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Swedish Radiation Safety Authority

Research

Region-specific radio-ecological evaluation of accidental releases of radionuclides from ESS

2021:21

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SSM perspective

Background

To strengthen the material science research in Europe, the European Spallation Source (ESS) is being built in Lund in the southern part of Sweden. In the spallation process, neutrons are generated when the accelerated protons hit the tungsten target. In addition, a broad range of radioactive by-products are produced that differs from the fission products produced in a nuclear reactor. In case of an accident scenario target material, particles could be released to the atmosphere and contaminate the surroundings. The particles, such as the alpha-emitting ^{148}Gd, are found in various eco-systems and possibly introducing exposure pathways to humans from several food chains. The knowledge on how to estimate the nuclide concentrations in environmental samples is a significant part of the dose assessment. Another important aspect in the dose assessment is the knowledge of the local food production and the consumption patterns for representative persons.

Therefore, there is a need to enhance the knowledge of the behaviour of radionuclides such as gadolinium and other rare earth elements (REEs) in the terrestrial environment before the ESS facility starts to generate neutrons in a few years. The Swedish Radiation Safety Authority has found monitoring and modelling the transfer of key radionuclides in the environment and, in particular, in the Lund region of great importance to support. Moreover, the support reflects the ambition to prioritize radioecology, since it is one of the critical research areas in retaining future competence.

Results

The project embarked with a literature review of the available radioecological models and their appropriateness to model REEs and further elements related to ESS, in the region-specific environment. The conclusion is that most models are applicable for these elements; instead, the limiting factor is the lack of available radioecological parameter values. The authors indicate the Ecolego software as a candidate for future developments due to its flexibility and previous uses. In addition, an investigation of available radioecological parameter values for the region-specific environment led to the conclusion that the equilibrium concentrations in various environmental media need more attention since most values for gadolinium are missing.

Furthermore, the authors identified major food production in eight different areas nearby ESS, a very fertile agricultural area, together with possible representative persons. Samples collected in the areas are from the radiation survey the licensee ESS is obliged to make before operation (zero point assessment). The report includes a description of two extractions methods (5-step Wiche *et al* method and 4-step Mittermüller *et al* method) for REEs in soil and a pseudo-total extraction with aqua regia. The latter method is also compared with the extraction result of an REE reference material, an estuarine sediment. In the project, the methods were applied to four of the top soil samples before being prepared and analysed with inductively coupled mass spectrometry (ICP-MS). The 4-step method and ICP-MS seems to be suitable to assess gadolinium and other REEs in environmental samples. However, it is too early to say that ICP-MS together with multi-collector and selective extraction is the best method in quantifying ^{148}Gd.

The authors also conclude that the baseline for the REE concentration around ESS seems to follow the levels in similar soils, even if more REE content might have been introduced from fertilizers in cultivated areas.

Relevance

This report is preceded by the related report *Identifying radiologically important ESS-specific radionuclides and relevant detection methods* (2020:08) by the same research group and presents the results of a pilot study on sample preparation and mass spectrometry measurements of REEs in a number of environmental samples. The final goal is to obtain knowledge on how to collect representative samples, how to prepare and measure radionuclides in case of an accident with a release from ESS. Moreover, knowing how to model environmental transfer with relevant input data will help assessing doses from intake of food contaminated by these radionuclides. These studies are therefore part of the emergency preparedness and response as a basis for decisions taken on implementing food restrictions or other protective actions.

Need for further research

Today, the measuring techniques, radioecological models and parameter values focusses on dominant fission products in the context of dose contribution such as radioactive isotopes of iodine and cesium seen in large scale accidents as Chernobyl and Fukushima Daiichi. When data for environmental transfer are missing for a specific nuclide, modellers are referred to use an analogue (isotope/element/species) although is it not the preferred approach. Future research should strive to fill those gaps to achieve more reliable dose assessments. The authors indicate analytical methods for assessing radioactive gadolinium concentration in various environmental samples as a field for future research. It is essential to identify suitable analytical methods combined with optimal methods for sample extraction, separation and preparation before deducing any radioecological parameter values with the help of experimental studies.

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Summary

Gadolinium-148 is one of the radionuclides of most concern that will be produced in the tungsten target of the European Spallation Source (ESS), as a by-product of the spallation reaction used by the facility to produce neutrons. Since ^{148}Gd a pure alpha emitter, it is both very radiotoxic and difficult to measure. With its half-life of 75 years, it will remain in the environment for a long time if released from the facility during normal operation or after an accident. There are still uncertainties regarding the amounts that actually will be produced by spallation in the tungsten targets of the facility [1], [2]. As ^{148}Gd does not occur naturally in the environment, there is no information available about its analysis in environmental samples [3] but a few studies provide data from irradiated target material analysed by alpha spectroscopy [4], [5] or inductively coupled plasma mass spectrometry (ICP-MS) [6]. This report is a continuation of the SSM project as described in the SSM report 2020:08, entitled “Identifying radiologically important ESS-specific radionuclides and relevant detection methods” [3] that focused on the ESS-related radionuclides that will be the most relevant to study and monitor in the environment as well as the analytical techniques to detect them. The present report focuses on the rare earth elements (REEs), including their radioactive isotopes, in particular ^{148}Gd, and is intended to highlight the knowledge gaps that exist regarding their fate in the specific environment of the ESS area.

In the first part of the report, the available literature on radioecological models was reviewed, with emphasis on ESS-related radionuclides. The existing modelling programmes were surveyed as well as the most relevant environmental parameters and experimental radioecological data required to build models specific to the ESS.

In the second part of the report, the area in the vicinity of the ESS was surveyed to identify the important producers of foodstuff, what plant species are grown in the area and also the local husbandry and hunting practices, in order to identify critical pathways after a radioactive dispersion into the environment in connection with a potential accident at the ESS.

In the third part of this report, after a thorough literature review and preliminary assays, we propose to investigate the use of ICP-MS for assessment of ^{148}Gd in the event of an accidental release, knowing that this analytical technique is already used for the measurement of stable Gd and REEs in the environment. The existing methods to extract REEs from environmental samples (soil, water, plants, and animal products) and to properly assess their concentration are described in the form of a literature review. The presented examples of methods were selected to fit the type of environment found around the ESS facility and the local agricultural and horticultural practices. A pilot study was also conducted to test extraction and measurement methods on the specific type of soil around ESS. These results are presented at the end of this report.

Sammanfattning

Inom några år kommer en av världens mest kraftfulla neutronstrålkällor, den Europeiska spallationsanläggningen ESS, som är belägen i utkanten av Lund, att tas i drift. Den gör det möjligt att studera material på molekylär och atomär nivå med tillämpningar inom många olika forskningsområden. I anläggningen kommer protoner att accelereras till 2 GeV mot ett strålmål av volfram, och då generera ett flöde av neutroner som i sin tur kommer att användas för de olika forskningsprojekten. Som icke önskvärda biprodukter till neutronerna bildas många hundra olika radioaktiva ämnen, bland annat ”svårmätbara” isotoper av grundämnen som Gd, Sm, Hf, m.fl.

En av de radioaktiva isotoper som är av störst intresse från ett strålskyddsperspektiv är gadolinium-148. Det är en ren alfastrålare med 75 års halveringstid och hög radiologisk toxicitet om den frigörs och får möjlighet att internkontaminera personal och allmänhet. En svårighet är att vi – till skillnad mot de radionuklider som produceras i våra kärnkraftverk – har mycket begränsad erfarenhet av hur gadolinium-148 uppträder i omgivningen och i människokroppen. Det är därför av stor betydelse att skaffa mer kunskap kring denna radionuklid och kring andra radionuklider bland övriga sällsynta jordartsmetaller för att bättre kunna hantera och minimera konsekvenserna av en eventuell olycka vid ESS.

På uppdrag av Strålsäkerhetsmyndigheten har vi inom detta projekt (SSM2019-1010) börjat undersöka några av de komponenter som kommer att vara betydelsefulla vid händelse av en olycka vid ESS som skulle kunna leda till ett större utsläpp av gadolinium-148 till omgivningen utanför anläggningen. Dessa komponenter omfattar dels modellberäkningsverktyg för gadolinium i ekosystem rent generellt, dels den specifika miljön runt ESS, men också hur svårmätbara radionuklider som gadolinium-148 bättre kan separeras från miljöprover som växter och jord, och analyseras.

Projektet har varit indelat i tre olika förberedande och undersökande delar. Alla delarna kommer att sammankopplas för att framöver kunna utveckla bättre kunskap och beredskap för en oförutsedd olycka vid ESS. I den första delen av rapporten undersöks befintlig litteratur med avseende på tillgängliga modelleringsverktyg med fokus på ESS-specifika förhållanden. Dessa verktyg utvärderades utifrån vilka grundläggande data som för närvarande finns tillgängliga och som krävs för att modellera en potentiell olycka vid ESS. I den andra delen av rapporten beskrivs den specifika miljön i området med avseende på jordbruk, trädgårdsodling och annan markanvändning samt befolkningsstruktur i området. I den tredje delen av rapporten beskriver en bred genomgång av befintlig litteratur inom vad som tidigare gjorts med avseende på mätningar av sällsynta jordartsmetaller. I denna del inkluderas även förslag på nya metoder att extrahera gadolinium från olika typer av prover samt analysresultat av dessa.

I sin helhet utgör denna rapport ett viktigt underlag för fortsatta studier av betydelsefulla ESS-radionuklider för stråldosbestämningar för representativa personer i händelse av en allvarlig olycka vid anläggningen.

Abbreviations and notations

BCR: European Community Bureau of Reference

d.w.: dry weight

ESS: European Spallation Source

IAEA: International Atomic Energy Agency

ICP-AES: Inductively Couple Plasma Atomic Emission Spectroscopy

ICP-MS: Inductively Coupled Plasma Mass Spectrometry

LA-ICP-MS: Laser Ablation Inductively Coupled Plasma Mass Spectrometry

LOD: Limit of detection

ppb: Parts per billion

ppm: Part per million

REE(s): Rare earth element(s) (including Sc, Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu)

RSD: Relative standard deviation

SKB: Swedish Nuclear Fuel and Waste Management Company

SSM: Swedish Radiation Safety Authority

w.w.: wet weight

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1. Introduction

This introductory chapter gives the background to and the objectives of the project SSM2019-1010, financed by the Swedish Radiation Safety Authority (SSM).

1.1. Background

The European Spallation Source (ESS) is currently being constructed ~4 km northeast of the centre of Lund, in the south-western part of Sweden. The 5 MW proton accelerator is planned to start operation within the next few years, but full operation of the neutron research facility may not be expected for yet some years. The experience from operation of high-power spallation sources like ESS is very limited. The operation of the facility will generate a wide variety of radionuclides, resulting from nuclear processes in the target itself, but also through activation of air, cooling media, accelerator materials, building materials and surrounding soil. The radionuclides formed in ESS may be released to the environment during normal operation [7], during maintenance as well as at potential incidents and accidents [8]. According to directives from the Swedish Radiation Safety Authority (SSM), the total annual effective dose should not exceed 0.1 mSv y^{-1} for members of the public from the operation of ESS [9]). Thus, it is important to rigorously assess the potential environmental consequences of the ESS-site operation.

In the tungsten target, radionuclides with lower as well as higher atomic numbers than the target material itself will be generated during operation. Examples of elements produced by spallation, fragmentation and high-energy fission are isotopes of Ta, Hf, Lu, Yb, Tm, Er, Ho, Dy, Tb, Gd, Eu, Sm and Pm. From a modelling study of the radionuclides produced by proton irradiation of the tungsten target using FLUKA [1], [3], it is shown that most of the low atomic number radionuclides anticipated in previous assessments [2] will occur. However, the different models foresee very different ^{148}Gd production. Indeed, Barkauskas et al. [1] as well as Kókai et al [10] estimate its production to be an order of magnitude higher than the estimation of the ESS Preliminary Safety Analysis Report from [11] and Mora et al. [2]. Therefore, it is of special importance to obtain more knowledge on the behaviour of gadolinium and other rare-earth elements in the environment, to ensure that the consequences of any radiological accident are understood and can be minimised.

Despite the significant impact of radiological accidents, it must be kept in mind that the probability of this occurring is very low. However, during normal operation ESS will constantly produce radionuclides in e.g. the soil [12] and hence, the behaviour of radionuclides such as ^{24}Na, ^{32}P, ^{35}S, ^{55}Fe should also be assessed, as they are expected to be constantly produced in the soil outside the ESS accelerator tunnel.

Gadolinium:

Gadolinium (Gd) belongs to the group of rare earth elements (REE), and globally exists with an abundance of 6.1 ppm in surface soils on the Earth's crust [13]. The corresponding concentrations in Swedish top soils are estimated to be somewhat lower, ranging from 1.0 to 4.8 ppm [13]. Typical concentrations of stable Gd in plants vary from 30 – 60 ppb in Northern European biomass, such as forest plants, grass, leaves, cabbage and moss, and concentrations of most REE elements in the above-ground biomass of vascular plants are usually quite low (e.g. transfer factors of 0.04-0.09 have been reported from German forests [14]). No other group of elements in the periodic system displays such a great similarity as the REEs, in particular regarding their chemistry. The REEs are found, usually several together, in a variety of accessory minerals (chemical forms can be e.g. phosphates, carbonates, fluorides and silicates).

