assumptions on exposure scenarios and modelling parameters) suitable for a generic (i.e. valid at European level for unspecific range of situations) and conservative (i.e. the estimated effective dose is surely higher than the real one) assessment in the context of NORM disposal at conventional landfill.

Literature relevant to the topic (e.g. Anderson and Mobbs 2010; Jeong *et al.* 2018; Mora *et al.* 2016), results from e-NORM survey (Mrdakovic Popic *et al.* 2023) and expertise from Task 2.8 participants indicate that additional to the RP 122 Part 2 (European Commission 2002) also the software RESRAD-ONSITE and RESRAD-OFFSITE (Yu 2006) will be used.

This study begins by outlining the European regulatory framework for NORM disposal in conventional landfills. Subsequently, it details the exposure scenarios, pathways, assumptions, and parameters employed for dose assessment. The rationale underpinning the derivation of Operational Levels and the estimation of annual admissible mass for conventional landfill disposal will then be explained. Finally, the results are presented and discussed. A paper detailing all the methods and results has been published in the Journal of Environmental Radioactivity (Venoso *et al.* 2024b).

2.1.2 Regulatory framework for disposal of conventional waste at European landfills (including some examples)

Directive 1999/31/EC establishes the EU's foundational framework for managing conventional waste disposal (European Commission 1999). It categorizes waste into inert, non-hazardous, and hazardous types. Specific requirements include geological barriers (especially for non-inert waste landfills), drainage layers, and sealing liners during operation to prevent groundwater contamination from leachate. Notably, a cover system is implemented post-closure. For some layers, requirements in terms of thickness and permeability K are established (see Figure 1).

	Layer	Requirements of 1999/31/EC	
Put in place after the closure	Top-soil cover	Thickness > 1 m	
	Drainage layer	Thickness > 0.5 m	
	Impermeable mineral layer		
	Artificial sealing liner		
	Gas drainage layer		
	Waste layer		
	Drainage layer		
	Artificial sealing liner	Thickness > 0.5 m Necessary if the requirements for the geological barrier are not met	
	Geological barrier	*For hazardous waste* Thickness > 5 m K ≤ 1.0 10^{-9} m/s	*For non-hazardous waste* Thickness > 1 m K ≤ 1.0 10^{-9} m/s

Figure 1. Conventional landfill typical layers according to the requirements of the 1999/31/EC Directive

The aftercare phase is extensive, typically spanning 30 to 70 years, requiring long-term safeguarding and controls to prevent lasting negative effects without further technical intervention. This includes mandatory, regular monitoring of leachate and groundwater over three decades post-closure.

According to the general requirements for conventional landfills, Member states have integrated NORM residue management into their national regulations, revealing significant variations in regulatory approaches. A few examples are given below.

- **Germany**: Governed by the Radiation Protection Act and Ordinance, Annex 7 specifies clearance conditions (in kBq kg^{-1}) for joint landfilling of NORM residues. If residues meet these conditions, 1 mSv y^{-1} dose criterion is not exceeded, allowing disposal in conventional landfills.
- **France**: Following its 2018 regulatory updates, NORM waste with activity concentrations below 1 kBq kg^{-1} can be disposed of in conventional landfills based on chemical properties. For concentrations between 1 and 20 kBq kg^{-1}, disposal is permitted in conventional landfills specifically authorized to receive NORM, depending upon impact assessments (on workers, the public, and the environment) and the implementation of radiological provisions. Activity concentrations above 20 kBq kg^{-1} necessitate disposal in nuclear waste repositories.
- **Italy**: NORM residues are considered exempted and suitable for conventional landfills if activity concentrations are below specific clearance levels (e.g., 0.5 kBq kg^{-1} for U-238/Th-232 chains, 5 kBq kg^{-1} for K-40, and 2.5 kBq kg^{-1} for Po-210/Pb-210 if radioactive equilibrium is broken). Residues exceeding these levels can still be placed in conventional landfills if a dose assessment confirms compliance with the 0.3 mSv y^{-1} public exemption level. Non-exempted material requires dedicated NORM landfills, which must meet hazardous waste standards and receive specific authorization, including procedures like daily clay covering.
- **Norway**: NORM management is addressed under a "Pollution Act," requiring assessments for human health, non-human biota, and the environment. A graded approach with numerical values for specific radionuclides is employed. NORM waste between 1 kBq kg^{-1} and 10 kBq kg^{-1} can be disposed of in hazardous or non-hazardous landfills with permits from the radiation authority. If the NORM waste is also chemically hazardous, it must go to sites authorized by the Environment Agency for hazardous waste. A dedicated NORM site exists for the oil and gas industry.
- **Spain**: Conventional landfill disposal is allowed if NORM concentrations are below 1 kBq kg^{-1} for U-238/Th-232 chains (assuming secular equilibrium). Conventional management is

considered acceptable if estimated annual effective doses are ≤ 1 mSv y^{-1} for the public and ≤ 6 mSv y^{-1} for landfill workers.

2.1.3 Exposure scenarios

This section details the exposure scenarios used for dose assessment of workers and members of the public. While primarily derived from the RP 122-II guidelines (European Commission 2002), these scenarios incorporate specific assumptions regarding exposure pathways and parameters, which are further elaborated in subsequent sections.

2.1.3.1 Worker handling NORM waste at an active Landfill – *worker scenario (Sc1)*

Workers handling NORM waste during regular landfill operations may encounter the following exposure situations:

a. **Landfill Operator**: Performing tasks such as controlling waste upon arrival, unloading it from trucks, de-bagging, placing waste in storage cells, compacting waste layers, and covering the compacted layers.
b. **Truck Driver:** Transporting waste materials to the landfill (over short or long distances) and involved in loading and unloading these materials.

Calculations in RP 122-II (European Commission 2002) indicate that scenario (a) leads to maximum annual effective doses considerably higher than those in scenario (b). For scenario (a), it is assumed that the landfill is uncovered during active operations, and the worker is inside an excavator cabin, which shields external radiation by a factor of 2. Therefore, this study will focus solely on situation (a), designated as *worker scenario*.

2.1.3.2 Members of the public living near an active landfill – *members of the public scenario (Sc2)*

In this scenario, members of the public are living near the active landfill, which is assumed to be an above-ground heap, according to RP 122-II (European Commission 2002). Similarly to RP 122-II, it is also assumed that the residential area starts at the edge of the landfill with the garden being located 20 m away and walls of the housing being located 25 m away. The distance of a residential area from the landfill site assumed in this study is likely to be an underestimate of the minimum distance. In fact, despite a minimum distance from the landfill for a residential area is not provided in the European Directive for landfill (European Commission 1999), in some national regulations the mandatory minimum distances are much higher than 25 m (e.g. in France residential areas cannot be closer than 200 m).

The members of the public are subject to various possible exposure pathways. In particular:

- the exposure pathway indoors, i.e. the inhalation of dust coming from the outside of the building;
- the exposure pathway in the garden, i.e. the inhalation of dust while staying outdoors;
- consumption of food stuff grown in the garden contaminated by deposition of dust coming from landfill;
- the (lateral) external radiation from the heap during the time spent in the garden and at home.

In contrast with the assumptions made by RP 122-II, *members of the public scenario* does not envelope the scenario of recreational person and of the contamination of groundwater.

The scenario of the recreational person, modelled by RP 122-II with an individual on the top of the heap, was not considered since no access to the landfill is generally possible during the active lifetime (operational and after-care phase) of the landfill. In the next phase the landfill is covered by soil and/or artificial layers which prevent external radiation and intake.

The contamination of groundwater and the exposure due to contaminated drinking water or contaminated food via root uptake is not considered due to the requirements for landfills in European countries. It is in fact highly improbable – due to compulsory monitoring of leachate formation and any possible percolation into deeper soil layers and aquifer during active lifetime of a landfill including the aftercare phase – that any contamination may occur reaching the members of the public. A systematic overview of situations at landfill in relation to NORM residues' disposal has been carried out recently in Belgium (Pepin *et al.* 2021). By means of monitoring activities, it was found that no increased levels of activity concentration of NORM exist near the active landfills and that only slightly increased levels of activity concentrations are found in groundwater near closed landfills, which have been considered as being of no radiological relevance (Pepin *et al.* 2021). Similarly to *worker scenario*, also for this scenario it is assumed conservatively that landfill is not covered during its operative phase. In Table 1, an overview of the exposure scenarios related to landfilling activities and NORM, main assumptions and relevant exposure pathways considered in the present study to evaluate the annual effective dose is given.

Table 1. Summary of exposure scenarios related to landfill disposal, relevant exposure pathways, time frames considered for dose calculations as well as age groups considered in the dose assessment procedure.

Exposure scenarios	Representative person	Time frame	Exposure pathways
Worker scenario	Worker (not occupationally exposed)	Operative phase of a landfill (35 years)	- External radiation - Inhalation of dust - Inadvertent ingestion of soil
Members of the public scenario	Members of the public (6 ICRP age groups)	Operative phase of a landfill and post-operational phase without loss of control by the authorities (150 years)	- Consumption of food stuff grown in the garden contaminated by deposition of dust coming from landfill - Inhalation of contaminated dust indoors and outdoors - (Lateral) external radiation from the heap while in the garden and in the house

2.1.3.3 Members of the public *long-term* scenario (Sc3)

Long-term scenarios pertaining to the disposal of NORM wastes are occasionally addressed in the literature (Anderson and Mobbs 2010; Mora *et al.* 2016). A notable example is the "resident farmer" scenario, an intrusion event considered for the post-closure phase of a landfill. This scenario presumes the construction of a residential dwelling on the former landfill site long after closure, once institutional control and societal memory of the waste's presence have diminished. Such intrusion scenarios are typically evaluated in safety analyses for low-level waste disposal sites.

In such a scenario, exposure pathways related to dust inhalation and ingestion of food contaminated via root uptake or foliar deposition would likely become significant only following substantial erosion of the protective soil cover. However, this is considered a low-probability event, as excavation activities for construction would likely reveal the presence of waste if the cover thickness is less than 1 meter (Anderson and Mobbs 2010). Consequently, the primary radiation exposure for members of the public residing in such an area and producing a significant portion (at least half) of their food (vegetables, fruit, milk, meat) on-site would predominantly arise from the contamination of groundwater utilized for drinking, crop irrigation, and livestock watering. Additionally, inhalation of radon, originating from its exhalation from underlying waste layers, could also contribute to exposure.

The modelling of this long-term scenario is critically dependent on the methodologies employed for the groundwater pathway assessment and necessitates specific assumptions regarding landfill dimensions, capacity, geographical location, and local environmental characteristics (e.g., hydrogeological conditions). These factors are inherently site-specific, rendering generic modelling of the groundwater pathway challenging and characterized by substantial uncertainty, particularly for extended timeframes. Specifically, the uncertainties associated with requisite input parameters (which would need to be representative of generic European conditions) can lead to unrealistic or extreme outcomes. For instance, the application of default parameter values within modelling tools such as RESRAD can result in Operational Limits (OLs) for Ra-226+ of approximately 0.03 kBq kg^{-1}, a value comparable to typical background concentrations of Ra-226 in soils. Furthermore, significant hydrogeological changes can occur over the protracted timeframes considered (i.e., >1,000 years). Unlike the case for artificial radionuclides, the modelling of groundwater pathways for NORM to evaluate clearance levels necessitates a site-specific approach that is not readily generalizable.

Moreover, at the European level, the reliance on private wells for drinking water is not a common practice for a representative individual. Therefore, this "resident farmer" intrusion scenario was not incorporated into the quantification of the annual admissible mass for landfill disposal in the present study. This decision aligns with the conventional approach to landfilling, wherein considerations regarding the temporal evolution of contamination are typically confined to the active and aftercare phases, as detailed in Section 2.1.2.

2.1.4 Exposure pathways and dose assessment

The dose assessments in this work utilized models outlined in RP 122 Part 2 (European Commission 2002) alongside those implemented in the RESRAD ONSITE and OFFSITE software (collectively referred to as RESRAD; (Yu 2006)). For each radionuclide, assessments were conducted at a nominal activity concentration of 1 kBq kg^{-1}, enabling straightforward re-scaling of the findings to different concentration levels. Dose evaluations were carried out for each segment of decay chains listed in Table 2.

Table 2. Segments of the natural radionuclide series considered and list of radionuclides that are assumed to be in secular equilibrium. Segments and radionuclides are the same in RP 122 Part 2.

Segment of decay chain	Radionuclides in secular equilibrium
U-238sec	U-238, Th-234, Pa-234m, Pa-234 (0.3%), U-234, Th-230, Ra-226, Rn-222, Po-218, Pb-214, Bi-214, Po-214, Pb-210, Bi-210, Po-210 (and 4.6% of U-235sec)
Unat	U-238, Th-234, Pa-234m, Pa-234 (0.3%), U-234, U-235 (4.6%), Th-231 (4.6%)
Th-230	Th-230
Ra-226+	Ra-226, Rn-222, Po-218, Pb-214, Bi-214, Po-214
Pb-210+	Pb-210, Bi-210
Po-210	Po-210
U-235sec	U-235, Th-231, Pa-231, Ac-227, Th-227 (98.6%), Fr-223 (1.4%), Ra-223, Rn-219, Po-215, Pb-211, Bi-211, Tl-207 (99.7%), Po-211 (0.3%)
U-235+	U-235, Th-231
Pa-231	Pa-231
Ac-227+	Ac-227, Th-227 (98.6%), Fr-223 (1.4%), Ra-223, Rn-219, Po-215, Pb-211, Bi-211, Tl-207 (99.7%), Po-211 (0.3%)

Segment of decay chain	Radionuclides in secular equilibrium
Th-232sec	Th-232, Ra-228, Ac-228, Th-228, Ra-224, Rn-220, Po-216, Pb-212, Bi-212, Po-212 (64.1%), Tl-208 (35.9%)
Th-232	Th-232
Ra-228+	Ra-228, Ac-228
Th-228+	Th-228, Ra-224, Rn-220, Po-216, Pb-212, Bi-212, Po-212 (64.1%), Tl-208 (35.9%)
K-40	K-40

Note: The percentage in parenthesis of some radionuclides refers to the activity concentration

Results obtained for the U-235 series are purely informative as the chain U-238sec is assumed to be in natural isotopic ratio with the U-235sec chain, and thus its chain members have lower activity concentrations by a factor of about 20.

For the estimation of annual dose from NORM disposal, and in alignment with the defined exposure scenarios, the representative persons considered are landfill workers and members of the public. The latter group includes individuals residing near the landfill during its operational phase and those potentially inhabiting the site long after closure. Dose assessments for members of the public encompass all six age groups: infant, 1-year-old, 5-year-old, 10-year-old, 15-year-old, and adult (ICRP 2007).

Effective dose is determined by radionuclide concentrations in environmental media (e.g., plants, milk, meat, water, soil, dust) following their transfer from a contamination source via environmental pathways (see Figure 2). Dose assessment models, which include assumptions, parameters, and mathematical equations, must be suited for conservative calculations. This means the calculated dose should intentionally overestimate the actual dose, while minimizing excessive conservatism.

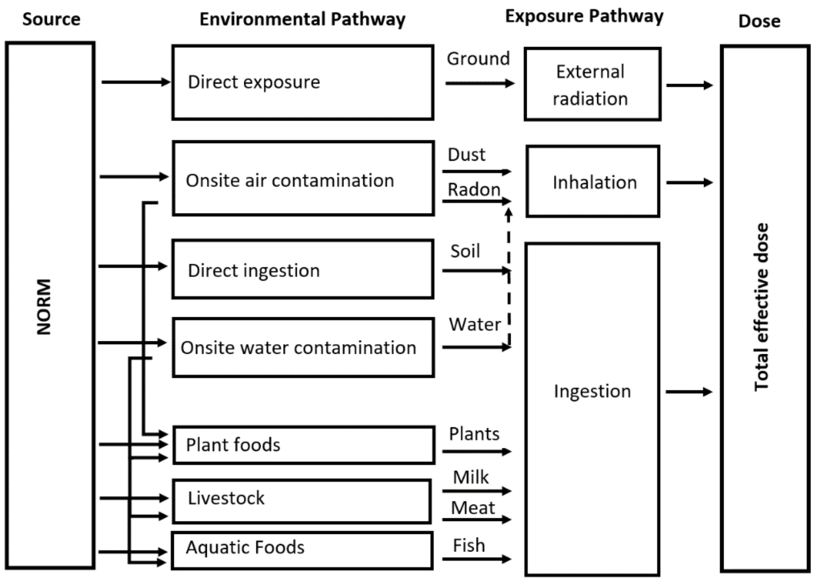


Figure 2. Relevant environmental and exposure pathways for evaluating possible exposure due to NORM.

In the following, the details of how the effective dose has been calculated for the main exposure pathways will be presented.

2.1.4.1 Dose from intake

In scenarios Sc1 and Sc2, which pertain to workers and members of the public respectively, the calculations employed to estimate the dose resulting from various sources – namely, inhalation of dust, inadvertent ingestion of soil, and ingestion of foodstuffs contaminated by dust – operate under the assumption of a constant waste concentration, equivalent to that which occurred at the time of landfill disposal. In summary, the impact of decay and ingrowth over time is not taken into consideration. It is therefore implicitly assumed that fresh material is constantly brought to the landfill, and this process determines steady concentrations of nuclides.

According to the RP 122 Part 2 approach for the worker scenario (Sc1), the annual dose from inhaling dust ($D_{i,inh}$) is calculated per unit activity concentration of NORM waste of each segment of decay chain *i*.

$D_{i,inh}$ is expressed in [(Sv y^{-1})/(kBq kg^{-1})] and quantified as follows:

$$D_{i,inh} = DC_{i,inh} \cdot c_{dust} \cdot B_{rate} \cdot t_e \cdot f_k \qquad \text{(Eq. 1)}$$

where:

$DC_{i,inh}$ is the **inhalation** dose coefficient for worker for segment of decay chain i (in Sv Bq^{-1});

c_{dust} is the amount of dust at the contamination site or offsite (g m^{-3});

B_{rate} is the breathing rate (in m^3 h^{-1})

t_e is the exposure time (in hours per year)

f_k is the fraction of activity that binds to the small particles causing the activity to concentrate in the fine dust fraction and is set to 1 in this work.

For the member of the public scenario (Sc2), the annual effective dose from inhaling dust during time spent outdoors in the garden ($D_{i,inh,garden}$) and indoor ($D_{i,inh,house}$) is quantified per unit activity concentration for each segment of decay chain *i* and for each representative person group (see Section 3.2). $D_{i,inh,graden}$ and $D_{i,inh,house}$ are expressed in [(Sv y^{-1})/(kBq kg^{-1})] and quantified as follows:

$$D_{i,inh,garden} = DC_{i,inh} \cdot c_{dust,garden} \cdot B_{rate} \cdot t_{ingarden} \cdot f_k \qquad \text{(Eq. 2)}$$

$$D_{i,inh,house} = DC_{r,inh} \cdot c_{dust,house} \cdot B_{rate} \cdot t_{inhouse} \cdot f_k \qquad \text{(Eq. 3)}$$

where $DC_{i,inh}$ is the inhalation dose coefficient (see Table A1) for segment of decay chain *i* (in Sv Bq^{-1}); $t_{ingarden}$ and $t_{inhouse}$ are the exposure times (hours per year), i.e. the time the representative person spends outdoors and indoors, respectively:
B_{rate} $(m^3\ h^{-1})$ is the breathing rate of the representative person per age group;

$c_{dust,house}$, $c_{dust,garden}$ $(g\ m^{-3})$ are the dust concentrations for each segment of a decay chain indoors and outdoors, respectively;

f_k is the concentration factor for the activity in the inhalable dust fraction and is set to 1.

According to the RP 122 Part 2 approach for the worker scenario (Sc1), the annual dose due to inadvertent **ingestion** of soil ($D_{i,ing}$) per unit activity concentration for each segment of decay chain *i* is calculated. $D_{i,ing}$ is expressed in [(Sv y^{-1})/(kBq kg^{-1})] and quantified as follows:

$$D_{i,ing} = DC_{i,ing} \cdot r_{ing} \cdot t_e \qquad \text{(Eq. 4)}$$

where $DC_{i,ing}$, are the dose coefficients for worker (see Table A2) for segment of decay chain *i* (in Sv Bq^{-1});

r_{ing} is the soil ingestion rate (in g/h)

t_e is the exposure time (in hours per year).

For member of the public scenario (Sc2), the equation to quantify the annual effective dose from ingesting food products ($D_{i,ing,dust}$ in Sv y^{-1}) grown in the own garden and contaminated by dust coming from the landfill, is:

$$D_{i,ing,dust} = DC_{i,ing} \cdot (1 - r_{market}) \cdot (FC_{leafy} + FC_{other\,veg}) \cdot r_{prep} \cdot C_{crop,dust} \qquad \text{(Eq. 5)}$$

The food consumption rates FC_{leafy} and $FC_{other\,veg}$ $(kg\,y^{-1})$ are for leafy vegetables and other products grown onsite, respectively, and are provided in Table A5 of Supplementary materials. $r_{market} = 0.5$ is the relative contribution of food that is bought at the market, $r_{prep} = 0.5$ is the preparation reduction factor for food.

The concentration $C_{crop,dust}$ (in Bq kg^{-1}) of each segment of decay chain in the crop following deposition of dust is calculated as:

$$C_{crop,dust} = C_{A-dust} \cdot c_{dust,garden} \cdot \left(v_{dep} \cdot 86400\,\frac{s}{days}\right) \cdot \left(\frac{1-e^{-\lambda_w t_{dep}}}{\lambda_w}\right) \cdot f_{dil} \cdot f_{ret} \cdot f_{tr} \qquad \text{(Eq. 6)}$$

where:

$C_{A-dust} = 1\,kBq\,kg^{-1}$ is the unit activity concentration of the decay segment present in the dust.

$C_{dust,garden} = 10^{-4}\,g\,m^{-3}$ is the dust concentration outdoor.

$v_{dep} = 0.001\,m\,s^{-1}$ is the deposition velocity.

$\lambda_w = 0.05\,days^{-1}$ is the environmental removal rate for all plant surfaces.

$t_{dep} = 60$ days is the duration during which dust deposition can occur on crops

$f_{dil} = 1$ is the dilution factor, i.e. fraction of NORM present in waste.

$f_{ret} = 0.4$ is the retention factor which characterizes the fraction of dust that is incorporated into the plants.

$f_{tr} = 0.1\,m^2\,kg^{-1}$ is the translocation factor, which account for the translocation of the radionuclides from foliage to the edible tissues of the vegetation.

It is worth noting that λ_w accounts for the reduction in the concentration of material deposited on plant surfaces due to processes other than radioactive decay. Since its value is significantly higher than the typical decay constants of NORM radionuclides, the removal due to radioactive decay can be considered negligible.

2.1.4.2 Dose from external radiation

The RESRAD software suite (Yu 2006), which embeds equations for quantifying annual effective doses from external radiation, was employed for this exposure pathway. Its dose library was updated for this study using the coefficients detailed in 2.1.5.6. RESRAD facilitates the modification of input parameters, selection of age-specific dose coefficient libraries, and can track the temporal evolution of contamination, including decay and progeny ingrowth. Default dose coefficients assume exposure to gamma radiation from a semi-infinite volume source (person centred 1 m above ground). These are adjusted using radionuclide-specific depth-and-cover correction factors to account for finite contaminated soil depth and potential cover. Additionally, area-and-shape factors, derived via the point-kernel method (Kamboj *et al.* 2002), model geometries differing from the semi-infinite volume.

For the worker scenario, exposure was modelled with the worker positioned 1m above a uniformly contaminated ground surface (named 'contaminated zone' in RESRAD-ONSITE). A constant waste concentration, equivalent to that at the time of landfilling, was assumed. Consequently, decay and ingrowth were not considered, based on the rationale that older, more deeply buried material does not significantly contribute to surface external radiation. An instantaneous dose (at t=0) was calculated by setting RESRAD's time integration to a single point.

Conversely, for the public scenario, RESRAD-OFFSITE modelled the landfill as an above-ground, uniformly contaminated heap. Public exposure to lateral external radiation was assessed at distances of 20 m (garden to heap) and 25 m (house wall to heap). In this scenario, the temporal evolution of radionuclide concentrations was considered, as the above-ground landfill configuration means material on its sides may have been deposited years prior. Therefore, equilibrium within decay chains (see Table 2) was assumed only at the initial disposal time (t=0).

2.1.5 Assumptions and parameters used in this study

Parameter values were primarily adopted from RP 122 Part 2 (European Commission 2002), with any modifications explicitly stated herein. For RESRAD calculations, software default parameters were retained unless otherwise specified (see Table A3 and Table A4). Assumptions and parameters were generally selected to reflect European lifestyle patterns.

2.1.5.1 Landfill size and characteristics

For radiation protection assessments, landfill structures are typically simplified compared to actual engineered facilities. This study incorporates minimum layer thickness and hydraulic conductivity requirements from the European Directive for conventional landfills (European Commission 1999) but excludes advanced engineering features (e.g., leachate collection, gas extraction). The landfill was modelled as a homogeneous repository. These are the landfill sizes and characteristics chosen for each scenario:

- *Worker Scenario (Sc1)*: Landfill volume was 10^6 m³, with a 10 m waste thickness, 10^5 m² area, and a waste density of 1500 kg m^{-3}. This results in a total capacity of 1.5 x 10^6 Mg.
- *Member of the Public Scenario* (Sc2): the landfill was modelled as an above-ground heap with a height of 10 m, an area of 10^5 m², and a length of 300 m.

2.1.5.2 Exposure times

The exposure time of workers (Sc1) is set to 1800 hr y^{-1}. Exposure times of the members of the public living nearby during the active life of the landfill (Sc2) are 1760 hr y^{-1} outdoors and 7000 hr y^{-1} indoors, respectively, i.e. a person spends 80% indoors and 20% outdoors near the contaminated area.

2.1.5.3 External gamma penetration factor in the house

For assessing indoor exposure from the heap in Sc2, RESRAD-OFFSITE's external gamma penetration factor, representing the fraction of outdoor gamma radiation penetrating house walls, was set to 0.25. This value, deviating from the default 0.7, was derived from a MicroShield® assessment considering higher-energy radionuclides (K-40, Tl-208) and a 20 cm concrete wall. This configuration provides >80% effective dose reduction, rendering a 0.25 penetration factor sufficiently conservative.

2.1.5.4 Dust concentration in air

Airborne dust concentration (C_{dust}) was set to 1 mg m^{-3} at the landfill site during operations (Sc1, assuming no cover) and 0.1 mg m^{-3} outside the landfill perimeter (Sc2). These are conservative estimates accounting for periods of both high mass loading and normal activity.

2.1.5.5 Food consumption rates and breathing rates

Annual food consumption rates were adopted from BMUV (2020), representing very conservative German values, even if it is acknowledged that consumption patterns may vary geographically. The inadvertent soil ingestion rate (r_{ing}), combining outdoor soil and indoor dust, was 18 g y^{-1} for all age classes except infants (0 g y^{-1}).

Annual breathing rates for public age groups were sourced from ICRP (1994). For Sc1, the worker breathing rate (1.2 m^3 h^{-1}) was higher than the adult residential rate, reflecting greater physical exertion during work.

2.1.5.6 Dose coefficients

This study employs **inhalation** dose coefficients (DCs) for landfill workers based on an activity median aerodynamic diameter (AMAD) of 5 µm, consistent with the recommendations of RP 122-II (European Commission 2002). The specific DCs utilized herein are derived from the most recent International Commission on Radiological Protection (ICRP) publications concerning Occupational Intake of Radionuclides (OIR) series, specifically ICRP Publication 137 (Paquet *et al.* 2017), ICRP Publication 141 (Paquet *et al.* 2019), and ICRP Publication 151 (Paquet *et al.* 2022).

For each radionuclide, inhalation DCs are dependent on its chemical form, which determines the rate of absorption into the bloodstream via the respiratory tract. Accordingly, ICRP typically provides DCs for three material types, categorized by their solubility and subsequent Lung Clearance Class: Type S (Slow), Type M (Medium), and Type F (Fast). In cases where specific information to assign a material to an absorption type is unavailable, ICRP recommends a default type, which is predominantly Type S. It should be noted, however, that the DC for the default type does not invariably correspond to the highest value and, consequently, may not represent the most conservative choice. The present work follows the approach recommended by the ICRP for default type selection. The lung absorption class utilized for each radionuclide in this study is detailed in Appendix A.

For members of the public, inhalation DCs corresponding to an AMAD of 1 µm are applied, which also aligns with the methodology outlined in RP 122-II (European Commission 2002).

Regarding **ingestion** pathways, DCs for workers are sourced from the ICRP-OIR publications, while those for adult members of the general public are obtained from ICRP Publication 72 (ICRP 1995). The coefficients from ICRP Publication 72 are also consistent with those employed in RP 122-II.

For **external radiation**, this study incorporates DCs from the recent ICRP report, "Dose coefficients for external exposures to environmental sources" (Petoussi-Henß *et al.* 2020). The ICRP's calculation methodology for a semi-infinite soil volume source is analogous to that of the U.S. Federal Guidance 15 (Bellamy M. B. *et al.* 2019), although the ICRP model further incorporates electron transport (Satoh and Petoussi-Henss 2024). These external radiation DCs are applied to both *worker and members of the public scenarios*, considering all relevant ICRP age groups. These DCs were used to update the RESRAD dose library as previously mentioned (see Section 2.1.4.2)

A comprehensive list of the ICRP publications from which the dose coefficients for each radionuclide were derived for this study is provided in Table 3.

Table 3. Dose coefficients used in the present study for both the considered scenarios

Representative Person	External radiation	Inhalation AMAD 5 μm	Inhalation AMAD 1 μm	Ingestion
Worker	ICRP 144	*ICRP IOR 2022*: ICRP 141, ICRP 137, ICRP 151	—	*ICRP IOR 2022*: ICRP 141, ICRP 137, ICRP 151
Member of the public	ICRP 144 (6 ICRP age groups)	—	ICRP 72	ICRP 72

In the Appendix A, tables regarding dose coefficients for inhalation, ingestion and external exposure are reported.

2.1.6 Methodology for estimating Operational Levels (OLs) and annual admissible mass disposable at the landfill

The dose criterion is defined as the increment of annual effective dose, above the prevailing natural background, that an individual may experience as a result of exposure to NORM in a given scenario. In this study, the dose criterion was set at 1 mSv y^{-1}, which is the most widely employed dose criterion in Europe (García-Talavera *et al.* 2021). This dose criterion was also applied to landfill workers. These individuals are not considered to be occupationally exposed but rather constitute a subgroup of the general public.

In accordance with the specifications outlined in RP 122 Part 2, an extended dose criterion of 1.037 mSv y^{-1} was employed, inclusive of a background value of 0.037 mSv y^{-1}, for the exclusive application in the context of the worker scenario. It is important to note that, in this particular scenario, the presence of NORM leads to a reduction in the dose attributable to background radiation (European Commission 2002).

For each chain segment of radionuclides reported in Table 2, the operational level (OL) for a given dose criterion is calculated using the following formula:

$$\mathrm{OL}_{\text{Dose criterion}}\left(\frac{\mathrm{kBq}}{\mathrm{kg}}\right) = \frac{\text{Dose criterion}}{\text{Annual dose per unit of activity concentration}} \qquad \text{(Eq. 7)}$$

where the annual dose per unit activity concentration is estimated for the limiting scenario, which represents the scenario that gives rise to the maximum dose among those considered in this study.

The Operational Limits (OLs) denote the average annual activity concentration (in kBq kg^{-1}) for each segment of the chain that can be disposed of annually to comply with the dose criterion. When multiple disposals of NORM waste occur within a year, the OL is calculated as the weighted average of the activity concentrations of each disposal, with the mass of the waste as the weighting factor.

The methodology employed in this study is outlined as follows. For each chain segment of radionuclides (see Table 2):

- The annual effective dose per unit activity concentration for each exposure scenario considered in this study was estimated.
- The scenario that gave rise to the highest dose among those considered in this study was identified. This scenario is referred to as the 'limiting scenario'.
- Operational levels (OLs) were calculated based on the annual dose per unit activity concentration of the limiting scenario
- The quantity of NORM that can be disposed of at the landfill on an annual basis in accordance with the dose criterion was estimated.

For NORM residues with a given activity concentration $C_{res,i}$ of decay chain segment *i*, the annual admissible amount of NORM waste ($M_{res,i}$) that can be disposed of at a landfill is estimated by using the following relation:

$$M_{res,i}(Mg) = M_{landfill\ per\ year}(Mg) \cdot \frac{OL_i\left(\frac{kBq}{kg}\right)}{C_{res,i}\left(\frac{kBq}{kg}\right)} \quad \text{(Eq. 8)}$$

This equation relates the annual admissible amount of NORM residues $M_{res,i}$(Mg) and the annual capacity of the landfill $M_{landfill\ per\ year}(Mg)$, which can be estimated as the total capacity $M_{landfill}(Mg)$ divided by the active lifetime (in years). It is important to note that Operational Levels (OLs) do not impose constraints on the activity concentration of individual waste amounts, as they are based on an annual average. To meet the dose criterion of 1 mSv y^{-1}, the actual constraint is the annual activity (in MBq) disposable in a landfill, which is calculated as the product of the annual landfill capacity ($M_{landfill\ per\ year}$) and the OLs. This can be defined as the Annual Operational Level (AOL) expressed in MBq. Therefore, an individual amount of waste may exceed the OL in terms of activity concentration, provided that the total activity of the waste disposed of within a year remains below the AOL.

