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ENVIRONMENTAL RADIOLOGICAL STUDIES OF KVARNTORPSHÖGEN

Dose and Radiological Risk Assessments for Humans and Biota

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Essay/Thesis:	30 hp
Program and/or course:	Medical Physics Programme
Level:	Second Cycle
Term/year:	Fall 2020
Supervisors:	Rimon Thomas, Francisco Piñero García, Mats Isaksson
Examiner:	Magnus Båth

Abstract

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Background: The pile Kvarntorpshögen and the lakes Surpölen and Norrtorpssjön are the remains of an industry which mined and processed oil shales to extract oil during World War II. The pile mainly consists of shale ash, a waste from the production, while the lakes are nowadays water-filled mining pits. Both are by-products of an industry with "Naturally Occurring Radioactive Material" (NORM), making them "Technologically Enhanced NORM" (TENORM). The Kvarntorp area thus has somewhat increased concentrations of radionuclides, and the radiation exposure that may arise to humans and biota have to be assessed, to protect against radiation risks, both now and in the future.

Aim: The aim of this project was to carry out a radiological characterisation on and around Kvarntorpshögen by measurements of ^{238}U, ^{235}U, ^{234}U and ^{210}Po in soil, water and plants in the area, and calculating the transfer of ^{210}Po and U-isotopes in the soil-root-plant system. A second aim was to perform a radiological risk analysis for humans and biota by estimating effective doses and absorbed doses, respectively.

Method: During the summer of 2020, samples of water, shale ash, soil and plants were collected on and around Kvarntorpshögen. The U-isotopes and ^{210}Po were analysed by their alpha emission and the samples were prepared through radiochemistry, to be measured by alpha spectrometry. Transfer factors were calculated by ratios of activity concentrations in root and soil, plant and soil, and plant and root. The radiological risk analysis for humans was performed by conservatively estimating effective dose rates through ambient dose equivalent rates and comparing to average yearly effective doses. The radiological risk analysis for biota was performed by estimating absorbed dose rates with the ERICA Tool (Environmental Risk from Ionising Contaminants: Assessment Tool) Tier 1 and Tier 2.

Results and Conclusions: External dose rates in the Kvarntorp area are highest by exposed alum shale walls at the pit lakes and by shale ash at Kvarntorpshögen. The activity concentrations of ^{238}U, ^{235}U, ^{234}U and ^{210}Po was seen to decrease in the soil-root-plant system at the pile. ^{238}U, ^{235}U, ^{234}U and ^{210}Po are in equilibrium in soil and roots, but not in plants, probably due to atmospheric deposition of ^{210}Po. The activity concentration of ^{238}U, ^{235}U, ^{234}U and ^{210}Po in plants at the Serpentine ponds are in equilibrium, and are slightly lower than in plants at Kvarntorpshögen. For water in the Kvarntorp area, the highest activity concentrations were found mainly in Surpölen because of its acidity, second highest in Norrtorpssjön and lowest in the Serpentine ponds. In the water, ^{210}Po was not in equilibrium with the U-isotopes due to differences in solubility. In the Serpentine ponds a decrease of activity concentration was seen the further away the ponds extend from the pile, showing that the treatment dams are working to reduce the concentration of radionuclides in the water.

The radiological risk to biota at Kvarntorpshögen, the Serpentine ponds, Surpölen and Norrtorpssjön cannot be concluded as being of negligible concern. Further assessments are necessary to establish the risk. However, the radiological risk to humans at any site in the Kvarntorp area is of limited concern compared to average yearly effective doses.

Sammanfattning

Bakgrund: Kvarntorpshögen och sjöarna Surpölen och Norrtorpssjön är kvarlevor från en industri där man bröt alunskiffer under andra världskriget i syfte att utvinna olja. Högen består mestadels av rödfyr (rester från produktionen), medans sjöarna är vattenfyllda stenbrott. Båda är biprodukter av en industri med naturligt förekommande radioaktivt material (NORM), vilket gör dem till teknologiskt förhöjt NORM. Kvarntorpsområdet har därför något förhöjda halter av radionuklider. Den radiologiska risken för människor, växter och djur bör undersökas, för att förhindra risk från strålning, både nu och i framtiden.