The ecotoxicity of REE has mainly been studied in aquatic systems although some data can be found for terrestrial plants such as rice, wheat and tomato [15]. The mobility and the bioavailability of REE from soils to plants is a complex issue that can be influenced by many parameters including the chemical form of the REE, the type of soil, the soil microorganisms and the plant species. The nitrates, chlorides, and sulphates of REEs are usually soluble, while their carbonates, phosphates, and hydroxides are usually insoluble. The amount of organic material in the soil is of great importance to the mobility of the REEs, and their solubility is strongly dependent on pH. The retention and release of the REEs are also influenced by high contents of clay minerals and presence of secondary REE minerals such as certain phosphates and high concentrations of phosphate or sulphate in the soil solution. The soil microorganisms are of great importance for the mobility of REE through the root system of the plant, and the root uptake is also specific to each plant species. Solutions containing REEs sprayed on crops may be translocated in the plant tissues, and there are even indications that movements of REEs may also take place from leaf to root. Tyler [13] also states that it is often difficult to discriminate between amounts possibly present in the plant from fractions that are not easily washable surface dust contamination or REEs contained in plaque around roots. All these parameters seem to have greater influence than the actual concentration of REE in the soil i.e. a higher contamination of the soil does not automatically imply a higher bioaccumulation.

Modern pollution by stable isotopes of Gd is due to its use in contrast agents injected to patients before Magnetic Resonances Imagining examinations. Once excreted by the body, the contrast agents end up in the waste-water system from which they are spread in the environment. An interesting observation is that the speciation of this anthropogenic Gd is different from that of natural Gd and suggests a long environmental half-life for the Gd complexes used as contrast agents [16]. Rare earth elements are also increasingly used in agricultural and zoo-technical applications, as fertilizers and feed additives in particular in China [17].

The most common techniques for the analysis of stable Gd are Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and Inductively Couple Plasma Atomic Emission Spectroscopy (ICP-AES). Both techniques have very low detection limits and allow to measure, in a single analysis, not only Gd but the full series of REEs. Measurement of the full REE series is valuable for estimating concentration anomalies in environmental samples. This method has been used to study erbium concentrations around Chernobyl [18] and is also the method used to estimate the contribution from Gd contrast agents to the total concentration of Gd in environmental samples [19].

ICP-MS is one of the techniques that might be used to analyse the abundance of difficult-to-measure radionuclides in environmental samples after accidental releases by ESS. Indeed, the ESS-specific alpha emitters such as ^{148}Gd, ^{146}Sm or ^{154}Dy are REEs. ICP-MS coupled with high-performance liquid chromatography

(HPLC) has previously been used e.g. to determine the nuclide abundance of lanthanide metals produced via spallation reactions in tantalum [20]. The knowledge of the current level of stable REEs could help to provide more accurate results when estimating releases of ESS specific isotopes, e.g. both neodymium and samarium have stable isotopes of mass 148 that could interfere with the quantification of ^{148}Gd by mass spectrometry.

*Radioecological model*s:

There are existing models for radioecological transfer of radionuclides in terrestrial and aquatic environments such as CROM [21] and ERICA [22]. The ERICA Tool is a software system that has a structure based upon the tiered ERICA Integrated Approach. It is used to assess radiological risk to terrestrial, freshwater and marine biota, and is largely based on transport models taken from the International Atomic Energy Agency (IAEA) [23]. In a recent assessment by Ene et al. [7], models from IAEA have been used for dose calculations during operational releases from ESS, combined with a number of internally elaborated ESS models [24]–[27]. From these calculations, it was estimated that the total annual effective dose from continuous airborne releases will be dominated by radionuclides such as ^{13}N, ^{11}C, ^{41}Ar, ^{15}O, ^{125}I and ^{3}H, and by ^{3}H, ^{22}Na and ^{35}S from exposure to groundwater around the accelerator tunnel area.

The safety assessment carried out by SSM in 2018 was used as a basis for the classification of the ESS facility in terms of emergency preparedness category according to the IAEA safety standards [28]. It provides estimates of exposures to humans in case of a pessimistic accident scenario to evaluate the need for immediate protection measures. In that assessment it was concluded that in 90% of the local weather conditions, sheltering up to 1.5 km from the ESS stack release point will be a sufficient countermeasure, and other more resource-demanding measures such as evacuation may not be warranted. Lund municipality have decided an emergency preparedness zone of about 700 m from the ESS stack release point, which corresponds to 80% of the local weather conditions in the SSM safety assessment, with the same protective measures as previously mentioned i.e. planning for evacuation or distribution of stable iodine is not justified. The assessment is based on model calculations of the effective dose to an unprotected person during 7 days (release phase) due to atmospheric dispersion and subsequent cloud shine, inhalation doses from the plume and ground shine from the deposited releases. However, not mentioned in the report is the long-term transfer and internal dose contribution to humans, and other biota, by means of ingested crops and pasture and other contaminated foodstuff, or from local water consumption. Preliminary calculations, including uncertainty analysis, are being made on the impact on human and nonhuman biota using the ERICA tool [29] based on representative persons and reference ecosystems defined by ESS themselves.

IAEA has issued a handbook of parameter values for prediction of radioecological transfer in terrestrial and freshwater environments [30]. The handbook provides soil mobility distribution coefficients, K_d (L kg^{-1}), interception coefficients, a (m^2 kg^{-1}), soil-to-plant transfer, F_v (dimensionless), freshwater concentration ratios, CR (dimensionless) etc. (see Table 1 for a selection). However, none of the models currently have any exhaustive parameter sets explicit for two of the most relevant radionuclides in connection with an accidental airborne release from the ESS stack: ^{148}Gd and ^{172}Hf. Parameter values exist for some chemical analogues such as Ce and La. Surveying the IAEA handbook, it is evident that there is a consistent lack of data for the transfer model parameters listed in Table 1 and Table 2. For instance, there is no data on interception of either wet or dry atmospheric deposition, for any type of vegetation, for any of the elements of most concern in case of an accidental ESS release.

Table 1: Selection of available ecological parameters for terrestrial species in the IAEA handbook relevant for accidental releases of radionuclides from ESS [30]

Parameter	Hafnium	Gadolinium*	Tungsten
Translocation factor, f_{tr} (%)	No data	No data (Ce=0.01-7.8 for wheat, barley and rye)	No data
Interception factor, α (m^2 kg^{-1})	No data	No data	No data
Mass interception factor, f_B (m^2 kg^{-1})			
Distribution coefficient, K_d (L kg^{-1})	$2.5 \cdot 10^3$ (range: $4.5 \cdot 10^2$ – $8.5 \cdot 10^3$)	No data Ce=$1.2 \cdot 10^2$ - $2.0 \cdot 10^4$; Dy=$8.2 \cdot 10^2$ - $2.1 \cdot 10^3$; Ho=$2.4 \cdot 10^2$ - $3.0 \cdot 10^3$; Lu=$5.1 \cdot 10^3$, La=$5.3 \cdot 10^3$; Sm=$2.4 \cdot 10^2$ - $3.0 \cdot 10^3$; Tb=$5.4 \cdot 10^3$ - $6.6 \cdot 10^3$; Tm=$3.3 \cdot 10^2$	No data
Soil to plant transfer, F_v (kg d.w. kg^{-1} d.w.)	No data	No data (Ce=$2.4 \cdot 10^{-4}$ -$2.0 \cdot 10^{-2}$ for all grain and $2.0 \cdot 10^{-2}$ - 3.5 for pasture; La=$6.0 \cdot 10^{-6}$ - $6.0 \cdot 10^{-3}$ for a number vegetation species; Pr=$2.0 \cdot 10^{-2}$ for cereals and leafy vegetables; Ce for fruit=$4.4 \cdot 10^{-4}$ – $6.2 \cdot 10^{-4}$)	$1 \cdot 10^{-1}$
Transfer factor, F_m (kg L^{-1})	No data	No data (Ce= $3.2 \cdot 10^{-3}$ for cow's milk)	No data
Feed transfer coefficient to cow's milk, F_m (day L^{-1})	No data	(Ce=$2.0 \cdot 10^{-6}$ - $1.3 \cdot 10^{-4}$)	$3.4 \cdot 10^{-5}$ - $6.8 \cdot 10^{-4}$
Feed transfer to beef, F_f (d kg^{-1} d.w.)	No data	(La=$1.1 \cdot 10^{-4}$ - $1.5 \cdot 10^{-4}$)	No data

*Gadolinium analogues: Ce, Dy, Er, Eu, Ho, La, Lu, Nd, Pr, Sm, Tb, Tm, Y, Yb.

Table 2: Description of the parameters

Parameter	Description
F_m	Concentration factor for milk
F_f	Concentration factor for meat
F_v, crops (kg w.w. kg^{-1} d.w.)	Concentration factor for crops in d.w. units
F_v, forage (kg w.w. kg^{-1} d.w.)	Concentration factor for forage in d.w. units
VT	Total deposition coefficient

1.2. Objectives

The project is divided in three main tasks defined in agreement with SSM:

- **Task 1.** Review the literature and current radioecological models (e.g. ERICA) for elements that may be released at ESS. Special attention will be given to gadolinium. *Sub-task 1.1*: Identify gaps in the existing radioecological models for the relevant elements, and to which extent they are relevant for the regional specific conditions outside the ESS-site. *Sub-task 1.2*: Evaluate how current models can be adapted to site-specific conditions outside the ESS-site and the city of Lund. The research should highlight strengths and weaknesses of the available radioecological models, including place specific parameters in the region. The research should focus mainly on accident scenarios.
- **Task 2.** Review and evaluate the local food production and consumption for identifying important existing pathways and critical groups. Part of this study will also be based on the previous baseline study (also referred to as zero-point measurements by Bernhardsson et al., 2018 [31]), where a network with members of the public and local farmers was established, enabling assessments of local farming, local household production as well as access to private wells. As an additional outcome of these assessments, it will also be possible to identify the amount of export of local products outside of the area in order to estimate the total (global) radiological impact of ESS-radionuclides taken up in agricultural products. The research should have main focus on accident scenarios including seasonal variations.
- **Task 3.** Conduct a measurement survey of the existing concentrations of stable Gd in ESS-local samples. In addition to a selection of samples (soil profiles, bioindicators, grass, crops, forage, milk, sewage sludge, ground and surface water), already available in the zero-point measurement sample bank, a number of new relevant samples will be taken. At least 50 samples will be sent for ICP-MS assay at Department of Geology (LU) for assay of stable Gd, Hf and Sm. *Sub-task 3.1*: Bibliographic review of the methods for the evaluation of the stable Gd and REE levels in the environment (sample preparation, analytical procedures by ICP-MS and calculations). *Sub-task 3.2*: Selection of samples from the previous zero-point project, and collection of additional samples (e.g. soil and associated crops). *Subtask 3.3*: Developing a method for extraction of Gd and other REEs from the newly collected samples as well as from those collected during the previous sampling campaigns before analysis by ICP-MS at the Geology Department (LU). The research should focus on method development.

Through the course of the project the scope of some tasks was slightly modified according to the outcome of the literature review and the experimental results.

2. Radioecological models in the context of the ESS

The European Spallation Source (ESS), which is being built near Lund, will generate a wide variety of radionuclides, resulting from its operation [1]. The radionuclides produced in ESS may be released to the environment during operation, as well as during potential incidents or accidents. The impact assessment of ESS operation to the environment is part of ensuring radiological safety of the installation. Apart from the facility being justified and optimised, operation of ESS also imply dose constraints that apply to the general public and staff, which should follow the SSM regulations, corresponding to an annual effective dose limit of 0.1 mSv y^{-1} for the general public. In order to, on beforehand of operation, quantify these doses the concentrations of radionuclides in the environment should be evaluated by a combination of radioecological modelling and environmental measurements. Although the radiological impact on the environment from operational and accidental releases of the most important radionuclides produced in a nuclear power plant is relatively well-known [32], [33], ESS is another case, due to higher importance of rare radionuclides with much less documented data regarding their environmental and biological behaviour. The aim of this part of the report is to review radioecological models used to evaluate radionuclide transfer in ecosystems, their strengths and weaknesses in the light of ESS operational and accidental releases.

2.1. Radioecological models

Mathematical models are used to describe and predict radionuclide transfer in different ecosystems and to evaluate their radioecological impact. The earliest radioecological models dates back in 1960s-70s [34], [35]. There are different methods to perform such studies: deterministic or probabilistic. Radioecological models may also be separated into equilibrium, dynamic and dynamic models with equilibrium data [36]. There are models which primary focus is on phenomenology of the chemical and biological processes occurring in the plants and in the soil. Such models are very useful to understand and quantify microscopic processes. On the other hand, there are models that does not include any microscopic processes and focuses only on radionuclide transfer with known transfer coefficients. The simplest models consist of transfer functions and describe the addition and losses of radionuclides in the specified object or system.

Examples of generic models that are widely accepted and used are presented in e.g. IAEA safety reports series No.19 [23]. At least several modelling tools use information provided in the IAEA Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments [30] as a basis – ERICA [22], CROM [21], etc. The IAEA report [23] describes simple methods for calculating doses arising from radioactive discharges into the environment. The scope of this project does not include analysis of dose coefficients and their role in the final dose assessment, but the focus is rather on the radionuclide transfer in the environment. Generic methods are described for estimating radionuclide concentrations in air, water, terrestrial and aquatic food products. Although the models are generic, the number of radionuclides covered in the publication is limited: the only isotopes of the lanthanides covered are ^{141,}

^{144}Ce, $^{154,\ 155}$Eu and ^{147}Pm, i.e. the set dose criteria do not cover the elements associated with ESS, such as Gd, Sm, etc.