If more than one type of NORM residues is delivered or in one NORM residue different segments of decay chains are present, a summation formula can be used for accounting the various contributions, namely:

$$M_{res}(Mg) = M_{landfill\ per\ year}(Mg) \frac{1}{\sum_{i=1}^{n} \frac{C_{res,i}\left(\frac{kBq}{kg}\right)}{OL_i\left(\frac{kBq}{kg}\right)}} \quad \text{(Eq. 9)}$$

2.1.7 Results

Results of the dose evaluation per kBq kg^{-1} for *worker scenario (Sc1) and member of the public scenario (Sc2)* for each segment for decay chain are reported in Table 4.

Table 4. Annual effective doses (in mSv y^{-1}) estimated for Sc1 and Sc2 (values are given with 2 significant digits). The values in **bold** are the maximum dose values for each segment of decay chain.

Segments of decay chain	Sc1 Worker	Sc2 Infant	Sc2 1 year	Sc2 5 years	Sc2 10 years	Sc2 15 years	Sc2 Adult
U-238sec	**0.43**	0.18	0.18	0.18	0.18	0.17	0.17
Unat	**0.06**	0.01	0.01	0.01	0.02	0.02	0.02
Th-230	**0.03**	0.03	0.03	0.03	0.03	0.03	0.03
Ra-226+	**0.29**	0.15	0.15	0.14	0.14	0.13	0.12
Pb-210+	**0.03**	<0.01	<0.01	<0.01	<0.01	0.01	0.01
Po-210	**0.01**	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
U-235sec	**0.35**	0.09	0.12	0.13	0.15	0.16	0.16
U-235+	**0.04**	0.01	0.02	0.02	0.01	0.01	0.01
Pa-231	**0.11**	0.04	0.05	0.04	0.05	0.05	0.05
Ac-227+	**0.20**	0.08	0.10	0.08	0.10	0.15	0.15
Th-232sec	**0.58**	0.23	0.26	0.24	0.24	0.24	0.24
Th-232	0.12	**0.21**	0.21	0.19	0.19	0.18	0.18
Ra-228+	**0.18**	0.14	0.12	0.12	0.12	0.10	0.11
Th-228+	**0.28**	0.13	0.14	0.13	0.13	0.14	0.13
K-40	**0.02**	0.02	0.02	0.02	0.02	0.02	0.01

For Sc1, the annual effective dose to a worker is mainly due to the external radiation and inhalation of dust (Figure 3) and for each segment of a decay chain it never reaches the dose criterion of 1 mSv y^{-1}. Only for the U-238 and Th-232 series in secular equilibrium, the annual dose is greater than 0.4 mSv y^{-1}, mainly attributable to external radiation. Instead, as expected, for chain segments with radionuclides with negligible gamma emissions (e.g. Th-230, Pb-210, Po-210, Th-232), the dose contribution comes mainly from inhalation. For K-40, instead, it is only external radiation that plays a role.

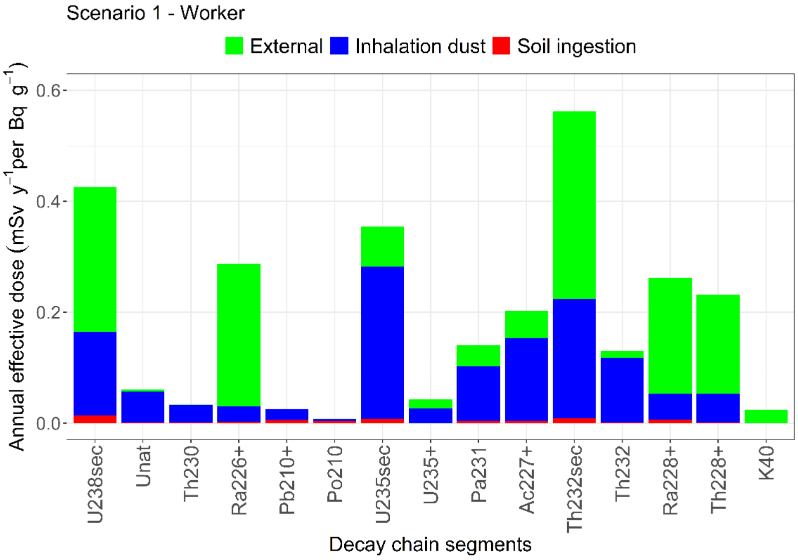


Figure 3. Impact of the various exposure pathways on the estimated annual dose for the worker scenario (Sc1).

As regards Sc2, the external radiation (green) and the inhalation of dust (blue) are the dominant exposure pathways (see Figure 4). The consumption of contaminated food due to the deposition of dust on crops is negligible as it is 10^{-6} mSv y^{-1} at most.

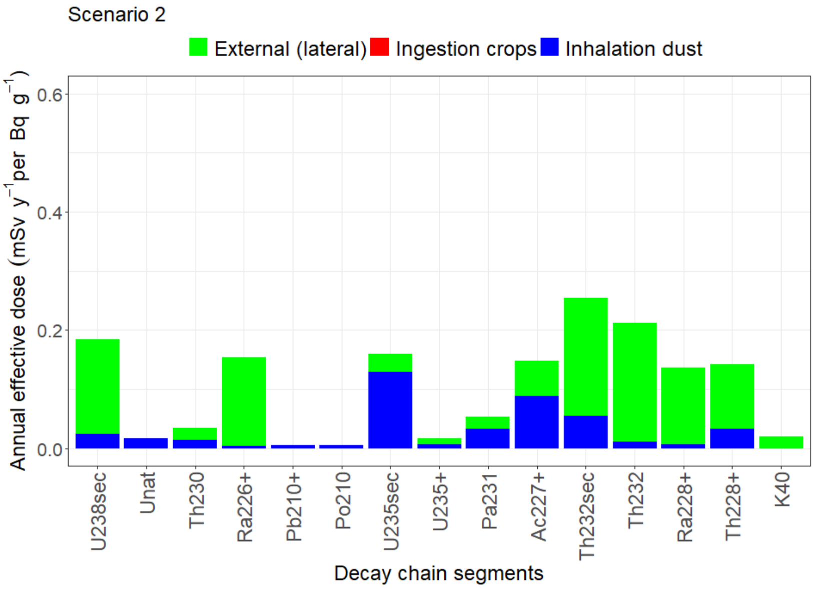


Figure 4. Contribution of the various exposure pathways to total annual effective dose for the most exposed age class for Sc2. The ingestion of contaminated crops is negligible (< 10^{-5} mSv y^{-1}), i.e. not visible on this y-scale.

Operational Limits (OLs) for all segments of the decay chains under consideration are presented in Table 5. The estimated OLs range between 1.9 kBq kg^{-1} (for Th-232sec) and approximately 150 kBq kg^{-1} (for Po-210). Within the U-238 decay chain, the segments yielding the lowest OLs are U-238sec (OL = 2.5 kBq kg^{-1}) and Ra-226+ (OL = 3.7 kBq kg^{-1}). For the Th-232 decay chain, in addition to Th-232, the most critical segments are Th-228+ (OL = 3.8 kBq kg^{-1}) and Th-232 (OL = 5.1 kBq kg^{-1}). Notably, for Pb-210, Po-210, and K-40, the calculated OLs exceed 40 kBq kg^{-1}. It is important to note that the maximum specific activity of K-40 in elemental potassium is approximately 31 kBq kg^{-1}. Consequently, the disposal of K-40 in landfills, even at concentrations approaching its natural abundance in potassium-bearing materials, is generally considered to pose a negligible radiological concern.

Another important consideration is that all the OLs estimated in this study are higher than the Clearance Levels reported in the Council Directive 2013/59/Euratom (European Commission 2014).

The admissible mass at landfill for each segment of decay chain is given in Table 5 for exemplary purposes for different levels of initial activity concentration – C_{res}(kBq kg^{-1}) – that can be present in residues to be disposed of. These values, however, can be rescaled (see section 2.1.6) for any other amount of initial activity concentration.

Table 5. OLs and percentage of exploitable landfill annual capacity for different values of initial activity concentration present in residue Cres (kBq kg^{-1}) delivered at a landfill complying the dose criterion of 1 mSv y^{-1}

Segment of decay chain	OL (kBq kg^{-1})	% of annual admissible mass based on C_{res} (kBq kg^{-1})				
		0.3	1	5	10	20
U-238sec	2.5	100	100	50	25	13
Unat	18	100	100	100	100	90
Th-230	32	100	100	100	100	100
Ra-226+	3.7	100	100	74	37	19
Pb-210+	41	100	100	100	100	100
Po-210	151	100	100	100	100	100
U-235sec	3.0	100	100	60	30	15
U-235+	24	100	100	100	100	100
Pa-231	10	100	100	100	100	50
Ac-227+	5.3	100	100	100	53	27
Th-232sec	1.9	100	100	38	19	10
Th-232	5.1	100	100	100	51	26
Ra-228+	6.1	100	100	100	61	31
Th-228+	3.8	100	100	76	38	19
K-40	44	100	100	100	100	100

Note: OL values have been rounded to one decimal place if they are less than 10 kBq/kg. Otherwise, the values have been rounded to the nearest unit.

As illustrated in Table 5, for Pb-210+, Po-210, K-40, and Th-230, even at the maximum activity concentration level that has been considered (i.e. 20 kBq kg^{-1}), the full capacity of the landfill can be utilised. Conversely, in relation to the remaining segments, the admissible mass shall be set at 100% provided that the waste material to be disposed of has an activity concentration that is less than or equal to the respective OL. In all other instances, the admissible mass shall be reduced in accordance with the parameters delineated in Eq. 8.

In the event of multiple segments being present in the waste, Eq. 9 would need to be applied in order to evaluate the annual admissible mass. As demonstrated in Figure 5 for Th-232sec, the scaling process is dependent on the initial activity concentration in NORM.

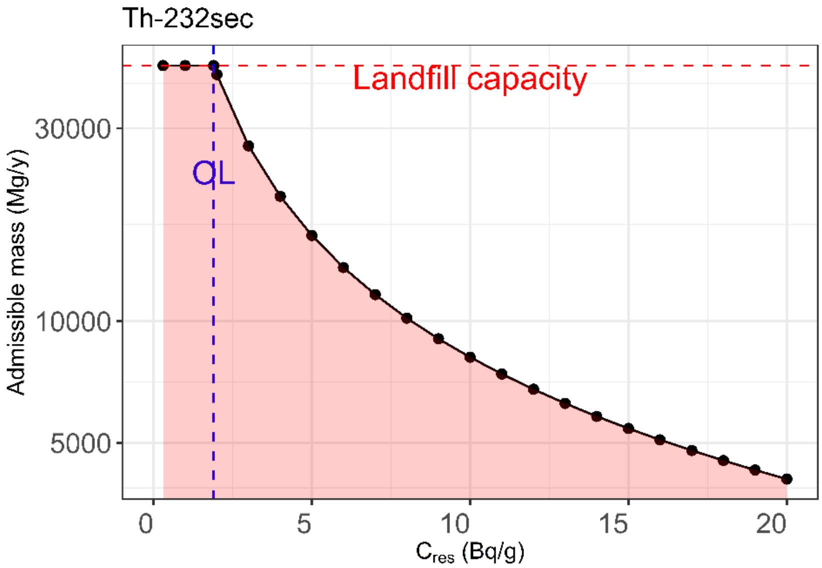


Figure 5. Amount of NORM residues containing Th-232sec that can be disposed of at the landfill. The capacity of the landfill can be fully exploited if $C_{res} \leq$ OL. If $C_{res} >$ OL, the annual admissible mass reduces according to eq. 8.

It is crucial to emphasize that values reported in the Table 5 are provided for illustrative purposes only. This caveat is particularly relevant considering that in numerous European countries, NORM residues exhibiting activity concentrations significantly below 20 kBq kg^{-1} are often precluded from disposal in conventional landfills and are instead classified as radioactive waste (Mrdakovic Popic *et al.* 2023). Furthermore, it should be noted that the Operational Limits (OLs) estimated in this investigation were derived from scenarios assuming exposure to substantial quantities of homogeneously distributed material. Consequently, the annual admissible masses detailed in Table 5 are applicable to large waste volumes (e.g., thousands of megagrams (Mg) for the specific landfill model considered in this study).

2.1.8 Discussion

As shown in the preceding sections, the worker scenario resulted to be the limiting scenario. In accordance with its definition, the annual dose attributable to radionuclide intake pathways within this scenario is independent of the overall dimensions of the landfill. It can be demonstrated that this also pertains to the case of external radiation. In fact, sensitivity analyses demonstrate that modifying the thickness of the landfill model from its nominal value of 10 metres down to 0.5 metres results in negligible alterations to the external dose estimates generated using RESRAD-ONSITE. This finding indicates that the radioactive material situated within the uppermost tens of centimetres of the landfill's surface layer is the primary contributor to the external exposure experienced by the worker. Furthermore, comparable outcomes are observed with variations in the landfill's surface area. Alterations to the landfill area over a range of 10^4 m^2 to 10^6 m^2 result in annual dose variations of less than 4%.

Therefore, it can be concluded with a high degree of confidence that the results presented in this study demonstrate a significant degree of independence from the specific physical dimensions (i.e. thickness and area) of the landfill.

The present study was compared with the UK-based study by Anderson and Mobbs (2010), which was published in 2010 and is widely considered to be significant in this field. Both studies established common ground in several areas. They identified the worker scenario as the limiting one for the majority of decay segments considered. Furthermore, both studies applied the same dose criterion of 1 mSv per year for these workers and found that their calculated Operational Levels (OLs) – or permissible activity concentrations – were generally higher than the standard EU Clearance and Exemption Levels for NORM (European Commission 2014). However, a key divergence emerged in the quantitative results, particularly concerning the OLs. The recent study's estimated OLs were approximately twice as high as those reported by Anderson and Mobbs (2010) for the most critical decay segments, such as Th-232 in secular equilibrium (see Table 6). This discrepancy is primarily attributed to the use of more up-to-date external radiation dose coefficients in the newer research, specifically those from Satoh and Petoussi-Henss (2024). These newer coefficients are roughly double the value of those utilized by Anderson and Mobbs (2010) (see Table A6), leading to a lower calculated dose per unit of activity and, consequently, higher permissible OLs. It was also noted that while the recent study encompassed all ICRP age groups, the UK study focused solely on adults.

Table 6. Comparison of the OLs and % of annual admissible mass based on an activity concentration of 5 kBq kg^{-1} for the most critical decay segment estimated in the present study and in the UK study (Anderson and Mobbs, 2010).

Decay segment	Operational Levels (kBq kg^{-1})		% of annual admissible mass based on a concentration of 5 kBq kg^{-1}	
	This study	Anderson and Mobbs (2010)	This study	Anderson and Mobbs (2010)
Th-232sec	1.9	1	38%	20%
U-238sec	2.5	2	50%	40%
Ra-226+	3.7	2	74%	40%
Th-228+	3.8	2	76%	40%
Ra-228+	6.1	3	100%	60%

Regarding methodologies for determining admissible mass, the UK study proposed a conservative graded approach. For NORM waste with an overall activity concentration below 5 kBq kg^{-1} (whatever the mixture of radionuclides), Anderson and Mobbs (2010) suggested that up to 20% of the landfill's annual capacity could be utilized, a figure derived from their OL for Th-232sec (see Table 6). If the OLs from the present study were applied to this scenario, the permissible capacity for similar NORM would be around 40%. For NORM exceeding 5 kBq kg^{-1}, Anderson and Mobbs (2010) recommended a summation rule to ensure compliance. This approach bases its conservatism on the OL of the most restrictive nuclide.

The rationale behind the Anderson and Mobbs (2010) approach, as highlighted in the comparison, aligns with the optimization principle in radiation protection and aims to simplify control procedures for landfill operators, potentially enabling the definition of a single annual mass limit for NORM waste, irrespective of the specific mix of decay chain radionuclides.

2.1.9 Conclusions

This study developed a robust methodology estimating Operational Levels (OLs) for NORM residue disposal in conventional landfills fulfilling the requirements of the European Directive 1999/31/EC. These OLs ranged from approximately 2 kBq kg^{-1} for the most limiting segments (e.g., Th-232sec) to significantly higher values for others. Notably, the derived OLs are substantially independent of landfill dimensions, enhancing their generic utility. The worker exposure scenario was frequently found to be dose-limiting.

Practically, these OLs facilitate the estimation of annual admissible NORM mass and support a graded approach for managing wastes with multiple NORM segments. The findings align broadly with existing research, with observed differences often attributable to the application of updated ICRP dose coefficients.

2.2 Development of a methodology for the dose-assessment for the use of NORM sludge in agriculture

2.2.1 Introduction

The commitment of European Union to a Circular Economy promotes the reuse of industrial residues, including those containing NORM. A key application involves the agricultural use of sludge, particularly from groundwater filtration facilities, as fertilizer. However, existing regulatory frameworks, such as Council Directive 86/278/EEC (Council of the European Union 1986) on sewage sludge in agriculture, and European guidelines *Radiation Protection 122 Part II* (European Commission 2002), lack specific provisions or methodologies for assessing the radiological impact of NORM-containing sludge used in this manner. This is a notable omission, given that groundwater filtration is recognized under Council Directive 2013/59/Euratom (European Commission 2014) as an industrial sector where NORM residues can be of concern.

This study details the development of a methodology to evaluate the annual effective dose to resident agricultural populations (farmers and their families) arising from the long-term application of NORM-containing sludge as fertilizer. As for the previous study, the primary objective is to derive screening values, named Operational Levels (OLs), that represent the activity concentrations (in kBq kg^{-1}) of natural radionuclides in sludge that align with established dose criteria.

2.2.2 Exposure scenarios

The study focused on a "resident farmer" exposure scenario to assess the radiological impact of utilizing NORM-containing sludge as agricultural fertilizer. This scenario was designed to be representative of an individual (adult agricultural worker) and their family members (members of the public) across various ICRP age groups: infant, 1 year, 5 years, 10 years, 15 years, and adult. These individuals were assumed to reside on or near arable land where the sludge is applied and to consume locally produced food.

Two principal temporal variations of this scenario were investigated:

- Scenario 1 (Sc1): **Occasional Application**: This scenario modelled a single, isolated application of NORM-containing sludge as fertilizer to the agricultural land within a one-year period. Dose assessments were then projected over a 100-year timeframe following this single application.

- Scenario 2 (Sc2): **Continuous Annual Application**: This scenario considered the repeated, annual application of sludge fertilizer to the same tract of agricultural land over an extended duration, specifically 50 years, consistent with practices evaluated in similar European and US studies (EPA (USA) 2004). The dose assessment for this scenario also extended over a 100-year timeframe, accounting for the accumulation of radionuclides in the soil. Notably, Sc2 was also applicable to individuals inhabiting dwellings constructed on land previously subjected to such long-term agricultural sludge application, assuming comparable dietary habits and exposure conditions.

2.2.3 Exposure pathways and dose assessment

For both Sc1 and Sc2, the primary exposure pathways evaluated encompassed external irradiation from radionuclides in the soil, inhalation of resuspended contaminated dust (excluding radon and its progeny, which were assessed separately), direct ingestion of soil, and ingestion of contaminated foodstuffs (plants, meat, and milk) grown or raised on the treated land. The contamination of these foodstuffs was modelled via water-independent pathways, namely direct root uptake by plants and foliar deposition of dust. Water-dependent pathways, such as the contamination of drinking water, irrigation water, or aquatic foods through leaching of radionuclides, were excluded from the primary dose calculation. This exclusion was based on the assessment that the 100-year modelling period was considered insufficient for these pathways to contribute significantly to the overall dose under the defined conditions. The contribution of indoor radon concentrations resulting from radium present in the applied sludge was also specifically evaluated.

2.2.3.1 Specific activity in soil following sludge application

RESRAD-ONSITE accepts the activity concentration in soil A_{soil} (in kBq kg^{-1}) as an input parameter. Therefore, this quantity is derived from the activity in dry sludge (A_{sludge}) using the following formula:

$$A_{soil}\ (\mathrm{kBq\ kg^{-1}}) = \frac{A_{sludge} \cdot S}{d \cdot r} \qquad \text{(Eq. 10)}$$

where:

A_{sludge} is the activity concentration in dry sludge (in kBq kg^{-1})

S is the annual application rate of dry sludge matter per m^2 (in kg m^{-2});

d is the tillage depth or soil mixing thickness (in meters);

r is the soil density (in kg m^{-3}).

A_{sludge} was set equal to 1 kBq kg^{-1} to obtain annual doses per unit of sludge activity concentration.

In Table 7, the values used for the above-mentioned parameters are reported.

Table 7. Parameter values used to calculate soil activity concentration

Parameter name	**Unit**	**Value used in the work**
Annual application rate of dry sludge (*S*)	kg m^{-2} y^{-1}	0.5
A_{sludge}	kBq kg^{-1}	1
Tillage depth (*d*) = thickness of contaminated zone	m	0.3
Soil density (*r*)	kg m^{-3}	1300

The determination of the annual application rate of dry sludge (S) was guided by existing regulatory frameworks. Specifically, the Italian legislative framework, which transposes Council Directive 86/278/EEC concerning the agricultural use of sludge, stipulates a maximum cumulative application of 15 tonnes per hectare (equivalent to 1.5 kg m^{-2}) over a three-year period. This implies an average annual application rate of 5 tonnes per hectare per year, or 0.5 kg m^{-2} year^{-1}, which was the value adopted for S in this study.

The relationship between sludge application and soil activity concentration (Eq. 10) indicates a direct proportionality between the application rate (S) and the resultant soil activity concentration (A_{soil}), assuming constant tillage depth (d) and soil density (ρ). It is worth to note that Council Directive 86/278/EEC itself does not impose restrictions on the total duration (number of years) over which sludge may be applied as fertilizer, a factor considered in the scenario developed in this study.

The selected values for tillage depth (0.3 m) and soil density (1300 kg m^{-3}), as presented in Table 7, are consistent with parameters employed in comparable radiological assessment studies (EPA (USA) 2004; Ugolini *et al.* 2020). By applying these parameters and an assumed unit activity concentration in the dry sludge (A_{sludge}) of 1 kBq kg^{-1}, the derived activity concentration in soil (A_{soil}) is 0.00128 kBq kg^{-1}. This calculated A_{soil} value subsequently served as the primary input for the radiological dose assessments performed using the RESRAD-ONSITE software.

2.2.3.2 Application of sludge for over more than one year

For Scenario 1 (Sc1), which modelled a single, occasional application of fertilizer within one year, the initial soil activity concentration (source term) was determined as reported in the previous section. Subsequent RESRAD calculation estimated annual effective doses (in mSv y^{-1}) for all relevant age categories over a 100-year period. This timeframe was selected to ensure a conservative dose assessment while reasonably limiting uncertainties inherent in long-term predictions of environmental parameters (e.g., meteorological conditions, soil characteristics, human habits) that may exhibit significant temporal variability.

As previously reported in Section 2.2.2, Scenario 2 (Sc2) addressed the continuous annual application of sludge, resulting in a time-dependent source term due to radionuclide accumulation. An iterative methodology, consistent with previous approaches (EPA (USA) 2004; Ugolini *et al.* 2020), was employed. While RESRAD calculates the dose consequence of a single application event at a specific time, the cumulative annual effective dose at year k, $D_{n\ years}(k)$, resulting from *n* annual sludge applications, was determined by summing the contributions from each individual year's application. This can be expressed as:

$$D_{n\ years}(k) = \sum_{j=0}^{n} D_{1\ year}(k-j) \qquad \text{(Eq. 11)}$$

where $D_{1\ year}(k-j)$ is the annual effective dose attributable to the sludge spreading occurring at year j. This approach effectively superimposes the dose contributions from each successive annual application, accounting for radioactive decay and ingrowth over time.

In order to obtain a conservative estimate, it was assumed k = n, that is the annual effective dose is estimated after the last year of sludge spreading.

In the present work, the Operational Levels (OLs) were calculated assuming an annual sludge application during 50 years, similar to the approach adopted by previous studies (EPA (USA) 2004; Harvey and Simmonds 2002)

2.2.4 Assumption and parameters used in this study

The parameterization of the RESRAD-ONSITE model for scenario and pathway analysis predominantly utilized default software values. However, specific adjustments were implemented for age-dependent parameters (detailed in Table 8) and contaminated zone characteristics (as outlined in Table 7).

Age-class specific data for food consumption rates, inhalation rates, and occupancy factors (time spent indoors and outdoors) were incorporated as presented in Table 8. Inhalation rates were derived from ICRP Publication 72 (ICRP 1995). The adult farmer receptor was assigned a proportionally greater outdoor occupancy time on the arable land compared to other household members, reflecting typical agricultural work patterns.

Table 8. Values of age-dependent parameters used for the RESRAD-ONSITE dose estimates

Parameter	Infant	1 year	5 years	10 years	15 years	Adult	Worker
Ingestion							
Fruit/vegetable/grain consumption (kg y^{1})	72	132	220	250	260	240	240
Leafy vegetables consumption (kg y^{-1})	3	6	7	9	11	13	13
Milk consumption (l y^{-1})	45	160	160	170	170	130	130
Meat and poultry consumption (kg y^{-1})	5	13	50	65	80	90	90
Soil ingestion (g y^{-1})	0	18	18	9	15	15	44
Inhalation							
Inhalation rate (m^3 y^{-1})	1100	1900	3200	5640	7300	8100	9600
Occupancy							
Indoor time fraction (at home)	0.8	0.8	0.8	0.8	0.5	0.5	0.5
Outdoor time fraction (in the contaminated area)	0.2	0.2	0.2	0.2	0.25	0.25	0.5

As for the previous study (see Section 2.1.5.5), food consumption rates for the representative individual were primarily sourced from German national legislation BMUV (2020). This legislation provides tabulated values specifically established for estimating doses to members of the public in the context of notifying or authorizing practices involving radiation exposure.

The dose coefficients (DCs) used for are the same as those used in the previous study (see Section 2.1.5.6). Notably, these DCs were used to update the dose coefficients of RESRAD library.

2.2.5 Methodology for estimating Operational Levels (OLs)

The methodological framework for quantifying radiological impact and deriving Operational Levels started with the selection of an appropriate dosimetric modelling tool, i.e. RESRAD-ONSITE (v. 7.2) software package.

Recognizing that radionuclides within the U-238 and Th-232 decay series are frequently not in secular equilibrium in environmental matrices like water (Dinh Chau *et al.* 2011) and, consequently, in sludge derived from water treatment processes due to differential solubilities, dose estimates were performed for some segments of these decay chains (Table 9). These segments, consistent with those delineated in the European Commission's RP 122 Part II guidance document, were individually modelled to provide a more realistic assessment. The U-235 decay series was excluded from consideration owing to its typically negligible activity concentrations in such materials.

Table 9. Decay chain segments considered for dose assessment

Parent nuclide(s)	Nuclides considered in secular equilibrium
U_{nat}	U-238, U-234, U-235 (4.6%) and their short decay products
Th-230	Th-230
Ra-226+	Ra-226, Rn-222, Po-218, Pb-214, Bi-214, Po-214
Pb-210+	Pb-210, Bi-210
Po-210	Po-210
Th-232	Th-232
Ra-228+	Ra-228, Ac-228
Th-228+	Th-228, Ra-224, Rn-220, Po-216, Pb-212, Bi-212, Po-212, Tl-208
K-40	K-40

For each identified decay chain segment, the calculation involved determining the annual effective dose per unit of activity concentration present in the dry sludge. This normalized dose was expressed in units of (mSv y^{-1}) per (kBq kg^{-1}) of dry sludge.

Subsequently, Operational Levels (OLs) were derived using the same approach described for the previous study, using the Eq. 7. The primary dose criteria considered were 1 mSv y^{-1}, aligning with the dose limit for members of the public in the EU-BSS, and a more conservative 0.3 mSv y^{-1}, reflecting the value used in some Member States (García-Talavera *et al.* 2021).

2.2.6 Results

For Scenario 2 (Sc2), which modelled the annual application of fertilizer over 50 years, the maximum annual effective doses per unit of activity concentration were calculated for each age class and decay chain segment (Table 10). Analysis revealed that the infant age class generally represents the critical receptor group, exhibiting the highest annual effective doses for most decay chain segments. Exceptions were noted for Po-210, where the 1-year-old age group was critical, and Th-232, for which the 15-year-old age group received the highest dose.

Operational Levels (OLs) were subsequently derived for each radionuclide segment. These OLs were calculated utilizing the maximum dose coefficients (reported in bold in Table 10) using two distinct dose criteria: 1 mSv y^{-1} and 0.3 mSv y^{-1}. For Ra-226+, Ra-228+, and Th-232, the derived OLs were consistently of the order of a few kBq kg^{-1} or less. The OL for Th-232 corresponding to the 0.3 mSv y^{-1} dose criterion was found to be lower than the General Clearance Levels (GCLs) estimated by RP 122 Part II (European Commission 2002).

Table 10. Annual effective doses for each age class and relative operational levels (based on a dose criterion of 1 and 0.3 mSv y^{-1}) calculated for spreading of fertilizers over 50 years. In bold the highest annual effective doses and relative OLs.

Segment of decay chains	Annual effective dose per kBq kg^{-1} of dry sludge (mSv y^{-1} per kBq kg^{-1})							Operational Level (kBq kg^{-1})		General Clearance Levels
	Infant	1 Year	5 Years	10 Years	15 Years	Adult	Worker	Dose criterion 1 mSv y^{-1}	Dose criterion 0.3 mSv y^{-1}	Dose criterion **0.3** mSv y^{-1}
U_{nat}	**2.9E-03**	2.8E-03	2.7E-03	2.5E-03	2.1E-03	1.8E-03	1.8E-03	340	102	5
Th-230	**7.0E-03**	4.9E-03	5.2E-03	5.9E-03	6.3E-03	5.4E-03	2.5E-03	144	43	10
Ra-226+	**2.3E-01**	1.6E-01	1.6E-01	1.6E-01	2.0E-01	8.9E-02	9.7E-02	4.3	1.3	0.5
Pb-210+	**4.6E-02**	4.1E-02	3.9E-02	3.3E-02	3.1E-02	1.4E-02	5.0E-03	22	7	5
Po-210	3.9E-04	**4.2E-04**	3.9E-04	2.6E-04	1.9E-04	1.5E-04	2.6E-05	2395	719	5
Th-232	1.3E-01	1.5E-01	3.8E-01	1.8E-01	**5.5E-01**	1.5E-01	2.0E-01	1.8	0.5	5
Ra-228+	**1.7E-01**	8.2E-02	7.8E-02	9.2E-02	1.1E-01	3.0E-02	3.1E-02	5.7	1.7	1
Th-228+	**7.7E-03**	7.0E-03	6.7E-03	6.2E-03	4.7E-03	4.5E-03	6.3E-03	130	39	0.5
K-40	**1.3E-03**	1.2E-03	1.1E-03	1.1E-03	7.9E-04	7.7E-04	1.1E-03	774	232	5

For each segment of decay chains, the maximum annual effective dose per kBq kg^{-1} is reported in **bold**.

Conversely, Operational Level (OL) for the remaining radionuclides, whether assessed individually or as decay segments, are substantially higher. These OLs surpassed the GCLs of RP 122 Part II by one to three orders of magnitude. For illustrative purposes, RP 122 Part II specifies GCLs of 5 kBq kg^{-1} for Po-210 and 0.5 kBq kg^{-1} for Th-228+.

For the critical infant age class, particularly when considering the dose contributions from Th-232 and Ra-226+ (which yielded the highest doses per unit activity), pathway analysis (as shown in Figure 6) indicated that external radiation (most notably for Th-232) and water-independent plant ingestion were the principal contributors to the total effective dose.