Syfte: Syftet med det här arbetet var att utföra en radiologisk karaktärisering på och runt Kvarntorpshögen genom mätningar av ^{238}U, ^{235}U, ^{234}U and ^{210}Po i jord, vatten och växter i området samt att beräkna överföringen av ^{210}Po and U-isotoperna i jord-rot-växt-systemet. Ett andra syfte var att genomföra en radiologisk riskanalys för människor, växter och djur genom uppskattning av effektiva doser respektive absorberade doser.

Metod: Under sommaren 2020 samlades prover av vatten, rödfyr, jord och växter in på och omkring Kvarntorpshögen. U-isotoperna och ^{210}Po analyserade genom deras alfasönderfall. Proverna förbereddes genom radiokemi för att sedan mätas med alfaspektrometri. Överföringsfaktorer beräknades genom kvoter av aktivitetskoncentrationen i rot och jord, växt och jord, och växt och rot. En radiologisk riskanalys för människor genomfördes genom mätningar av miljödosekvivalent, vilket användes som en konservativ uppskattning av effektiv dos som sedan jämfördes med årliga effektiva doser. Den radiologiska risken för växter och djur genomfördes genom beräkning av absorberad dos med ERICA Tool (Environmental Risk from Ionising Contaminants: Assessment Tool) Tier 1 och Tier 2.

Resultat och Slutsatser: Extern dosrat i Kvarntorpsområdet är högst vid exponerade alunskifferväggar vid sjöarna och vid rödfyren på Kvarntorpshögen. Aktivitetskonentrationerna av ^{238}U, ^{235}U, ^{234}U och ^{210}Po minskade i jord-rot-växt-systemet på högen, och jämvikt hittades bland radionukliderna i jord och rötter men inte i växtstammen och bladen. Detta är troligtvis p.g.a. atmosfärisk deposition av ^{222}Rn som är en sekundär källa till ^{210}Po. Aktivitetskoncentrationen av ^{238}U, ^{235}U, ^{234}U och ^{210}Po i växter från serpentindammarna är i jämvikt och är något lägre än i växter på Kvarntorpshögen. För vatten i Kvarntorpsområdet återfinns den högsta aktivitetskoncentrationen i Surpölen, därefter i Norrtorpssjön och minst i serpentindammarna. I vattnet är ^{210}Po inte i jämvikt med U-isotoperna då de beter sig kemiskt olika. I serpentindammarna minskade aktivitetskoncentrationen i vattnet som en funktion av avståndet från högen, vilket visar på reningsdammarnas uppgift att rena vattnet som passerar.

Slutsatsen kan inte dras att den radiologiska risken till växter och djur på Kvarntorpshögen, serpentindammarna, Surpölen och Norrtorpssjön är försumbar. Ytterligare undersökningar behövs för att fastställa risken. Däremot är den radiologiska risken för människor på alla platser i Kvarntorpsområdet försumbara jämfört med årliga effektiva doser.

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1. Introduction

1.1. Naturally Occurring Radioactive Materials (NORM)

Several radionuclides occur naturally and have a half-life long enough to still be present from when the earth was formed (^{238}U, ^{235}U, ^{232}Th and ^{40}K), or are decay products of these (1). Presence of such radionuclides, as well as other components in bedrock, depend on the local geology and the rock's depositional environment (2) (i.e. the processes related with a sediment's deposition and its lithification to becoming a solid rock). Materials containing naturally occurring radionuclides goes by the name "Naturally Occurring Radioactive Material" (NORM).

The alum shale is a type of sedimentary rock which name was introduced over 300 years ago when the shales was mined for its alum (potassium aluminium sulphate). The shale contains relatively high concentrations of NORM, especially uranium, and is rich in organic matter. It was deposited on the seafloor in an oxygen-deficient environment about 500 million years ago and the organic matter in the shale are the remnants of dead animals and plants from that time. It also contains sulphide and a number of heavy metals, and has lenses of limestone throughout its depth (2, 3). In Sweden, the presence of alum shale on land is greatest in the south and along parts of the mountains in the north, but also occurs in Gävlebukten and the Baltic sea, see left side of Figure 1.

1.2. Mining the Alum Shales in Kvarntorp for Oil

During World War II import of goods to Sweden were cut off, one being the import of crude oil. To keep up with demand the industry had to find a way to get a hold of oil in Sweden. The country's largest fuel reserve was found in the alum shales in Kvarntorp, a small town in the municipality of Kumla in the province of Närke, see red marking on left side of Figure 1. The shales were located close to the surface, allowing extraction through open pit mining and oil production in Kvarntorp began in 1942 and lasted until 1966. Thereafter the production was no longer beneficial due to cheaper import products which became available again after the war. The oil was extracted from the mined shales through pyrolysis. In this process the rocks are heated to release volatile organic compounds in gaseous form, which then through condensation and distillation results in liquid oil. This extraction method is still used today in some places (3). During the latter years of the oil production, extraction of uranium was also carried out in Kvarntorp due to the high concentration in alum shales (4).