Validation and testing of these models are vital to ensure their suitability and to understand limits of their applications. Between 2009 and 2011, the IAEA conducted a program dedicated to the development, comparison and testing of environmental assessment models for estimating radiation exposure of humans and radiological impacts on flora and fauna due to releases of radionuclides [37]. When testing different calculation models for many combinations of radionuclide and reference organism, the variability in predictions was found to range over three or more orders of magnitude. It was also found that the largest contribution to the variability between different model predictions was due to the differences in the way the transfer pathways were parametrised (comparisons among different radiological models can be found in the IAEA report [37]). The importance of subjective judgement must be stressed: the personal approach of the modeller and the parameter selection. If the scenario is not well defined, the modeller will tend to use a subjective interpretation based on his or her previous experience [38].

The report of the Swedish Nuclear Fuel and Waste Management Company (SKB) from Avila et al. [34] describes a model of the long-term behaviour in temperate and boreal forests of radionuclides entering the ecosystem with subsurface water. Radionuclides covered therein are important for high level radioactive waste management. The report is very detailed and covers several relevant references, questions, descriptions and discussions about processes [34]. The authors published another article discussing a similar topic with additional useful references [39].

In a book chapter by Seaman and Roberts [40], different components of generic soil are discussed, chemical processes governing radionuclide behaviour in soil, transfer mechanisms to the biota, theoretical models of radionuclide transfer in soil and water. The behaviour of the most important nuclides is also described. The authors stressed the importance of site-specific data for the development of both empirical and mechanistic models.

There are lots of examples of modelling applications, only a selection is given for illustration: a dynamic compartment model was developed to evaluate the transport of accidentally released radionuclides onto rice-fields [41]. Existing radiocaesium soil-to-plant transfer models were categorised and discussed [42]. The reliability of model results is defined by its uncertainties. The paper by Kirchner and Steiner [43] discusses sources of uncertainties in radioecological models and provides guidelines to improve their reliability.

Analogues, preferably a stable isotope of the same element, are often used to estimate the movements between various ecological compartments in absence of experimental data of the considered radionuclide. However, the use of analogues may still require extensive experimental data of the parameters governing its transport in a given ecosystem in order to describe the corresponding behaviour of the radionuclide. The timescales of the transport processes of the analogue as well as the biological, physical and chemical properties of the mixing and transport media are thus needed [44]. In addition, the metabolic behaviour in man of the analogue will be needed to make dose assessments from the radionuclide intake. A culprit in interpreting data from a stable analogue can be that the half-life of the considered radionuclide may be too short to reach chemical equilibrium in the mixing media into which it is released, and hence the equilibrium values found in the various compartments of the stable element can be misleading. With its long physical half-life of 75 y, the alpha emitter ^{148}Gd may, however, if released be more likely to reach transient equilibrium in the environmental transfer processes than compared with most short-lived ESS-nuclides. The ESS safety report [7] used radioecological data on La and Ce as analogues for the corresponding transfer of Gd-isotopes. In terms of ionic radius (1.06 and 1.03 vs 0.93 nm) and valence (all mostly form stable 3+ ions) they appear to be fairly similar with that of Gd [45]. Due to the abundance of La and Ce in nature and the larger amount of available data compare to other lanthanides it is not unreasonable that they have been used as Gd-analogues. A factor indicating against the use of La and Ce as analogues may be their lower atomic mass, and the fact that Ce can also form 4+ ions unlike Gd. A radioecological transport model based on stable Gd should, if possible, be used for a more accurate prediction of radioactive Gd transfer from atmospheric fallout to terrestrial species in which a fractionation of the REE can occur according to their mass [45].

The statistician George Box once said “All models are wrong, but some are useful”. Various simplification techniques are used to perform modelling when all the data needed does not exist: extrapolation, use of analogues etc. Beresford et al. [46] summarised that all necessary data will never be available to populate all of the potential radioecological modelling parameters for the assessments. Therefore, we need robust extrapolation approaches which allow us to make best use of our available knowledge to predict the fate of hitherto unmapped ESS radionuclides. Although most of the models are using linear transfer, one should not forget that the reality is more complex, especially in case of contamination with high radionuclide concentrations. A publication by Tuovinen et al. [47] discusses the non-linear effects and a way to include them in the modelling. Even though the use of chemical analogues of uncharted radionuclides is not an accurate way of modelling, it must occasionally be used if there is no other data available. Analogues can be used to provide input values for model parameters as default values, although care has to be taken not to push such analogies beyond their legitimate domain of applicability. Thorough knowledge is required of the system for which an analogue is sought to derive parameter values. Relevant processes and features have to be known, such as time scales of these processes, physical, chemical and biological properties of the environment and relevant media [48]. This essentially means that the local environmental conditions around the ESS vicinity must be characterised in such a way that the fate of dispersed rare radionuclides (e.g. ^{148}Gd and ^{146}Sm) can be predicted with sufficient accuracy to ensure compliance with the predefined dose constraints under normal operation (0.1 mSv y^{-1}), or whether an accidental release of such radionuclides will exceed a dose criterion (e.g. 1 mSv y^{-1} internal dose from intakes of contaminated foodstuff).

Studies suggest that there are strong similarities among all the lanthanides, though there are some subtle differences due to variations in the oxidation states along the series. Also, there are analogies between the lanthanides and higher actinides [13], [48].

2.2. Software

There are several programs suitable for radioecological modelling. They provide the opportunity to perform modelling in a clear and transparent manner, they may contain necessary parameters and models, although not all of them have this feature. Several of these programs are presented below.

The ERICA Tool [22] is a software system dedicated for assessment of the radiological risk to terrestrial, freshwater and marine biota. This tool allows its user to perform necessary calculations to estimate doses for animals and plants. ERICA allows assessment using approaches of different complexity (Tiers). The simplest one is dose assessment when knowing the concentrations in the media, while the most complex option is to run the assessment probabilistically if the parameter probability distribution functions are known. The tool has simple transport models included that are based on the IAEA Safety reports series No.19 “Generic models for use in assessing the impact of discharges of radioactive substances to the environment” [23].

RESRAD-BIOTA is another tool for radiation dose evaluation in biota, developed in Argonne (USA). There are three tiers of analysis implemented in this code. Both external and internal radiation exposure are included in the dose calculation. The code includes the possibility to estimate the transfer of radionuclides to animals. However, it includes quite a low number of radionuclides that are available for evaluation.

The CROM 8 [21] is a tool for the integrated assessments of effective doses for humans and absorbed doses for biota. As input, it requires contamination levels of environmental media such as air, freshwater, soil, etc. The software was developed using the IAEA Safety reports series No.19 [23] as basis, but some additional improvements were performed as well.

AMBER is commercial software developed in the UK that is used to study contaminants in environmental, biological and engineered systems using compartment models. The software is fully probabilistic and allows its users to simulate contamination from historic, routine, accidental and long-term release situations. AMBER may be used for modelling the transfer of radionuclides in the environment and through the food chain, following releases of radionuclides. AMBER can also be used to model the transfer of contaminants within the human body, following their ingestion using ICRP models.

GoldSim is another software dedicated for radionuclide transport studies, mostly focusing on transfer through engineering barriers. It has a Contaminant Transport Module that allows Monte Carlo modelling of mass transport processes in engineered and/or natural environmental systems.

Ecolego is a commercial software tool for creating dynamic models and performing deterministic or probabilistic simulations using compartments. It automatically handles nuclide decay and ingrowth. Ecolego is very flexible and transparent tool that allows the user to easily make own models, parameters and equations can be easily entered and modified as well. The user-friendly interface also allows time-dynamic graphical presentation of models and calculation results. Ecolego has already been used by SKB and ESS to perform safety assessments for the facility.

All the existing software programmes for a radionuclide transfer modelling has different flexibility (from the use of existing build-in models to a possibility to create own models), but the REE transfer coefficients are not included in these programmes. However, it is possible to add necessary data in the software and track transfers of interest. To sum up, the main obstacle for modelling REE transfers in the environment is not the software, but the lack of relevant experimentally justified transfer coefficients. Therefore, motivation of choosing a specific software for further development might be based on other factors, e.g. if it is recently developed, if the software was successfully used for the licensing and is accepted by the regulator. The experience in using software for the region-specific conditions might also be a factor. Ecolego fulfils these criteria: the newest version was released at the end of 2020 [49], it was accepted as a proper tool in licensing spent nuclear fuel repository in Sweden [50], Ecolego was also used by ESS in simulating radionuclide transfer to groundwater [51].

2.3. Radioecological data

A number of parameters that characterise radionuclide specific transfer rates between environmental systems that have importance for the aggregate radioecological transfer to man and biota [30] are listed in Table 1. Radionuclide transfer to plants depends on numerous factors including physical and chemical forms of the radionuclide, soil properties, plant species, plant compartment, farming practices, etc. Such factors result in a high variability, and the individual parameter values themselves can vary with more than three orders of magnitude.

Soil types were analysed and grouped in four major groups in the IAEA Tecdoc-1616 [48]: sand, loam, clay and organic. The soils were grouped according to the sand and clay mineral percentages, and the organic matter (OM) content in the soil (see Table 3). We propose that the best correspondence of these soils and the soils existing in southwest Scania/Lund environment is loam [52].

Table 3: Usual range of values for various soil parameters for four soil groups, from IAEA [48]

Soil group	pH	% organic matter	Cationic exchange capacity, CEC ($cmol_c\ kg^{-1}$)	Sand content in the mineral matter fraction	Clay content in the mineral matter fraction
Sand	3.5-6.5	0.5-3.0	3.0-15.0	≥65	<18%
Loam	4.0-6.0	2.0-6.5	5.0-25.0	65-82	18-35
Clay	5.0-8.0	3.5-10.0	20.0-70.0	-	≥35
Organic	3.0-5.0	≥20	20.0-200.0	-	-

Assessment of soil-to-plant transfer values, F_v, based on literature sources is always associated with many shortcomings, and very often considerable judgment must be performed in evaluating the available data. Such data are normally based on studies that were not originally intended for assessments of transfer factors and the experimental design may deviate greatly from the transfer factor definition.

For atmospheric deposition of Gd, the parameters interception and translocation in plants are important for the aggregate transfer to man. Interception is defined as the fraction of a radionuclide deposited by dry and wet deposition that is initially retained by the vegetation. Due to weathering, a fraction of the radionuclides moves to the soil, but another fraction is intercepted directly to the plant, and this fraction might be the cause of high initial activity concentrations in the plant. Data on interception factors for elements of fission products, such as ^{137}Cs, ^{131}I, ^{90}Sr etc. are abundant, whereas there is little information on the interception of atmospherically deposited REE elements [53]. In the ESS safety assessment [54], a generic factor of 0.3 for crops and 3 for forage was assumed for all elements.

Translocation is the process that describes how the radionuclide is transferred from the contaminated area to the other parts that have not been directly contaminated, e.g. transfer from root to edible parts of a plant. IAEA presents translocation factors, f_{tr}, in terms the ratio of the activity over a ground surface of the edible part of a crop at harvest time (Bq m^{-2}) to the foliage activity of the crop at the time of deposition (Bq m^{-2}) expressed as a percentage. In other words, this parameter describes what fraction of the intercepted radionuclides will end up in the edible parts of various plants.

The translocation factor represents the ratio between the transferred activity in the various parts of the plant that were not contaminated directly and the deposited activity on the plant itself. Its expression and, therefore its meaning, vary among authors [48]. There are also many experimental protocols and results from them are very heterogeneous. However, the ESS safety assessment bypassed IAEAs definition of translocation by instead characterising the uptake of elements in crops by the soil-to-plant factor, F_v, defined by IAEA [48]. The soil-to-plant factor directly relates the concentration in edible parts of a given crop to the concentration in the top soil layer [23]. As an example, ESS used values for the chemical analogue La for leafy vegetables as an estimation for Gd uptake in crops (a central estimate of 0.000456 kg w.w. kg^{-1} d.w.). This value is almost 90 times lower than the corresponding factor for the alkali metal Cs. An estimate of the parameter F_v could be experimentally obtained if comparing the Gd equilibrium concentration in crops with that of the wet and dry mass soil concentration at different layers, e.g. the top 5 cm soil layer of cropland.

The review in IAEA Tecdoc-1616 provides the state of knowledge about the values of the translocation factors, defined as the ratio between total activity in the edible parts of the plant and activity retained on its foliage (percentage) [48]. The analysis of the scientific literature concerning this parameter of transfer highlighted an important lack of knowledge. Especially the interaction of translocation and plant development is poorly investigated for many types of crops and radionuclides. Furthermore, there are practically no data on translocation factors in chronic contamination throughout the vegetative cycle of a crop. Also, the majority of the available data relate to caesium and strontium. Radioisotopes of some other elements (Mg, Co, Fe, Ru, Sn, Ce, Ba, Zn, Te, Hg, Cr, Na, Ca, Be and Pb) have been investigated, but the data are far from sufficient to obtain reliable values for all plant types. Furthermore, there is no data mentioned on the ESS-related elements Gd, Sm and Hf. The translocation factor, as well as the soil-to-plant factor for chronic contamination could be experimentally tested by sprinkling irrigation studies on representative crops such as wheat or sugar beets (see section 3 of this report) using trace radionuclides such gamma emitting ^{153}Gd for the estimate of F_v and f_{tr} for ^{148}Gd.