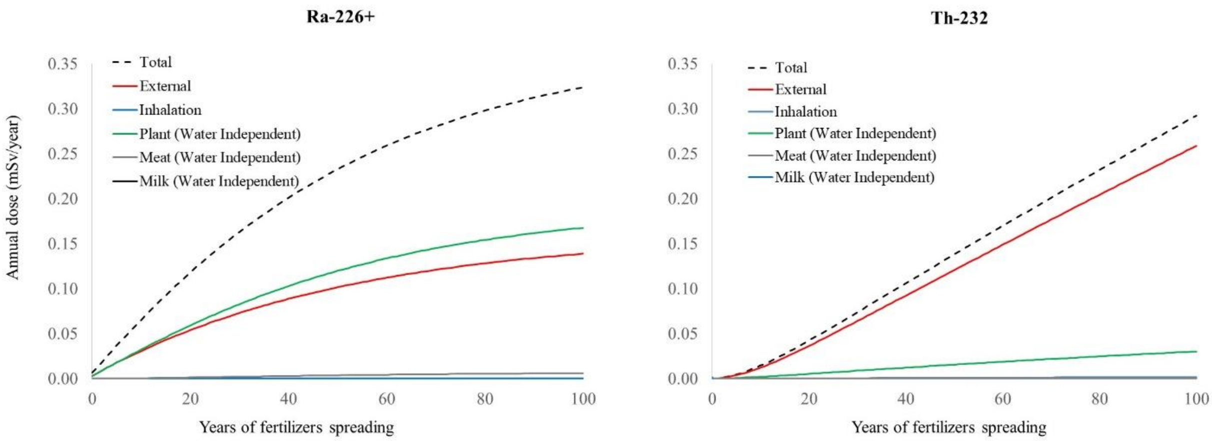


Figure 6. Exposure pathway contributions to the annual effective dose for infant *versus* years of sludge spreading, in case of Ra-226+ and Th-232

Notably, for Th-232, the dose is attributable mainly to the ingrowth of its daughters (Ra-228 and Th-228), contrary to Ra-226+ decay segment, for which the contribution due to the ingrowth of Pb-210 and Po-210 is negligible (see Figure 7). It is interesting that the annual dose determined by Th-232 continuously increases with time. This effect is due to the very high default value of distribution coefficient K_d (60000 cm^3 g^{-1}) adopted by RESRAD-ONSITE. It is worth noting that annual doses even after 100 years of sludge spreading do not exceed 1 mSv per year.

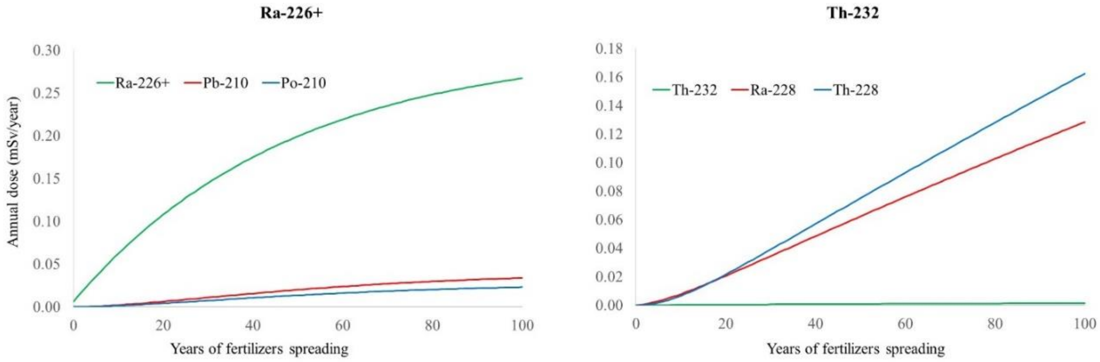


Figure 7. Contributions of the daughter ingrowth for Ra-226+ (left box) and Th-232 (right box) to the total annual effective dose for infant.

Regarding the estimation of **radon activity concentrations**, at 1 kBq kg^{-1} in dry sludge, Ra-226+ (over 50 years) produces ~20 Bq m^{-3} of indoor radon, and Th-230 yields <1 Bq m^{-3}. These levels are substantially lower than 100–300 Bq m^{-3} reference levels recommended by ICRP for dwellings and workplaces. Consequently, the OL values for Ra-226 and Th-230 (Table 10) would also lead to radon concentrations well under 100 Bq $m^{-3.}$

Continuous sludge application over years, versus a single spreading, increases annual effective doses and consequently lowers OLs for the critical age group (Table 11). Specifically, over 1–20 years, OLs for U-238 series radionuclides (except Po-210) decrease 15–20 fold. Po-210 OLs remain high and stable due to its ~180-day half-life. Th-232 OLs show a pronounced decrease (factor of ~200) due to Ra-228 ingrowth, whereas other radionuclides/segments exhibit OL reductions by a factor less than 10.

Table 11. Operational Levels (in kBq kg^{-1}) based on a dose criterion of 0.3 mSv y^{-1} for the critical age classes for different numbers of years of sludge spreading.

Segment of chains	Critical age class	Number of years of sludge spreading						
		1	5	10	20	50	75	100
U_{nat}	Infant	2832	598	319	181	102	87	81
Th-230	Infant	3179	605	286	130	43	26	19
Ra-226+	Infant	48	10	5	3	1.3	1.0	0.9
Pb-210+	Infant	151	29	16	10	7	6	6
Po-210	1 Year	837	719	719	719	719	719	719
Th-232	15 Years	338	16	5	1.7	0.5	0.4	0.3
Ra-228+	Infant	14	4	2	1.8	1.7	1.7	1.7
Th-228+	Infant	128	47	40	39	39	39	39
K-40	Infant	1145	342	259	235	232	232	232

2.2.7 Discussion

The derived operational levels (OLs) for agricultural application of sludge containing NORM were compared with the results of previous modelling studies and empirical data on NORM activity concentrations from existing water treatment plants.

When compared with other modelling studies, particularly a comprehensive US assessment (EPA (USA) 2004) and an earlier UK evaluation (Harvey and Simmonds 2002), notable differences and convergences in OLs were observed (Table 12). The US study generally reported lower OLs (implying higher dose estimates per unit activity) for several radionuclides. This was primarily attributed to its adoption of a significantly longer assessment timeframe (1000 years versus 100 years in the present work) and the inclusion of groundwater contamination pathways (e.g., drinking and irrigation water), which become more influential over extended periods and were not considered in the current study. Despite these methodological variances, OLs for key radionuclides such as Ra-226 were broadly comparable, typically in the order of 1 kBq kg^{-1}. For other radionuclides, discrepancies could often be traced to differing assumptions, such as the inclusion or exclusion of radon contributions in summary OLs (as in the case of Th-230 in the US study) or variations in model parameters like the assumed thickness of the contaminated soil layer, which impacts the dilution of applied sludge (especially for Pb-210 and Po-210 differences with the UK study). For K-40, derived OLs were consistently high, suggesting its radiological impact in this context is negligible. A comparison with general clearance levels (GCLs) from European guidance RP 122 Part II (European Commission 2002) indicated that these GCLs were largely applicable for the sludge-as-fertilizer scenario, with notable exceptions for Th-232 and Pb-210+, where specific OLs are generally more restrictive.

Table 12. Comparison of OLs (kBq kg-1) obtained by the different studies with a dose criterion of 0.3 mSv y^{-1}. A spreading of 50 years of sludge was considered.

Decay chain segments	Harvey and Simmonds (2002)	EPA US (2004)	Present study	RP 122-II General Clearance Levels
U_{nat}	22*	13**	102	5
Th-230		0.4§	43	10
Ra-226+	0.9	0.5	1.3	0.5
Pb-210+	0.7	2.2	7	5
Po-210	6.4	935	760	5
Th-232		0.4	0.4	5
Ra-228+		2.4	1.7	1
Th-228+		23	39	0.5
K-40		115	232	5

*Sum of contributions of U-234 and U-238. ** Sum of contributions of U-234 and U-238 and 4.6% of U-235
§It includes the contribution of radon to the effective dose

In addition, the OLs calculated in this study were compared with a collection of reported NORM activity concentrations measured in sludge samples from operational water treatment plants, mainly drinking water plants.

This empirical comparison revealed that for certain radionuclides, most significantly Ra-226 and Ra-228, the measured average or maximum concentrations in some investigated sludges (Baeza *et al.* 2014; Dilling *et al.* 2012; Kallio *et al.* 2023; Kunte 2012) approached or, in some instances, exceeded the newly derived OLs. This suggests that radium isotopes are potentially limiting factors for the agricultural use of these specific sludges. In contrast, measured Th-232 concentrations were typically well below its corresponding OL. For Pb-210, only isolated cases in the literature reported concentrations exceeding the derived OL (Kunte 2012), and often this sludge was not recommended for agricultural use in its region of origin. More details about this comparison are reported in (Venoso *et al.* 2024a)

Collectively, this comparative analysis highlights that radium isotopes (Ra-226 and Ra-228) are likely the most critical radionuclides for water treatment sludge. It is also important to underline for this study that the derived OLs provide a screening framework, but if measured concentrations in specific sludges approach these values, more detailed, site-specific assessments will be required.

2.2.8 Conclusions

This work developed a methodology for assessing radiological doses from agricultural NORM sludge application, deriving Operational Levels (OLs) for NORM radionuclides. Radium isotopes (Ra-226+, Ra-228+) showed to be the most radiologically significant, with OLs around 1 kBq kg^{-1} for a 0.3 mSv y^{-1} criterion considering sludge spreading on the same land over 50 years. These OLs offer conservative screening tools, aiding decisions that balance radiation protection with circular economy goals.

Future work could extend this methodology to related scenarios, such as wastewater treatment plant sludges receiving NORM-containing backwash water. The results may contribute to the re-assessment of discharge levels, namely to a possible update of the RP 135 guidelines.

3. Feasibility study for use of complex water flow models for NORM situations

When assessing the potential impact of NORM over the long-term on the environment, the groundwater transport of radionuclides needs to be considered. Indeed, contaminants can be suspended and dispersed by surface water and or via transport through non-saturated zone to groundwater. For regulatory activities in the context of NORM very simple water flow and solute transport models are considered and suffice (European Commission 2002; IAEA 2005). However, in situations in which specific sites are considered and measurements are available, it is feasible and may be beneficial to use more accurate and realistic models in order to understand physical, chemical and biological mechanisms driving not only the present but also the future distribution of contamination at the site. Consideration of more complex models may also allow for improved insight of impact of natural heterogeneity on radionuclide distribution in the environment. This work deals with the application of process-based water flow models and solute transport models at NORM-involving sites, in order to better understand the importance of hydrogeochemical and chemical processes for radionuclide transfer in the environment and to consider possible advantages of using such models for understanding the behaviour of radionuclides at specific NORM situations. Within this context, it was proposed to evaluate the potential influence of water-related transport processes (via water flow, solute transport, geochemical speciation) models for NOR at specific sites.

In the frame of task 2.8, the primary focus was to define the models that could be used and to find an interesting site to apply these models. Such research focused primarily on legacy sites with sufficient valuable data. Additionally, the research also focused on sites that may be affected by leaching to groundwater.

As a case study, it was decided to select an area located downstream a former uranium mine: the Rophin site, located in the central part of France. Indeed, the interest of this site is double. Firstly, dose rates 20 to 30 times higher than background are measured along the Gourgeat stream and in a so-called “wetland area” where a source of contamination is present (due to successive discharges from settling ponds during the operational phase of the mine). Secondly, this site is already monitored and used for studies in the frame of RadoNorm Task 2.6 (focussing on the transfer of NOR to plants) and 2.7 (focussing on the geochemistry underlying the transfer of NOR from soil to water and vice versa). It was thus decided to apply process-based water flow-geochemical models to the Rophin site in order to understand and describe:

- the fate of U and Ra in the wetland area;
- the link between radionuclides' concentration in the subsurface whitish layer of the wetland area (which is a source of contamination) and in the stream that crosses the watershed;
- the possible transfer of radionuclides to the groundwater.

However, the more complex the model, the more parameters related to the site are needed. Most of the time parameters are difficult to obtain and/or are scarce. It is therefore necessary to know the capabilities (strengths and weaknesses) of the codes/software available, their implemented equations and corresponding applicability. Furthermore, a well-defined modelling strategy is required to allow relevant conceptualization of the site and the use of the available hydrodynamic and geochemical existing data on site.

3.1 Rophin site description

"Zones-Ateliers" is a French network of scientists and local regulators focussed on long-term Physical and Socio-Ecological Research Observatories, addressing a wide range of environmental problems (Bretagnolle *et al.* 2019). Within "Zones-Ateliers", the "Zone-Atelier Territoires Uranifères" (ZATU) has been created in 2015 and the activities fall within the context of a strong societal concern regarding the characterisation and prediction of the short and long-term risks of the exposure of socio-ecosystems to natural radioactivity reinforced by bio- geo- chemical processes or by human action. It provides as well a place of dialog among stakeholders of uranium mining sites, *i.e.* non-profit organisations, academic laboratories, French national research organisms (BRGM, ASNR, CEA).

Located in the department of Puy-de-Dome of the region Auvergne-Rhone-Alpes, the Rophin site (old uranium mine, one site studied by the ZATU) has been exploited for uranium mining from 1947 to 1956. At the end of the exploitation, mine tailings and waste rocks were stored *in natura.* Rophin is one of the 15 sites of uranium waste storage in France (MIMAUSA database[1]). The site is now an uninhabited place where vegetation grows on mine tailings as well as downstream from the storage area (Figure 8).

[1] https://mimausabdd.irsn.fr/

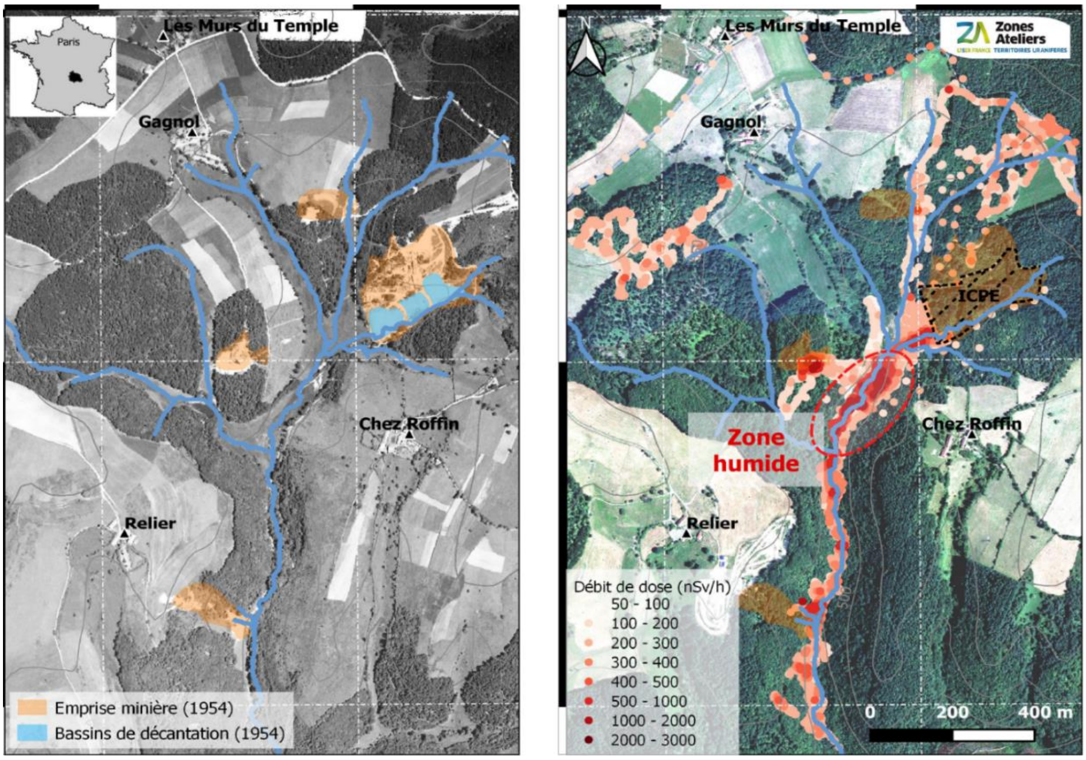


Figure 8. Left: location of the mines (yellow, 1954) and the settling pond (blue) at the Rophin mine (1954). Right: radiological mapping with focus on the wetland (red circle). *Kindly provided by ZATU*.

A description of the watershed, which includes the wetland and the Gourgeat stream area, is presented in Figure 8. Gamma-ray surveys have shown “high” radiation levels alongside a creek (the Gourgeat stream) downstream of the storage site, especially in a wetland (WL) area at around 200 meters from the storage site. Drill cores in this area show uranium concentrations up to 2000 ppm in a whitish, clayey layer near the surface with a thickness varying from a few centimetres to a few tens of centimetres. The origin of the white layer is connected to the active period of the site. It was formed by the sedimentation of fine clay particles, rich in radionuclides, transported in suspension in the water following episodes of overflow from the settling ponds associated with former mining activities (Martin *et al.* 2020). Since the closure of the mine (more than 60 years ago), the progressive vegetation of the site and the development of the wetland have led to changes in the speciation and lability of the radionuclides in this horizon (Martin *et al.* 2021).

The watershed has been equipped by BRGM since 2019 with piezometers and surface sensors to monitor the water (electrical conductivity, temperature and water flow). The site is also equipped with a weather station. The analysis of the first results (from measuring campaigns in 2021 and 2022) confirms the presence of a groundwater table in the wetland, that is rather close to the surface and varies depending on meteorological conditions. The consortium has also more than 70 photos of soil cores that have been taken from the wetland (organic-rich surface layer, whitish layer, and paleosol layer).

3.2 Available data

3.2.1 Soil / sediment layers

3.2.1.1 Core drilling

The various layers of the wetland have been analysed on the basis of 40 core drill measurements distributed all over the wetland area and near the water streams as indicated in Figure 9.

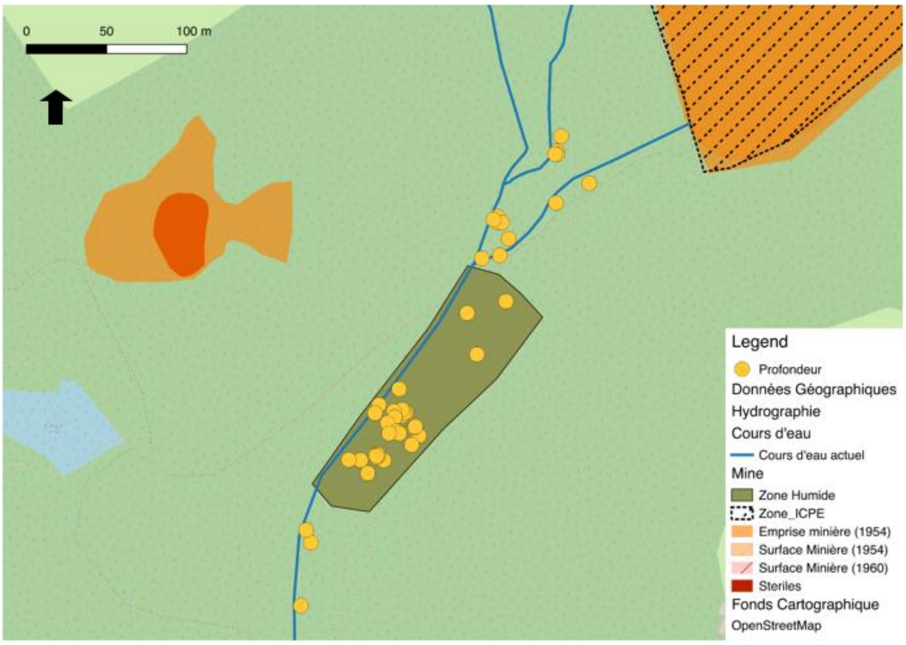


Figure 9. Drilling locations used to assess characteristics of the soil layers in the Rophin watershed (Courtesy of SUBATECH).

The organic-rich layer and whitish layer have an average thickness of 19 cm and 9 cm, respectively as it can be seen in Figure 10. The paleosol layer consists of the entire layer below the whitish zone and could be more than 100 cm thick. This quaternary lithosphere rests on a granitic basement on the entire area (most of the core drill did not reach the granitic layer).

The GPS data provided show that several core drill locations have identical coordinates despite layer depths differing by several tens of centimetres. As a result, we cannot build a precise picture of the spatial heterogeneity of the different layer thickness.

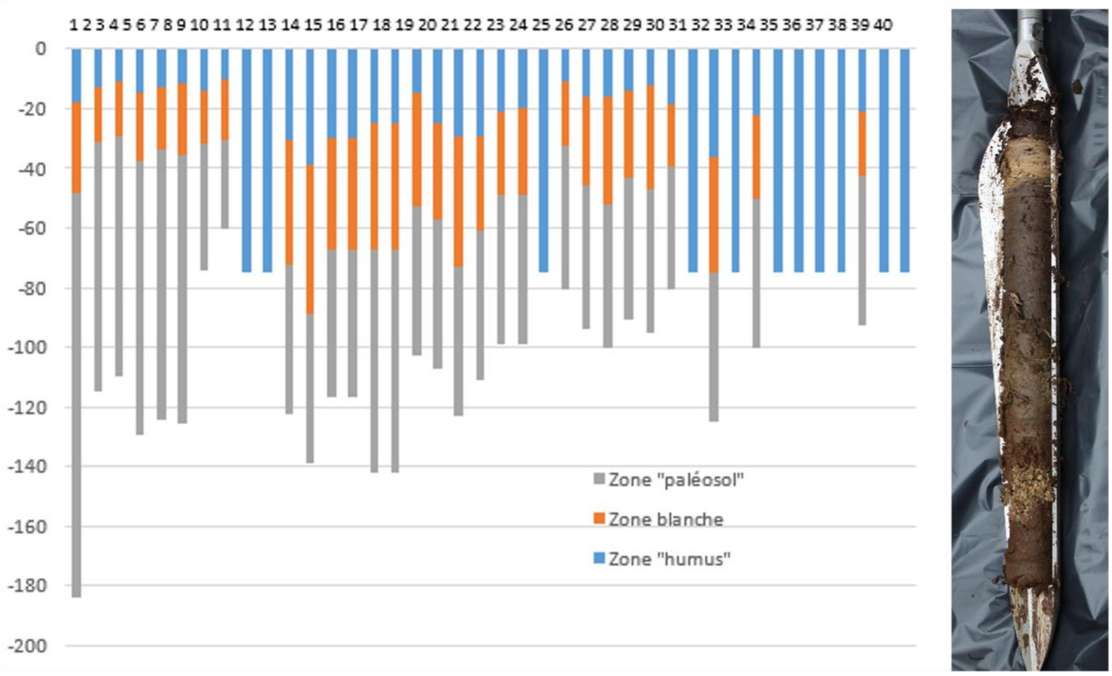


Figure 10. On the left: thickness of the different soil layers (organic-rich layer in blue, whitish layer in orange and paleosol layer in grey) in wetland obtained by core drilling over the wetland area. On the right: Photograph of a 50 cm long core sample. Courtesy of SUBATECH.

3.2.1.2 Geophysics campaign

A geophysical study using resistivity tomography and induced polarization was carried out by NAGA Geophysics (2023) on the Rophin wetland. A set of six transects were placed in the study area as shown in Figure 11.

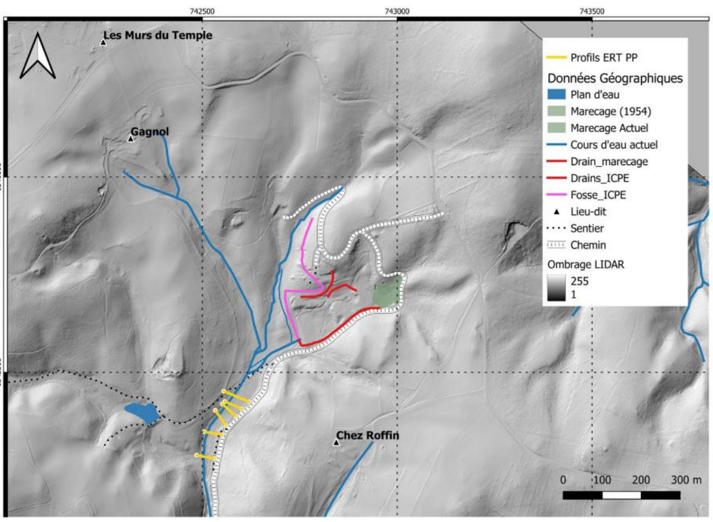


Figure 11. Profiles positions (yellow lines) downstream of the old mine (red and pink lines). Source: NAGA Geophysics (2023)

The aim of these six geophysical profiles was to use spatial interpolation to approximate a range of hydrodynamic data, as well as the depths of different layers.

NAGA Geophysics also provided an estimate of paleosol, and bedrock depths calculated by a statistical method called clustering (Figure 12). Clustering is the statistical discrimination of data according to results and it is used to associate the depths of defined layers with the raw profile data. These depths are then interpolated spatially between the profiles.

3.2.1.3 Combining methods

Comparing the results of the geophysical survey with the boreholes drilled, we note that the geophysical method used does not allow us to differentiate the humus layer from the whitish layer (Figure 12). In conclusion, it will not be possible to produce maps showing the depth of different layers according to geographical coordinates. Consequently, for this study, average thicknesses from core drill will be used.

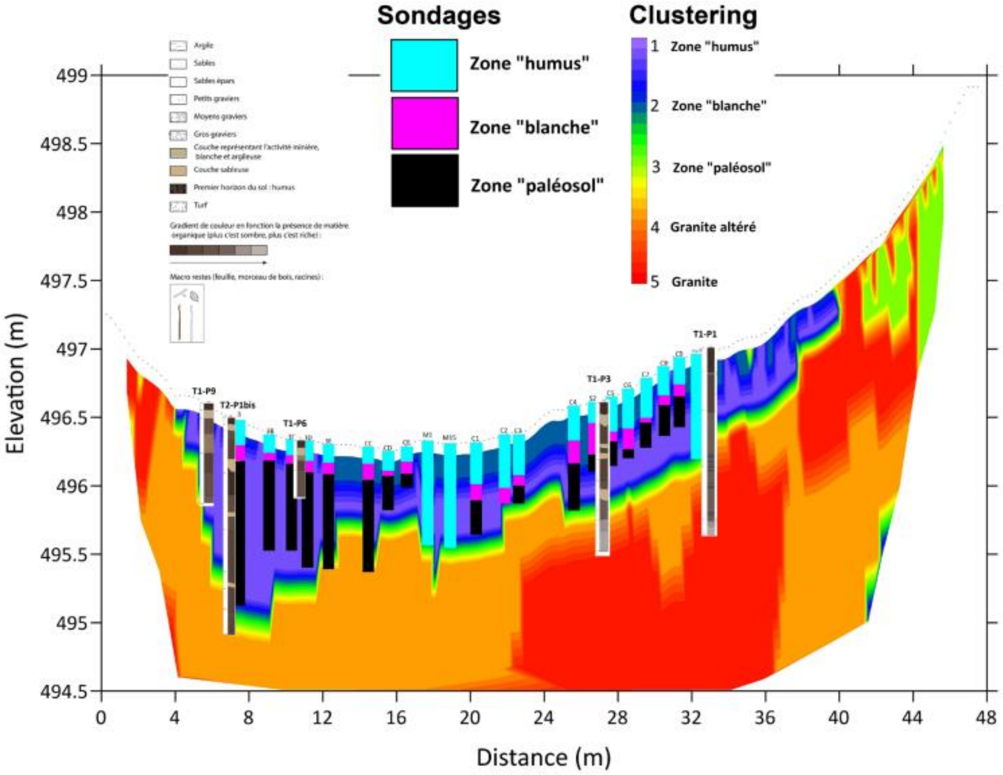


Figure 12. Clustering results for the ROP1 profile and previously core drill (approximate positions). Source: NAGA Geophysics (2023) modified

3.2.2 Hydrodynamic and hydraulic parameters

3.2.2.1 Hydraulic conductivity and porosity

The permeability of the medium, and in particular of the three layers, is essential and difficult to obtain. The geophysical study carried out by NAGA Geophysics (NAGA 2023) presents data derived from post-processing of geophysical measurements, including intrinsic permeability as a function of altitude z NGF (where NGF refers to the General Levelling of France). As it was not possible to identify the three layers in this study, the hydraulic conductivities measured were assigned to the different layers according to depth. The values obtained are respectively 4.10^{-4}, 3.10^{-4} and 1.10^{-4} m s^{-1}.

As the soil is saturated, the percentage of water determined corresponds to the total porosity (Table 13).

Table 13. Water saturation rate, organic matter content and density of the three layers

	"Humus" zone			"Whitish / Impacted" Zone			"Paléosol" zone		
	Number of sample	*Mean value*	*Median value*	*Number of sample*	*Mean value*	*Median value*	*Number of sample*	*Mean value*	*Median value*
Water (%) (saturated sample)	*10*	73	75	*10*	50	53	*10*	74	85
Organic matter (%) (dry sample)	*3*	33	31	*3*	7,1	3,8	*3*	47	64
Density (g/cm3)	*1*	0,5		*1*	1,0		*1*	0,7	

3.2.2.2 Hydraulic gradient

Eight piezometers were installed in the wetland area by BRGM (Figure 13). The first four piezometers (Pz1, Pz2, Pz3 and Pz4) present data from 2020 onwards. These piezometers are not placed along the major direction of groundwater flow, but rather perpendicular to it. The other four piezometers (Pz6, Pz7, Pz8 and Pz9) were installed in October 2024 in the direction of flow and at both ends of the wetland. We do not yet have data on these new piezometers. They will be essential for the continuation of the study presented here.

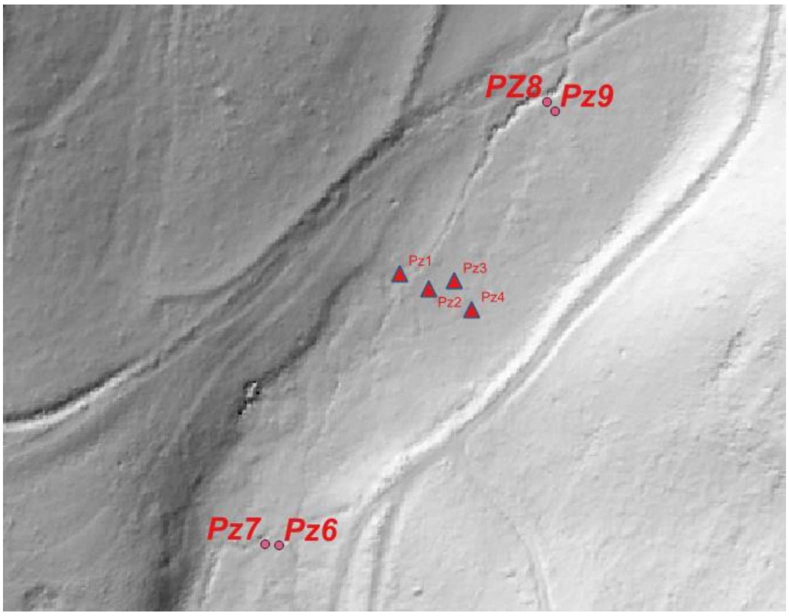


Figure 13. Location of the 8 on-site piezometers

The exact location of the four first piezometers was provided by the “Ordre des Géomètres”, as shown in Figure 14.

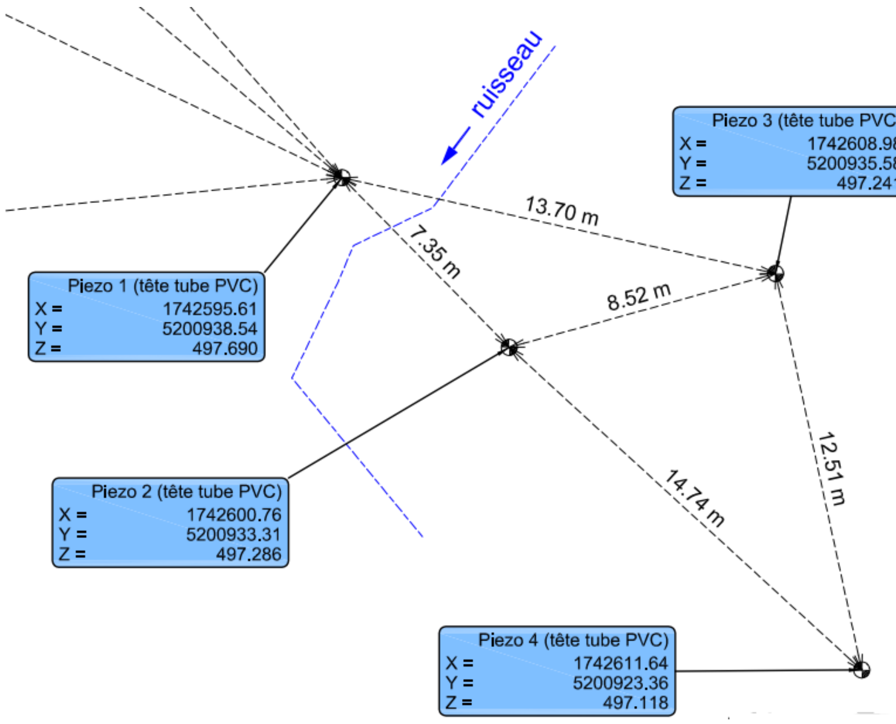


Figure 14. Piezometers localisation on the Rophin site, extract from the « Plan de repérage de la commune de Lachaux CNRS », measurements taken on 31/08/2023, Ordre des géomètres-experts.

N.B. In this document (Plan de repérage lieu - dit "chez Rophin", relevés alimétriques de 4 piézomètres) the mode of calculation and transformation of longitudinal and latitudinal coordinates given is Lambert 93-CC46, but the coordinates correspond to another system: RGF93-CC46, of which Lambert 93 is the projected version. Adding these data to a geographic information system therefore requires first importing these coordinates in RGF93-CC46 and then projecting them in Lambert 93.