The waste products from the oil production are crushed but not further processed alum shale ("fines"), processed burned alum shale ("coke") and processed alum shale burned to ash ("shale ash") (3). In Swedish, shale ash is called *rödfyr* because of its strong brick red colour and is among other things used as ground layer on tennis courts. Un-processed shale has a dark colour because of its organic matter contents (1), see right side of Figure 1.

1.3. Kvarntorpshögen – Technologically Enhanced NORM (TENORM)

Some of the waste from the oil production was put back into the open pits (nowadays water-filled) when the mining ended, while the rest was deposited on the meadows of Kvarntorp, forming the approximately 100 m high and 700 m wide hill that today is known as *Kvarntorpshögen*. The pile consists of 2 million tons of coke (mainly in the bottom), 3 million tons of fines and 23 million tons of shale ash (3), as well as lime waste, heavy metals and sulphides (5).

Kvarntorpshögen and the pits are by-products of an industry with NORM, making them Technologically Enhanced NORM (TENORM). The Kvarntorp area has therefore somewhat increased concentrations of radionuclides. There are many piles like Kvarntorpshögen in Sweden, but no one greater than the one in Kvarntorp, with its 40 million cubic meters of burnt alum shale with uranium concentrations of a few kBq/kg, or a few hundred ppm, while world average is in the order of 3 ppm (6, 7). This makes Kvarntorp one of the most contaminated areas in Sweden with regard to NORM. However, to our knowledge, little is known regarding the distribution of uranium nuclides in the Kvarntorp area, as well as the presence of other naturally occurring radionuclides.

Kvarntorp is also one of Sweden's most contaminated areas with regard to heavy metals and oil. However, mobility of heavy metals through leaching from infiltration of water is limited at the moment (8). This is due to the high inner temperature of the pile, a result from when hot shale ash was deposited and caused exothermic oxidation processes of sulphides and organic matter. Even with extensive covering, the oxidation processes are still ongoing in the pile today, resulting in Kvarntorpshögen having temperatures of up to 70°C on the surface and 700°C just 15 m below the surface (3). Once the pile cools (estimated a century from now) the leaching will be of major concern to the environment (8). Leaching of radioactive isotopes, however, is low in shale ash (4).