Radionuclide mobility in plants, and thus in the food chain, is strongly affected by its sorption in soils, which can be estimated by the quantification of the solid-liquid distribution coefficient (K_d). This parameter may vary within various orders of magnitude depending on the radionuclide and soil type. A K_d database with around 2900 records of 67 elements can be found in [48]. This data come from field and laboratory experiments, with various contamination sources, considering mainly the scenario of soils contaminated by radionuclides. There are still evident gaps of K_d values for a substantial number of radionuclides and soil types. In some cases, values originate from just a single reference. For these gaps, the use of analogues (data on other elements or media, such as pure soil phases or sediments) is an option, but must be undertaken with care considering similarities and differences in the chemical properties of the analogue. K_d values for the element Gd in various soils outside ESS can be estimated by determining the ratio between the Gd concentration in liquid phase with that of Gd in dried soil.

The ESS safety assessment modelled the uptake in crops and fodder by combining the aforementioned concentration factors, F_V, with another parameter, V_T, that translates the daily rate of atmospheric deposition (sum over both wet and dry deposition) density to a top soil concentration ((Bq m^{-2} d^{-1}))/(Bq m^{-3})) (=m d^{-1}) defined in reference [23] assuming generic values of 1000 m d^{-1} for most type of elements.

Transfer to animals is important considering doses to humans. The transfer to meat and milk can be expressed by the IAEA defined variables F_f (d kg^{-1}) and F_m (d kg^{-1}), respectively, which are the ratios between the activity concentration of a radionuclide in beef or fresh milk, respectively, compared with the corresponding concentration in the daily ingested fodder. For most radionuclides, the concentration ratio data compiled varies little between the species considered (sheep, goats, cattle, horses and poultry). Therefore, concentration ratios derived for one species could be applied to another. Unfortunately, many authors that report transfer coefficients do not provide the information required to estimate concentration ratios. A number of radionuclides have recently been identified for which there is currently inadequate information for modelling or assessment including ^{36}Cl, ^{237}Np, ^{99}Tc, U isotopes, Th isotopes, ^{241}Am, ^{59}Ni, ^{94}Nb and ^{60}Co [48]. In addition, there is still no data on F_f for the ESS-specific elements Gd, Sm and Hf. ESS used values for the analogue La for estimating F_f for the element Gd in meat, and the REE Ce for transfer to milk, F_m. Generally, the transfer to milk and meat for Gd is anticipated to be about more than 300 times less than for the alkali metal Cs [55].

In general, the suitability of a particular definition of a transfer parameter depends on the purpose of the radioecological model, the manner in which the data used to calculate the transfer parameter have been obtained and the level of knowledge of the ecosystem to be modelled. The latter is often limited by the availability of field data, either obtained directly by measurement or from the literature.

In conclusion, the approach for mapping the knowledge gap concerning the relevant transfer factors for the REE Gd should be to first determine ratio between equilibrium Gd-concentrations in the various compartments associated with terrestrial agriculture practices in vicinity of the ESS (e.g. top soil, top soil water, dry and fresh mass of crops and fodder, cow’s milk and meat, etc.). These values should then be compared with the values for chemical analogues used by the ESS safety assessment to estimate what potential impact variations in the transfer parameters may have on the concentration levels in foodstuff and associated internal dose to human consumers. The temporal variation in the uptake and release of Gd in these compartments can, however, only be determined through trace studies which require additional effort in terms of accessibility of test husbandry, management of used gamma emitting tracers, ethical approval, etc. Lysimeter[1] and sprinkle studies in analogy with what have been used for studying the translocation and transfer of radiocaesium to edible parts of crops [56], may also contribute valuable knowledge on what contamination levels can be anticipated from a given atmospheric dispersion of radioactive gadolinium, and for which scenarios radical food restrictions may be necessary. The next section shows the results from a survey of the local food production around ESS and potential risk groups, illustrated in a map centred around ESS.

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[1] **1:** Experimental set-up dedicated to the study of the relations between water, contaminants and biota in soil

3. Evaluation of the food production around the ESS

After an accidental release, particles of irradiated target material would be spread on the soil and plants surrounding the facility. The uptake of this material by the plants and animals that are part of the food chain could later lead to human contamination. To identify important transfer pathways, the specific area around ESS was surveyed. This section contains a summary of the identified local ecology and populations.

The environment around ESS in the northern part of Lund is dominated by arable lands, which are classified 8-10 on a ten graded scale of the lands yield capacity [57]. These uniquely fertile soils are mainly found in southwest Scania. Within a radius of about 1.5 to 2.5 km from ESS there are today a limited number of private houses, with the small village of Östra Odarslöv in the northeast direction from ESS. The area around ESS is expanding and more inhabitants are expected in the future when ESS is in operation, for example at the Science village that is to be built in-between MAX IV and ESS.

To evaluate the current food production and risk groups, in case of a severe accident at ESS, the area has been surveyed by maps and by several visits in the area. As one important part of this, a questionnaire has been developed and sent to some key persons and organisations that have been identified as representative for farming and the population near ESS. The interviews were done in-person or by telephone and were based on eight different questions related to the food production (e.g. what, how much, where, end consumer), animals and hunting. Approximate positions of the main farmlands and producers close to ESS (most of which are included in the ongoing Zero Point Assessment program [31]) as well as current and future key populations are shown in Figure 1.

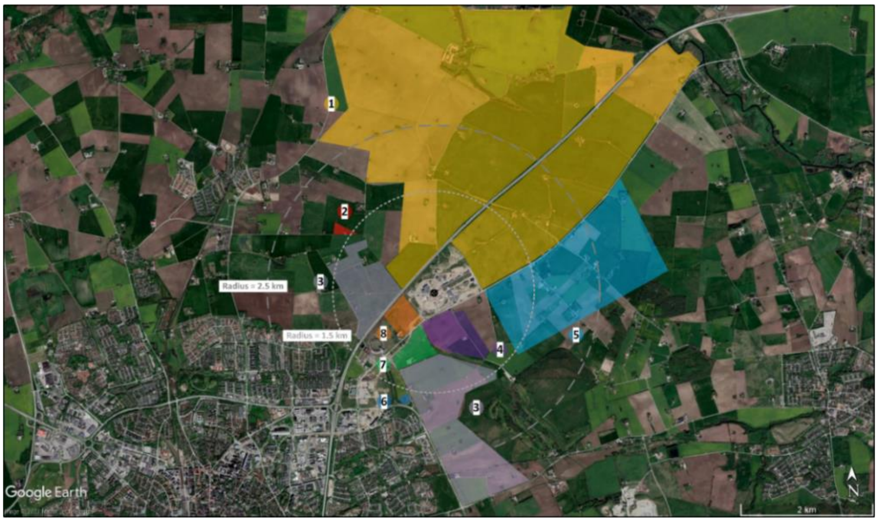


Figure 1: Illustration of identified areas around ESS that are relevant and representative for the food production and population in the area. The coloured areas are approximate locations of: 1) farmlands belonging to Svenstorps estate management, 2) Glorias apple orchard, 3) farmland owned by the Swedish Church, leased and operated by Lundaslättens drift, 4) a private farmer, 5) part of Östra Odarslöv (mainly private houses and some farmlands), 6) Brunnshögsodlingen (urban cultivation), 7) Kunskapsparken (recreation area and future location of the Brunnshögsodlingen), 8) Science village (prel. start of construction 2021).

Although the areas identified and highlighted in Figure 1 do not represent the complete number of local food producers in the area, they provide a representative picture of what is regularly grown in the area, with the main producers 2020 included. Below is a summary of the different areas that are relevant for the local impact of a radioactive deposition outside the ESS premises:

1. **Svenstorps estate management:** is the largest privately owned farming industry in Sweden with about 1500 ha (1 ha = 10^4 m^2) of farming land in Lund. Crop rotation of wheat, barley, rape, and small amounts of corn is applied. The annual yield from these farmlands is 8-9 tonnes of wheat per ha, 7 tonnes of barley per ha, and 3.5-4.5 tonnes of rape per ha. The majority of what is produced is sold as seeds (Sv. “utsädes”) to Lantmännen, but it is up to Lantmännen to decide the final use of the grains, and it may be distributed to different parts of Sweden. The corn is sold to Swedish Agro. For the coming years it is planned to change the corn production to growing of sugar beets. No animals are kept by the estate, and no land is leased out for keeping cattle. There is a local team of hunters that are active on the land.
2. **Gloria’s apple orchard:** this privately owned farm grows apples (Discovery, Katja, Aroma, Rubinola, Santana, Elstar, Ingrid Marie and Frida) on about 12 000 trees distributed over 3.24 ha, plus some 100 trees with pears. About 20-25% of the annual yield is sold as “self-picking”, another part is sold in various forms in the farm shop, and one part is sold to a local cider factory. Each year bees are hired for pollination. The honey is sold to a local reseller (Honungsgården).
3. **Lundaslättens drift:** is the second largest farmer in the area with about 270 ha of fields. On the fields west of ESS there are peas, and on the other fields (south of ESS) grow wheat, malt grains, rape seeds and sugar beets. Their annual approximate yield from these farmlands is 9 tonnes of wheat per ha, 4 tonnes of rapeseed per ha, 70 tonnes of sugar beets per ha, 7 tonnes of malt grains per ha. The harvest is sold for food production to e.g. Karlshamn grain reception (rapeseed), Nordic sugar (sugar beets), Lilla Harrie vallskvarn (wheat), Pågens bageri, Foodhills AB (peas).
4. **Private farmer:** the house of this private farmer is located in the middle of the 43 ha farmlands, and a small part of the farmland is currently not in use. This farmer is applying crop rotation of autumn wheat, bread wheat, malt grains, and grows sugar beets each year (on various fields). The annual yield is approximately 90 tonnes of malt grains, 200 tonnes of wheat and 500 tonnes of sugar beets. The crops are sold to Örtofta sugar mill and to Svenska Foder (Knästorp), for production of sugar, flour and beer.
5. **Part of Östra Odarslöv:** apart from the north-eastern parts of Lund, the majority of the private houses are located in this section of the map (also in the main wind direction from ESS). The population in Östra Odarslöv (18 ha), as of 2015, was 70 persons [58]. Within the marked area is also some small farmlands and an apple orchard, although not in full operation yet.
6. **Brunnshögsodlingen:** this urban community farming has 100 m^2 of land for individuals to rent a part of, or for groups of individuals, for private farming. Among other things, there is a large green house,

bushes with berries and a chicken farm. Soon, this land lot will be moved to the park area of the Science village (Kunskapsparken).

7. **Kunskapsparken:** The Brunnshögsodlingen will be moved to the southern part of the park area of the Science village and will cover approximately 1.9 ha. The park is estimated to be finished in 2021-2022.
8. **Science village:** this 18 ha area is to expand with the start of the ESS. As part of the research infrastructure between MAX IV and ESS it will hold facilities such as University and college branches, guest houses, fitness centre, restaurants and many other services. The initiative was presented in 2009 and is expected to develop for many years.

The interviews revealed only one team of hunters that hunts for game in the area (shooting mainly rabbits and deer). Apart from private gardens, bees are kept privately and have also been kept at e.g. MAX IV for pollination of nearby fields and production of honey. At the south-eastern part of the map (next to area “3” in Figure 1) there is a grazing area for cattle.

The overall outcome of the survey shows that there are several actors that are important for what is produced in the area, except for what is grown in private gardens. The main identified crops in 2020 was: wheat, barely, rape, malt grains, sugar beets, peas and apples. Limited amount of corn was grown in 2020 but this production will be exchanged to sugar beets in the future. It can be noted that the majority of what is grown in the area (from Svenstorps estate “1” in Figure 1), is currently not sold for food production but as seeds. There are no cattle in the area, apart from grazing cattle in the south-eastern part of the ESS. Hunting of game is ongoing but rather limited, partly due to the infrastructure in the area.

Based on this survey some key potential pathways for transferring ESS dispersed radionuclides from plant to man have been identified. Today there is one defined group that can be considered for defining the *representative person* in the area, consisting of the inhabitants of the private houses along Odarslövsvägen (area “4” in Figure 1), only a few meters south of ESS. In the future, there will be several guest houses located in the Science Village (area “8” in Figure 1), some hundreds of meters southwest from ESS. Considering that the private dwellings all have garden plots with home grown vegetables and fruits, are located very close to ESS, these few houses will be of particular interest also in the future for defining the representative person around ESS. This together with current and future farming in the area is of importance for the next steps, as mentioned before, in determining specific transfer and uptake factors, of the area specific plants and soils, of REEs and in particular gadolinium.

4. Assessing the gadolinium levels in the surroundings of the ESS

This section contains a literature review on the existing methods to extract and assess REE in the environment focusing on the data relevant to the study of the ESS site followed by a pilot study performed on a soil sample from the ESS vicinity.

4.1. Background and context

According to the existing data, the natural REE levels are expected to be rather low around the ESS. The maps produced by the Global Geochemical Baselines Programme for the Forum of European Geological Surveys (FOREGS) provide values for many of the REEs in soil, sediment and water for the whole continent. The range of concentration in topsoil obtained from those maps for the area surrounding the ESS are reported in Table 4 [59]. It is worth noticing that the closest measurement point used for producing theses maps seems to be 50 km north of the ESS (potentially with different soil types). The other available set of topsoil data comes from Tyler and Olsson [60] that analysed forest samples collected in Scania, but from soil conditions that are very different than around the ESS. Those values are also reported in Table 4.