One of these piezometers, Pz1 (see Figure 14), is located on the other side of the wetland and cannot be considered as in continuation of the three others. Only piezometers no. 2 and no. 3 are relatively parallel to the watercourse (and present data) and can therefore provide an approximation of the hydraulic gradient in the study area (pending data from the four recently installed piezometers). The difference in piezometry between these piezometers is around 0.193 m, corresponding to a hydraulic gradient of 0.023 m m^{-1}, with the distance between the two piezometers being 14.74 m.

Table 14. Manual piezometry measurements carried out on Pz1, Pz2, Pz3 and Pz4 (*Courtesy of BRGM*).

	Pz1 (m NGF)	**Pz2 (m NGF)**	**Pz3 (m NGF)**	**Pz4 (m NGF)**
ground-level lidar measurement	497.02	496.69	496.58	496.41
22/12/2020	496.79	496.44	496.7	496.49
16/04/2021	496.65	496.56	496.6	496.38
06/08/2021	496.28	496.46	496.64	496.22
20/12/2021	496.74	496.63	frozen	frozen
20/10/2022	496.25	496.23	495.84	495.22

3.2.2.3 Stream flow

From the measuring campaigns carried out by BRGM in 2020-2022, there is indication that water circulation at the site occurs between the Gourgeat stream and the dam and between the Gourgeat stream and the wetland.

Following the summer drought in 2022, no water flow was observed in October 2022. Hence the piezometric levels in the area in this time frame were low (low water flux). On the opposite, in summer 2021, due to heavy rains, piezometric levels were almost at the soil surface (indicatively at around ~20 cm depth) leading to high water flux indicating that the watershed is almost always in saturated conditions. Except from times when there is drought. An important conclusion from first measurements is that the water flow upstream is always higher than at downstream and 'missing' water is most probably flowing into the groundwater in the watershed. This can be seen in Table 15. Location and values of measurements for October 2022 is given in Figure 15.

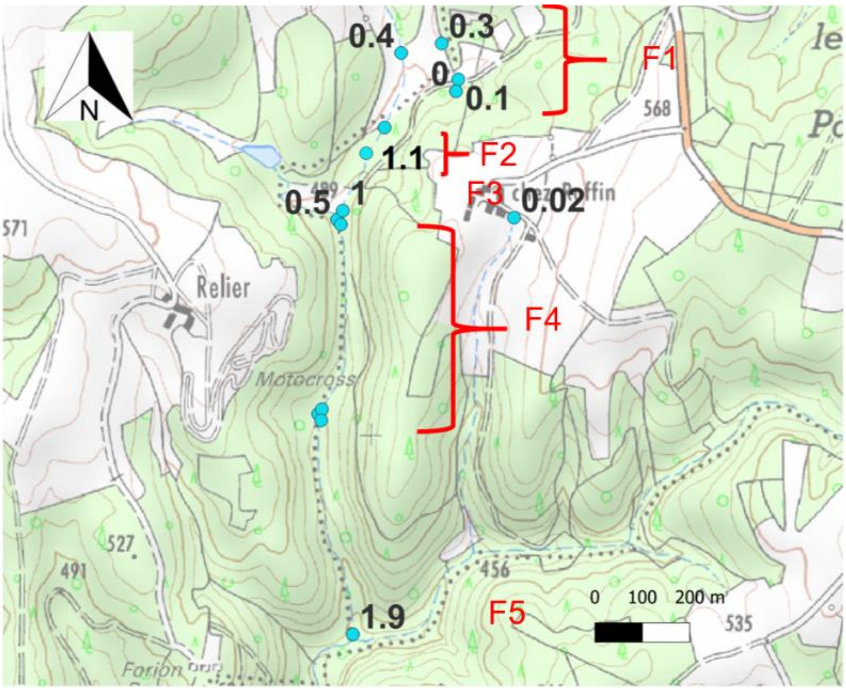


Figure 15. Locations of water flow measurements upstream (1), in the wetland (2), after the wetland (3) and from tributaries downstream (4). The values in black are the measurements obtained in October 2022. (courtesy of BRGM).

Table 15. Measurement of water flow F1, F2, F3, F4, F5 (L s^{-1}) at various location in the Rophin watershed (*Courtesy of BRGM*).

Flow measurements (L s^{-1})					
	Dec. 2020	Apr. 2021	Aug. 2021	Dec 2021	Oct 2022
F_1 upstream	8.4	3.5	9.7	8.4	0.8
F_2 wetland		2.3	7.2	7.5	1.1
F_3 after wet			7		1
F_4 tributary	5.8	3.7	4.9	6.3	0.5
$F_{(1+4)}$	14.2	9.5	28.8	22.2	3.4
F_5 - downstream	15	7.6	16.3	17.9	1.9

The groundwater has a temperature of 8-12 ℃ and electrical conductivity in surface water has values ranging between 69-130 µS.cm^{-1} upstream and 53-123 µS.cm^{-1} downstream. Higher conductivity values upstream indicate that different types of water mix most probably upstream.

The main conclusion, on the basis of first hydrological analysis, is that when there is low flow, the wetland and the stream are disconnected (i.e. in the months August, September, October 2022 / information from BRGM) whereas when there is high flow, there is connection between surface water and groundwater (i.e. in the months February, March 2022 / information from BRGM).

3.2.2.4 Rainfall

Rainfall data for the area of interest can be estimated from data available via Météo France from 1958 to 2020. Three weather stations are located close to the Rophin site, in Clermont-Ferrand, Roanne and Vichy. In addition, a meteorological station has been installed on the Rophin site in 2021. The data have not been processed as the recording period is still too short. As part of this study, CIEMAT processed rainfall data from the Vichy and Clermont-Ferrand stations for the period between 1994 and 2020. A seasonal pattern can be seen in Figure 16 a winter with little variation in precipitation and rather dry, and a summer marked by more intense and successive rainy episodes. Evapotranspiration has also been taken into account.

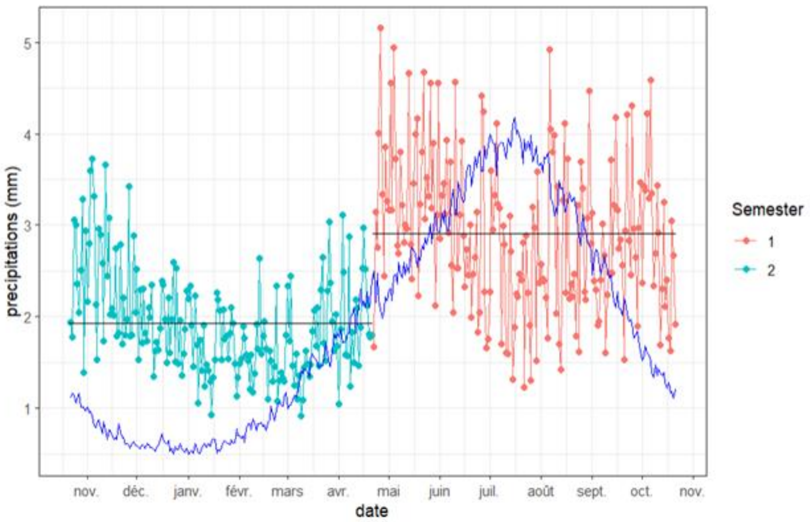


Figure 16. Precipitation averaged over one year using Vichy and Clermont piezometers between 1994 and 2020. Blue curve: evapotranspiration.

Table 16. Temperature, precipitation and evapotranspiration averaged per month using Vichy and Clermont piezometers between 1994 and 2020

Month	T (°C)	Precipitations (mm)	Precipitations min (mm)	Precipitations max (mm)	Etp (mm)	Etp min (mm)	Etp max (mm)
January	2,1	52,2	14,0	131,8	18,5	10,0	27,8
February	3,0	44,5	2,9	118,5	24,4	12,4	44,4
March	5,7	55,0	6,6	131,8	44,1	27,8	63,6
April	8,5	71,9	6,8	178,9	61,9	37,7	93,2
May	12,5	107,6	29,9	210,4	82,2	51,0	115,3
June	16,1	86,5	3,9	191,9	102,3	67,5	156,0
July	18,4	77,8	20,5	250,0	120,1	81,1	183,3
August	17,8	87,9	28,8	180,6	100,3	71,8	137,9
September	14,4	83,2	5,6	242,8	66,9	46,9	100,7
October	10,5	83,1	8,6	180,2	39,8	21,7	61,1
November	5,6	73,5	7,7	155,9	22,3	11,5	36,3
December	2,8	59,7	7,4	143,3	18,0	9,7	33,7

3.2.3 U and Ra transfer parameters

As part of task 2.7 of the RadoNorm project, a set of geochemical data was determined in the wetland: chemical composition of water and soil, sediment mineralogy, partition coefficient (K_d and smart K_d), as well as information on the interaction of colloids with U. Table 17 presents data on the concentration and mobility of U and Ra for the different layers; the concentration is the initial condition for carrying out the transport calculations.

Table 17. Mineralogical data and radionuclide concentrations

		"Humus" zone			"Whitish / Impacted" Zone			"Paléosol" zone		
		Number of sample	*Mean value*	*Median value*	*Number of sample*	*Mean value*	*Median value*	*Number of sample*	*Mean value*	*Median value*
	Quartz (%)	*1*	38	38	*1*	51	51	*1*	17	17
	Kaolinite (%)	*1*	10	10	*1*	8,8	8,8	*1*	5,4	5,4
	Orthoclase (%)	*1*	6,0	6,0	*1*	7,1	7,1	*1*	3,5	3,5
	Albite (%)	*1*	6,1	6,1	*1*	7,2	7,2	*1*	3,6	3,6
	Muscovite/Illite (%)	*1*	9,4	9,4	*1*	18	18	*1*	6,4	6,4
	Thickness (cm)	*28*	21	19	*28*	10	9	*28*	30	21
Uranium	[U] (mg/kg dry matter)	*13*	1206	533	*13*	1650	936	*13*	737	363
	[U] Bq/g (dry)		8,24			42			19	
	% labile	*6*	0,9	1,0	*6*	3,1	2,7	*1*	1,4	1,4
	Kd (L/kg)	*6*	62	29	*6*	35	30	*1*	86	86
Radium	[Ra] (ng/kg dry matter)	*1*	518	518	*1*	2099	2099	*1*	183	183
	[Ra] Bq/g (dry)		5,12			77			7	
	% labile	*1*	3,2	3,2	*1*	4,7	4,7	*1*	0,7	0,7
	Kd (L/kg)	*1*	413	413	*1*	890	890	*1*	33	33

The humus layer and the whitish layer have a mineralogical composition close to granitic composition that could be found in the host rock, in particular the rocks from the old mine. As expected, the humus layer has a much higher organic matter content. The paleosol beneath the white layer is described as the initial soil prior to anthropic activity (Martin *et al.* 2021).

The white layer appears to be the main source of U in the area. However, the organic matter present, particularly in humus, favours the trapping of U, especially that coming from water enriched by contact with tailings that have run-off from the old mine (Martin *et al.* 2021).

Mallet *et al.* (2024) carried out Ra measurements on the top 2 cm of the wetland. The result was 1.8 Bq g^{-1} on saturated sediments, compared with 5.12 Bq g^{-1} for measurements carried out on the entire humus layer by Task 2.7. This difference highlights the heterogeneity of the humus layer. It is even more marked for U, with 1.7 Bq/g in the first 2 centimetres compared with 8.24 in the Task 2.7

measurements (Table 17). This variation in U content with depth is measured in Martin *et al.* (2020). The latter study highlights U concentration peaks at the white layer-humus and white layer-paleosol interfaces.

3.3 Modelling features

3.3.1 Modelling strategy

The modelling strategy will depend on the type of available data and will be strictly linked to conceptual model. It will provide information on how to approach the available data and models so to obtain the results we aim at.

In general, a modelling strategy consists of the following steps (Lege *et al.* 2013):

1. Problem formulation
    - Define receptor point at which evaluation of model results versus experimental data should occur
    - Define requirements that model results need to have (e.g. maximum concentration, spatial and temporal variability)
2. Creation of a conceptual hydrogeological model
    - Description of the system (data acquisition)
    - Abstraction and schematization of the system (conceptual model)
3. Use of a modelling tool, i.e. numerical model/ software or analytical solution for the water flow/ radionuclide transport
    - Choice of the tool
    - Apply tool to specific situation
    - Obtain results (model output at the specific receptor points)
4. Validation and analysis of results
    - Presentation of the results

3.3.2 Conceptual model

A conceptual model aims at providing an abstract physical representation of the site to the extent to which it is reasonable and useful for the purpose of the study. The complexity of such a conceptual model depends on

- data availability
- the complexity of the hydrogeologic system at the site

Conceptual model building begins with the interpretation of hydrogeological primary data, whose schematisation leads to the hydrogeological model, and ends after successful selection of a suitable computational program with the actual simulation model (mathematical-numerical model) for the flow and transport processes to be investigated.

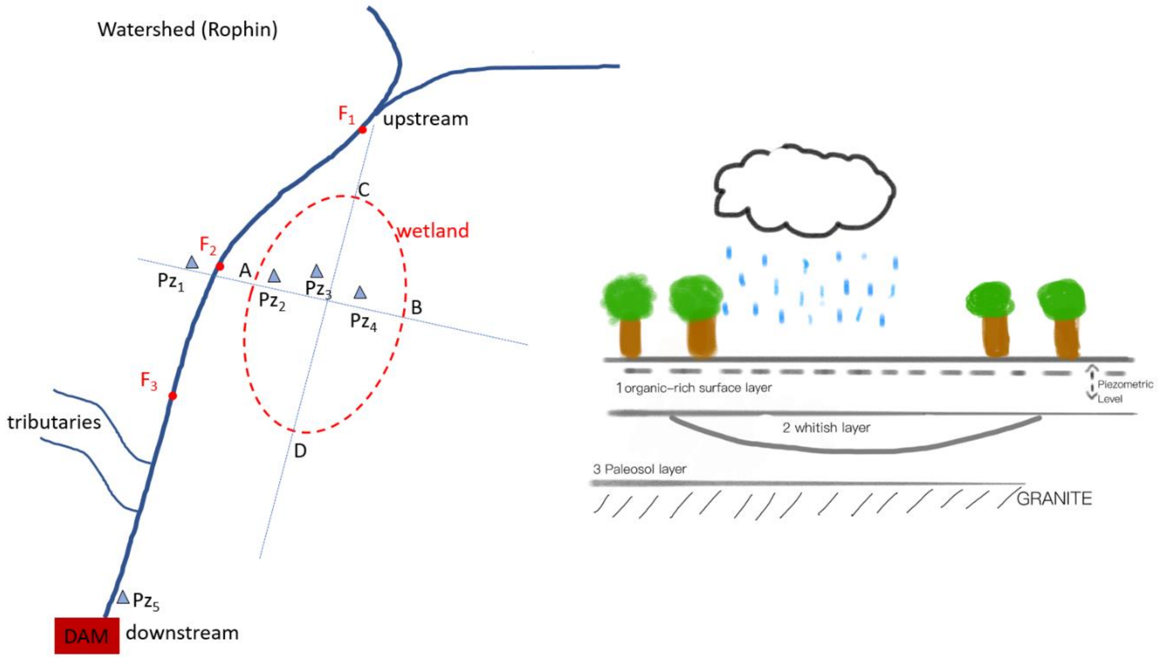
The conceptual model for water circulation and solute transport at the Rophin site is given in Figure 17 and will be the basis for carrying out 1 D and 3 D simulations.

The soil column consists of the three layers of the wetland, i.e. the organic-rich surface layer, the whitish layer and the paleosol layer. The section of the wetland is considered in the longitudinal direction (from points C to D in Figure 17). For establishing proper boundary conditions for the hydrological calculations, an assumption has to be made about the water flow in the wetland going from upstream and downstream, since piezometers pz_2, pz_3, pz_4 situated in the wetland are positioned along the latitudinal layer. However, by using the information about water flows at F_1, F_2 and F_3, the assumption can be made that the difference between the flows at the various locations along the stream provides an average water flow in the longitudinal direction in the wetland. This average flow together with difference in pressure head at points C and D (Figure 17) will provide suitable boundary conditions for the hydrologic calculations with the available models. As a check for the validity of this assumption, experimental pressure head at pz_3 may be recovered via calculations.

When using 3D models, in order to consider the simplest possible mesh for calculations, the centre of the wetland will be considered, in which the thickness of the whitish layer is rather constant. The use of 1D models may be possible only by making assumptions on horizontal drainage form the three soil layers of the wetland towards the stream. The different lithological layers have a uniform thickness in the conceptual model. Surface topology is not considered. Thus, the surface of the conceptual model is at z= 0m.

Calculations will be carried out until the system reaches an equilibrium state (in terms of water flow from and to the watershed). For solute transport calculations, the source term considered will be based on information about radionuclide contamination that was historically introduced in the wetland, i.e. in the whitish layer. The K_d approach will be used based on information available from Task 2.7 for the 3 soil layers. For both flux regimes, saturated soil will be considered, i.e. no need to consider the presence of an unsaturated zone since even in the low flux regime the water table is expected very close to the surface (at a depth of about 20 cm).

Models' output will be the U and Ra concentration in groundwater. By means of mass balance analysis, it will be also possible to quantify the radionuclide concentration in the Gourgeat stream.



Note: On the left the schematisation from above: the oval is the wetland. Piezometers (Pz_{1-5}) locations are indicated as well as measurement locations of the water flows along the Gourgeat stream (F_{1-3}). Points A and B indicate transversal direction to the wetland, points C and D indicate longitudinal direction to it. On the right, the section of the wetland along the longitudinal direction in which the three layers (1, 2, 3) of the wetland are indicated

Figure 17. Conceptualisation of Rophin site.

3.3.3 Modelling tools

3.3.3.1 CIEMAT Model

The CIEMAT Model (Pérez-Sánchez and Thorne 2014) is a 1 D compartmental model built with the Software AMBER (Quintessa Ltd). It considers a simplified modelling approach to soil hydrology to estimate seasonally varying water contents and fluxes in a multi-layer soil model. The soil is divided into a column of up to 10 compartments as shown in Figure 18. The flow calculations are required to provide water flows into and between these soil compartments and water content for each of them. This soil column is considered either to drain from its base or to receive an upward groundwater flux from below and root-depth distribution and root-depth uptake are accounted for.

Modelling locations specific situations required consideration of timescale shorter than one year, as it is seasonal variations in hydrological conditions that cause the migration of contaminants, up and down the soil column. In various dose-assessment tools (e.g. RESRAD-ONSITE), only averaged annual fluxes of water are considered, but such models cannot capture the non-linear effects that arise from multiple interactions, i.e. changing water flows, water contents, root uptake rates, sorption coefficients and volatilisation rates. In CIEMAT model a monthly representation of the hydrological cycle is adopted because this captures in most cases adequately the main seasonal variations and because climate station data are readily available on a monthly basis.

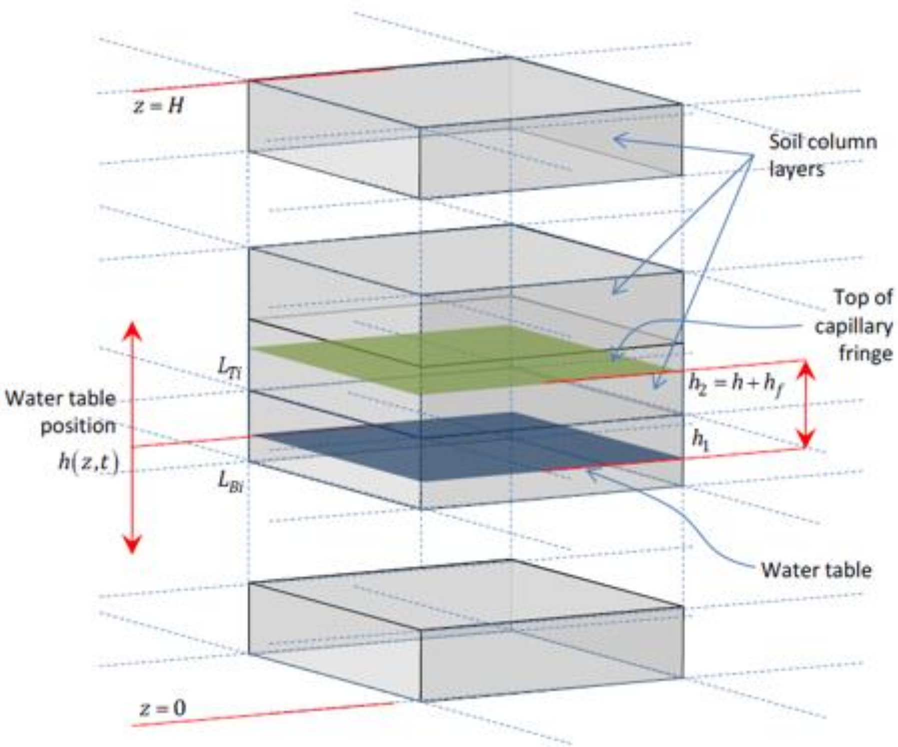


Figure 18. Compartmentalization of soil column in CIEMAT model (Pérez-Sánchez and Thorne 2014)

In the model, the soil column is considered either to drain from its base or to receive an upward groundwater flux from below. The former is appropriate to well-drained agricultural soils whereas the latter is appropriate to discharge zones (e.g. riparian areas). The fundamental equation used to represent the drainage situation is:

$$dh/dt = [R_i - ah^b - S_i PE_i]/\Delta\theta \quad \text{(Eq. 12)}$$

Where: h (m) is the height of the water table at time t; R_i (m y^{-1}) is the average precipitation/irrigation rate in month *i*; PE_i (m y^{-1}) is the potential evapotranspiration rate for month i; *a* and *b* are calibration constants; S_i is the fraction of the potential evapotranspiration (PE) that constitutes actual evapotranspiration (AE); Δθ is the difference in water content between soil that is fully saturated and soil that contains only some minimal water content.

The adopted transport model treats each of the soil layers as a well-mixed compartment with transfers governed by first-order kinetics. The effective volume of each compartment includes the effects of sorption via a K_d model. The model is normalised to a reference soil area, *A*, of 1 m^2. Thus, the effective volume of each soil layer i, V_i (m^3), is given by:

$$Vi = A \cdot d_i(\theta_i + \rho K d_i) \qquad \text{(Eq. 13)}$$

Where: d_i (m) is the user-specified depth of the soil layer; θ_i (-) is the average water content of that soil layer obtained from the hydrological model; ρ (kg m^{-3}) is the dry bulk density of the soil; Kd_i (m^3 kg^{-1}) is the distribution coefficient for that soil layer.

Above the capillary fringe, oxic conditions prevail and one value of the distribution coefficient (Kd_2) applies. Below the water table, anoxic conditions prevail, and a second distribution coefficient (Kd_1) prevails. In between, a linear interpolation is assumed, i.e. at height x, where $h_2 > x > h$:

$$Kd(x) = Kd_2 + (Kd_1 - Kd_2)(h_2 - x)/(h_2 - h) \qquad \text{(Eq. 14)}$$

K_d values computed as described above are vertically averaged over each soil layer. Henceforth, where K_d values are cited, it is these vertically averaged values that are meant.

Radionuclide transport rates between the soil layers and across the top and bottom surfaces of the soil column are calculated using the water fluxes, *F*, thus:

$$\lambda_{ij} = A\, F_{ij}/V_i \qquad \text{(Eq. 15)}$$

where λ_{ij} (y^{-1}) is the rate coefficient for transfer from layer *i* to layer *j* (defining layer 0 as a sink for surface discharge and layer *ij* as a sink for drainage through the base of the soil column).

3.3.3.2 MELODIE

The MELODIE software is a numerical tool developed by ASNR (Mathieu *et al.* 2008) to model the transport of radionuclides in the context of radioactive waste disposal facilities. MELODIE describes the release and migration of radionuclides from the repository through the geosphere to the outlets, under saturated and unsaturated conditions. The code is based on the hypothesis that the natural environment is a continuous porous medium. Hence, the physical properties of the medium are considered as average variables conveying the effects of the heterogeneities of the rock on the scale of the representative volume element (RVE) chosen. The design of MELODIE is based on finite volume finite element (FVFE) (Amaziane *et al.* 2008; Dymitrowska *et al.* 2007) method for numerical approximation of flow and transport equations.

Regarding radionuclide transport, the MELODIE software considers convection, dispersion and anisotropic diffusion mechanisms, together with the physical and chemical interactions with the medium, radioactive decay and the properties of daughter products. The physical and chemical mechanisms incorporated are currently limited to adsorption and desorption using the K_d model, and to the consideration of the solubility limits of the radionuclides. Taking all the mechanisms described above into account results in the following global (Eq. 16) for the hydro-dispersive transport of a RN in a saturated medium:

$$\mathrm{Div}\left[\left(\bar{\bar{\alpha}} \times |\vec{\mathrm{U}}| + \omega \mathrm{D_p}\right)\overrightarrow{\mathrm{grad}}\mathrm{A} - \vec{\mathrm{U}}\mathrm{A}\right] = \omega' \mathrm{R}\frac{\partial \mathrm{A}}{\partial \mathrm{t}} + \omega' \lambda \mathrm{RA} - \omega' \mathrm{R_p} \lambda \mathrm{A_p} \qquad \text{(Eq. 16)}$$

where $\vec{U}$ indicates the Darcy velocity (m y^{-1}), $\bar{\bar{\alpha}}$ the dispersion tensor (m), D_p the pore diffusion coefficient (m^2 y^{-1}), ω' refers to the kinematic porosity where the fraction of immobile water bound to the solid phase is assumed to be only very slightly penetrated by the transported elements; ω the porosity accessible to the radionuclide (-), A represents the specific activity of element in the liquid phase (Bq kg^{-1}), Ap the specific activity of the parent element, at time t (Bq kg^{-1}), λ is the radioactive decay constant (year^{-1}), R is the retardation coefficient (-), ρ_s refers to the density of the porous medium (kg m^{-3}). The three terms at the right side of Eq. 16 represent the accumulation, the radioactive decay, and the creation of daughter products, respectively. The retardation coefficient R is reported in Eq. 17 where the distribution coefficient K_d (L kg^{-1}) is considered.

$$R = 1 + \frac{1 - \omega}{\omega'} \rho_s K_d \qquad \text{(Eq. 17)}$$

3.3.4 Inputs and parameters needed for CIEMAT and/or Melodie

Ideally the following information is required at the observations points and in each compartment considered (based on conceptual model presented in Section 3.3.2)

Hydrological parameters

- geographical location of the watershed area from which water is reversing into the wetland and Gourgeat stream, location of observation points (piezometers pz_1, pz_2, pz_3, pz_4, pz_5) as well location of other measurements (water flow measurements F_1, F_2, F_3), location at which the Gourgeat stream sediments were characterised;
- average depth of each of layer considered separately in the wetland (organic-rich surface soil layer, whitish layer, paleosol layer);
- location of piezometric level, i.e. water table during high flow regime and low flow regime;
- source term, e.g. water flow rate in the stream (L s^{-1});
- soil permeability, dispersivity, diffusion coefficient, porosity;
- hydraulic head (m) (at piezometers' locations) and at locations C, D (Figure 17);
- soil density, type of soil (%loam, %sand, %clay) and porosity.

Meteorological parameters

- hourly/daily precipitation rates [mm hr^{-1}],
- daily sunshine duration [hr d^{-1}],
- relative humidity [%], wind velocity [m s^{-1}],
- air temperature [degree]

This data may be obtained from the closest meteorological weather station.

Geochemical parameters

- Inventory of radionuclides and total concentration present in the layers of the wetland
- Inventory of radionuclides and total concentration present in the sediments of the Gourgeat stream
- Radionuclide concentration in groundwater
- Information on the quantity of radionuclides originally dismissed in the wetland (source term)
- Heavy metals present in soil solution [mg l^{-1}], pH, redox potential, electrical conductivity, ionic strength, dissolved organic carbon (DOC) in water (river stream / percolating water / porewater)
- Ionic elements concentrations in water (river stream / percolating water / porewater)
- Mineral composition and mass fraction [mmol kg^{-1}], organic carbon [%]
- CEC for clay minerals, Fe-oxides [mmol kg^{-1}], Al-oxides [mmol kg^{-1}]

Vegetation parameters:

- Description of plants present at the surface
- Depth of their roots.
- Number of plants per m^2.

This data may be necessary if evapotranspiration is modelled in more detail. However, simple models exist with generic assumptions for which this information may not be necessary and only information on air temperature is required.

3.4 Results

3.4.1 CIEMAT

These results consider only the hydrological regime in the site, not consider the lateral flow and surface runoff process that would be consider in next calculations.

The radionuclide transport model adopted treats each of the soil layers as a well-mixed compartment, with transfers governed by first-order kinetics.

Apart from the quantities defined in description of the site, parameter values for the following quantities are required to run the model:

- time series of potential evapotranspiration;
- time series of precipitation;
- depth to the field drains and their location;
- depth to the water table;
- saturated hydraulic conductivity for each layer;
- saturated and residual water content for each layer;
- values of the Van Genuchten parameters α and m for each layer.

Mean monthly potential evapotranspiration and precipitation values were provided by meteorological data in spreadsheet format. These values were converted to rates using the number of days in each month. This also gives the period in days during the first year for which these rates apply. For second and subsequent years, the periods are obtained by adding an integer multiple of 365 to the listed values. Note that leap years are omitted as a simplification. The unit is set in mm d^{-1} whereas in the model, these rates are specified in units of m d^{-1}. The depth to the field drains is taken to be -2.5 m and they are taken to be located laterally 10 m from the modelled area (i.e. L = 10 m). In practice, in the illustrative calculations, the soil column remains significantly under-saturated, so drainage to these field drains does not occur.

The water table is taken to lie at -10.0 m. The important point is that it is located a substantial distance below the soil column, so that vertical drainage is maintained. Its exact position is not of any great significance. Thus, the results obtained are generally applicable to any soil of the texture studied that is well-drained to depth. The model also requires initial soil moisture conditions. These were set at 87.3% of θ s. However, the soil rapidly drained from this initial wet condition, so the exact degree of initial saturation is of little significance.

Degree of saturation is shown in Figure 19. The seasonal cycle is very clear, with wetting of the soil in winter and drying in summer. Note also that the soil rapidly dries from its initially wet state, such that by 100 days results are similar to those at 465 days, i.e. one year later.

In winter, the soil is wettest near the surface, as precipitation exceeds evapotranspiration. However, in summer, the surface layers dry out and the lower layers are wettest, though they also exhibit a small degree of summer drying. As expected, soil moisture characteristics vary much more strongly with time in the superficial layers of the soil than in the deeper layers. Even in winter, in this rather arid climate and with a deep water table, the soil never approaches full saturation. This means that actual evapotranspiration is reduced below potential evapotranspiration to a greater degree than would be expected in northern European conditions.

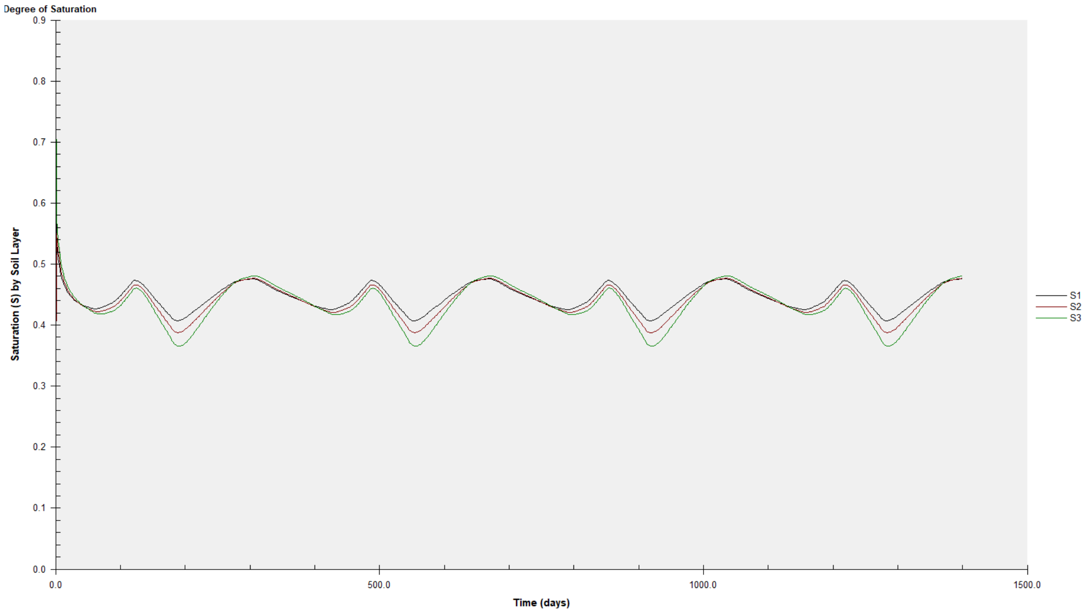


Figure 19. Time-variation in degree of soil saturation in different soil level.