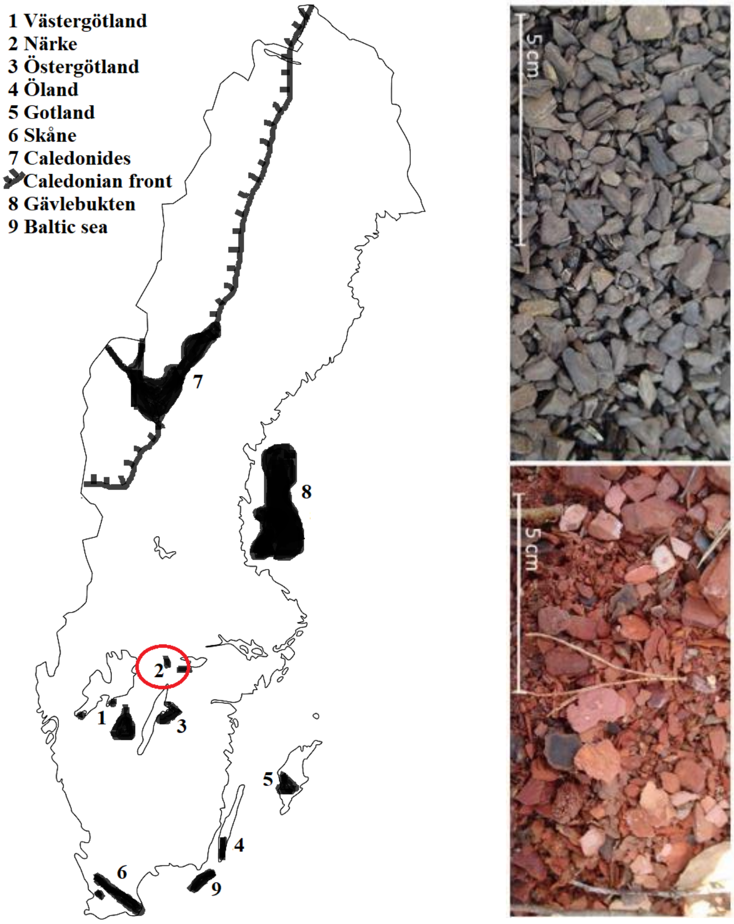


Figure 1 - **Left**: Occurrence of alum shale in Sweden, where the red marking indicates the province of Närke. Modified after Murase (2). **Top right:** Fines from Kvarntorpshögen. **Bottom right**: Shale ash from Kvarntorpshögen. Taken from Åhlgren *et al.* (9), photographer Kristina Åhlgren.

1.4. Importance of Radiological Risk Assessment

Kvarntorpshögen is a well-visited place during all seasons for recreational purposes. During the summer, visitors climb a 427-step staircase to the top, bike in the terrain and enjoy a walk among the art exhibition on the top of the pile. During the winter visitors also use the ski slope, and Kvarntorpshögen is home to various species of animals and plants as well. The number of visitors during 2020 (38 000 by October 8th) has also increased by 48% since earlier years, presumably due to the corona virus covid-19. The radiation risk that may arise to the exposed public and environment from the elevated concentrations of naturally occurring radionuclides in the Kvarntorp area have to be assessed, to protect people and the environment against radiation risks, both now and in the future (10).

1.5. Aims

The aim of this project was to carry out a radiological characterisation on and around Kvarntorpshögen by measurements of ^{238}U, ^{235}U, ^{234}U and ^{210}Po in soil, water and plants in the area, and calculating the transfer of ^{210}Po and U-isotopes in the soil-root-plant system. A second aim was to perform a radiological risk analysis for humans and biota by estimating effective doses and absorbed doses, respectively.

2. Method

2.1. Collection and Preparation of Samples

During the summer of 2020, July 7th-8th, samples of soil, water and plants were collected on Kvarntorpshögen and in its surroundings, see Figure 2. The Serpentine ponds is a complex with treatment dams below Kvarntorpshögen, and Surpölen and Norrtorpssjön are nearby lakes (water-filled mining pits). The different sampling sites at Kvarntorpshögen and the Serpentine ponds can be seen in Figure 3 and the samples collected at each site in Table 1. To obtain site averages, soil samples were collected in a triangular manner, with three cores (Ø = 6 cm, depth = 10 cm) in the corners of an equilateral triangle with side lengths of 50 cm (11). Plant samples were collected at each site in proximity of the sampled soil in order to calculate transfer factors in the soil-root-plant system. The most abundant species were collected in volumes fitting in normal sized plastic and paper bags. Water samples (surface water) were collected from the shore in cans of a volume up to 5 litres.