Table 4: Ranges of concentration of REEs in topsoil (in mg kg^{-1})

Element	Geological maps Salminen et al [59]	Scanian forest soil Tyler and Olsson [60]
Scandium (Sc)	6.32-8.21	0.92-5.16
Yttrium (Y)	13.0-17.0	4.9-17.6
Lanthanum (La)	10.7-18.1	5.5-33.2
Cerium (Ce)	28.3-36.8	11-68
Praseodymium (Pr)	2.5-4.3	1.3-7.5
Neodymium (Nd)	12.2-15.7	9.3-53
Samarium (Sm)	1.8-2.3	0.9-4.6
Europium (Eu)	0.6-0.8	0.22-0.83
Gadolinium (Gd)	2.4-3.0	1.0-4.8
Terbium (Tb)	0.37-0.46	0.15-0.65
Dysprosium (Dy)	2.10-2.60	0.9-3.7
Holmium (Ho)	0.53-0.68	0.20-0.74
Erbium (Er)	1.6-2.0	0.63-2.2
Thulium (Tm)	0.23-0.30	0.09-0.33
Ytterbium (Yb)	1.99-2.40	0.60-2.3
Lutetium (Lu)	0.30-0.35	0.09-0.34

REE concentrations in several types of plant samples can be found in the work of Tyler et al. The authors measured REEs in grass from central Scania [61], moss from Scanian forests [13], the leaves of 9 plants species such as lily of the valley collected in Scanian forest [62] and beech forest litter (dead leaves) from Hässleholm (Scania, Sweden) [63].

The literature lacks examples of measurement of ^{148}Gd or other radioactive REEs in the environment [3] . However, several articles present ICP-MS measurement of radioactive lanthanides including ^{148}Gd in irradiated target materials. Kerl et al. [20], Day et al. [64] and recently Chiera et al. [6] have demonstrated that it is possible to extract and separate radioactive REEs contained in irradiated tantalum targets and to analyse the isotopic composition of each element using ICP-MS coupled to chemical separation techniques. It may be difficult to apply methods developed for concentrated material once there are spread and diluted in soils, water and biota. Hence, there is a knowledge gap regarding the suitability of ICP-MS to measure a ^{148}Gd contamination in the event of an accidental release by the ESS.

The following chapter contains, first a short literature review of the methods for the evaluation of stable Gd and the REE levels in the environment (sample preparation, analytical procedures by ICP-MS and calculations) and some applications of interest in the context of the ESS. The second part of the chapter presents experimental tests of methods that were performed on soil samples collected around the ESS to investigate their REE content, with emphasis on the existing isotopes with an atomic mass of 148.

4.2. Extraction of REE

This section describes the different methods that can be applied to extract REE from various types of environmental samples and the possible outcome from such sample preparation techniques.

4.2.1. Extraction of REE in soils

4.2.1.1. Total extraction and pseudo-total extraction

Total extraction refers to the extraction of the full amount of an analyte from the sample matrix. In the case of soils or sediments, it implies the dissolution of all the minerals contained in a sample, which is not an easy task. This is most commonly used for geological studies where the purpose is to obtain the complete composition of the sample. Ramos et al. [65] report many examples of the use of acid mixtures or alkaline fusion for total extraction of REEs from soils before analysis by ICP-MS. Acid mixtures include hydrofluoric acid combined with nitric, hydrochloric or perchloric acids put under high pressure and temperature in Teflon vessels [66]. The other possibility, named alkaline fusion, is based on the addition of salts such as $LiBO_2$ or Na_2CO_3 to the sample before heating at high temperature in metal crucibles [66].

The alternative to the total extraction is the pseudo-total extraction. Using this method, only the fraction of the sample that can be dissolved under strong (but less extreme) extraction conditions is obtained. The remaining insoluble fraction is considered to be very inert and will not interact with the environment. Therefore, this is not the most relevant part of a sample and its contribution can be neglected. Pseudo-total extraction procedures usually mix the sample in a solution of aqua regia that is heated. The heating can also be performed in pressurised vessels in a microwave to shorten the time or to increase the yield of the extraction [60], [67].

4.2.1.2. Sequential extraction

The principle of sequential extraction means that more information can be obtained from complex samples such as soil, as compared to the total concentration of metals only [68]. The same aliquot of a sample can be successively extracted by solutions of increasing strength (increasing acid concentration) or by extractants of different nature (oxidising agents, reducing agents, etc.) in order to provide data on its different components, also called fractions. In terms of environmental studies, another important parameter to consider is the bioavailability [69]. All the metal-trace content of a soil is not entirely available to living organisms. The most easy-to-extract trace metal fractions are the most relevant to study because they have the highest bioavailability (i.e. being more easily transferred to plants, animals and microorganisms).

In the 1980’s and 1990’s, the lack of uniformity in sequential extraction methods for trace metals led the European Commission to start a collaborative group to work on standardization and production of reference material [70], [71]. The task was managed by a European programme called at the time the Community Bureau of Reference (BCR), and then, the Standards, Measurements and Testing Programme (SMT) [72]. The outcomes of the programme were inter-comparison exercises and a standardised three-step extraction method, further referred as BCR method [73]:

- Fraction 1: acetic acid extractable metals
- Fraction 2: acidic hydroxylamine extractable metals
- Fraction 3: acidic hydrogen peroxide extractable metals

The BCR method was later improved to compensate some flaws of the original design [72], [74] as well as to include additional steps such as the study of the extraction residues [75].

Another popular approach is the method developed by Tessier et al. [68] that was developed for the extraction of particulate trace metals. It was initially tested on sediment samples and is composed of four extraction steps:

- Fraction 1: exchangeable
- Fraction 2: bound to carbonates
- Fraction 3: bound to iron and manganese
- Fraction 4: bound to organic matter

Both the BCR and Tessier inspired extraction methods have been designed more specifically for REE assessment in soils and sediments. Thus, it is possible to find examples of methods from the literature suitable for various types of soils. Other studies have a stronger focus on the bioavailability of the REE and their authors modify the standardised extraction methods according to this purpose. Rao et al. [76] developed a BCR 3-step method to analyse Indian soils, including some samples from a site contaminated by mining operations and accidents. Šmuc et al [77] analysed paddy soil using a Tessier 4-step method. Mittermüller et al [67] based their work on BCR but split the first step in two in order to assess the role of phosphates in the bioavailability of REE in soils and tailing materials. Wiche et al [78] used a 6-steps method suitable for both germanium and REE, that they applied on soils in mining areas in Germany. To get a better understanding of the REE bound to Fe, Mn or Al, they split the third step of the Tessier method into non-crystalline and crystalline oxides.

4.2.2. Extraction of REE in biological samples

An extraction method is usually more straightforward when living organisms are analysed. Indeed, the purpose of such analysis is to measure the content of REEs in the full organism or in some of its organs and tissues. It is thus unnecessary to sequentially extract the samples.

Common chemical extraction methods are applied to leach the REE from the samples. It usually involves a mineralisation step: the mixing of the sample in a strong acid solution while heating. This type of extraction can also be performed in pressurised vessels heated in a microwave oven to accelerate the process or to increase the extraction yield. This method is usually referred to as microwave digestion. Since REEs tend to accumulate differently in each part of the plant (root > stem > leaf > flower > fruit and seed, according to Liang et al. [79]), many authors analyse the different parts of the plant separetely. If the plant samples grew in the environment (instead of being fed with REE in a laboratory), the soil from the sampling site is also usually extracted and measured [80]. Wiche et al. described such a microwave extraction on rapeseed, barley and maize: their samples were dried, ground and digested in a mixture of nitric and hydrofluoric acid [78].

The same type of procedure can be applied to animal samples. Perrat et al. [81], [82] describe the following procedure: the samples (small aquatic organisms) are dried and then mineralised in nitric acid (Figure 2). The obtained solution is then diluted and stored at 4°C before analysis by ICP-MS. Macmillan et al. [83], that analysed various types of animal samples, were adapting their method to the sample nature: an acid mineralisation for invertebrates at low temperature and a microwave digestion for the mammal and fish tissues. Similar methods can also be used to determine REEs in honey, Squadrone et al. [84] used a mixture of 70% HNO_3 and 30% H_2O_2 (v/v) for microwave digestion of honey samples.

Figure 2: Example of extraction procedure from Perrat [81]

There is no difference between the methods that are used for REE contamination tests performed on plants and animals in labs or for samples collected in the environment.

4.2.3. Extraction of REE in water samples

Water samples do not usually require specific pre-treatments before analysis by ICP-MS and can thus be analysed directly after a dilution in acid (typically a 2% nitric acid solution) [19]. However it is possible to purify or pre-concentrate a sample by evaporation liquid-liquid extraction or extraction chromatography [18], [85].

4.3. Analysis of REE by ICP-MS

4.3.1. Method

4.3.1.1. Instrumentation

ICP-MS is a widely used technique for the analysis of traces, ultra-traces and isotope ratios of both stable elements and radionuclides [86]. The technique is fast and can provide simultaneous determination of all trace and ultra-trace elements in aqueous solution. It has an excellent sensitivity, good accuracy and very low limits of detection: 0.001–0.1 pg mL^{-1}. These low limits of detection (LoD) combined with precise low volume injection systems enable the assessment of very small levels of analyte (ng to fg). The precision of the trace element determination is ±2–5%. In the case of isotope ratio measurements, the precision can go down to 0.001% using multicollector-ICP-MS [86].

One of the drawbacks of this analytical technique is the difficulty to analyse ions with the same mass-to-charge ratio (m/z) or the formation of molecular ions (metal with H^+ for example). These phenomena are also called isobaric interferences and are of particular importance in the case of REEs since there are several isotopes with the same masse in this series of elements. Therefore, it is important to select the isotopes of each element that could be measured without interferences. Tyler et al. [62] selected the following isotopes for their REE measurement: ^{89}Y, ^{139}La, ^{140}Ce, ^{141}Pr, ^{142}Nd, ^{147}Sm, ^{151}Eu, ^{153}Eu, ^{158}Gd, ^{159}Tb, ^{164}Dy, ^{165}Ho, ^{166}Er, ^{169}Tm, ^{174}Yb, ^{175}Lu. If one wants to obtain the isotopic composition of an element of the REE series, it is necessary to chemically purify the sample before analysis [6], [87].

4.3.1.2. Normalisation of the REE results

The rare earth elements follow the Oddo-Harkins rule: the even-numbered elements are more abundant than the odd-numbered ones [88]. This phenomenon leads to a “saw-tooth” pattern in the abundancy plots (see Figure 3). When the full REE series is measured, it is possible to normalise the data to obtain a smooth curve that makes the comparison between elements easier. The REEs have very homogenous chemical properties and usually form trivalent REE^{3+} ions in solutions. There are however two exceptions: Eu^{2+} and a partial oxidation of Ce to Ce^{4+}. Those two elements are therefore exposed to additional chemical processes that make their concentration less predictable [89]. It is also worth noticing that promethium does not exist in nature, and thus do not normally appear in REE series measurement of environmental samples [88].

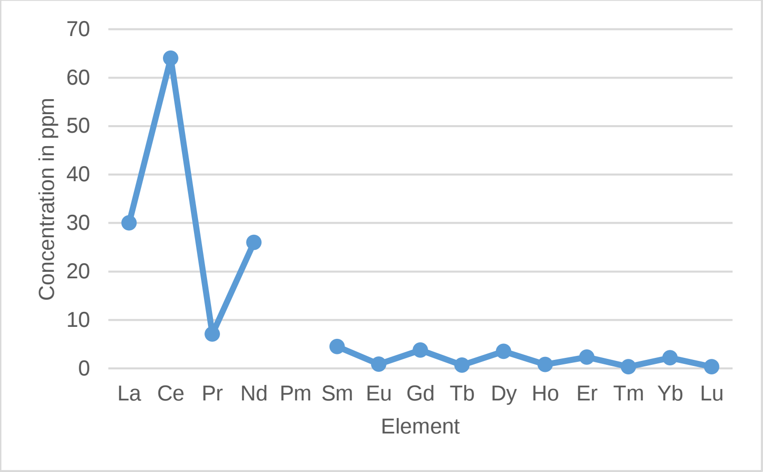


Figure 3: Composition of the upper continental crust from McLennan [90]

In terms of soils, the normalisation is performed using data measured in certain types of sediments (shale) that are representative to the composition of the Earth upper crust [90]. Values commonly used for this purpose are the PAAS (Post Archean Australian Shale) [91], the NASC (North-American Shale Composite) [92] and the EUS (EUropean Shale) [92]. For water and biological samples, the normalisation can be performed using shales but also with the local sediment or soil values. Those local values allow a better visualisation and detection of an excess or lack of a particular element since the plants or animals were in contact with this particular soil or sediment. Those excesses or lacks are called concentration anomalies and are often due to human activities.

4.3.2. Applications of ICP-MS measurements in solution in the context of the ESS

Many examples of the use of ICP-MS to analyse REE can be found in the literature with a very wide range of purposes and outcomes. The following paragraphs present a selection of studies that may be relevant in the event of an accidental release of ESS target materials in the environment, however, with first a need of assessment of the current levels of REE in the soil, water and locally grown plants, then the knowledge of the transfer factors to those plants in case REE contamination and eventually the decontamination of soils.

The first domain of application is environmental monitoring. Here, the purpose is to investigate the concentration of the REE series or the isotopic composition of a specific element. Depending on the purpose of the study, the analysis of one type of sample may be sufficient. Information on natural background levels before a contamination (zero-point assessment) or anthropogenic contaminations can be obtained that way as well as the evolution of REE concentrations over time.