The following Figure 20 illustrates the variations in matric potential. Note that matric potential represents suction and is therefore negative. During winter, the matric potential is close to zero, indicating relatively moist soil conditions. The lowest soil layer exhibits a much higher suction than the others, reflecting the downward pull of water toward the local water table. In summer, the upper layers of the soil dry out considerably, leading to much more negative matric potential values. These calculated values fall within the typical range observed in natural soils. Notably, layers 1 to 3 experience drying earlier in the summer than layer 4. The limited drying observed in layer 1 may be due to the relatively small proportion of evapotranspiration assigned to this layer and the ongoing input of precipitation, which is averaged on a monthly basis. The figure clearly demonstrates the consistent annual patterns in these variations, highlighting the stability and repeatability of the results year after year.

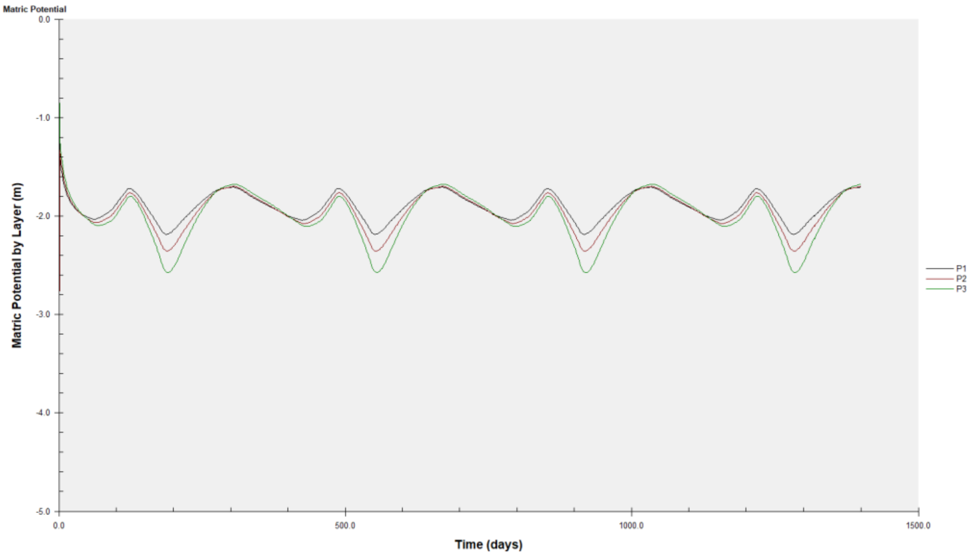


Figure 20. Matric potential in considered soil layer.

CIEMAT model have limitations of applying one-dimensional models to large-scale groundwater systems. The one-dimensional (1D) models are useful tools for understanding groundwater behaviour in simplified or localized contexts, their application to large-scale territories presents significant limitations. The primary challenge lies in the inability of 1D models to represent lateral flow and spatial variability—key characteristics of real-world aquifer systems.

In extensive areas, groundwater movement is influenced by three-dimensional heterogeneities in geology, topography, recharge patterns, and hydraulic properties. 1D models, by definition, can only simulate flow in a single vertical or horizontal direction, and therefore fail to capture lateral groundwater interactions, flow convergence/divergence, and complex boundary conditions typical of large aquifers.

Moreover, large territories often include multiple hydrogeological units, variable land uses, and diverse recharge/discharge zones, all of which require a more detailed spatial representation to accurately simulate water balances, contaminant transport, or aquifer responses to external pressures.

As a result, while 1D models are valuable for preliminary assessments, vertical infiltration studies, or local-scale unsaturated zone processes, they are generally inadequate for regional-scale groundwater management and decision-making, where multi-dimensional approaches are necessary to capture the complexity of the system.

The CIEMAT model has proven to be a reliable and effective tool for describing the accumulation and transport processes of U and Ra in soil. Its capacity to simulate the influence of sorption properties, water flux variability, and long-term dynamics makes it particularly suitable for studying redox-sensitive radionuclides under realistic environmental conditions. The model's outputs align with expected behaviours of U and Ra based on their geochemical characteristics, supporting its application in safety assessments and environmental impact studies involving contaminated soils.

The Figure 21 show the simulation results obtained using the CIEMAT model, which was specifically developed to assess the behaviour of redox-sensitive radionuclides in soil environments. These results highlight the contrasting transport and accumulation patterns of U and Ra, driven largely by differences in their sorption properties, as described by the distribution coefficient (K_d).

For U, which exhibits a lower K_d, the model predicts a series of inter-annual peaks and troughs in concentration over time, superimposed on a general increasing trend. This behaviour reflects U's relatively high mobility in the soil and its sensitivity to changes in water fluxes, such as those induced by seasonal irrigation or precipitation. Over a period of about ten years, the system approaches a steady-state condition, although annual fluctuations persist. These results demonstrate uranium's potential for vertical migration, with significant implications for leaching and groundwater contamination under certain conditions.

In contrast, Ra, which has a higher K_d, shows a different pattern in the model output. Although an initial increase in concentration occurs during irrigation periods - similar to uranium - the accumulated Ra remains stable throughout the simulation. Its strong sorption affinity ensures that Ra is not mobilized by varying water fluxes, and as such, it does not exhibit the same seasonal variability. The model thus indicates that radium tends to accumulate near the surface, with limited vertical transport, highlighting a risk of long-term surface contamination but low potential for leaching.

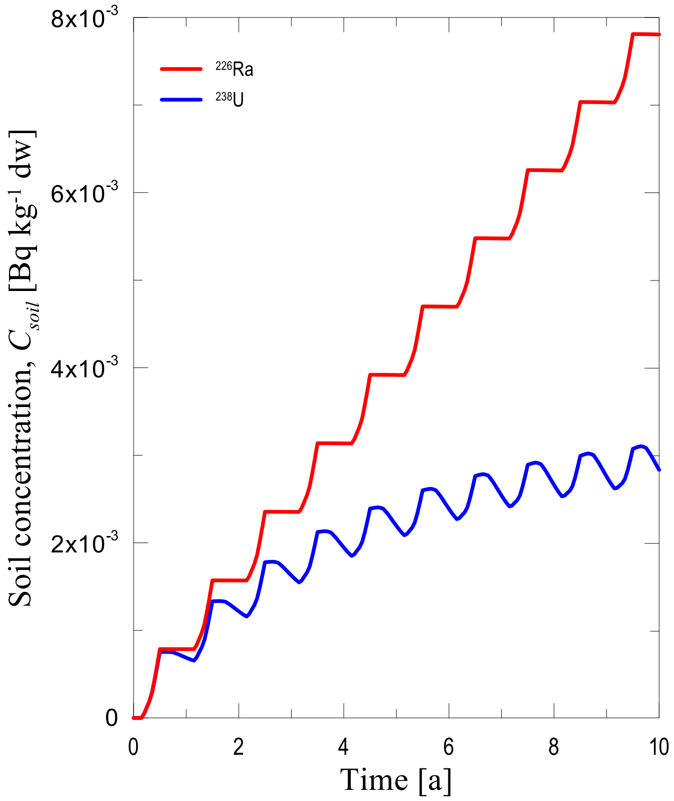


Figure 21. Behaviour of the accumulation pattern of Ra-226 and U-238 radionuclides in soil environments

The Figure 22 compares the behaviour of Ra and U as simulated by two different modelling approaches: a simplified one-compartment model and the more detailed multi-layer model developed by CIEMAT. Both models were used to evaluate long-term accumulation in soil, with particular attention to the impact of sorption (K_d) and hydrological fluctuations.

The results reveal that in the long-term, Ra exhibits similar accumulation patterns in both models. This is largely due to its high K_d value, which ensures that sorption dominates over other dynamic processes such as water table fluctuations or seasonal changes in water flux. As a consequence, Ra becomes immobilized in the upper soil layers, showing limited vertical migration regardless of the model complexity.

However, while the accumulated concentrations are comparable, the one-compartment model tends to slightly overestimate Ra retention. This discrepancy can be explained by the spatial distribution of activity in the CIEMAT multi-layer model: in the latter, Ra is not confined to a single uniform zone but instead is distributed across the ten-layer soil profile, leading to a more realistic (and somewhat diluted) representation of its accumulation.

This comparison highlights the robustness of the CIEMAT model in reproducing realistic behaviour under complex environmental conditions, while also emphasizing the limitations of simplified models when it comes to accurately representing spatial distribution and retention processes in soil. It supports the use of multi-layer modelling in safety assessments, particularly when precise predictions of contaminant localization and long-term behaviour are required.

In contrast to Ra, U exhibits a more dynamic behaviour due to its lower distribution coefficient (K_d), which implies weaker sorption to soil particles and greater sensitivity to changes in hydrological conditions. The comparison between the one-compartment model and the multi-layer CIEMAT model reveals key differences in how U is accumulated and transported through the soil profile.

The multi-layer model shows that U concentrations increase over time but eventually reach a steady state, as sorption and transport processes reach equilibrium. Seasonal fluctuations in water flow, such as those caused by irrigation, are reflected in small oscillations in the accumulation curve. These fluctuations are smoothed out over time, but they are essential in capturing U's partial mobility and potential for redistribution within the soil column.

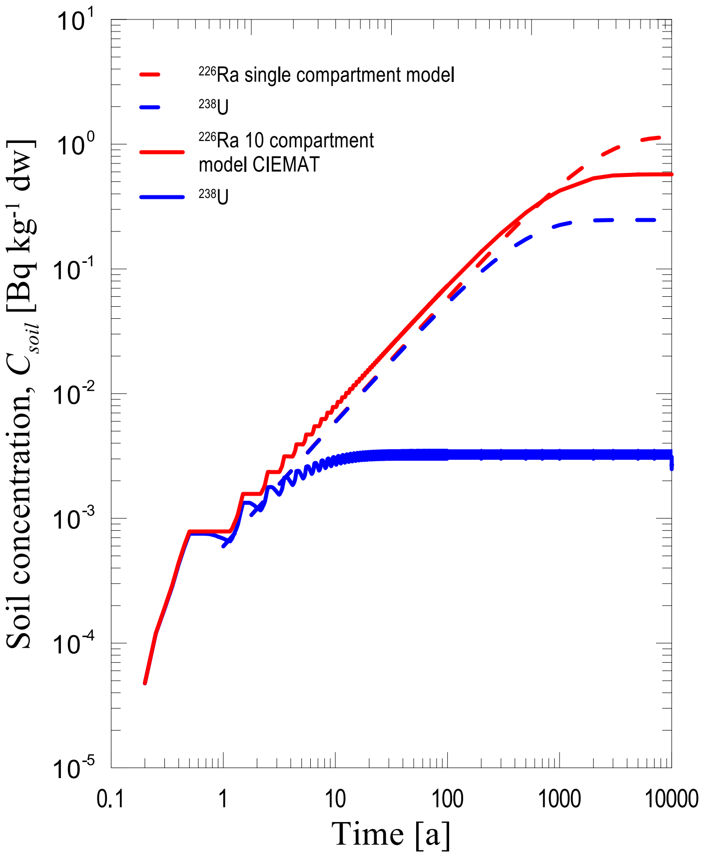


Figure 22. Comparison of one-compartment and multidimensional compartment models for vertical distribution of U-238 and Ra-226 at long term.

In the simplified one-compartment model, U accumulation appears more linear and slightly overestimated compared to the multi-layer results. This overestimation stems from the lack of vertical resolution in the single-compartment approach, which does not account for U redistribution across multiple soil layers. In reality, as modelled by the CIEMAT simulation, U is gradually spread throughout the profile, especially in the upper layers, due to moderate sorption and downward transport.

The CIEMAT vertical (1D) groundwater model is often suitable for simulating infiltration, recharge, and vertical flow processes in the unsaturated zone. However, when it comes to modelling contaminant transport, especially over large areas or in complex hydrogeological settings, such a model presents significant limitations.

As already said, CIEMAT models have limitations of applying one-dimensional models to large-scale groundwater systems. The one-dimensional (1D) models are useful tools for understanding groundwater behaviour in simplified or localized contexts, their application to large-scale territories presents significant limitations. The primary challenge lies in the inability of 1D models to represent lateral flow and spatial variability key characteristics of real-world aquifer systems.

In extensive areas, groundwater movement is influenced by three-dimensional heterogeneities in geology, topography, recharge patterns, and hydraulic properties. 1D models, by definition, can only simulate flow in a single vertical or horizontal direction, and therefore fail to capture lateral groundwater interactions, flow convergence/divergence, and complex boundary conditions typical of large aquifers.

Moreover, large territories often include multiple hydrogeological units, variable land uses, and diverse recharge/discharge zones, all of which require a more detailed spatial representation to accurately simulate water balances, contaminant transport, or aquifer responses to external pressures.

A purely vertical groundwater model presents several important limitations that restrict its applicability for simulating contaminant transport in real aquifer systems. One of the main shortcomings is the lack of representation of lateral flow. Vertical models are unable to capture horizontal transport processes, which are often critical in the migration of contaminants through aquifers. As a result, ignoring the lateral dimension can lead to inaccurate predictions regarding the dispersion of contaminants, the direction and extent of plume movement, and the potential arrival of pollutants at receptors such as wells, springs, or rivers.

In addition, vertical models tend to oversimplify the complex mechanisms involved in transport. Accurate contaminant modelling requires the integration of multiple physical and chemical processes, including advection, dispersion, diffusion, sorption, and decay. These processes interact across all three spatial dimensions in heterogeneous geological environments. A 1D vertical model cannot realistically simulate how these mechanisms operate simultaneously, nor can it properly represent the interaction between flow and reactive transport.

Another critical limitation is the inability of 1D models to capture spatial heterogeneity. Natural aquifers often consist of multiple geological layers with varying hydraulic conductivities, porosities, and geochemical properties. This heterogeneity greatly influences contaminant transport behaviour, especially in terms of preferential flow paths and retention zones. Vertical models, by averaging or simplifying these features, fail to account for local variations that may significantly affect the fate of contaminants.

Finally, vertical models offer limited capacity to define boundary conditions and source terms with the necessary spatial and temporal precision. In real-world applications, contamination sources may be distributed horizontally or may enter the system from lateral boundaries, and such configurations are difficult to implement in a vertical-only framework. Consequently, the reliability of transport simulations based solely on vertical models is insufficient for comprehensive risk assessment or remediation planning in complex hydrogeological settings.

3.4.2 Melodie – Flow modelling

Two conceptual models have been defined, the first focusing on the wetland and the second representing the wetland and the adjacent river.

These two models have in common the absence of interaction between the wetland and the underlying groundwater table.

3.4.2.1 Model 1 - Wetland stricto sensu

The first model represents the wetland with the river represented via a boundary condition. The model has a surface dimension of 200 m x 50 m and a depth of 1.3 m. The layer thicknesses are 20 cm, 10 cm and 100 cm respectively for the humus layer, the whitish layer and the paleosol.

The hydraulic gradient is not explicitly defined. Boundary conditions setting the inflow and outflow, and a node with an imposed potential determine the flow conditions. In zone A, to the north of the model, the inflow (F1-F2) corresponding to the inflow of water from the river into the wetland is defined as the boundary condition. For zone B, which represents the boundary between the river and the wetland, the flow (F5-F4-F2) can be positive or negative. This is because, depending on the season, the river feeds the wetland or vice versa. Zone C represents the model output with a flow rate equal to F5-F4-F1. (Flow explained part 3.2.2.3)

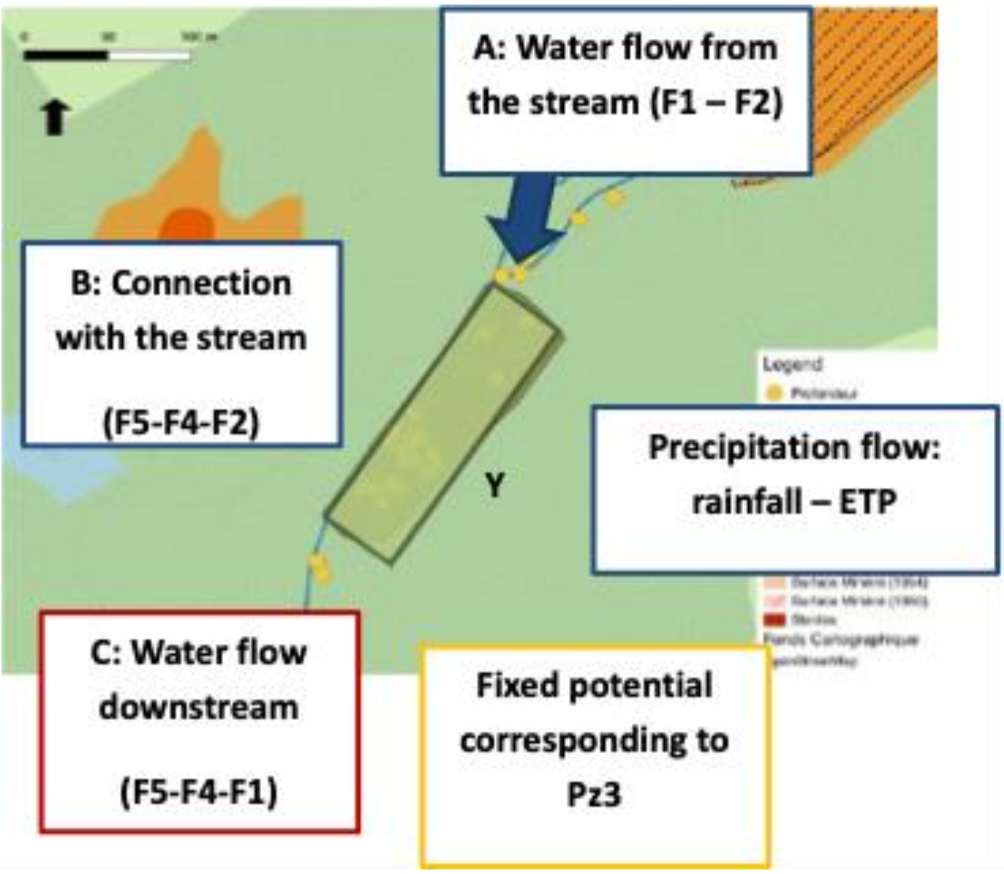


Figure 23. Model 1 boundary conditions

In order to solve the partial differential equations, it is necessary to fix a hydraulic load on at least one node of the model. After several tests, it appears that the position with the least impact on the gradient is in the middle of the model, on a node at the surface. The potential is set equal to the Z value of the most central node in the model, at the surface.

The study was carried out considering three flow configurations in the river, using data from August, October and April, which are the only months with fully usable data (see Table 15 above). Note that the flow in zone B, representing the connection between the wetland and the river, is negative for the month of October. This indicates that, unlike other periods, water has then flowed from the wetland to the river. The different parameter values for A, B and C of the model 1 are shown in Table 18.

Table 18. Calculated flow rates for Model 1 boundary conditions.

	Boundary condition	Flow (L s^{-1})	Surface (m^2)	Flow ((m s^{-1})/node)
Model 1 August	A	1.7	567	2.99E-06
	B	2.5	1825	1.36E-06
	C	4.2	567	7.41E-06
Model 1 October	A	0.6	567	1.05E-06
	B	-0.3	1825	-1.64E-07
	C	0.3	567	5.29E-07
Model 1 April	A	0.4	567	7.05E-07
	B	1.2	1825	6.57E-07
	C	1.6	567	2.82E-06

3.4.2.2 Model 2 - Wetland and river

The second conceptual model explicitly models river flow (Figure 24). To achieve this, the previous model's footprint is doubled in the direction of the Y width (200 m x 100 m x 1.3 m). The mesh density is identical to Model 1. Concerning boundary conditions, water flows in the river are imposed with F1 at input A and F5-F4 at output C. Inlet B at wetland level corresponds to flow 'F5 - F4 - F1'. The hydraulic gradient of the model is imposed with hydraulic loads upstream and downstream of the river section. No constraints are imposed between the river section and the wetland section (blue parallelepiped).

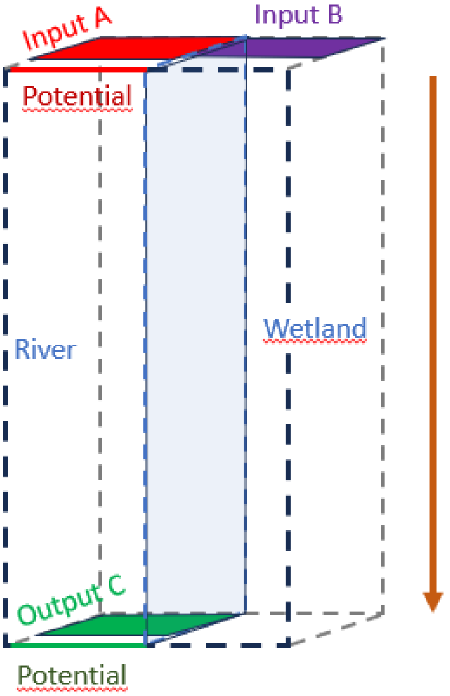


Figure 24. Model 2 boundary condition

The same three periods will be studied: April, August and October. Details of input and output values are given in the Table 19.

Table 19. Calculated flow rates for Model 2 boundary conditions

	Boundary condition	Flow (L s^{-1})	Surface (m^2)	Flow (($m\ s^{-1}$)/node)
Model 1 August	A	9.7	525	1.84E-05
	B	1.7	450	3.77E-06
	C	11.4	525	2.17E-05
Model 1 October	A	0.8	525	1.52E-06
	B	0.6	450	1.33E-06
	C	1.4	525	2.66E-06
Model 1 April	A	3.5	525	6.66E-06
	B	0.4	450	8.88E-07
	C	3.9	525	7.42E-06

3.4.2.3 Model 1 - Flow results

The main direction of flow is from north to south, as expected, as shown in Figure 25.1. The river feeds the wetland during this period. The differences in permeability between the different layers (from Naga geophysics study) are reflected in the Darcy velocities: the respective velocities in the humus, whitish layer and paleosol layers are of the order of 1.6×10^{-5} m s^{-1}, 1.3×10^{-5} m s^{-1} and 4×10^{-6} m s^{-1} respectively (Figure 25.2).

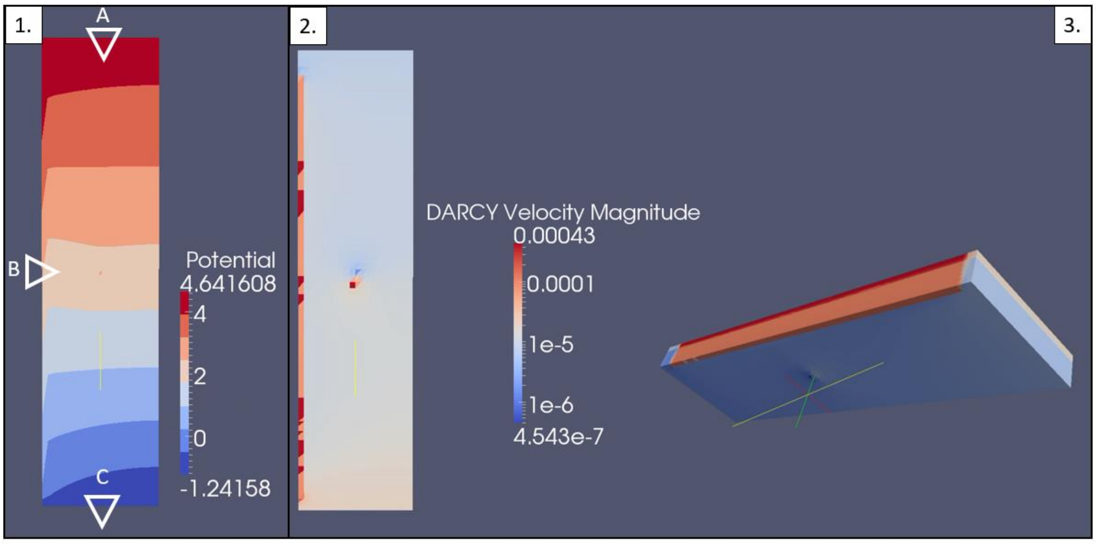


Figure 25. 1. Potential field; 2. Darcy velocity field (top view (left) and side view (right)-*August data*

N.B.: On all Melodie figures, the orange vertical line in the middle corresponds to the y axis.

The node with an imposed potential impacts the flow in its immediate environment, as shown in the Figure 25 and Figure 26.

The water level in the model corresponds to the difference between the potential and the Z coordinate (altitude) and is shown in Figure 26. The water level is highest upstream of the model. An unsaturated part appears downstream.

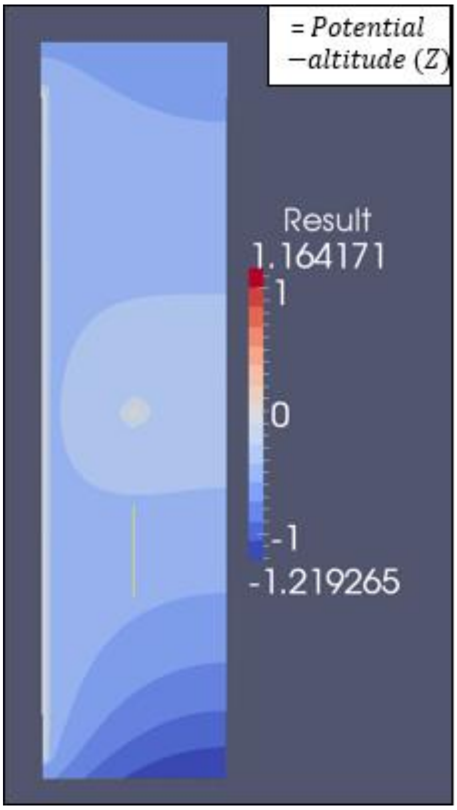


Figure 26. Water level distribution according to XY, model I, August data.

3.4.2.4 Model 2 - Flow results

The potential gradient and flows imposed on the river section explain the homogeneity of the potential and Darcy velocity results observed on the left-hand side of the model (Figure 27).

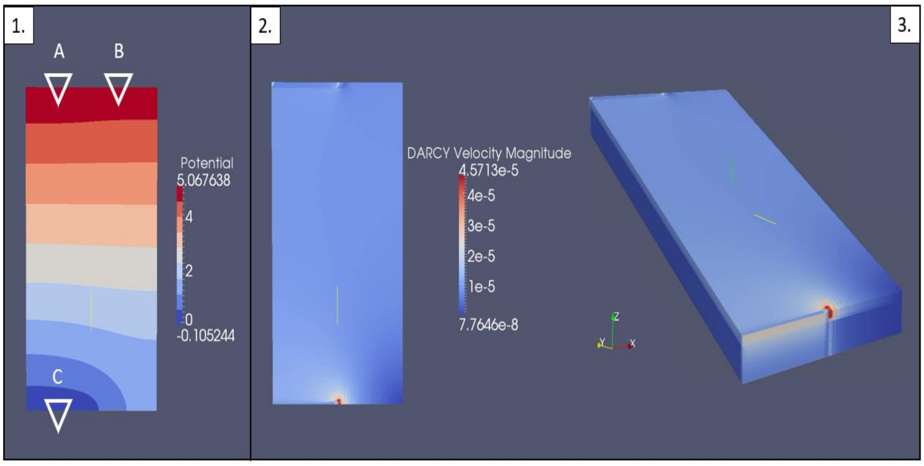


Figure 27. Potential and Darcy velocity field for model II. August data

Observed water levels are high in the bottom right of the model Figure 28. The absence of a boundary condition on the lower right edge causes a blockage of water downstream of the wetland, the only possible exit being the river zone.

August data show that the river feeds the wetland upstream, and the reverse downstream.

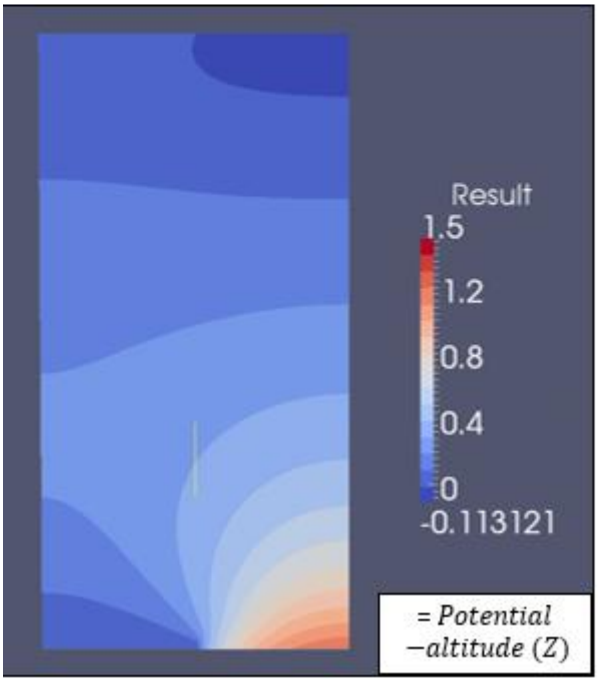


Figure 28. Water level for model II August data

The results for the April and October periods are similar in terms of potential fields, Darcy velocities and water heights, but differences in absolute values are observed. Interactions between the wetland and the river also vary depending on the month studied.

3.4.2.5 Conclusion – flow part

At this stage, we are not in a position to discriminate the most appropriate conceptual model between the two that have been designed. To do this, we will need to gain a better understanding of the flows within the study area via:

- piezometers installed in the main flow direction (these were installed in October 2024)

- a better understanding of flows within the watershed to set the boundary conditions of our model (interaction between the groundwater table and the wetland?). For this, a watershed-scale flow model could be the solution to know the boundary conditions to apply in this model centred on the wetland.

At the same time, it is essential to obtain specific permeabilities for each layer, as this parameter has a significant impact on flow velocities.

3.4.3 Melodie - Transport model

The transfer of RNs in the Rophin wetland is linked to water flow and is therefore mainly convective. Furthermore, it is considered that the solubility of U and Ra in the medium is driven solely by sorption onto mineral surfaces and not by the precipitation of a mineral phase. Thus, retention phenomena in the different layers are only described with a K_d.

U and Ra have a labile (minority) part and a non-labile (majority) part (Table 17). This parameter is not taken into account in the model. When interpreting transport results, it is therefore important to keep in mind that we are only looking at the behaviour of the labile part of radionuclides and for this reason we have chosen to carry out model comparisons by changing parameters such as boundary conditions or permeability, rather than discussing absolute values of activity flows over time.

For a first approach, we have chosen to carry out transport modelling with the “Model 1”.

Radionuclides are introduced into the red zone shown in Figure 29 based on the data presented in part 3.2.3 and we will start with a study of uranium transport.

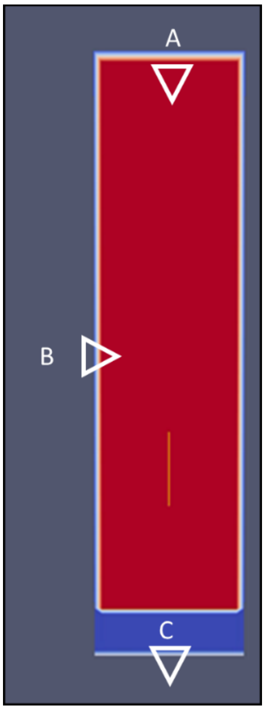


Figure 29. Initial state of radionuclide distribution, XY view

3.4.3.1 Boundary conditions test

In a first step, the sensitivity of radionuclide transfer to transport boundary conditions is investigated. Four configurations of boundary conditions are studied:

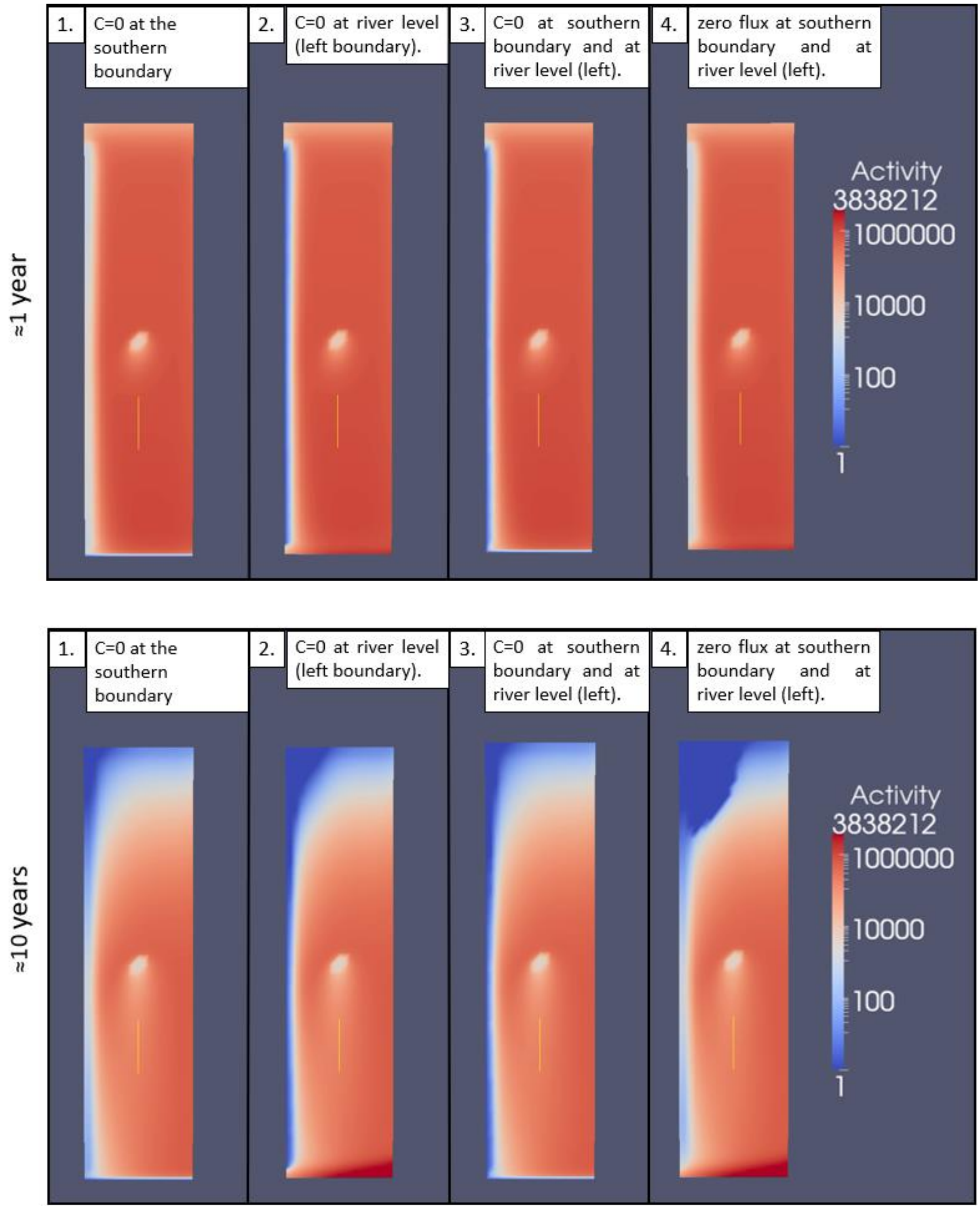
1) C=0 at the southern boundary.
2) C=0 at river level (left boundary).
3) C=0 at southern boundary and at river level (left).
4) zero flux at southern boundary and at river level (left).