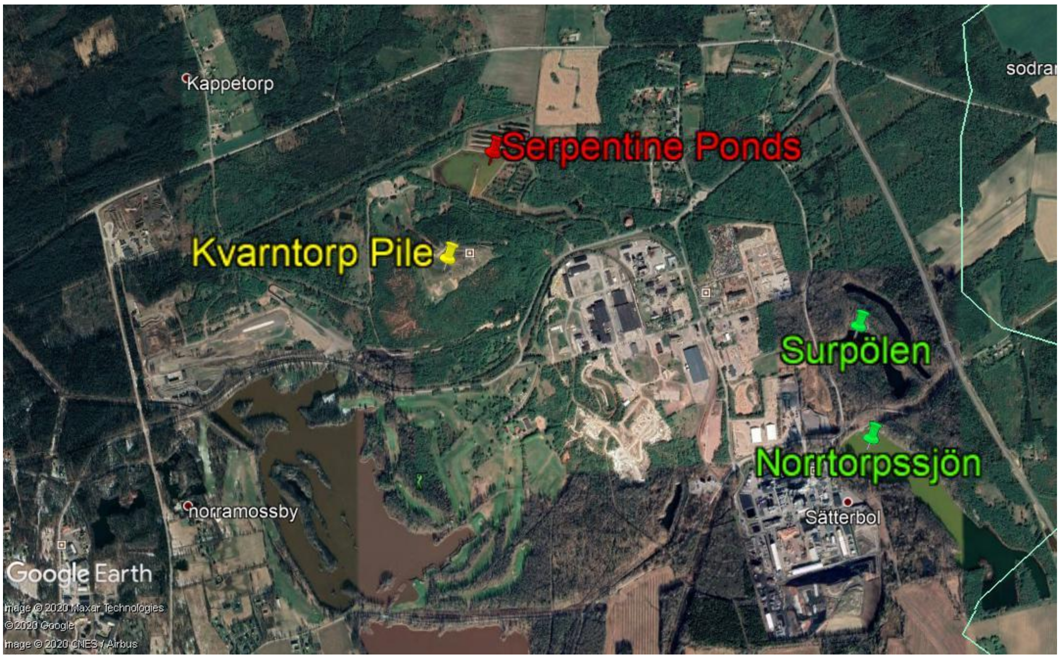


Figure 2 - Overview of the area surrounding Kvarntorpshögen, depicting an area of about 13 km^2. Samples were collected on the pile, from the Serpentine ponds, and from Surpölen and Norrtorpssjön. Map data ©2020 Google.

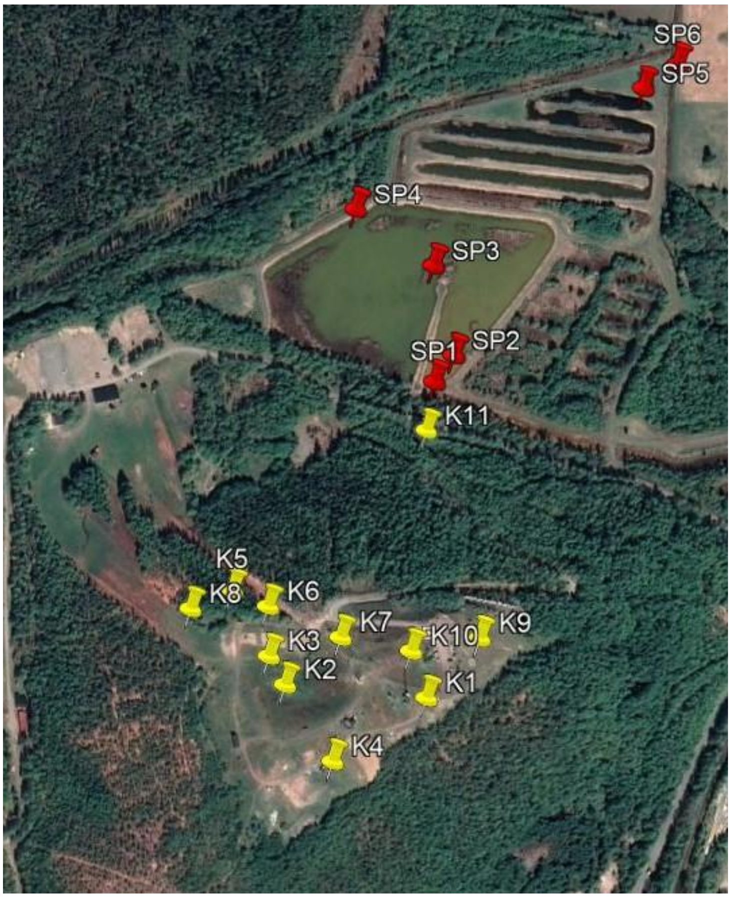


Figure 3 - Sampling sites at Kvarntorpshögen and the Serpentine ponds. K = Kvarntorpshögen and SP = Serpentine Ponds. Map data ©2020 Google.

The U-isotopes and ^{210}Po were analysed by their alpha emission and the samples were therefore prepared through radiochemistry to be measured by alpha spectrometry. The soil samples were dried and then sieved to <0.063 mm, and the plant samples were separated as root and plant (stem and leaves), washed with cold water, dried, and then mixed. Plant samples were digested through microwave assisted digestion with a mixture of nitric and hydrochloric acid (dissolution of organic material), and soils with an addition of hydrofluoric acid to achieve total digestion (dissolution of silica particles). Water samples were coarsely filtered (10-12 µm) to remove visible debris (SP1 SP2, SP4 and SP6).

A tracer (chemical analogue to the element of interest) was added to all samples to account for any loss of the nuclides during the chemical preparations, and to calculate the yield of the chemical procedure. Thereafter, the radionuclides were concentrated through Fe co-precipitation, followed by a chemical separation through liquid-liquid extraction with TBP (tri-n-butyl phosphate). Lastly, a thin source for measurement of each sample was obtained by CeF micro-precipitation for U, and by spontaneous deposition of Po on copper discs in hydrochloric acid.