As mentioned in the section 4.1, many local [60] or global [59] geological surveys of REEs in soils have been performed. The need of reliable and large scale data mapping arose after the Chernobyl accident in 1986 when it was realised that the baseline of pollutant (including radioactive ones) could not be defined [59]. There are also examples of studies focusing on the detection of anthropogenic contaminations by REE in the environment and their evolution: Plausinaitis et al. [18] analysed water samples from Chernobyl area and found a positive concentration anomaly of erbium in local rivers. The same method is used by Hatje et al. [19] to monitor the increasing concentration of Gd in water samples in the San Francisco bay over the years due to the increasing use of Gd as a contrast agent in MRI.

Once a baseline has been established and an enhanced level of contamination has been detected, the scope of the ICP-MS applications extends to the field of ecotoxicology which can be defined as "the science of contaminants in the biosphere and their effects on constituents of the biosphere, including humans" [45]. The REEs originating from the natural soils or human activities can be transferred to the biosphere with potentially detrimental consequences. There is no consensus on the effect of low levels of REE on plants even though they are used sometime as fertilisers [45], [65], [93], [94]. On the other hand, their toxicity for mammals and humans (in particular Gd toxicity) has already been demonstrated [15], [45], [65], [93]. One of the relevant question regarding the fate of REE in the environment is whether they can enter the human food chain and what is the threshold of their toxicity [45], [65]. When studying biological samples, it is thus important to consider the transfer of REEs within the environment and to the living organisms. In order to do so, the transfer factors from soil to plant or from water to aquatic animals need to be calculated. Such data can then be used to estimate the bioaccumulation of REEs in organisms or in some of their organs.

ICP-MS is a relevant tool to estimate what fraction of the REEs in soil is available to the plants and other soil organisms (i.e. the REE bioavailability). As described in section 4.2, authors have extensively studied the fractionation of metals, and in particular of REE, depending of the extraction conditions [67], [76]–[78]. All of these studies relied on ICP-MS measurement first being used to develop their extraction methods and secondly to assess the REE concentrations in each extraction fraction obtained from environmental samples. For example, Wiche et al. [78] investigated the REE levels in different types of soil in a mining region in Germany.

In the case of plants or animals that belong to the human food chain, it is desirable to study whether the species exhibit significant bioaccumulation capabilities as this would influence the risk of REE transfer from foodstuff. Once again, this type of study on transfer factors can be performed in a controlled environment of a laboratory with plants fed by solutions of REEs or in the field by sampling plants grown on soil containing high levels of REEs. For example, Rezaee et al. [95] fed pak choi (*Brassica rapa*) and sunflowers wit La and Nd to study their accumulation and toxicity for the plant while Wiche et al. [78] analysed rapeseed, barley, maize grown in mining areas in Germany. In the context of the ESS, it means that limited data already exists on foodstuff species cultivated around the facility such as apples [96] or honey [84] and that the methods mentioned in these articles could be replicated on other plant grown in the vicinity of the ESS. The literature on REE in the species of plants grown close to ESS is summarised in Table 5. It is interesting to note that there is no data available on some of the main species cultivated around the ESS area.

Table 5: Literature data on REE levels in local foodstuff

Foodstuff grown near ESS	Articles of REE in foodstuff	Reference
Wheat	D'Aquino et al. (2009); Ding et al (2005)	[97], [98]
Barley	Wiche et al. (2017)	[78]
Rapeseed	Wiche et al. (2017),El-Ramady (2008)	[78], [99]
Corn	Wiche et al. (2017); El-Rhamady (2008); Diatloff et al. (1995)	[78], [99], [100]
Apples	Sager et al. (2015)	[96]
Honey	Squadrone et al. (2020)	[84]
Green peas	No data	
Sugar beet	No data	

The plants that accumulate REEs, thanks to higher soil-to-plant transfer factors, can be used to efficiently decontaminate a polluted soil by applying o-called phytoremediation. Phytoremediation is the use of plants to make metal contamination in soil nontoxic thanks to the accumulation of the contaminant in the plant. It is a low cost, and rather fast, strategy for soil rehabilitation that can also allow recycling of the extracted metals [101]. Authors have chosen to study the bioaccumulation in plants with known affinity for other metals, or to investigate the properties of plants widely distributed in contaminated areas. Mikołajczak et al. [102] investigated the bioaccumulation properties of common herbaceous plants such as the common dandelion growing next to roads. Zhenggui et al. [103] followed another approach by studying a fern species known for its bioaccumulation in a region contaminated by REE.

Global studies of all the components of an ecosystem are possible but rarer due to the complexity of the systems and the number of samples to consider. MacMillan et al [83] made studies in fresh matter, marine and terrestrial environment in eastern Canadian Arctic. Their studies include soil and sediments, water, plants and animal (including species hunted for food like trout). Another approach is to compile sets of data from different articles in a review study. Liang et al. [79] used this method to provide an overview of the REE levels in the environment and humans working and living in Chinese mining areas.

4.3.3. Laser ablation

ICP-MS is not limited to the analysis of solutions. Laser ablation ICP-MS (LA-ICP-MS) is an alternative technique that allows its users to analyse the surface of solid samples directly without chemical extraction [104]. This method works according to the following principle: the surface of the sample is ablated by a laser beam and the aerosol produced is then transported by a carrier gas to the plasma where it is ionised and finally to the mass spectrometer where it is analysed. The LA-ICP-MS can also map the chemical composition of the surface and provide depth profiles up to several hundreds of µm [104]. Like all ICP-MS techniques, LA-ICP-MS is sensitive to interferences and matrix effects. Proper calibration with standards is thus mandatory to properly perform quantitative analysis [104].

LA-ICP-MS is mainly applied to geological samples including those for REE analysis [104], [105]. For example, Neves et al. [105] analysed REE in soil cores (depth = 35 cm) from cultivated and uncultivated sampling sites. They observed a trend of higher REE concentration in the cultivated soils.

The LA-ICP-MS is not limited to soil samples. it is also possible to apply the method to plants. As an example, Kötschau et al. [106], [107] applied both type of methods to study the accumulation of metals, including REE, in sunflowers. The authors used mineralisation to study individual parts of the plant [106] but also laser ablation to map concentration in metals on the whole area of the leaves [107]. Lingott et al. [108] also mapped gadolinium concentrations in several aquatic and terrestrial plants such as cress to determine the transfer pathway of the Gd uptake in the plants analysed.

4.4. Test of extraction and analysis of REE in ESS soils

In this pilot study, methods for the extraction of REE and their measurement by ICP-MS were tested in order to answer the following questions. Is ICP-MS suitable to assess the baseline levels of REEs around the ESS? Is it possible to establish a baseline of the isotopes of mass 148 before an event of contamination by ^{148}Gd? Can sequential extraction method provide relevant data for modelling purposes?

4.4.1. Selection of samples

Four soil samples were selected among the archived samples collected during a previous project [31]. Those samples are 20-centimeter-deep top soil samples collected around the ESS. The location of the samples sites is presented in Figure 4.

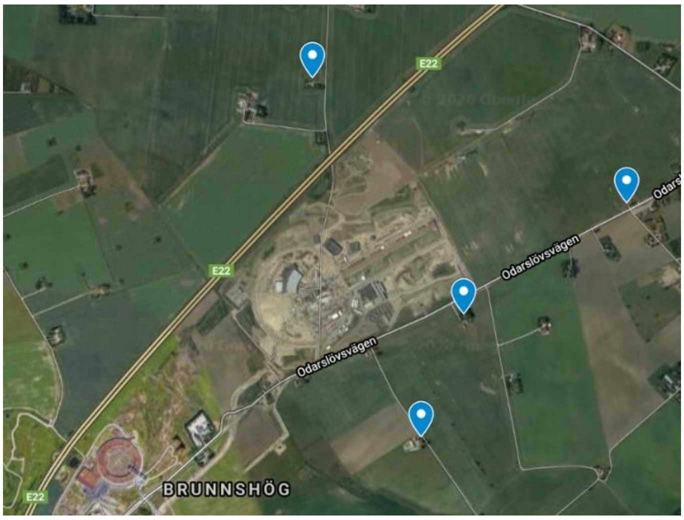


Figure 4: Location of the sampling sites around the ESS

The sampling was performed in cultivated grounds or near roads, and include two types of sites that may present elevated levels of REEs compare to undisturbed locations due to the car trafficking and the use of fertilisers [65], [102], [105]. Therefore the REE levels could already be higher around the ESS facility than what the existing data show in the rest of the region (c.f. Table 4). For comparison purpose and to validate the method developed hereafter, a reference sample of estuarine sediment with certified values of REEs (BCR-667, JRC, IRMM) was also analysed.

4.4.2. Test of extraction methods

Four soil samples were randomly selected among the archive samples collected during a previous project [31]. Those samples are 20-centimeter-deep top soil samples collected around the ESS.

The selected samples were first extracted sequentially based on Wiche et al [78] method:

- Fraction 1: mobile exchangeable soluble materials (1 M ammonium acetate pH = 7)
- Fraction 2: acid soluble elements (1 M acetate buffer pH = 5)
- Fraction 3: element bound to oxidisable matter (acetate buffer pH = 5 + oxygen peroxide at 45°C)
- Fraction 4: non crystalline fractions, Fe-, Mn-, Al-oxide (0.2 M ammonium oxalate pH = 3.2)
- Fraction 5: crystalline Fe-, Mn-sesquioxides (0.2 M ammonium oxalate + 0.1 M ascorbic acid, 45°C)

Other aliquots of each soil sample were extracted according to the method developed by Mittermüller et al. [67]:

- Fraction 1: easily soluble, ion exchangeable (0.05 M calcium nitrate)
- Fraction 2: carbonate bound and mobilised by complexation (0.1 M citric acid)
- Fraction 3: reducible fraction (0.5 M hydroxylamine hydrochloride pH = 2 or 0.1 M nitric acid)
- Fraction 4: acid soluble fraction (1.4 M nitric acid)

Those extraction methods were compared to a pseudo-total extraction with aqua regia (aqua regia 60°C to dryness and redissolved in 2% nitric acid).

4.4.3. Inter-comparison exercise

In order to assess the proficiency of several international laboratories performing ICP-MS measurements, an intercomparison exercise was organised using seaweed samples already measured in the past as well as a certified seaweed reference material and a certified sediment reference material. The exercise is still on-going and all the laboratories have not yet provided the results for the reference materials. The participants were asked to measure several metals such as lead, mercury and cadmium but also one REE, gadolinium. The obtained results for this element are presented in Table 6. The full results will be available in another report (Mattsson et al. SSM2020-797 [109]).

Table 6: Gd values in seaweed samples measured in part per billion (ppb) by participating ICP-MS laboratories and the relative standard deviation in % (%RSD)

Concentration (d.wt.), ppb ± %RSD (lab. A), ± SD (lab. C)			
Element	Gd		
Laboratory ID	A	B	C
Sample 1	47.4±4.6%	15	85.4±0.2
Sample 2	89.5±4.0%	21	77.5±0.01
Sample 3	132±3.4%	20	135.1±1.8

The values provided by the participants differ significantly, in particular the ones from the laboratory B. In anticipation of the full results, the laboratory that was the closest to the known values of the seaweed samples was selected for the further test of extraction methods presented hereafter. If discrepancies are observed in the future between the results provided by this laboratory and the reference values, corrections will be applied.

4.4.4. Test of REE analysis by ICP-MS

4.4.4.1. Sample preparation

Sample preparation before analysis by ICP-MS was performed according to Rao et al. [76]. Samples were diluted in 1% HNO_3 with different dilution factors depending on the nature of the extractant used in each fraction to minimise interferences from calcium and sodium: 1:10 for all mild extracts (e.g. 0.1 M $NaNO_3$, 0.01 M $CaCl_2$, 1 M NH_4NO_3); 1:50 for complexing agents (e.g. 0.005 M DTPA, 0.05 M EDTA) and acid solution 0.43 M CH_3COOH; 1:100 for 1 M HCl and aqua regia.

4.4.4.2. Operating conditions

Samples were measure on a Bruker Aurora Elite ICP-MS instrument at the Geology Department (Lund University). The following list of nuclides were measured: ^{45}Sc, ^{89} Y, ^{139}La, ^{140}Ce, ^{141}Pr, ^{146}Nd, ^{148}Nd, ^{147}Sm, ^{153}Eu, ^{157}Gd, ^{159}Tb, ^{163}Dy, ^{165}Ho, ^{166}Er, ^{169}Tm, ^{172}Yb, ^{175}Lu. The concentration calculation for each element was performed assuming their respective isotopes were present in the sample at their natural abundance.

4.4.4.3. Quality assurance

A 2% nitric acid solution was used for blank measurements and a REE solution (Rare earth element mix for ICP, 16 elements, TraceCERT®, Supelco®) was used as a primary standard to calibrate the ICP-MS instrument. A calibration curve, using this solution diluted to 0.1; 1; 10 and 100 ppb in the same 2% nitric acid solution was established. A solution of ^{103}Rh was also used as an additional internal standard.

In addition, a certified reference material (BCR-667 estuarine sediment, JRC, IRMM) with certified concentrations of REEs was extracted according to the Mittermüller method and used to validate the method.

4.4.5. Results and discussion

The four soil samples collected around the ESS (labelled E4, E258, E331 and E344) were prepared by pseudo total extraction with aqua regia and two sequential extraction methods before ICP-MS analysis. The reference sediment sample followed the same aqua regia but only the second sequential extraction method was applied to it. The full analysis of these samples is presented in the following sections.