The boundary condition "C=0" means uranium concentration equal to zero. This is a Dirichlet condition, imposing the value of the unknown. In concrete terms, when a radionuclide reaches the model boundary, it is automatically removed from the model to respect the chosen boundary condition.

Imposing a zero flow or writing nothing on a boundary is interpreted in the same way by Melodie. This means that convective transport is blocked, and radionuclides will leave the model very slowly, as they will only diffuse.

Southern boundary corresponds to "C" on Figure 29 and river level to "B".

Uranium transfer results are shown in Figure 30 for the four boundary conditions, and for timeframes of 1 year, 10 years and 1000 years.


1.
C=0 at the southern boundary
2.
C=0 at river level (left boundary).
3.
C=0 at southern boundary and at river level (left).
4.
zero flux at southern boundary and at river level (left).
≈1 year
≈10 years
Activity
3838212
1000000
10000
100
1

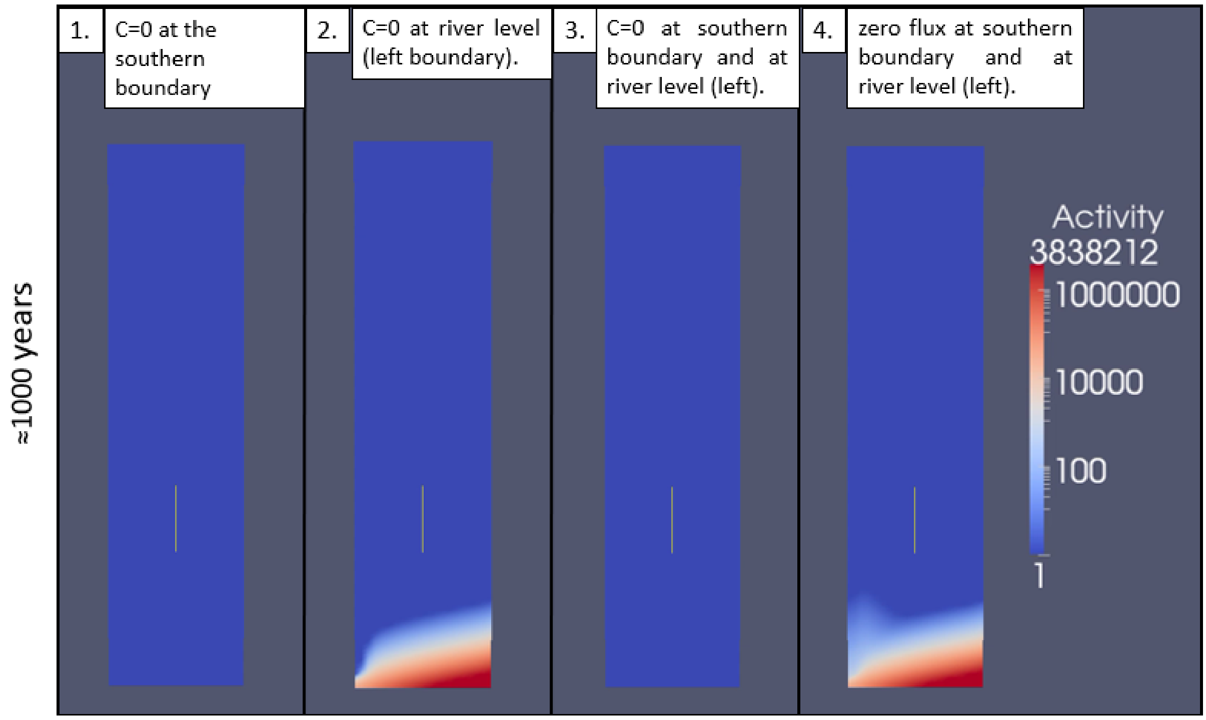


Figure 30. U activities (Bq) from transport calculations with different boundary conditions at 1, 10 and 1000 years.

Cases 2 and 4, with either no boundary conditions or a zero-flux condition for the southern boundary of the model, show that radionuclides remain trapped downstream of the wetland. This is not consistent with field observations, which do not show higher uranium concentrations downstream. In this respect, it is chosen for the rest of the discussion to not consider these cases. The difference between cases 1 and 3 is only visible in the short term (1 and 10 years) in the area of interaction with the river. Our knowledge of the wetland is not, at this stage, sufficient to determine which of these two cases is closer to reality. Arbitrarily, for the following tests, the boundary conditions of Case 1 will be used.

3.4.3.2 Comparison of U and Ra transport

Transfer within the wet zone for Ra is slower than for U, as shown in Figure 31. U and Ra transfer results show that the retention of Ra is greater than that of U in all layers. The difference in transfer rate between U and Ra is explained by the difference in K_d between these two radionuclides.

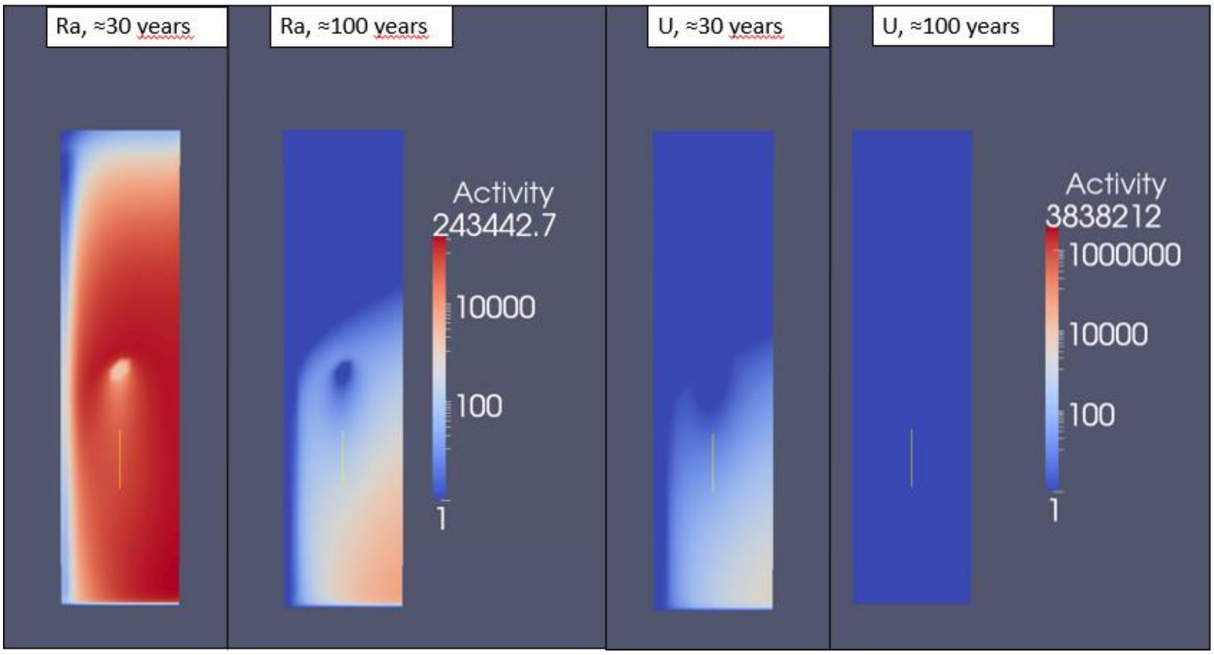


Figure 31. Evolution of Ra and U activities (Bq) in Model I and for case 1 (August).

As transport is mostly convective, the temporal extension of the plume of the two radionuclides is driven by the downward flow of water within the wetland and the flow of water from the river. In this model, the impact of the potential fixed in the middle of the wetland is visible on the radionuclide plume.

3.4.3.1 Seasonal variability

Simulations were carried out using river flows measured in August (the reference month in this report), October and April. The results show that for the month of April, transfer is slower than for the other two months, demonstrating a seasonal variability in U transport within the wetland as a function of hydrological conditions (Figure 32).

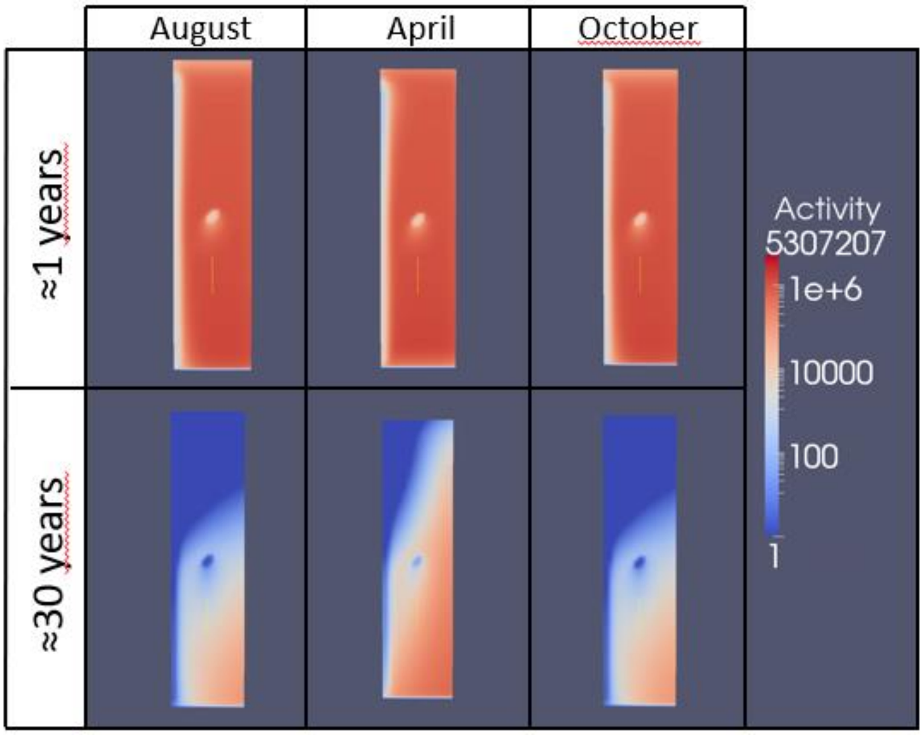


Figure 32. Comparison of U plumes at 1 and 30 years for the months of August, April and October.

3.4.3.2 Sensitivity test on the permeability of the whitish layer

A sensitivity test was carried out on the model to assess the impact of permeability on the transfer of radionuclides, as this parameter is highly uncertain in relation to the NAGA Geophysics geophysical results that could not distinguish the humus layer and the whitish layer.

The test is performed on U. The model used is the same as the initial model, with the permeability of the whitish layer divided by 100, i.e. $3x10^{-6}$ m s^{-1}. This value was defined arbitrarily but corresponds to the same order of magnitude expected for a layer with a silty granulometric composition, as the whitish layer approximates this granulometry in certain respects (Martin *et al.* 2020).

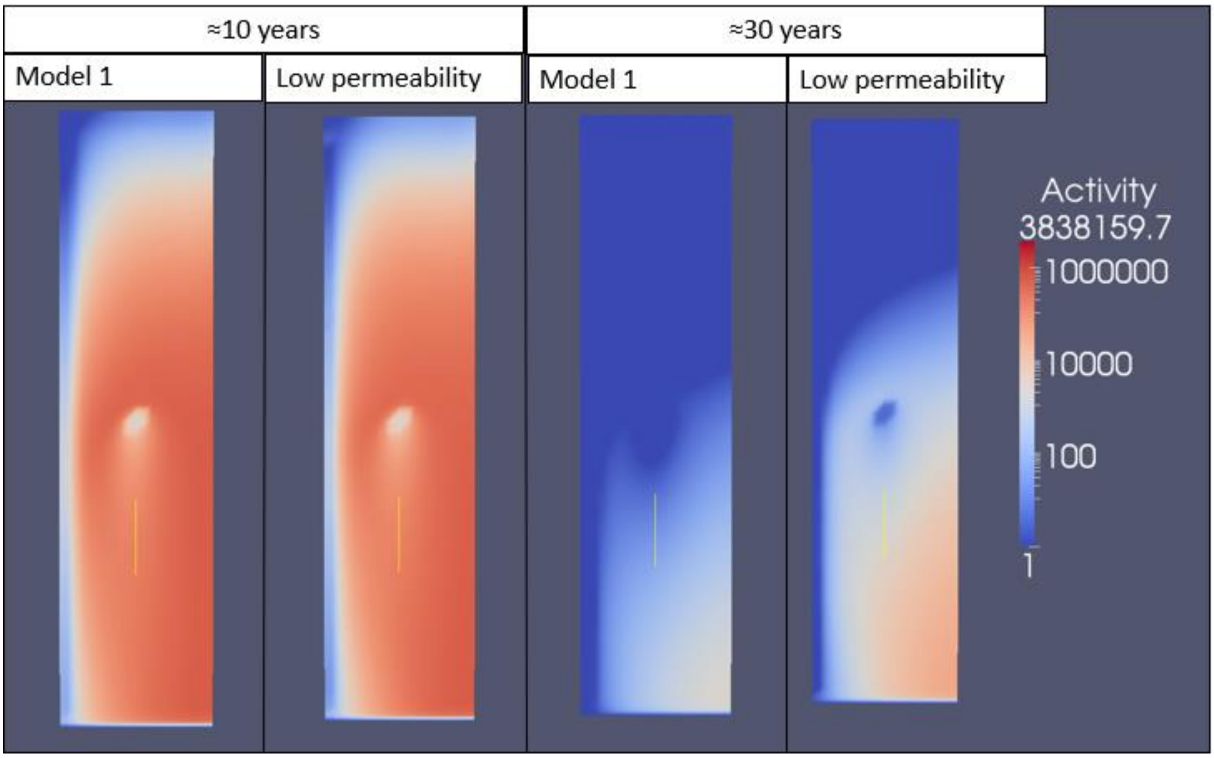


Figure 33. U transfer with K = 3.10^{-4} m/s (Model 1) and K = 3.10^{-6} m/s (Low permeability) for the whitish layer (August).

The permeability of the white layer influences the rate of U transport, as shown in Figure 33, which presents the U activity plume at 10 and 30 years but the trend remains the same. Depending on how the model will be applied, the need for precise knowledge of the permeabilities of the different layers may vary. If we wish to understand the mechanisms driving radionuclide transport, a permeability taken from literature may perhaps be sufficient. On the other hand, if we wish to obtain the activity fluxes leaving the wetland over time and integrate them into impact studies, additional investigations will have to be carried out.

3.4.3.3 Conclusion – transport part

The boundary conditions chosen can have a major impact on the results, as shown by the tests carried out. These parameters must be chosen with care, and a cross-check between model and field data is essential at the validation stage.

U is transferred more rapidly than Ra, in the order of fifty years. This is due to the delay factors of the two upper layers (humus and whitish layers), which are lower for the U than for the Ra.

Knowledge of certain key parameters such as permeability is essential for transport modelling.

3.5 Conclusions

Groundwater modelling can be approached using different levels of spatial complexity, depending on the objectives of the study and the available data. One-dimensional (1D) models are typically used for simplified analyses where flow occurs primarily in a single vertical or horizontal direction, such as in soil column studies or infiltration through unsaturated zones. These models are computationally efficient and useful for assessing vertical water movement, recharge processes, or solute transport in stratified soils.

In contrast, three-dimensional (3D) models provide a more realistic representation of groundwater systems by accounting for flow in all spatial directions (x, y, z). They are essential for simulating complex aquifer systems, interactions between surface water and groundwater, and heterogeneous hydrogeological conditions. 3D models can incorporate variations in hydraulic conductivity, boundary conditions, and sources or sinks such as wells, rivers, and recharge zones.

While 3D models offer greater accuracy and spatial detail, they also require significantly more data, computational resources, and calibration effort. The choice between 1D and 3D modelling depends on the scale of the problem, the degree of heterogeneity, and the specific questions being addressed.

4. Review of approaches and tools for assessing impact of NORM on non-human-biota.

4.1 Introduction

In the past, exposure assessments to ionising radiation were mainly focused on humans, based on the premise that if human beings were adequately protected, non-human-biota and the environment were also protected. However, the International Commission on Radiological Protection (ICRP), in its recommendations published in 2007 (ICRP 2007), included within its system of radiation protection (RP) the objective of protecting the environment in an explicit and dedicated way.

When dealing with NORM, situations may be encountered in which the risk of impact of radiation on non-human-biota may arise. Various tools and methods are available and are being further developed to tackle the issue of NORM exposure and radiation risk to non-human-biota.

The Commission in its Publication 142 (Lecomte *et al.* 2019) has provided guidance on radiological protection of workers, public and the environment for NORM-involving industries. These industries are diverse and may involve exposure of the environment where protective actions need to be considered. NORM industries may give rise to multiple hazards, and the radiological hazard is not necessarily dominant. Therefore, an integrated and graded approach is recommended by ICRP for the protection of the environment, where both non-radiological and radiological hazards are included, and the approach to protection is optimised (graded). According to the characteristics of the exposure situation and the magnitude of the hazards, the exposed organisms in the environment should be identified and appropriately derived reference levels (DCRLs) should be used to inform decisions on options for the control of exposure.

Also, with implementation of new regulations (for example in Europe with the Directive 2013/59/EURATOM (European Commission 2014) and increased environmental protection awareness due to the impact of climate change, further considerations are seen as necessary within the RP community to address the RP of non-human-biota from a less anthropocentric perspective. Hence more studies are being implemented that tackle these issues.

An overview of the available approaches and modelling tools to quantify exposure and risk for non-human-biota was collated. In particular, literature collected regarding NORM and terrestrial/marine/freshwater ecosystems and different types of NORM-involving industries is reviewed and lessons-learned from these are summarised.

4.2 Approaches for radiological protection of the environment

4.2.1 ICRP approach for radiological protection of the environment

The ICRP stated that their aim for environmental protection is '*preventing or reducing the frequency of deleterious radiation effects to a level where they would have negligible impact on the maintenance of biological diversity, the conservation of species, or the health and status of natural habitats, communities and ecosystems*'.

The approach for the protection of the environment must be proportional to the general level of risk, as well as complementary, analogous and parallel to the approach followed for the radiological protection of humans. In addition, it must be compatible with the approaches used in the protection of the environment against other hazards (e.g. chemical products) (ICRP 2008).

The ICRP approach focuses on impacts on biota but not upon detriments to the abiotic component of the environment or environmental compartments (soil, air, water, sediment). Thus, the approach to assess and control the effects of ionising radiation on flora and fauna is based on: (i) the use of Reference Animals and Plants (RAP) and Representative Organisms (ICRP 2007; 2008; Pentreath *et al.* 2014), consistent with the concepts of Reference Person and Representative Person for humans (ICRP 2007), and (ii) in dose rate criteria in the form of Derived Consideration Reference Levels (DCRL).

The RAPs are defined as hypothetical entities, with the assumed basic biological characteristics of a limited suite of animals and plants, which can be used to relate exposure to dose, and dose to effects for that type of organisms, and thus derive some form of numerical guidance (the DCRLs) to help decision making and manage the protection of the environment in different situations of exposure to ionising radiation (ICRP 2008) (Table 20).

Table 20. Wildlife groups, ecosystems they inhabit and DCRLs for the twelve RAPs currently included in the ICRP approach for radiological protection of the environmental (ICRP 2008).

Wildlife group	Ecosystem	RAP	DCRL, mGy d^{-1} (shaded)		
			0.1-1	1-10	10-100
Large terrestrial mammals	T	Deer			
Small terrestrial mammals	T	Rat			
Aquatic birds	F, M	Duck			
Large terrestrial plants	T	Pine			
Amphibians	F, T	Frog			
Pelagic fish	F, M	Trout			
Benthic fish	F, M	Flat fish			
Small terrestrial plants	T	Herb			
Algae	M	Brown alga			
Terrestrial insects	T	Bee			
Crustacean	F, M	Crab			
Terrestrial annelids	T	Earthworm			

Note: T, terrestrial; F, freshwater; M, marine.

The ICRP also gives information on environmental dosimetry in the form of dose conversion factors and on biological effects for the set of RAPs, considering that the biological endpoints of most relevance in individuals after radiation exposure will be those that could lead to changes in population size or structure. Among these endpoints are early mortality, morbidity, impairment of reproductive capacity and the induction of chromosomal damage (ICRP 2008).

The ICRP provides follow-up guidance in several subsequent publications. In Publication 114 (Clement *et al.* 2009), a methodology is outlined for the purpose of quantifying transfer from media (water or soil) to RAPs. The report concludes that equilibrium concentration ratios (CRs) are most commonly used to model transfer and would thus offer a comprehensive data coverage for use in ICRP's framework. Methods used to derive CRs are presented, and a means of summarising statistical information from empirical data sets described. The ICRP, within its general radiological system, makes a distinction between emergency, existing and planned exposure situations. In Publication 124 (Pentreath *et al.* 2014), the application of the ICRP's environmental protection approach to these types of exposure situation is explained, providing the key principles that are relevant; and hence how reference values (the DCRLs) can be used to inform on the appropriate level of effort relevant under various conditions. An update to environmental dosimetry is presented in ICRP Publication 136 (Ulanovsky *et al.* 2017) wherein dose coefficients, DCs, are rendered compatible with the most up-to-date radionuclide database, an extended range of organisms and locations is covered and an alternative methodology for considering the dose contributions from progeny in decay chains is elaborated. Finally, in Publication 148 (Higley *et al.* 2021) the Commission provides radiation weighting factors for alpha and low beta (tritium) radiations for RAPs. The basic outline of the approach is illustrated in Figure 34.

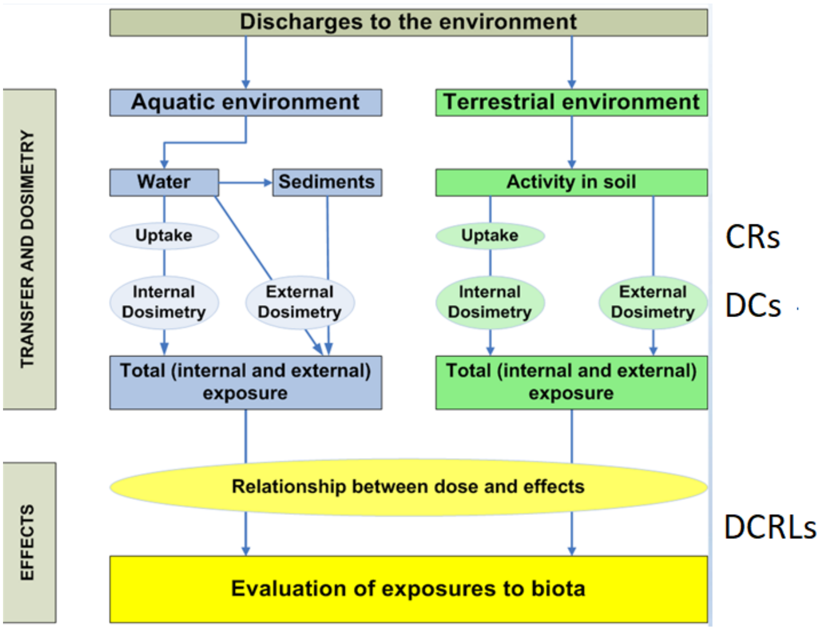


Figure 34. A schematic is showing the basic steps in an environmental radiological impact assessment according to the ICRP framework.

As mentioned in the introduction, the ICRP Publication 142, focused on RP from NORM in industrial processes, recommends using the ICRP approach for RP of the environment (use of RAPs, representative organisms and DCRLs) to the NORM case. The Commission indicates that, in the context of NORM, it is necessary to stay pragmatic in the application of these approaches and to consider radioactive and non-radioactive risks (usually NORM industries also produce non-radioactive contaminants).

4.2.2 IAEA Standards and consideration of radiological protection of the environment

For the explicit evaluation of RP of the environment, the IAEA provides in its Standard GSG-10 "Prospective Radiological Environmental Impact Assessment for Facilities and Activities" an example of a generic methodology for assessing exposure of flora and fauna, i.e. in the context of planned exposure situations applying the approach of the ICRP (concepts and use of RAPs, DCRLs, and Representative Organisms) (ICRP 2008). This integration of flora and fauna protection within the human RP can be considered, in practical terms, assuming a linkage in the exposure scenarios. The figure below, reproduced from Telleria *et al.* (2015) shows a scheme for estimating and evaluating exposures to humans and biota, considering the three exposure situations (i.e. planned, existing, and emergency) (Figure 35). The assessment of exposures to humans and flora and fauna is based on measured or estimated radionuclide activity concentrations in the environment. Using this approach, the managerial decisions related to the environment should be made based on considerations for the protection of humans and non-human-biota in an integrated manner.

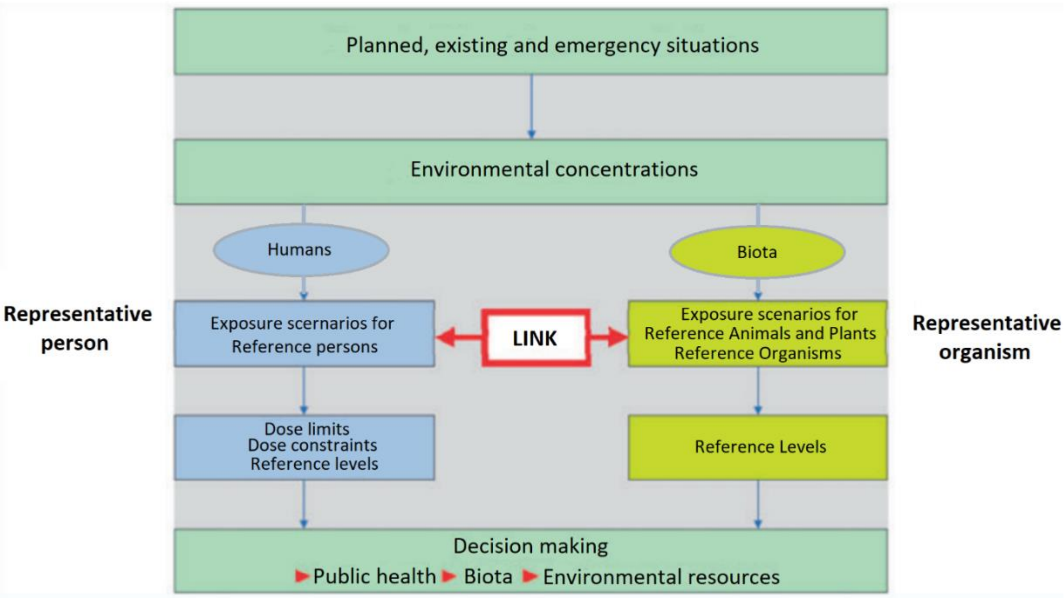


Figure 35. IAEA framework for impact assessment to non-human biota. Reproduced from (Telleria *et al.* 2015).

There is a general requirement to ensure that the application of the system of radiological protection is commensurate with the radiation risks associated with the exposure situation (IAEA 2018; ICRP 2006). This requirement extends to the level of detail and resources required in the assessment of doses associated with planned exposure situations (IAEA 2001; 2018). The standard approach for a radiological environmental risk assessment is to implement a tiered or graded approach. This can be evidenced in the graded approach reinforced by the US Department of Energy (USDOE 2002) and the ERICA integrated approach (Brown *et al.* 2008, 2016; Larsson 2008) which both adopt this methodology in determining the risk of harm to the environment arising from planned or existing exposure situations.

Further practical guidance for an international audience on components of a radiological assessment system for the environment has been provided or is under development by the IAEA (e.g. update of SRS 19). In publication *TRS-479* (IAEA 2014), the IAEA provides a statistical compilation of radionuclide CRs for common groups of organisms in terrestrial, freshwater and marine environments. As such, this data compilation may be seen as a broad extension of the above-mentioned ICRP Publication-114 to many wildlife groups beyond the quite limited suite of ICRP

RAPs. For the marine environment, IAEA TECDOC-1759 (IAEA 2015) provides guidance on integrated assessments elaborated upon the application of ICRP RAPs and associated parameters CRs and DCRLs. IAEA TECDOC-1759 provides a practical guidance regarding how to assess doses to workers and members of the public and adds a similar approach for assessing doses to marine flora and fauna. The publication was expected to be used mainly by national regulatory authorities responsible for authorising disposal at sea of candidate materials, as well as those companies and individuals applying to obtain permission from such authorities to dispose of these materials at sea. It also provides guidance to national radiological protection authorities who might become involved in determining whether candidate materials can be designated as *de minimis* for the purpose of the London Convention and Protocol.

IAEA will deliver in 2023 the new SRS methodology which will include guidance on conducting impact assessments for the environment including routine releases involving NORM.

4.2.3 European approach for radiological protection of the environment

In Directive 2013/59/Euratom of the European Union (European Commission 2014), it is stipulated that the recommendations of the International Commission on Radiological Protection (ICRP) will be followed, in particular those of Publication 103 (ICRP 2007) which, as noted above, include among the objectives of the radiological protection system the protection of the environment. In addition, the Directive states that "…. Given that the state of the environment can affect human health in the long term, a policy is needed to protect the environment against the harmful effects of ionising radiation", in addition to requiring environmental criteria for the protection of human health to be based on internationally recognized scientific data, such as those published by the European Commission, ICRP, UNSCEAR and IAEA. In the 2000s, Europe started to develop an approach for the radiological protection of the environment. The approach was finalised in the ERICA project (Beresford *et al.* 2007) building on the earlier FASSET (Larsson *et al.* 2004) and EPIC (Brown *et al.* 2003) projects. Furthermore, the application of the ERICA approach within a regulatory context was addressed in the PROTECT project (Copplestone *et al.* 2010; Howard *et al.* 2010).

Similar to the ICRP approach, the ERICA integrated approach is based on the use of reference organisms and the development of related parameter values, including radionuclide transfer factors and dose conversion coefficients based on occupancy factors and representative geometries. ERICA is a graded approach, which allows optimizing resources used for the radiological protection of the environment.

The ERICA integrated approach consists of three phases:

- i) management, applied in general terms for the decision-making process before, during and after the evaluation;
- ii) assessment, in which, by defining the exposure conditions, the exposure of the biota is estimated, and the radiation dose rates to the selected fauna and flora are calculated;
- iii) risk characterization, which includes the estimation of the probability and magnitude of the effects on biota, as well as the determination of the associated uncertainties.

As previously mentioned, the ERICA approach uses Reference Organisms defined as "a series of entities that provide a basis for estimating the radiation dose rate to a series of organisms that are typical or representative of a contaminated environment. These estimates, in turn, would provide a basis for evaluating the probability and extent of radiation effects." This approach is consistent with the ICRP approach.

In the ERICA project, dose rate reference values were derived, using a method that followed the recommendations of the European Commission (EU 2003b) for the estimation of the predicted no effect concentration (PNEC) for chemicals. For this, reliable (i.e. quality assured) data on effects on mortality, morbidity and reproduction collected in the FREDERICA database were used. The ERICA screening value is 10 µGy h^{-1}. The ERICA integrated approach has been implemented in the ERICA tool (Brown *et al.* 2008), which allows evaluations to be carried out at three levels with different degrees of conservatism.