4.4.5.1. REE series

The efficiency of the aqua regia (pseudo-total extraction) can be estimated by comparing the certified concentrations of REEs in the reference sediment sample with the result of its ICP-MS analysis. The results of this series of measurement are presented in Figure 5. The estimated efficiency is not homogeneous through the REE series. The higher values were obtained for the middle-REEs from Sm to Tb with a maximum for Gd (72%). This range of efficiencies is expected for heavy metal [110]. Indeed, an aqua regia treatment is not sufficient to dissolve all minerals but it is considered to be representative of the bioavailable fraction [110]. The comparison between the sediment and the soil samples is however limited by their different mineral composition.

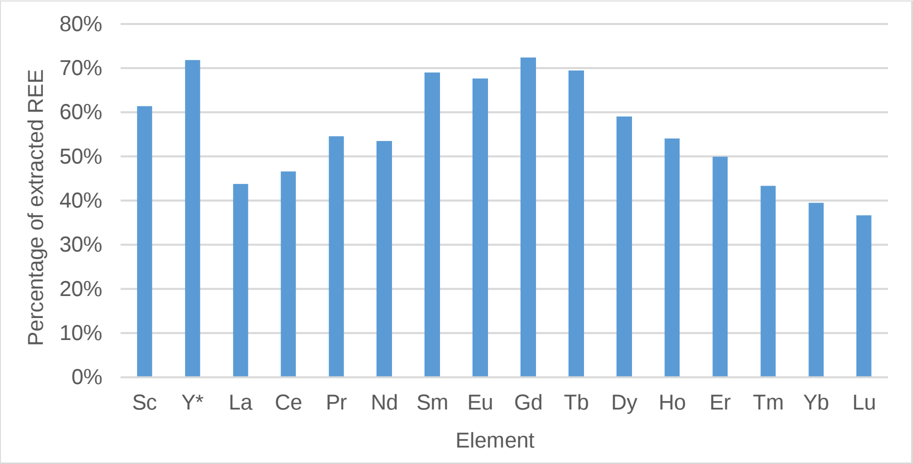


Figure 5: Percentage of REEs in a certified reference material, BCR-667 estuarine sediment, extracted by aqua regia (* = Y concentration is only indicative and not certified)

The range of concentration of REE extracted by aqua regia extraction in the ESS soil samples are presented in the Table 7.

Table 7: REE concentration range in aqua regia extracted ESS soil samples (mg kg^{-1}) compared to literature values

Element	ESS soil sample (N = 4)	Scanian forest soil Tyler and Olsson [60]
Scandium (Sc)	1.94-3.44	0.92-5.16
Yttrium (Y)	6.57-10.32	4.9-17.6
Lanthanum (La)	13.14-19.03	5.5-33.2
Cerium (Ce)	27.97-39.19	11-68
Praseodymium (Pr)	3.18-4.86	1.3-7.5
Neodymium (Nd)	11.66-18.43	9.3-53
Samarium (Sm)	2.13-3.46	0.9-4.6
Europium (Eu)	0.35-0.57	0.22-0.83
Gadolinium (Gd)	1.95-3.04	1.0-4.8
Terbium (Tb)	0.27-0.42	0.15-0.65
Dysprosium (Dy)	1.29-2.04	0.9-3.7
Holmium (Ho)	0.24-0.38	0.20-0.74
Erbium (Er)	0.66-1.07	0.63-2.2
Thulium (Tm)	0.08-0.14	0.09-0.33
Ytterbium (Yb)	0.51-0.84	0.60-2.3
Lutetium (Lu)	0.07-0.12	0.09-0.34

All REE concentrations measured are in the range determined by Tyler and Olsson on Scanian forest soils [60], with the exception of the lowest values of Tm, Yb and Lu which are slightly lower than the one measured in forest soils. No significant excess of REE can be observed in these samples, collected in a heavily cultivated area compared to the forest soils of the same region. This seems to indicate that the current local agricultural practices do not increase the REE content of the soils.

However, the plots of land themselves, where the samples were collected, are not cultivated. It would be interesting to compare these REE values to samples collected in actual fields near the ESS. The assessment of the REE concentration of cultivated and non-cultivated soil samples could form a strong baseline of the ESS surroundings that would stay relevant even if the agricultural practices change in the future (with the use of REE as fertilisers for example).

4.4.5.2. The case of Gadolinium-148

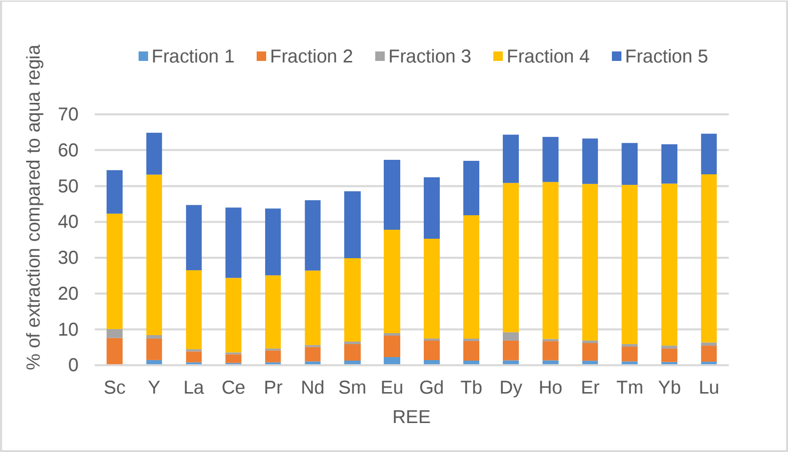
There is currently no ^{148}Gd in the environment around the ESS but the stable ^{148}Nd (5.756 % natural abundance) and the very long lived ^{148}Sm ($t_{1/2}$ = 7 x 10^{15} y; 11.24 % abundancy) may be found in today samples. Thus, it is worth investigating the natural content of isotopes of mass 148 in ESS soil samples. In the event of a release of ESS target material in the environment, ^{148}Gd would be introduced in soils together with other shorter lived radionuclides, namely ^{148m}Pm ($t_{1/2}$ = 41.3 d) and ^{148}Eu ($t_{1/2}$ = 54.5 d) [111]. It is also possible that some amounts of stable elements of mass 148 could be produced in the ESS target by spallation or decay of the spallation products. To our knowledge, data on the composition of stable elements of the target are not available and would require additional calculations.

Experimentally, the concentration of Nd in the sample can be calculated with the same accuracy based on the ^{146}Nd signal alone or based on the ^{148}Nd one after correction of the ^{148}Sm with concentrations of Nd of 14.57 and 14.56 mg kg^{-1}, in sample E4 for example. This suggest that ^{148}Nd and ^{148}Sm are present in the sample at their natural abundance with no particular enrichment of these isotopes. Thus, it is reasonable to assume that the sample contains between 0.66 and 1.05 mg kg^{-1} of ^{148}Nd and between 0.24 and 0.39 mg kg^{-1} of ^{148}Sm.

The presence of ^{148}Gd could hypothetically be detected with the same type of ICP-MS measurements that was performed in this report. An increase of signal at the mass 148 would be observed but it could not be quantified using external standardisation as it here was done for ^{148}Nd. Indeed, this calibration approach is based on the abundance of natural isotopes only. To quantitatively assess ^{148}Gd would therefore require a Gd material with a known ratio of ^{148}Gd for calibration based on isotopic dilution. However, such a material may not be easy to obtain. Another possibility would be to separate Gd from the other REE to determine isotopic ratios in the sample. Unfortunately, this type of separation is particularly difficult to perform due to the very similar chemical properties of all REEs [6], [87].

4.4.5.3. Sequential extraction

The two extraction methods described in section 4.4.2 were applied to the four soil samples. The objective of these selective extractions was to find a method that would accurately assess the fraction of REEs that is bioavailable. The **Figure 6** and **Figure 7** here after presents the percentage of REEs extracted by each step of the sequential extraction methods tested in this pilot study.


Fraction 1
Fraction 2
Fraction 3
Fraction 4
Fraction 5
% of extraction compared to aqua regia
70
60
50
40
30
20
10
0
Sc
Y
La
Ce
Pr
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Gd
Tb
Dy
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Tm
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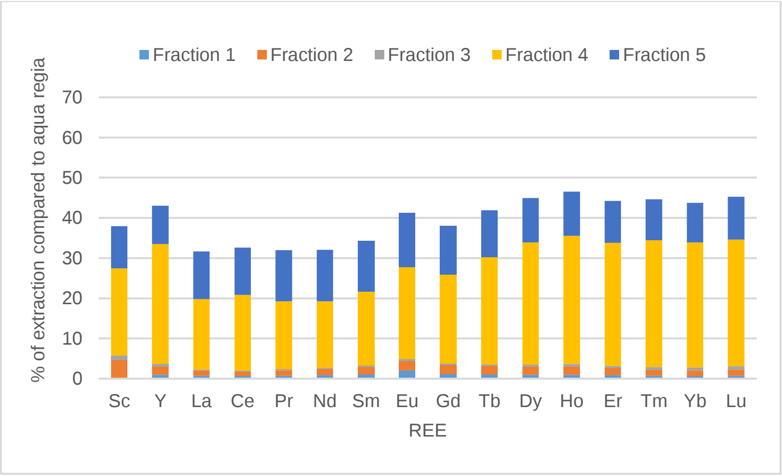
Lu
REE

a.


Fraction 1
Fraction 2
Fraction 3
Fraction 4
Fraction 5
% of extraction compared to aqua regia
70
60
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b.

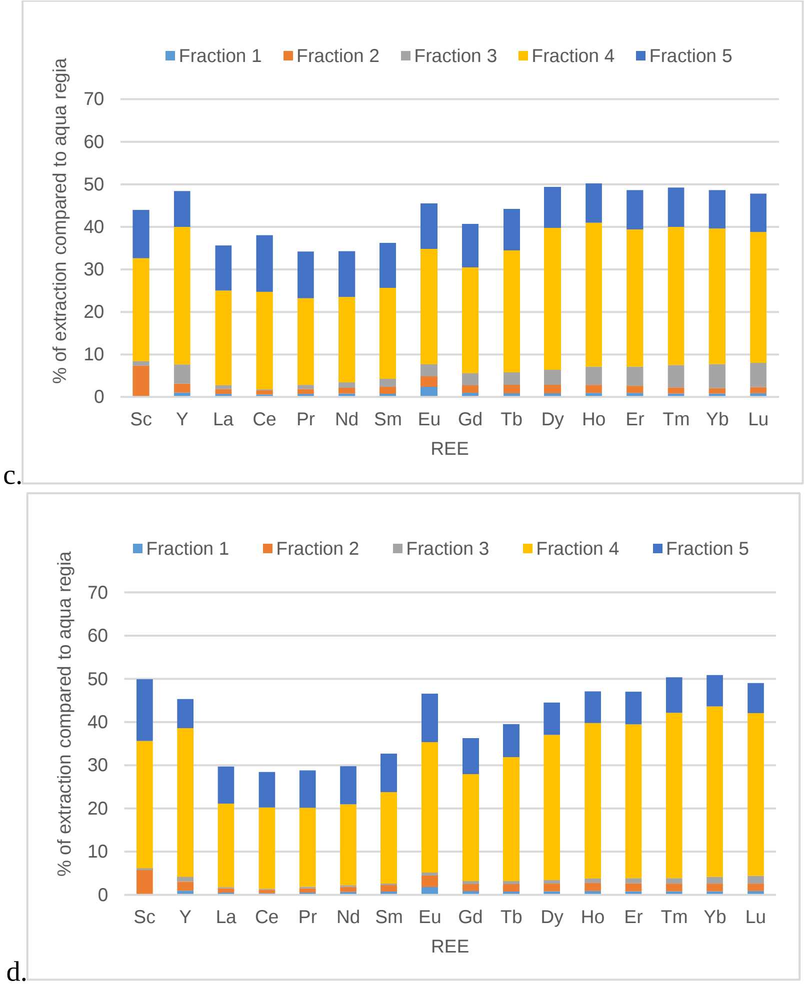
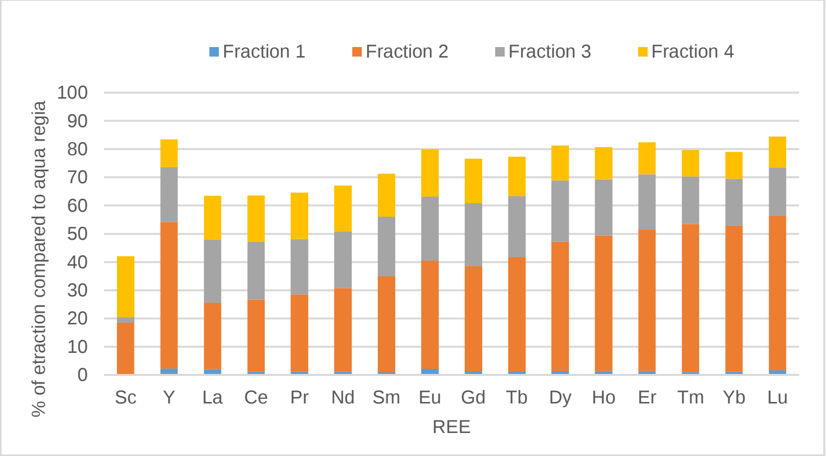


Figure 6: Sequential extraction of REE in ESS soil samples a) E4, b) E258, c) E331, d) E344 using the 5-step Wiche et al. method [78] (see section 4.4.2 for the detailed methods)

a.