4.2.4 Approach in the United States for radiological protection of the environment

The US Department of Energy in a technical standard of 2019 provides methods, models, and guidance to characterize radiation doses to aquatic and terrestrial biota within a graded/stepwise approach to protect animal and plant populations from harmful effects of anthropogenic ionising radiation (USDOE 2019). This approach follows the regulations of Order 414.1D and Order 458.1 of the DOE (USDOE 2020).

The stepwise approach has been implemented in the RESRAD-BIOTA code (USDOE 2004), specifically designed to complement the application of Biota Concentration Guides (BCG) contained in the aforementioned technical standard. In this methodology, aquatic and "coastal" animals and terrestrial biota (animals and plants) are considered as reference organisms, defining reference dose rate values for each group, which allows demonstrating that populations of animals and plants are adequately protected from the effects of ionising radiation:

- ≤10 mGy d^{-1} for aquatic animals or for terrestrial plants;
- ≤1 mGy d^{-1} for terrestrial and “coastal” animals.

These reference dose rate values are generally consistent with the DCRL bands for RAPs described by the ICRP (ICRP 2008).

The stepwise approach to assessing radiation doses to biota consists of three stages that guide the user from an initial conservative assessment to a more rigorous site-specific analysis when necessary:

1. Data collection, data from the environment is gathered defining the evaluation area (source terms, receptors, routes of exposure, concentrations of radionuclides in water, sediments and soil for subsequent screening).
2. Screening the maximum concentrations of the radionuclides measured in the medium are compared with the Biota Concentration Guides (BCG), which represent the limit concentration for each radionuclide that does not exceed the recommended dose rates for the biota in between. To carry out and document this phase, it is recommended to use the RESRAD-BIOTA tool, carrying out a level 1 evaluation in a terrestrial or aquatic ecosystem, with the maximum concentrations of each radionuclide.
3. Analysis. This stage consists of three steps, each time more detailed:
   i. Site-specific screening. This is carried out at level 2 of the RESRAD-BIOTA evaluation and involves the use of more realistic and representative bioaccumulation factors at the site of interest. Means of radionuclide concentrations may be used instead of peak values, considering the temporal and spatial dependence of contamination.
   ii. Site specific analysis. In this case, level 3 of the RESRAD-BIOTA evaluation is used with kinetic modelling applicable to riverside and terrestrial animals. The kinetic model employs allometric equations that relate body mass to parameters such as food/soil consumption rate, inhalation rate, lifetime, biological elimination rates.
   iii. Actual site-specific biota dose assessment, involving the collection and analysis of biota samples and subsequent assessment at level 3 using RESRAD-BIOTA.

For the application of the graded approach, it is emphasized that care must be taken if the possible radiological impacts on "endangered, threatened, rare or sensitive species" or commercially or culturally valued species are to be evaluated, since then the screening methodology may not provide correct answers to the questions being addressed.

4.3 Approaches to tackle the challenges, i.e. integrated assessment, population modelling, multi-contaminants

4.3.1 Integrated assessment for human and non-human-biota

There is a complete and relevant review of an integrated assessment for human and non-human biota by Vandenhove *et al.* (2018). The information below is primarily, but not exclusively, derived from this publication.

There are various differences between RP assessments for human and non-human-biota. First of all, the protection targets are different: for humans deterministic (tissue reactions) and stochastic effects on individuals are considered, whereas for non-human-biota, the target is the population, not the individuals, and therefore the relevant biological effects are those related with population viability (early mortality, reproductive impairment (fertility or fecundity) and morbidity (effects that can reduce the “fitness” of the animals and plants)).

Both human and environmental RP approaches, as promulgated by the ICRP, are based around points of reference in the form of the *Reference Person* and *RAPs*. For non-human-biota reference dosimetric models and reference data sets, together with what is known about the effects of radiation on these, or similar, types of animals and plants are considered. Physical radionuclide transport models, such as those describing the advection and dispersion of radionuclides, may be characterised as being independent of receptor and can, therefore, be the same for human and biota assessments. For example, the models from IAEA SRS-19 (IAEA, 2001), originally developed for human assessments, are implemented in the ERICA tool for non-human biota assessments (Brown *et al.* 2008). Biological transfer is also treated differently. For the human approach, elaborated biokinetic models are used to simulate the intake route and retention of a given radionuclide within a conceptualised reference person (Brown *et al.* 2014). For example, for an (oral) ingestion of a given radionuclide, *inter alia*, specific compartmental models are used, to simulate the retention within and depuration from the body over protracted timescales. The transfer to given organs is not normally reported explicitly, rather the biokinetic information is coupled to various components, such as dosimetric models, radiation, and tissue weighting factors to provide coefficients mapping an intake of activity (in Bq) onto an effective committed dose (in Sv). In contrast, transfer to non-human biota are normally derived explicitly though the application of whole-body to media concentration ratios (Brown *et al.* 2014). In fact, there are parallels here with human radiological assessments in the consideration that such transfer factors, i.e. concentration ratios, are applied in deriving radionuclide activity concentrations in plants and animals being consumed by people although the specific, underlying datasets may differ.

The general approach to estimate the dose (corresponding to biological harm) is therefore somewhat different for humans (where effective doses are normally determined) and non-human-biota (where weighted, absorbed doses are normally determined). Although similar radiation transport modelling approaches underpin determinations of absorbed dose and account is taken of the different qualities of radiation in causing damage for both categories of assessment, individual organ (equivalent) doses and the overall risk of detriment (committed effective dose) is uniquely calculated for humans.

More complex phantom models for non-human-biota are being developed (e.g. Caffrey and Higley 2013; Montgomery and Martinez 2020) congruent with those commonly applied for human dosimetry. However, such approaches are unlikely to be used in regulatory tools like ERICA because voxel models are unnecessarily complex (normally used to derive information at the organ level) and provide less conservative estimates of exposure than ellipsoids in the limited cases where such comparisons have been made (Ruedig *et al.* 2015).

The approach for assessing what calculated doses actually mean differs between human and environmental assessments. Human radiological protection is more elaborated relying on a system of interpretation, in many countries, based around dose limits, constraints and reference levels. Dose limits (and constraints) are often embedded within radiological protection regulations and have substantive legislative weight (e.g. they are used to set authorised discharge levels). In contrast, the benchmarks and DCRLs for non-human, with some notable exceptions (USDOE 2002), are not considered as dose limits and have been used more as a form of guidance to inform the potential for environmental impact. Examples include dose rate benchmarks of 40 µGy h^{-1} for terrestrial animals and 400 µGy h^{-1} for terrestrial plants and all aquatic species proposed by IAEA (IAEA 1992) and the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 1996) below which exposures are unlikely to be harmful to biota populations. The dose rate of 10 µGy h^{-1} used in the ERICA Tool is a no-effect benchmark used to screen out, i.e. remove from further assessment, those cases for which exposures can safely be assumed to be trivial. The DCRLs proposed by ICRP (2008) could provide some context to help an assessor make decisions with regard to optimisation of effort to be expended for protection of the environment.

4.3.2 Multi-contaminants analysis on the context of NORM

Radionuclides are commonly found in the environment together with other contaminants including but not limited to acidic or alkaline materials, heavy metals, hydrocarbons and pesticides. Multiple contaminant issues have been discussed for decades, but the number of published articles on mixtures, where at least one contaminant is ionising radiation, is relatively scarce (e.g. Vanhoudt *et al.* 2012) and international organizations, such as ICRP and IAEA, although highlighting the need to consider the risk of ionising radiation together with that of other contaminants present in the environment, do not provide guidelines on how this risk assessment should, in practice, be done.

There are many regulations for the protection of the environment (quality of air, water and soil) and the radioactivity present in the environment may not be the dominant issue.

Effects induced by a combination of contaminants can differ from the sum of the individual effects and compounds can exert effects in mixtures at concentrations where the single contaminants do not show effects. Cumulative effects can be additive, synergistic or antagonistic.

Interactions between pollutants in a mixture may occur at three levels: (1) Pollutants may influence each other's mobility in the environmental media and hence, each other's availability to organisms; (2) different pollutants may block or enhance each other's uptake into the organism; (3) once inside the organism, pollutants may block or enhance each other's detoxification, alter the nature of their toxic actions, and/or impact repair capacities. *The Biotic Ligand Model (BLM)* (Paquin *et al.* 2002) has been proposed as a tool to quantitatively predict the manner in which water chemistry affects the speciation and the biological availability of metals in aquatic systems.

Based on the dose-response curves and effect concentration (EC)-values of the individual toxicants, two generally accepted reference models *"Concentration Addition" (CA)* and *"Independent Action" (IA)* can be used for the prediction of the combined toxicity effects. The CA model (Loewe and Muischnek 1926, Loewe 1927) can be used for mixtures of toxicants that exert effects on similar toxicological processes (similar modes of action). The concept of IA assumes that the compounds of a mixture act on different physiological systems within the exposed organism. The CA and IA models only provide a description of the data and they do not consider interactions between co-contaminants that may affect the mobility, absorption, distribution, storage, biotransformation and elimination of other contaminants.

There has been great success in predicting the combined effects of chemical contaminants. However, the studies assessing the combined toxicity of ionising radiation with other contaminants using CA and IA approaches are scarce (Vanhoudt *et al.* 2012). From this literature review it could be observed that conclusions and terminology used in the papers should be considered with care since different wording can mean the same thing (e.g., no positive or negative interaction in CA-simple additivity- could be considered by authors evaluating the effect of two components in the mixture as synergistic or potentiating). The analysis of the data from literature showed that 58% of the interactions were considered positive (any effect that is higher than the effect induced following exposure of the single contaminant), 26% negative and 16% showed no interactions. In many cases, the interactions observed were dependent on dose or concentration, composition of mixture, life stage, exposure time, and endpoints studied. Hence, interactive effects are quite unpredictable but are clearly common.

Salbu *et al.* (2019), in a review paper on the multiple stressor concept, highlight that in mixed contaminated areas where organisms are exposed to multiple stressors (such as radionuclides, trace metals, organic compounds, non-ionising radiation), biological uptake and accumulation in organisms will depend on the interaction between stressors (toxicokinetics), while biological effects will depend on the competition between stressors to interact with key biological targets and interactions between different toxicity pathways leading to an adverse effect (toxicodynamics). Thus, not only the concentrations of the stressors, but also the physico-chemical forms of the elements released will be essential in the effect that will be caused. The authors state that although the number of articles dealing with multiple stressors where ionising radiation act as one of the stressors is limited, deviation from concentration (dose) additive responses should probably be expected, especially under field conditions where additional abiotic as well as biotic interactions take place. Uncertainties in the risk assessment of multiple contaminants could be reduced by using advanced technology to improve source characterisation (inputs for transport, dose, and impact models) and understanding of how exposure to multiple stressors affects toxicokinetics and toxicodynamics to cause deviation from additivity along the source-adverse outcome sequence.

Recently the ICRP set a working group (Task Group 125) to analyse possible benefits in incorporating *ecosystem services monitoring and assessment* in many contexts related to environmental protection and policy making. Ecosystem services are defined as the benefits humankind derives from the workings of the natural world, i.e., from ecosystems, and are crucial in human life for example by providing nutritious food and clean water; by regulating disease and climate; by supporting crop pollination and soil formation, and by offering recreational, cultural, and spiritual benefits. The assessment of ecosystem services, whether those services are regulating, supporting, provisioning, or cultural, have the potential to support a holistic approach to environmental radiological protection as developed in ICRP Publication 124 (Pentreath *et al.* 2014). The ICRP aims at providing recommendations concerning whether and how ecosystem services should be used in relation to the Derived Consideration Reference Levels (DCRLs).

4.4 Overview of existing models suitable for assessing exposure and risks to non-human-biota

There are various risk assessment tools (software packages) that have been used for performing environmental risk assessments (or components thereof) such as ECOMOD, EDEN 2, ERICA, K-Biota, LIETDOS-BIO, R&D 128 and RESRAD-BIOTA, SADA and PC-CREAM08 (for biota). The list is not exhaustive but covers the most widely used software and gives an indication of what is available.

A brief description of each model is shown in Table 21.

Table 21. Overview of existing models suitable for assessing exposure and risks to non-human-biota.

Model	Comments	Availability/Documentation
ECOMOD	Uses stable chemical analogues and ratios of radionuclides to determine the concentrations of radionuclides in aquatic biota and, as such, does not extend to terrestrial ecosystems. Nonetheless, the model is dynamic and can be used to make predictions under conditions of changing radionuclide concentrations. The model is particularly suited to radionuclides that are analogous to, or isotopes of, biologically active chemical elements. Concentration ratios, predominantly sourced from the Russian language literature, have also been used. ECOMOD employs previously published absorbed fractions across a range of ellipsoids to estimate unweighted absorbed doses for a limited number of radionuclides.	The software is not freely available. (Sazykina 2000)
EDEN 2	Elementary Dose Evaluation for Natural environment, with functions allowing for relatively complex dosimetric configurations but with less focus on environmental transfer and interpretation of dose-effects.	The software is not freely available. Code can be requested via: www.irsn.fr/EN/Research/ Scientifictools/Computer-codes/Pages/The-EDEN-computer-code-Elementary-DoseEvaluation-for-Natural-environment-2368.aspx (Beaugelin-Seiller *et al.* 2006)
ERICA Assessment Tool	It encompasses the entire impact assessment sequence from problem formulation and risk characterisation to dose calculation (weighted to account for the potential to cause harm of different radiation types) and interpretation of doses in terms of risk quotients and contextual information (e.g. link to dose-effects tables). The approach covers the 3 commonly considered ecosystem categories – terrestrial, freshwater and marine. Developments have continued to the present time: the latest version of the tool includes the updated dosimetry described in ICRP (2017) and draws upon the latest updates with regard to radionuclide transfer from the 'Wildlife Transfer Database'.	The software is freely available (as of March 2022, from https://erica-tool.com/) (Brown *et al.* 2008, Copplestone *et al.* 2013)

K-Biota	An approach developed in South Korea, which applies the methodology underlying the ERICA Tool for the dosimetric component. Plants and animals were selected based on ICRP recommendations, and the size of the selected organisms was determined from the corresponding Korean endemic species. External and internal dose conversion coefficients for the selected organisms and radionuclides were calculated by the uniform isotropic model or Monte Carlo simulation.	The software is not freely available. (Keum *et al.* 2011)
LIETDOS-BIO	Developed to address contamination issues associated with nuclear power production in Lithuania. The model used two CR databases: site-specific (used by preference) and generic (mostly based on Larsson et al. (2004) and data from the Russian language literature). A Monte Carlo transport code was used for DCC derivation. A bespoke method for describing phantoms allowed DCC values to be calculated for organisms of any size or shape.	The software is not freely available. (Nedveckaite *et al.* 2007)
PC-CREAM 08	Software developed by the UK Health Security Agency for the purpose of performing radiological risk assessments for routine and planned releases of radioactivity to the environment. The software comprises of several modules and allows, *inter alia*, the release and dispersion of radioactivity in the atmosphere and aquatic environments. Quite recently a new functionality has been added to the modelling suite to facilitate dose-rates to non-human biota to be determined. The module is heavily focussed on ICRP approaches drawing heavily on the transfer and dosimetric parameters given in the aforementioned ICRP report (108,114 and 136) and also DCRLs provided by the ICRP (in report 108 and 124).	The software is not freely available. (Anderson *et al.* 2022)

R&D 128 (England and Wales Environment Agency)	This model uses a similar approach to that of ERICA, although a smaller range of organisms and radionuclides is considered. DCCs are estimated using simple functions for energy deposition in a medium of unit density from point isotropic sources to represent the absorption of photons and electrons. Energy absorbed fraction functions are fitted separately for photons and electrons to provide a reliable interpolation between calculated values. These functions are then integrated numerically using a stochastic (Monte Carlo) algorithm to calculate the absorbed fraction. Guidance is provided on how to estimate CR values if they are missing for a given radionuclide–organism combination (later adapted for use within the ERICA approach).	The spreadsheet-based models, which this approach uses, are freely available (as of March 2022) from: https://wiki.ceh.ac.uk/pages/viewpage.action?pageId=115016183&msclkid=d8d698fbaaac11ec8ce310a95125316a (Copplestone *et al.* 2001, Copplestone *et al.* 2003)
RESRAD-BIOTA	The code was designed to be consistent with, and to provide a tool for, implementing the graded approach for biota dose assessment (USDOE, 2002). The code includes a kinetic-allometric approach (Higley et al. 2003) to estimate the transfer of radionuclides to animals. The internal and external dose conversion coefficients (DCCs; relating unweighted absorbed dose to media or biota activity concentrations) are estimated using a Monte Carlo transport code.	The software is freely available (as of March 2022) at https://resrad.evs.anl.gov/codes/resrad-biota/. Developments have continued to the present time.
SADA, Spatial Analysis and Decision Assistance	Developed for the United States Environmental Protection Agency and the United States Nuclear Regulatory Commission by the University of Tennessee. The tools within the SADA model include integrated modules for visualisation, geospatial analysis, statistical analysis, human health risk assessment, ecological risk assessment, cost/benefit analysis, sampling design, and decision analysis. Although primarily developed for non-radioactive contaminants, SADA can be applied to radioactive contamination for basic screening tier assessments by inclusion of USDOE biota concentration guidelines (BCG's) (i.e. predicted no-effects media concentrations). In effect this means that SADA can be used for assessments equivalent to Level 1 of the RESRAD-BIOTA package but with the added functionality of being able to consider the data within a spatial context.	A freeware version is available (as of March 2022) at: https://www.sadaproject.net/?msclkid=71bb4819aab111ecb1ba9a7918d6be5c.

In the broadest sense, all the above-mentioned assessment models, in part or in whole, adopt a similar conceptual approach wherein selected suites of non-human biota are represented by idealised geometries (often ellipsoids) and the assessment maps activity concentration (in these selected organisms and their environment) onto dose-rates that are interpreted by comparison with dose-effects relationships for these (or related) organisms. In other words, absorbed doses (weighted or unweighted to account for biological effect) constitute the metric by which risk is inferred (parallel to the radiological assessment approach for humans) and the approaches are reasonably congruent, at an overarching level, with the guidance given by the ICRP. Nonetheless, conceptual differences do exist when one considers the finer details of the models. For example, although many of the models (e.g. ERICA and EA R&D 128) adopt a steady-state transfer approach using the ratio of activity in biota to that in media, other models, notably ECOMOD, provide a dynamic approach whereby activity concentrations in (marine) biota are modelled via uptake and loss of radionuclides from the environment. RESRAD-BIOTA falls somewhere in between as, although a kinetic approach underlies many of the equations of transfer, this is normally used to derive lumped parameters that essentially equate to concentration ratios. There are also differences with regard to how spatial information is accounted for. Whereas most of the models are either set up to accept spatial information in a very rudimentary form, for example in ERICA one can simply enter location names, SADA allows for sophisticated geospatial analysis of datasets (albeit that the bespoke non-human biota exposure component of the SADA system is less well developed than other parts). There are also subtle differences in the dosimetric approaches employed. In some cases, as exemplified by EA R&D 128, functions are used to represent the energy deposition in matter by electrons and photons forming the basis of DCCs whereas for ERICA radiation transport codes were run, for selected geometries (in terrestrial ecosystems), and various interpolation fitting methodologies were applied to explore the effect of changing mass and shape of the concomitant ellipsoids. Nonetheless, all of the dosimetric approaches appear to be well-grounded in radiation physics and can be considered conceptually related. Commenting on whether the assessment of risk differs conceptually between approaches is problematic because, so few approaches address this issue in any depth. Where risk is determined, notably for ERICA and RESRAD-BIOTA, approaches are conceptually similar, this being achieved in a very simple way by dividing an exposure estimate based on measurement by an exposure benchmark. Finally, all the approaches are conceptually similar in the sense that the approaches were designed with planned and existing exposure situations in mind. The wide use of CRs is more amenable to steady state (underlying assumption in many models as noted above) and arguably only ECOMOD is suited to the consideration of an emergency situation albeit for the marine environment only. Nonetheless, with the publication of ICRP-124 (Pentreath *et al.* 2014) explaining how the system of environmental radiological protection may be extended to all exposure situations, efforts are afoot, notably for ERICA, to provide a dynamic transfer capability.

Some of the models listed above have been extensively employed in different parts of the world. Following its release, the ERICA Tool has been widely employed in various applications worldwide. Examples include: consideration of potential environmental impacts from deep geological disposal facilities in various European countries (e.g. POSIVA 2014; Robinson *et al.* 2010; Torudd 2010); scoping analyses in line with newly introduced environmental regulations (Hosseini *et al.* 2011); quantifying environmental impacts from operating and planned nuclear power stations (Li *et al.* 2015; Nedveckaite *et al.* 2011; Vandenhove *et al.* 2013); to derive radiological quality guidelines for Australian U mining sites (Doering and Bollhöfer 2016); assessments of the impact of near-surface radioactive waste repositories in Europe and Australia (Kabir *et al.* 2014), U mining impact assessments (ERA 2014); assessing releases from medical facilities (Carolan *et al.* 2011) and for exposure estimates for biota following accidents (Fuma *et al.* 2015).

RESRAD-BIOTA has been applied primarily for US based assessments and widely at USDOE sites but also elsewhere (e.g. Dieu Souffit *et al.* (2022) and R&D128 has also been applied in the implementation of EU's habitat's directive (Copplestone *et al.* 2003).

Many of the above-mentioned assessment tools have been included in international model-model inter-comparisons such as those undertaken under the auspices of the IAEA (see Beresford *et al.* 2008, Vives i Batlle *et al.* 2011).

Furthermore, some of the abovementioned models, but most extensively the ERICA Tool, have been used in inter-comparisons between model outputs and measurements made in the field. This has included:

- Dose estimates for small mammals exposed to elevated levels of (predominately) ^{137}Cs in the Chernobyl exclusion zone, where the correspondence between ERICA model predicted doses and doses measured using thermoluminescent dosimeters, given inherent uncertainties, was considered as being satisfactory (Beresford *et al.* 2008).
- Predictions, using different models, for naturally occurring radionuclide activity concentrations and dose rates arising from this for sites downstream of uranium mines and mills in northern Saskatchewan, Canada (Goulet *et al.* 2022). Various groups used ERICA extensively in this exercise. If anything, the inter-comparison showed that prediction using the same tool by different users can vary considerably owing to assumption taken and interpretation of scenario requirements.

4.5 Risk assessment for non-human-biota in the context of NORM

Each NORM-involving industry produces residues with activity concentration values that will depend on type of material involved, type of processing as well as natural variability of radionuclides present in the rock/raw material. The level and characteristics of the exposure will depend on environmental pathways involved and will of course depend on the type of industry as well as the technological processes implemented.

Some studies have made a generic appraisal of the limits associated with environmental impact assessments for NORM and the challenges encountered by practitioners in this field. For example, Michalik *et al.* (2013) considered some of the unique challenges associated with assessing environmental assessments for NORM compared to industrial anthropogenic sources. Of particular concern were the poorly elaborated regulations for NORM at the time of writing, noting that dramatic improvements have not been made in the last decade since this study was completed. Inconsistencies were apparent with regard to how regulations were applied dependent on the type of NORM material involved or the nature of the practice. The properties of NORM materials, often with concomitantly large volumes covering extensive areas and the complexity of radionuclide decay, ingrowth and divergent geochemistry, leading to fractionation of radioisotopes in long decay chains openly exposed to environmental processes (see Figure 36), created problems not normally encountered for anthropogenic wastes. Although some of these issues, such as consideration of long decay chains and varied geochemistry exist for industrial nuclear fuel cycle bi-products, these materials are normally closely controlled and highly contained. Michalik *et al.* (2013) provided no specific environmental assessment methodology, *per se*, but considered that the reductionist reference framework, recommended by the ICRP and others, might require supplementation with other approaches for NORM to account for a complex multi-stressor situation. Furthermore, the requirement to account for the interactions within ecosystems and insights into effects over a broad spectrum, covering many different levels of biological organisation, was considered important.

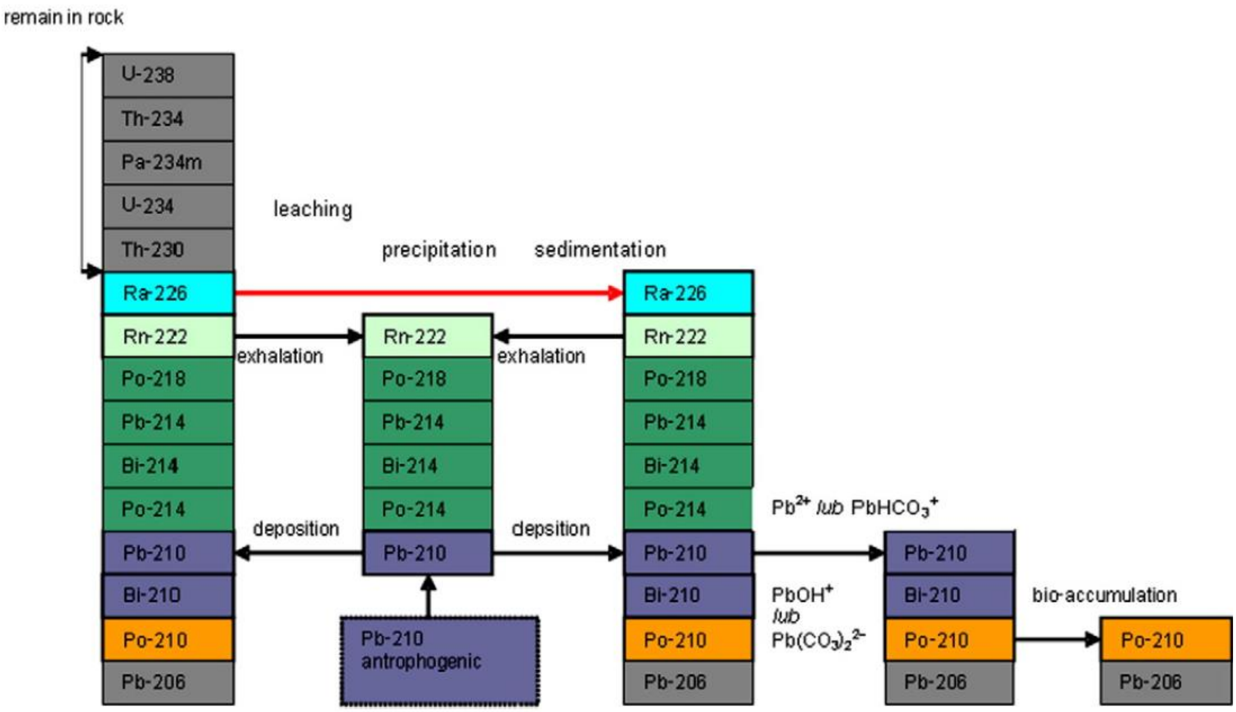


Figure 36. Fractionation and migration possibilities within uranium decay series; reproduced from (Michalik *et al.* 2013).

4.5.1 Literature with focus on NORM and impact on marine ecosystem

In the Northern European marine environment, radioactivity associated with the discharge of NORM from the oil and gas industry has received substantial attention owing to the potentially large inputs of radioactivity from this source as highlighted in the MARINA II study (EU 2003a) and the implementation of OSPAR Radioactive Substances Strategy which aimed at achieving 'concentrations in the environment near background values for naturally occurring radioactive substances'. The main releases of NORM are in the form of produced waters and a comprehensive overview of this subject can be found in Hosseini *et al.* (2012). The main findings of this review were that, in the few studies where the impacts from produced water in northern European waters were determined, the excess levels of NORM above background levels were not substantial and that environmental risks were correspondingly low. However, uncertainties associated with published assessments were considered to be large, noting that the multi-stressor context may be important, with chemicals, such as scale inhibitors, potentially influencing NORM radionuclide uptake to marine organism.

A review on the ecological effects associated with oil and gas installations was made by MacIntosh *et al.* (2022). The overview provided insights into the current level of knowledge with regard to the exposure of marine organisms to NORM and other stressors (organic chemicals and heavy metals). It was apparent that although some basic information exists, for example limited studies have been performed on uptake of and effects from selected radionuclides relevant to the oil and gas industry and initial assessments have been undertaken, there are still substantial limits to our understanding. MacIntosh *et al.* (2022) noted that the limited number of reviewed studies associated with the oil and gas industry where dose rates in exposed organisms had been determined did not indicate detrimental effects (including mortality, reproductive capacity or morbidity). Nonetheless, the uncertainties associated with such assessments were extremely large and no definitive conclusions regarding the potential impacts, in a more generalised sense, could be drawn. The authors recommended the need for future research on characterising NORM scale and assessing effects of scale to marine organisms through direct organism exposure experiments. Emphasis was also placed upon the need to incorporate ecotoxicology into environmental risk assessment for offshore petroleum decommissioning.

The impact of the metal mining industry on marine non-human biota in the North Aegean Sea, through monitoring the levels of heavy metals and radionuclides in sediments, was considered by Pappa *et al.* (2016). The concentrations of selected metals (As, Zn, Pb and Mn) were found well above guidelines set by various national (Australian, American and Canadian) organisations (regulators and councils), indicating at least a moderate impact from anthropogenic activities in the area. In contrast, the risk associated with radiological impacts was considered to be minimal with dose-rates falling several orders of magnitude below the benchmarks used by the IAEA and UNSCEAR (i.e. 400 µGy h^{-1}). The authors of this study opted to not use the ERICA screening benchmark although no explanation was given as to why they decided to do so. Heavy metals were considered to be more hazardous than radionuclides in this study although no numerical quantification of risk for the former was provided. The authors recommended the inclusion of ^{40}K in this type of assessment, as it may contribute substantially to external dose, although guidance on defining the incremental (above background) component was not provided.

Abbasi *et al.* (2022) estimated the radiological risk of natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) to marine ecosystem biota for a study site in Cyprus, the Mediterranean Sea using the RESRAD-BIOTA code. All estimated dose-rates to selected aquatic and riparian animals were low, falling substantially below applicable protective benchmarks and leading the authors to the conclusion that there would be no significant radiological impact.

4.5.2 Literature with focus on NORM and freshwater ecosystem

Specific modelling approaches have been applied to selected scenarios for freshwater ecosystems. Goulet *et al.* (2022) presented an inter-comparison exercise using some of the abovementioned models, notably RESRAD-BIOTA, ERICA and R&D128, for a site affected by uranium milling and mining. The exercise demonstrated that variability in model predictions for freshwater organisms could be extremely large and that models often poorly predicted measured NORM activity concentrations. Predictably, given the highly conservative nature of some model output, dose rates exceeded benchmarks by some margin in numerous cases. As noted earlier for terrestrial systems this, *per se*, does not indicate the presence of inevitable environmental impact and Goulet *et al.* (2022) provided no comment on this exceedance issue. More oppositely, much of the variability seemed to be introduced by an unconsidered selection of CR and K_d values and, with regards exposure estimates, differences in the way decay chains were treated. In many cases, assuming secular equilibrium between radionuclides along a given decay chain is not appropriate. The role of water chemistry, in terms of its influence on the transfer of NORM radionuclides, is often not given the consideration it should for such assessments. For this reason, the authors recommend a standardised best-practice approach to calculate weighted absorbed dose rates to freshwater biota to be applied when a facility is at the planning, operating or decommissioned stage. At the initial planning stage, the recommendation is to use conservative site-specific baseline activity concentrations in water, sediments and organisms and predict conservative incremental activity concentrations in these media by selecting concentration ratios based on species similarity and similar water quality conditions. At the operating and decommissioned stages, the recommendation is to rely on measured activity concentrations in water, sediment, fish tissue and whole-body of small organisms to further reduce uncertainty in dose rate estimates.

As part of an effort to establish whether radionuclides should be categorised as chemicals of mutual concern (between Canada and the USA) for the Great lakes, CNSC (2018) performed an ecological risk assessment for nuclear power plants and nuclear processing and waste-management facilities.

Although these assessments, using both RESRAD-BIOTA and ERICA, indicated no evidence that anthropogenic radionuclides within the Great Lakes posed currently an unreasonable risk to the environment, the calculations appeared not to include NORM in the environmental dose-rate estimates. This presumably reflected the conclusions drawn elsewhere in the CNSC (2018) report that there were no significant industrial activities involving NORM, on the Canadian side of the Great Lakes basin that would result in a measurable effect on water quality or the ecology of the Great Lakes.

4.5.3 Literature with focus on NORM industries and impact on terrestrial ecosystem

Both RESRAD-BIOTA and the ERICA approach were applied by Ćujić and Dragović (2018) in performing a radiological environmental impact assessment for a site contaminated by NORM emissions from a coal-fired power plant in Serbia. Differences in the resultant dose-rates from the two approaches could be seen but this was considered to be mainly attributable to the transfer parameters selected for use in the two models. For the ERICA tool, substantially elevated dose-rates, uniquely exceeding the 10 µGy h^{-1} benchmark, were observed for the lichen and bryophyte category although the authors highlighted the questionable relevance of a soil to organism transfer parameter for an organism deriving most of nutrients from the atmosphere. Furthermore, the limited dose-effects data available for lichen and bryophyte suggested that they were relatively radioresistant and, given the substantially lower dose rates for other biota categories, the authors were able to infer confidently that there would be no significant radiation impacts from the coal fired power plant on terrestrial biota.