Fraction 1
Fraction 2
Fraction 3
Fraction 4
% of etraction compared to aqua regia
100
90
80
70
60
50
40
30
20
10
0
Sc
Y
La
Ce
Pr
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Sm
Eu
Gd
Tb
Dy
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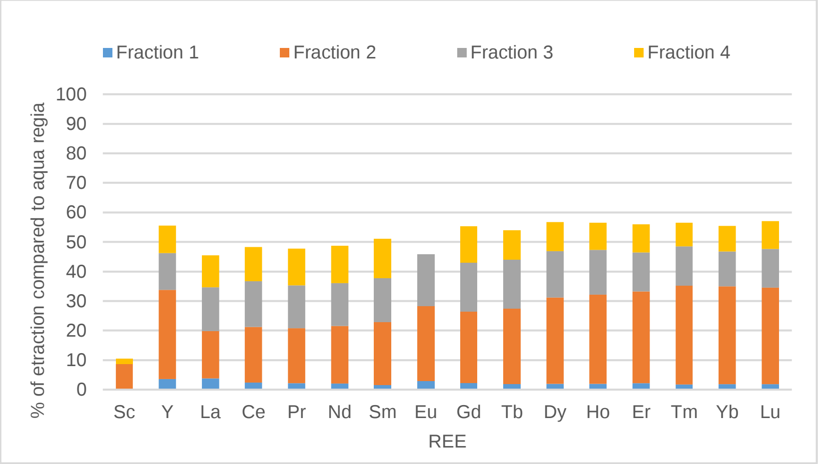
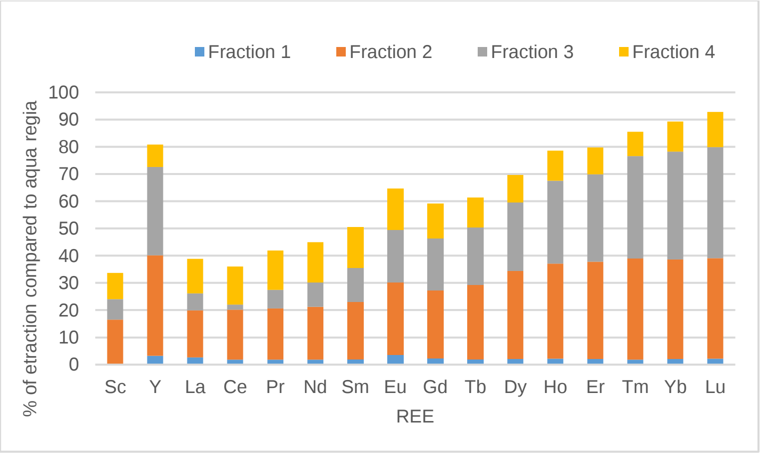
Tm
Yb
Lu
REE

b.


Fraction 1
Fraction 2
Fraction 3
Fraction 4
% of etraction compared to aqua regia
100
90
80
70
60
50
40
30
20
10
0
Sc
Y
La
Ce
Pr
Nd
Sm
Eu
Gd
Tb
Dy
Ho
Er
Tm
Yb
Lu
REE

c.



d.

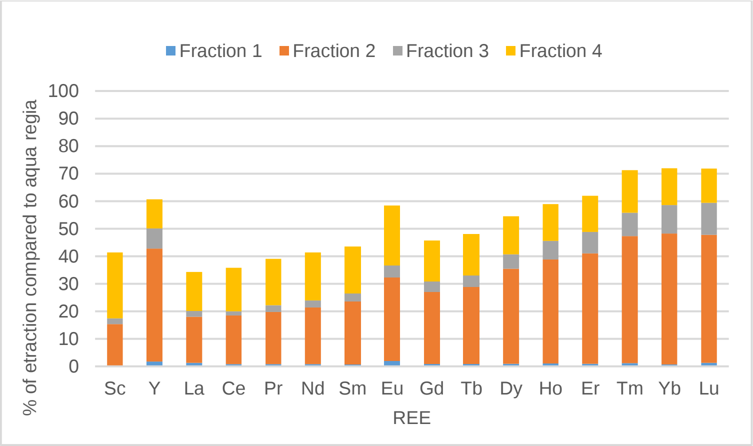


Figure 7: Sequential extraction of REE in ESS soil samples a) E4, b) E258 (where Eu in the fraction 4 was an outlier), c) E331, d) E344 using the 4-step Mittermüller et al. method [67] (see section 4.4.2 for the detailed methods)

The data obtained from the first method, in Figure 6, shows that the sequential procedure extracts 30 to 60% of the amount extracted by the pseudo-total extraction which is consistent with the results obtained by the authors of the method [78].The three first steps of the method (acetate buffer at pH = 7, pH = 5 and pH = 5 with hydrogen peroxide) extract less than 10% of the REE content. The third step (ammonium oxalate at pH = 3.2) seems to be the most efficient one of the methods, mainly due to the property of the oxalate ion to form metal complexes. It also seems to be increasingly efficient from the lighter REEs (about 20%) to the heavier ones (up to 45%). The last step (oxalate with ascorbic acid at 45°C) add another 15 to 20% to the sum.

The second sequential method, by Mittermüller et al. (Figure 7), lead to a more variable range of results but on average it extracts a higher proportion of the total lanthanide content than the first one, with values ranging from 35 to 90%. In some cases however, those numbers are actually higher than the ones obtained by the authors of the method [67]. The first step (calcium nitrate) barely extracts a few percent of the total amount. In the second step (citric acid), the efficiency is increasing with the mass of the lanthanide going from 15-25% of La and up to 55% of Lu. The third and fourth steps (0.1 and 1.4 mol l^{-1} of nitric acid) add the last 5 to 45% with a more homogeneous effect on all elements in two samples, but a similar increasing efficiency with mass for the two other. Citric acid is also a well-known complexing agent. Sc and Y behaviours differ from the rest of the series with a low extraction yield of Sc (only 10 to 40%) and a high extraction yield of Y (more than 50 to 80%).

Mittermüller et al. [67] explain that the three main fractions of REEs available to plants are: easily soluble and ion ex-changeable, mobilised by complexation, and mobilised under reducing conditions. From the results of both methods, we can say that the dominating process making REEs bioavailable in soil sampled in the ESS vicinity is the complexation. Indeed, in both case, most of the REEs are extracted by the complexing agents, namely oxalates and citric acid.

The results obtained during this pilot study indicate that extraction methods based on complexing agents seems to be the most promising way to assess the bioavailable fraction of REEs in soils and thus to produce experimental data for modelling purposes.

The use of a sequential extraction method for REE could also provide information about the nature of a contamination and its evolution in time by looking at the modification of the REE fractionation in soil. Such experiments could indicate if the contamination is easily soluble or bound to organic materials or certain minerals as it was done with radiocaesium after nuclear accidents [112].

4.4.5.4. Conclusions

From the results of this pilot study, we were able to answer some of the important research questions that are still pending regarding the environmental monitoring of radioactive REE releases from the ESS.

Is ICP-MS suitable to assess the baseline levels of REEs around the ESS?

- Yes, ICP-MS is a suitable for this purpose. However, our laboratory intercomparison highlighted the importance of a standardised calibration method to perform proper quantitative analyses. The measurements performed on soil samples collected in the vicinity of the ESS matched the levels measured in other parts of Scania – and do not seem to present any sign of existing contamination of REEs.

Is it possible to establish a baseline of the isotopes of mass 148 before an event of contamination by ^{148}Gd?

- Partially: the concentration calculation performed on ^{146}Nd and ^{148}Nd gave the same results which could indicate that ^{148}Nd and ^{148}Sm are present in the soil at their natural abundance. Unfortunately, this type of elemental analysis would not be sufficient to quantify the content of ^{148}Gd in a contaminated sample. The isotopic composition of Gd in a contaminated sample can however be performed by multi-collector ICP-MS instruments, although it requires a selective extraction of Gd beforehand (c.f. the method developed by Chiera et al. [6] or Olszewski et al. [87]).

Can sequential extraction method provide relevant data for modelling purposes?

- Yes, the tested sequential extraction methods provided interesting data on the bioavailability of REEs and their fractionation of REEs in these soils. The REEs were mainly extracted by complexation agents that can be produced by plants and thus contribute to their transfer from soil to biota. This phenomenon should be considered when building transfer models or design experiments to develop such models.

5. Summary and conclusions

There are many mathematical models that are well-established as tools for studying the fate of radionuclides in the environment. They have mostly been used to study radionuclides related to the nuclear fuel cycle and atmospheric nuclear fission releases and deposition, such as ^{137}Cs. However, nuclide specific radioecological model parameters are lacking for emerging REE contaminants such as ^{148}Gd. The currently existing software programmes, such as Ecolego, do not include REE transfer factors but they are usually flexible and such data may be added to them when available. The IAEA has established a list of relevant transfer parameters that are applicable in the context of the ESS radionuclides. To estimate these parameters, and to provide data for further detailed modelling of various accident scenarios at ESS, in-field and laboratory experiments may be conducted using either short-lived radioactive tracers of Gd, stable Gd itself or its chemical analogues, such as La or Ce.

A rough, but representative, mapping of the nearby ESS area in terms of land-use has been established based on networking with several key organisations and individuals operating or living close to ESS. The result from that survey has identified radiological important pathways in the food chain as well as current and future “critical groups” among the local inhabitants in the vicinity. For better understanding and modelling of the radiologically important radionuclide ^{148}Gd in the specific environment around ESS, this mapping hence provides a first basis. In order to make accurate predictions of doses to the critical groups identified it is important to first evaluate the observed levels of stable Gd in the main types of crops and fruits harvested in the area. Ultimately, it will be inevitable to experimentally study transfer parameters for crops and soil components to achieve new data on REEs, especially Gd, for better modelling the impact of a severe accident at ESS and its impact on critical groups.

ICP-MS was investigated as a possible technique to assess gadolinium and REEs in the environment and in particular in the type of environment found around the ESS site. Methods to extract REEs from water, soil and biological samples were presented. In the case of soil samples, sequential extraction can provide additional information on the relative mass distribution of the REEs in the different types of minerals. ICP-MS is a suitable method for the analysis of the elemental composition of samples or their isotopic composition after a chemical separation. ICP hyphenated with laser ablation can also enable direct quantitative assays of solid samples such as plants and to map REE elements.

The applications of ICP-MS in environmental studies span from geological assessments to environmental monitoring. The method has been used to study the transfer of REEs from soil to plants and animals and their bioaccumulation in those organisms. This literature review suggests that ESS-relevant plant species such as sugar beets should be studied to fill important knowledge gaps.

An experimental pilot study was conducted in order to test some of these analytical methods on ESS specific soil samples. The results confirm the potential of ICP-MS to quantitatively assess REEs in the context of ESS, both to establish a baseline of these elements and to provide data to build transfer models. The pilot study also identifies a direction for future research using ICP-MS to establish the baseline of the elements of mass 148 before a future contamination by ^{148}Gd from ESS target.

6. Outlooks

The studies and reviews conducted in this report point toward several ways to continue to reduce the knowledge gaps that still exist regarding the fate of the ESS-related radioactive REEs in the environment, with focus on the specific environment around the facility.

The determination of a baseline environmental level of REEs as well as their transfer from soil to biota could provide the lacking experimental data to model the behaviour of REEs in the local environment. Indeed, the software to build these models already exists but the transfer parameters are not often implemented and are instead calculated from analogues, which is a method that presents some limitations and uncertainties. The survey of the local food production around the ESS combined with the literature review of REEs in plants highlighted the lack of data on REEs in these products, and in particular Gd, in some locally relevant plant species such as sugar beets and peas. It would also be valuable to investigated REE levels in game, harvested in the area around the facility, since there are hunters active in the area. Interception experiment of sprinkled particles over a number of crops representative of ESS vicinity (barley, rye and rapeseed), and experimental design mimicking the one conducted by Gärdenäs et al. [56], [113] for wet deposition of ^{134}Cs could also be performed. Such interception experiments should be done using Gd or one of its analogues, such as La or Ce, in order to benchmark the similarity between the physicochemical behaviour of the elements, and thus providing an indication of the general agreement in environmental behaviour. Further studies on the size distribution and the chemical state of released oxidised tungsten particles, such as the TOAST-project (European Spallation Source (ESS) [114], [115], need to be conducted, in order to take into account the importance of these parameters in the transfer modelling

Before any experimental studies on the transfer parameters of gadolinium in soil and plant species can be done, efforts must first be done to identify suitable analytical methods for assessing radioactive gadolinium concentration in various matrices as well as the extraction and separation methods of ^{148}Gd (and other relevant REE isotopes) that would fit the best each analytical technique. For ^{148}Gd, three techniques seem to be the most promising: alpha spectrometry, inductive coupled plasma mass spectroscopy (ICP-MS) and neutron activation analysis (NAA). Alpha spectrometry of ^{148}Gd requires a more in-depth investigation, especially for separation and radiometry of low concentration, as would be the case in an accidental atmospheric release scenario at the ESS. In the case of ICP-MS, the focus should be on the preparation methods that are able to separate Gd from the other REEs, in order to provide isotopic composition of the gadolinium contaminated samples. Preliminary tests are currently planned to analyse stable Gd in samples of soil and plants using a new NAA set-up developed at Lund University. Thanks to the very high neutron capture cross section of Gd isotopes [116], it may be possible to measure and quantify these isotopes in environmental samples such as soil, but also in man [116], [117]. Finally, a comprehensive investigation of the detection limits for environmental concentrations of ^{148}Gd for all these analytical techniques should be made to provide a basis for prioritizing future emergency preparedness measurement strategies. In the case of alpha spectroscopy, this investigation should be conducted alongside with other alpha emitting ESS radionuclides such as ^{154}Dy and ^{146}Sm.

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