Nisti *et al.* (2022) considered the case of applying phosphogypsum (PG) as a soil conditioner. The ERICA Tool at tier 1 was used to determine risk coefficients (RQ). RQs associated with pure PG stacks were compared with soils amended with PG. Similar comparisons were made for leachates. Unsurprisingly, the RQs associated with PG amended soil and leachates were lower than those associated with PG. Nonetheless, the RQ associated with soils did not change drastically when PG was increased by a factor of 10 (probably reflecting the relatively small amounts of PG that are actually added). In fact, the soils with and without added PG did not exhibit very different RQs (the authors using this to say that the addition of PG was radiologically acceptable). It is of note that lichen/bryophyte again dictates the Tier 1 RQ often constituting the limiting organism for the selected NORM radionuclides. The paper is a novel application of ERICA but it seems unconvincing to argue that adding PG soil amendments somehow reduced risk. It is unclear that the complete removal of a PG stack to use as soil amendment would be acceptable and/or congruent with existing guidance/regulation.

Jannik *et al.* (2018) performed a biota dose assessment of small mammals sampled near several uranium mines in Northern Arizona. The authors used the RERAD-Biota code and calculated the dose-rates associated with isotopes of U, Th and ^{226}Ra. Estimates from the study showed that the potential biota doses were below the USDOE's biota dose standard of 1 mGy d^{-1}, indicating that population impacts would not be expected. In an article derived from the aforementioned report, Minter *et al.* (2019) refined further their original analysis by considering radionuclide enrichment. The results suggested that ^{226}Ra was more mobile than U and Th isotopes in the studied environment and bioaccumulated in the specified rodent species (e.g., in bones via the bloodstream). ^{226}Ra was also considered to be the radionuclide of most concern for rodent exposure and health.

A radiological impact assessment associated with contaminated soil from mining activity (Co-Ni deposit) in Eastern Cameroon was performed by Dieu Souffit *et al.* (2022). The assessment considered both impacts to plants and animals, using the RESRAD-BIOTA code, and to humans using RESRAD-ONSITE employing the same input datasets (measurements of selected NORMs in soils) and as such might be considered to be an integrated assessment. The dose rates derived for plants and animals fell substantially below the dose-rate benchmarks adopted from the USDOE, i.e. (1 mGy d^{-1} and 10 mGy d^{-1} for animals and plants, respectively). The use of the RESRAD-ONSITE code showed how the importance of different pathways for human exposure could be evaluated (direct external exposure was found to be important here cf. ingestion, radon exposure etc.) somewhat in contrast to the biota assessment which is limited to internal vs external. Furthermore, RESRAD-ONSITE allowed the users to explore the effect of remediation on human doses in the form of adding a cover layer to the deposit. How to provide a similar evaluation for non-human biota is not immediately clear, i.e. models don't exist in RESRAD-BIOTA or ERICA to consider the effect of shielding of the source on external doses to organisms.

An environmental risk assessment for a high background radiation area - Orrefjell in Norway – was undertaken by Maina (2018). A suite of naturally occurring radionuclides were determined from the ^{238}U decay series and ^{232}Th along with the heavy metals As, Cd, Cr, Cu, Ni, Pb, and Zn so that something might be inferred regarding the multi-stressor context. Calculated dose rates were substantially elevated although some reduction in estimated dose-rate was observed for some reference organisms (grass and herb but not shrub) if dose-rates were based on measured activity concentrations in the vegetation, *per se*, as opposed to CRs. Nonetheless, the ERICA screening benchmark of 10µGy h^{-1} was exceeded by some margin (registering several hundred µGy h^{-1} for some biota types) with the most elevated exposures observed, once more, for lichen/bryophyte. The transfer of NORM to this organism group may be an issue requiring follow-up, should developments (in ERICA) be requested. Secular equilibrium in the ^{238}U decay was clearly not present suggesting the need to, as far as possible, treat decay series members separately (in line with ERICA methodological recommendations).

Petrović *et al.* (2018) used the ERICA approach to calculate dose rates to plants and animals in Serbia arising from the presence of naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K). The range in dose rates was substantial spanning a range from <0.1 µGy h^{-1} (tree) to 5 µGy h^{-1} (lichen/bryophyte) although the latter value seems high for a natural background site and might reflect the application of default (natural decay series) CR values (some of which are known to be high for lichen/bryophyte). The authors consider their work to constitute a baseline study to establish whether anthropogenic activities will influence (human and environmental) exposures. Few conclusions can therefore be drawn in relation to application of environmental impact assessments at a NORM industrial complex.

Using the ERICA Tool, an evaluation of the potential radiological impact on non-human biota was performed in the NORM scenario of the Huelva phosphogypsum ponds (in the South of Spain), where the main radionuclide to consider is ^{226}Ra and its descendants. Two exposure situations were considered: (i) the biota lives on top of the pond; and (ii) the biota is 3 km from the pond. The results of these evaluations showed that the models used in ERICA (those included in the IAEA SRS-19) are not suitable for situations where large accumulations of NORM waste materials are found, as the models and approximations are extremely conservative. In these situations, it would be necessary to apply more refined models that are not currently programmed in ERICA (Mora *et al.* 2015).

4.5.4 Literature with focus on NORM but covering multiple ecosystems

In other cases, both terrestrial and aquatic systems have been studied in the same article. Vandenhove *et al.* (2015) performed environmental risk assessments (ERA) for environmental contamination resulting from the activities of 5 phosphate fertiliser plants (located in Belgium, Spain, Syria, Egypt, Brazil), a phosphate-mine and a phosphate-export platform in a harbour (both located in Syria). The study was designed to test the initial screening tier of the ERICA Tool and involved the collation of NORM activity concentration data for the aforementioned sites. It was found that RQ exceeded 1 in many cases leading to the recommendations that more detailed studies would be required in most cases. The authors considered that this may include (after selection of reference organisms more appropriate for the site studied) an ERA based on less conservative/ more realistic assumptions and more in-depth site monitoring for environmental concentrations or radionuclide concentrations in biota.

It was also considered that there was a further requirement, at the time of writing, to have more detailed information on effects at the dose rates of concern and that information on the environmental impacts of non-radioactive substances in the field would also provide useful context. The requirement to characterise incremental dose above background was discussed and led to the view that well defined local/regional levels of NORM would ideally be required to evaluate this credibly.

ERICA was applied by Hosseini *et al.* (2012) to selected NORM contaminated sites allowing dose-rates for a suite of organisms to be contextualised in terms of various screening and protective benchmarks. The study encompassed a phosphogypsum disposal site and settling ponds associated with mining in Poland and a legacy mining site, Søve/Fen, in Norway. The study allowed the more important radionuclides for environmental exposure to be revealed (e.g. ^{226}Ra was identified as being important for internal exposure at all sites) and the most exposed organisms (lichen and bryophytes) to be identified. The ERICA screening benchmark was exceeded in numerous cases but, in congruence with other assessments where exceedance has been an issue as note above, this did not lead to an immediate inference of radiological impacts on plants and animals.

The ERICA methodology was applied by Gomina *et al.* (2021) to both terrestrial and aquatic environments at a site in Nigeria focusing on natural radionuclides (^{226}Ra and ^{232}Th). Radionuclide activity concentrations in the environment had been compiled previously and the authors used maximum measured levels as input to ERICA to ensure conservatism. Some of the calculated dose-rates for the terrestrial environment (namely for lichen/bryophyte) and aquatic environments (including insect larvae, molluscs and zooplankton) substantially exceeded the ERICA screening benchmark of 10 µGy h^{-1}. The authors considered that their study could provide baseline data for the assessment of possible anthropogenic enhancement. It is challenging to draw any inferences beyond this consideration by the authors other than noting that substantial dose-rates can be derived, for natural environments, by simply selecting maximum environmental activity concentrations and applying default CRs. It should be noted that the methodological advice given for Tier 2 assessments in ERICA is to apply arithmetic mean values for media radionuclide activity concentrations.

4.5.5 Summary

In summary, it is challenging to say anything definitive about the potential impacts associated with particular industries. For the terrestrial environment, by way of example, it seems evident that environmental dose benchmarks have been exceeded in *ad hoc* assessments for some industries (e.g. coal-fired power plants and phosphogypsum stacks) but this does not immediately translate into the expectation of environmental impact from radioactivity. The choice of benchmark is of course important (ERICA screening vs RESRAD-BIOTA's population protective benchmark) and the arguably overly conservative radiological risk predictions for some organism groups (notably lichen and bryophytes) needs to be considered. It should, furthermore, be noted that similar issues concerning the exceedance of environmental benchmarks can be observed at high background radiation areas, which may be used to contextualise NORM industrial impacts.

Similarly, for marine and freshwater systems, the limited number of studies renders conclusive views on the potential impacts of industries difficult. There seems to be some evidence to suggest that environmental radiological impacts from mining and the phosphogypsum industry are possible at some locations but that the large uncertainties involved in modelling risk and the multi-stressor context for these exposure situations means that follow-up research would be required to concretise these impressions.

4.6 Conclusions

In conclusion, the authors of this report consider **the ERICA Tool to be a useful departure point for radiological assessments involving NORM**, although RESRAD-BIOTA provides a suitable, viable alternative. ERICA has been selected for further consideration for future case studies because:

- the tool lends itself to application to impact assessments concerning NORM as exemplified in Section 4.5 and noting that this is not the case for all the models covered in Section 4.4, and
- contributing authors have been directly involved in the development of this tool and thus the possibility exists to make changes in ERICA going forward. It is acknowledged that some adaptation to cases involving NORM may be necessary.

There appears to be broad support provided in relevant literature to implement a tiered or graded approach when undertaking an environmental impact assessment. Such an approach essentially attributes the level of detail and resources required for the assessment to the perceived risk associated with the hazard. Typically, the assessment would begin with a lower tier, screening assessment, where conservative assumptions are adopted (using simple models with generic parameters) to obtain pessimistic predictions of the radiological impacts. If at this stage it can be demonstrated that the risks are trivial, then there may be no need to perform more detailed calculations. However, where this is not the case, the assessment process should move to higher tiers where more site-specific details (and concomitantly more resources) are required to provide an estimate of the radiological impact. For the higher tiers, the additional resources might be spent on more detailed model parameterisation, more comprehensive data collation or model development. This approach is essentially the way assessments are organized in the ERICA integrated approach Figure 37 and **we recommend that this tiered/graded approach is adopted.**

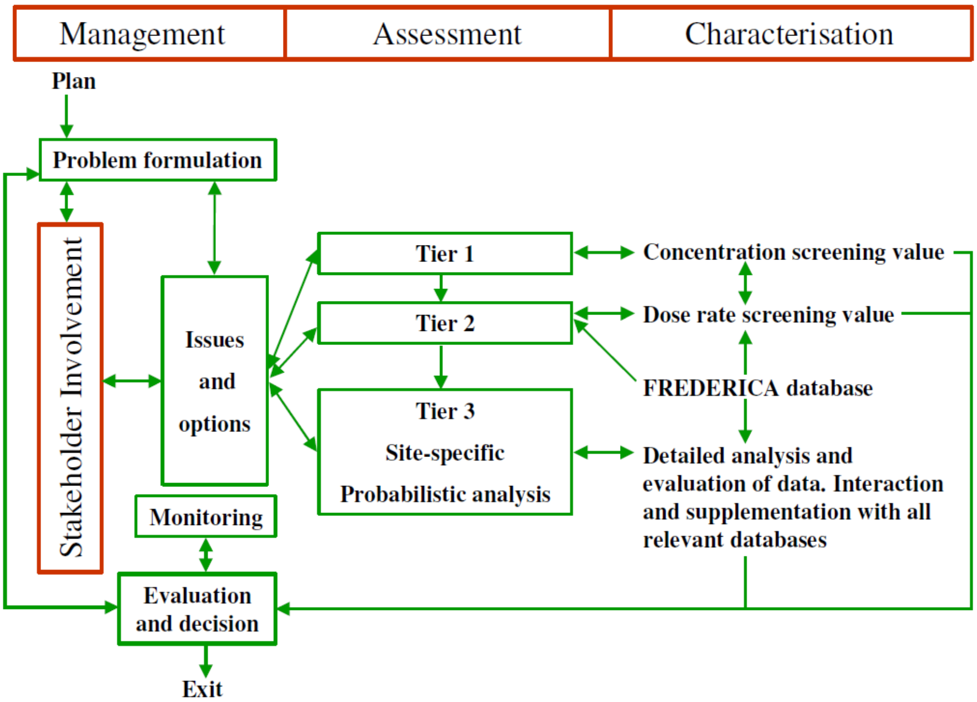


Figure 37. Overview of the ERICA Integrated Approach, outlining the interaction between assessment, risk characterization and management (Beresford *et al.* 2007).

As noted by Vandenhove *et al.* (2015), there is often a requirement to characterise levels of natural radiation ‘background’ levels because assessments are generally focussed on the incremental contribution from the NORM industry over such natural background radiation. This incremental contribution can be determined theoretically, through the application of models, but once measurement data become available, it is of importance, at least for the contextualisation of the exposure results, to determine the component of dose attributable to the natural background and the technologically enhanced part. **The guidance here is to collate information on regional background levels of natural radionuclides** either through bespoke monitoring campaigns or consideration of the literature (examples where regional background determination has been made for specific areas are given by Petrović *et al.* (2018) and Abbasi *et al.* (2022)). Failing to have information about local/regional background levels, more generic background levels of radionuclides may be considered (e.g. Hosseini *et al.* 2010). In the final stages of an assessment, the dose rates calculated will normally be contextualised through comparison with background dose rates and "benchmarks" (ERICA, ICRP, UNSCEAR, etc.).

NORM contamination scenarios often involve diffuse sources and often cover a much larger area than those typically associated with nuclear sites (e.g. nuclear power production and release). The modelling options available for tools like ERICA, for making prognosis concerning the advection and dispersion of radionuclides following release, normally concern point sources for discharge (see IAEA 2001). This functionality is suitable for some of the potential NORM assessment cases we might be interested in, for RadoNorm, such as those involving specific location, routine (and continuous) releases to the marine environment from e.g. oil and gas platforms. In such cases, input data would be required on the discharge rate of NORM radionuclides in terms of Becquerels released per unit time. **For cases that might not be considered as point sources, a recommendation may be to adopt tools available elsewhere,** examples for the terrestrial environment being NORMALYSA (Avila *et al.* 2018) and RESRAD-OFFSITE (Yu *et al.* 2009) **to provide input data concerning NORM activity concentrations in environmental media (soil, water and sediment).** These data can then be included directly within an ERICA assessment as inputs.

There are numerous examples in the published literature (e.g. Vanhoudt *et al.* 2012, Lecomte *et al.* 2019) where the issue of multiple stressors is highlighted as being important for cases involving NORM. Notwithstanding the fact that environmental impacts may be dominated by non-radiological hazards, it seems quite apparent that understanding the interaction of radionuclides with other stressors is a prerequisite for a robust environmental impact assessment. Nonetheless, no internationally accepted guidance for radioactivity in a **multi-stressor context** was available at the time of writing this report and it, therefore, seemed **premature to provide prescriptive guidance for the following analysis in the RadoNorm project**. For the case study, focus will be placed on the radiological assessment part of the analysis but recourse will be made to the developments in RadoNorm WP4 with a view to providing more concrete guidance in the future with regard to the interaction of multiple stressors.

A robust methodology is required to consider natural decay series decay chains and it is **recommended that the assessor understands the assumptions used with regards to the inclusion of daughter radionuclides in the model's dose calculations**. At Tier 2 in ERICA, when the user selects (at the assessment context stage) any of the parent radionuclides listed in the left hand-column of Table 22, the daughter(s), listed in the right hand column of the table (excluding radon and thoron – see below), are automatically selected and included in subsequent calculations. The assumption is then made that radionuclides in the selected decay chain are in secular equilibrium (<u>at the start of the simulation period</u>). In ERICA, the user has the option to change the default if this assumption is not valid by selecting and entering data for daughter radionuclides explicitly.

Table 22. Cases where, as default, decay chains in secular equilibrium are included when a parent radionuclide is selected

Parent nuclide	Daughter nuclides with physical half-lives under 10 days considered to be in equilibrium with parent nuclide						
Pb-210	Bi-210						
Ra-226*	At-218	Po-218	Bi-214	Pb-214	Rn-222	Po-214	
Ra-228	Ac-228						
Th-228*	Po-216	Tl-208	Bi-212	Pb-212	Rn-220	Po-212	Ra-224
Th-234	Pa-234m**						
U-235	Th-231						

*Excludes radon and thoron; ** Pa-234 not included as Pa-234m dominates completely as decay products – the yield of Pa-234 is relatively low. ICRP Publication 107 does not even include Pa-234 as a decay product.

When performing a Tier 2 assessment, the assessor should consider the entire natural series decay chain and include all radionuclides with physical half-lives greater than 10 days in the process of defining the assessment context. For example, for the ^{238}U series, the assessor would include ^{234}Th, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Po and ^{210}Pb. For the ^{232}Th series, the assessor would include ^{228}Ra and ^{228}Th.

For assessments that include ^{226}Ra or ^{228}Th, the contribution from radon (^{222}Rn) and thoron (^{220}Rn) in decay chains has been excluded because the primary dose contribution is due to inhalation. Dose rates from radon and thoron in the ^{226}Ra or ^{228}Th decay series therefore require the calculation of soil air concentrations and dilution of soil air in above ground air. The contribution of these radionuclides to dose rates can be evaluated following calculation of within and above air concentrations using the approach to calculating internal doses from radon and thoron described below. **The recommendation is that for terrestrial assessments, the bespoke functionality available in the ERICA Tool should be applied to determine, the environmental exposure associated with radon and thoron gases.** Radon and thoron data should be entered as radionuclide activity concentrations in air, allowing an immersion (i.e. above soil submersion in contaminated air), DCC to be applied. The DCCs are applied only to animals assigned either a fractional or fully 'on-soil' occupancy and immersion dose rates are not currently calculated for plants. It is assumed that animals are 100 % of the time present in contaminated air (irrespective of whether the organism is above or below ground), meaning that occupancy factors can be ignored.

For radon and thoron (^{222}Rn and ^{220}Rn), we need also to account for the contribution of these radionuclides (and more importantly their daughters) to dose rates arising from inhalation and deposition in the lung. In ERICA, this component of dose is treated as an 'internal' contribution to exposure and the values based on the methodology of Vives i Batlle *et al.* (2017) are applied. It is important to note that the units for internal DCCs, for ^{222}Rn and ^{220}Rn differ from the standard internal DCCs for all other radionuclides. Whereas the standard ERICA internal DCCs relate to the activity concentration in the (whole) body of the plant or animal, and thus have units of μGy h^{-1} per Bq kg^{-1} f.w., ^{222}Rn and ^{220}Rn are aggregated DCs and relate (the internal dose-rate) directly to the concentrations, and thus have units of μGy h^{-1} per Bq m^{-3}.

The various steps recommended for a Tier 2 ERICA assessment involving NORM are shown in Figure 38.

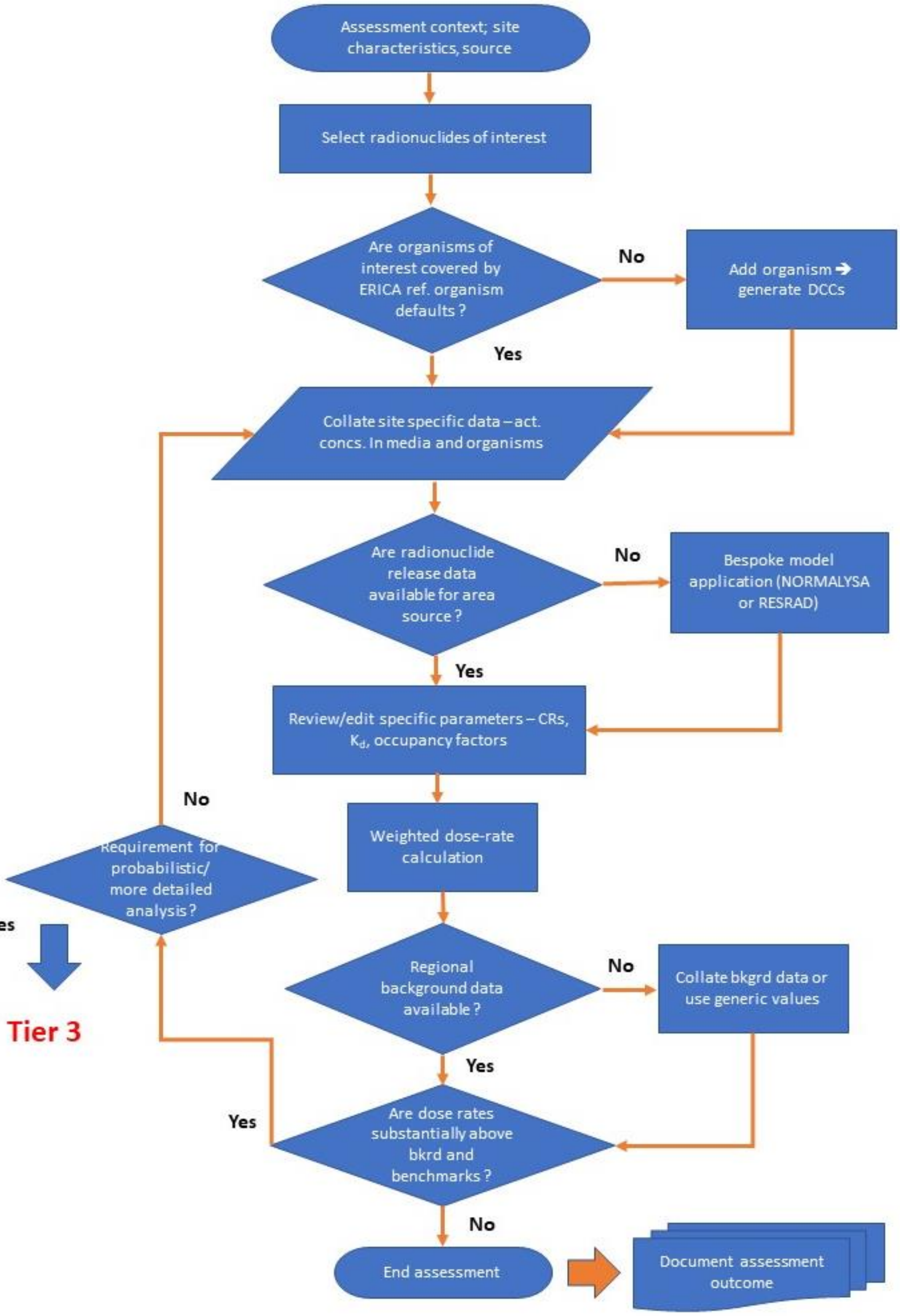


Figure 38. Schematic showing the various steps in a Tier 2 ERICA assessment for NORM

5. Conclusions

This document presents a series of studies aimed to develop dose assessment methodologies related to diverse NORM involving situations. Investigations across key NORM management areas, including residue disposal in landfills, agricultural reuse of NORM-containing materials, complex hydrogeological transport at legacy sites, and environmental impact on non-human biota, have yielded significant insights. The consistent application of updated dosimetric data and the evaluation of recognized modelling tools have been central to developing screening tools and identifying critical parameters for various exposure scenarios. The study underscores the importance of scenario-specific considerations, the value of tiered assessment strategies, and the continuous need for adapting existing tools and frameworks to the unique challenges posed by NORM.

Collectively, the methodological advancements established within this report provide a crucial and scientifically grounded foundation, and they are specifically intended to serve as input for the next RadoNorm deliverable D.19, "Dose-assessment procedure for NORM involving situations. Recommendations”.

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Appendix A. Supplementary materials

The following tables are reported in this document:

Table A1. Dose coefficients for **inhalation** used in the study (Sv Bq^{-1})

Table A2. Dose coefficients for **ingestion** used in the study (Sv Bq-1)

Table A3. Parameters used for the dose assessment for **worker scenario** (Sc1)

Table A4. Parameters used for the dose assessment for members of the public scenario (Sc2)

Table A5. Annual food consumption and breathing rates per age class

Table A6. Dose coefficients for external exposure (Sv h^{-1})/(kBq kg^{-1}) for a semi-infinite volume (infinite depth and lateral extent) at a distance of 1 m from the ground surface

Table A1. Dose coefficients for **inhalation** used in the study (Sv Bq^{-1})

Decay segment	Infant	1 y	5 y	10 y	15 y	Adult	Worker
U-238sec	1.91E-04	1.66E-04	1.09E-04	7.07E-05	6.15E-05	5.70E-05	6.99E-05
Unat	6.34E-05	5.52E-05	3.58E-05	2.25E-05	1.91E-05	1.78E-05	2.56E-05
Th-230	4.00E-05	3.50E-05	2.40E-05	1.60E-05	1.50E-05	1.40E-05	1.50E-05
Ra-226+	3.42E-05	2.91E-05	1.91E-05	1.20E-05	1.00E-05	9.53E-06	1.30E-05
Pb-210+	1.84E-05	1.83E-05	1.12E-05	7.33E-06	6.01E-06	5.69E-06	9.26E-06
Po-210	1.80E-05	1.40E-05	8.60E-06	5.90E-06	5.10E-06	4.30E-06	1.80E-06
U-235sec	3.95E-04	3.49E-04	2.33E-04	1.62E-04	1.45E-04	1.33E-04	1.27E-04
U-235+	3.00E-05	2.60E-05	1.70E-05	1.10E-05	9.20E-06	8.50E-06	1.20E-05
Pa-231	7.40E-05	6.90E-05	5.20E-05	3.90E-05	3.60E-05	3.40E-05	4.60E-05
Ac 227+	2.91E-04	2.54E-04	1.64E-04	1.12E-04	1.00E-04	9.07E-05	6.93E-05
Th-232sec	2.76E-04	2.38E-04	1.57E-04	1.06E-04	9.25E-05	8.46E-05	1.00E-04
Th-232	5.40E-05	5.00E-05	3.70E-05	2.60E-05	2.50E-05	2.50E-05	5.40E-05
Ra-228+	4.91E-05	4.81E-05	3.20E-05	2.00E-05	1.60E-05	1.60E-05	2.20E-05
Th-228+	1.73E-04	1.40E-04	8.83E-05	5.97E-05	5.15E-05	4.36E-05	2.42E-05
K-40	–	–	–	–	–	–	–

Table A2. Dose coefficients for **ingestion** used in the study (Sv Bq^{-1})

Decay segment	Infant	1 y	5 y	10 y	15 y	Adult	Worker
U-238sec	4.64E-05	1.43E-05	7.90E-06	5.83E-06	5.48E-06	2.57E-06	7.77E-07
Unat	7.71E-07	2.84E-07	1.87E-07	1.53E-07	1.49E-07	1.00E-07	6.81E-08
Th-230	4.10E-06	4.10E-07	3.10E-07	2.40E-07	2.20E-07	2.10E-07	6.00E-08
Ra-226+	4.70E-06	9.62E-07	6.21E-07	8.01E-07	1.50E-06	2.80E-07	1.30E-07
Pb-210+	8.42E-06	3.61E-06	2.20E-06	1.90E-06	1.90E-06	6.91E-07	3.21E-07
Po-210	2.60E-05	8.80E-06	4.40E-06	2.60E-06	1.60E-06	1.20E-06	1.80E-07
U-235sec	5.20E-05	5.70E-06	4.00E-06	2.96E-06	2.46E-06	1.97E-06	4.24E-07
U-235+	3.54E-07	1.33E-07	8.62E-08	7.17E-08	7.04E-08	4.73E-08	3.20E-08
Pa-231	1.30E-05	1.30E-06	1.10E-06	9.20E-07	8.00E-07	7.10E-07	1.80E-07
Ac 227+	3.86E-05	4.27E-06	2.81E-06	1.97E-06	1.59E-06	1.21E-06	2.12E-07
Th-232sec	4.12E-05	7.25E-06	4.36E-06	4.62E-06	5.86E-06	1.06E-06	4.76E-07
Th-232	4.60E-06	4.50E-07	3.50E-07	2.90E-07	2.50E-07	2.30E-07	7.00E-08
Ra-228+	3.00E-05	5.70E-06	3.40E-06	3.90E-06	5.30E-06	6.90E-07	3.40E-07
Th-228+	6.55E-06	1.09E-06	6.04E-07	4.31E-07	3.07E-07	1.43E-07	6.57E-08
K-40	–	–	–	–	–	–	

Dose coefficients for workers are taken from ICRP 137. Dose coefficients for members of the public are taken from ICRP 72.

Table A3. Parameters used for the dose assessment for **worker scenario** (Sc1)

Parameter	Unit	Value
Exposure time	h y^{-1}	1800
Inhalation pathway		
Dust concentration at the landfill (C_{dust})	mg m^{-3}	1
Breathing rate (B_{rate})	m^3 h^{-1}	1.2
Fraction of activity that binds to the small particles causing the activity to concentrate in the fine dust fraction (f_k)	–	1
Ingestion pathway		
Ingestion soil rate (r_{ing})	g y^{-1}	18
External radiation pathway (RESRAD-ONSITE parameters)		
Area of contaminated zone	m^2	10^5
Thickness of contaminated zone	m	10
Density of the contaminated zone	g cm^{-3}	1.5
Indoor time fraction	–	0
Outdoor time fraction	–	0.2
Time integration parameter (Maximum number of points for dose)	–	1

Table A4. Parameters used for the dose assessment for **members of the public scenario** (Sc2)

Parameter	Unit	Value
Exposure time indoor ($t_{in\ house}$)	h y^{-1}	7000
Exposure time outdoor ($t_{in\ garden}$)	h y^{-1}	1000
Inhalation pathway		
Dust concentration indoor ($C_{dust,\ house}$)	mg m^{-3}	0.1
Dust concentration outdoor ($C_{dust,\ garden}$)	mg m^{-3}	0.1
Breathing rate (B_{rate})	m^3 h^{-1}	See Table A7
Fraction of activity that binds to the small particles causing the activity to concentrate in the fine dust fraction (f_k)	–	1
Ingestion of food products contaminated by dust from landfill		
Relative contribution of food bought at the market (r_{market})	–	0.5
Preparation reduction factor for food (r_{prep})	–	0.5
Food consumption rates of leafy vegetables (FC_{leafy})	kg y^{-1}	See Table A7
Food consumption rates of other products grown ($FC_{other\ veg}$)	kg y^{-1}	See Table A7
External radiation pathway (RESRAD-OFFSITE parameters)		
Site Layout windows		
X dimension of primary contamination	m	300
Y dimension of primary contamination	m	330
Location (fruit, grain, non-leafy vegetable plot) *X-coordinate*	m	1000–1100
Location (fruit, grain, non-leafy vegetable plot) Y-*coordinate*	m	1000–1100
Location (Leafy vegetable plot) *X-coordinate*	m	1000–1100
Location (Leafy vegetable plot) Y-*coordinate*	m	1000–1100
Location (Pasture, silage growing area) *X-coordinate*	m	1000–1100
Location (Pasture, silage growing area) Y-*coordinate*	m	1000–1100
Location (Grain fields) *X-coordinate*	m	1000–1100
Location (Grain fields) Y-*coordinate*	m	1000–1100
Location (Dwelling site) *X-coordinate*	m	149.5–150.5
Location (Dwelling site) Y-*coordinate*	m	349.5–350.5
Primary contamination		
Thickness of primary contaminated zone	m	10
Dry bulk density	g cm^{-3}	1.5
Occupancy		
Fraction of time spent in offsite dwelling site (indoor)	–	0.8
Fraction of time spent in offsite dwelling site (outdoor)	–	0.2
Inhalation and external gamma		
External gamma penetration factor		0.25

Table A5. Annual food consumption and breathing rates per age class

Parameter	**Age Group**					
	Infant	**1 y**	**5 y**	**10 y**	**15 y**	**Adult**
Food type consumption rate (kg y^{-1})						
Other vegetable, fruit and grain $FC_{other\ veg}$	72	132	220	250	260	240
Leafy vegetable FC_{leafy}	3	6	7	9	11	13
Breathing rate B_{rate} ($m^3\ y^{-1}$)	1 100	1 900	3 200	5 640	7 300	8 100

Table A6. Dose coefficients for external exposure (Sv h^{-1})/(kBq kg^{-1}) for a semi-infinite volume (infinite depth and lateral extent) at a distance of 1 m from the ground surface

Decay segment	Satoh and Petoussi-Henß (2024) used in this study	Andersson and Mobbs (2010)
U-238sec	3.08E-07	5.48E-07
Unat	5.41E-09	9.98E-08
Th-230	2.90E-11	1.32E-10
Ra-226+	3.00E-07	5.40E-07
Pb-210+	1.51E-10	2.46E-10
Po-210	1.62E-12	2.71E-12
U-235sec	8.24E-08	2.47E-07
U-235+	2.01E-08	7.41E-08
Pa-231	4.51E-09	1.50E-08
Ac 227+	5.78E-08	1.58E-07
Th-232sec	4.10E-07	8.00E-07
Th-232	1.22E-11	5.68E-11
Ra-228+	1.44E-07	3.80E-07
Th-228+	2.66E-07	4.94E-07
K-40	2.86E-08	–