Table 1 – Sample description with the type of samples collected at each site. The species names of the plants are written in italics.

Kvarntorpshögen		
Site	**Type of Sample**	**Sample name**
K1	Soil	K1 Soil
	Common mugwort	K1 Plant A
	Hare’s-foot clover	K1 Plant B
K2	Soil	K2 Soil
	Common couch	K2 Plant
K3	Soil	K3 Soil
	Great mullein (withered)	K3 Plant
K4	Soil	K4 Soil
K5	Soil	K5 Soil
	Garden lupine	K5 Plant
	- Root	K5 Root
K6	Shale ash	K6 Rödfyr
K7	Soil	K7 Soil
	Root of *Shining cranesbill*	K7 Root
K8	Soil	K8 Soil
	Goat willow	K8 Plant A
	- Root	K8 Root A
	White bedstraw	K8 Plant B
	Canada thistle	K8 Plant C
	Parsnip	K8 Plant D
	Common couch	K8 Plant E
K9	Soil	K9 Soil
	Perforate St. John’s-wort	K9 Plant
	- Root	K9 Root
K10	Soil	K10 Soil
	Shale ash	K10 Rödfyr
	Japanese knotweed	K10 Plant A
	Grass	K10 Plant B
	Perforate St. John’s-wort	K10 Plant C
	Great mullein	K10 Plant D
	- Root	K10 Root D
	Hare’s-foot clover	K10 Plant E
	- Root	K10 Root E
K11	Shale ash	K11 Rödfyr

Serpentine Ponds		
Site	**Type of Sample**	**Sample name**
SP1	Water	SP1
	Wild strawberry	SP1 Plant
SP2	Water	SP2
SP3	Water	SP3
	Heath speedwell	SP3 Plant A
	Birch	SP3 Plant B
	Cyperus sedge	SP3 Plant C
	Sallow	SP3 Plant D
	Tansy	SP3 Plant E
	Common reed	SP3 Plant F
	Imperforate St. John’s-wort	SP3 Plant G
	Deergrass	SP3 Plant H
	Bittersweet	SP3 Plant I
	Selfheal	SP3 Plant J
	Perforate St. John’s-wort	SP3 Plant K
SP4	Water	SP4
	Common reed	SP4 Plant
SP5	Water	SP5
	Coltsfoot	SP5 Plant
SP6	Water	SP6
Surpölen		
S	Water	S
Norrtorpssjön		
N	Water	N

2.2. Determination of Activity Concentrations of Alpha Emitters

For the measurements, a sample chamber (Alpha Ensemble®) with ion implanted silicon charged particle detectors (ORTEC, USA) were used. Measurement time per sample was around 1-2 days and the spectra were analysed in GammaVision software (ORTEC, USA). Through the equation

$$A_{nuclide} = A_{tracer} \frac{N_{nuclide}}{N_{tracer}} \tag{1}$$

the activity A [Bq] of each nuclide was calculated, N being the net counts of the tracer and nuclide of interest and A_{tracer} being the activity of the tracer. This relation is derived from the equation

$$A_{nuclide} = \frac{N_{nuclide}}{t \cdot \mathcal{E} \cdot p \cdot Y} \tag{2}$$

where t is the measurement time [s], $\mathcal{E}$ the total efficiency, p the emission probability of the alpha particle and Y the chemical yield. Since all registered alpha particles from a nuclide were marked in the spectra, the emission probability was set to unity. Further, the yield can be calculated as

$$Y = \frac{N_{tracer}}{t \cdot \mathcal{E} \cdot A_{tracer}}. \tag{3}$$

In addition, the efficiency is assumed to be constant for alpha particles with energies between 4-8 MeV since they can be presumed to be fully absorbed once reaching the detector. Decays from both U-isotopes and ^{210}Po fall within this energy range.

Combining Equation 2 and 3 results in Equation 1 since the efficiency, measurement time and yield are the same for the nuclide of interest and the tracer. Further, by dividing Equation 1 with the mass of the sample, the activity concentration of the four radionuclides was calculated for the majority of the samples in Table 1. The samples whose activity concentrations were not calculated as above were the activity concentrations of ^{210}Po in K8 Soil, K9 Soil, K10 Soil and K10 Rödfyr, since having decayed too much before preparation and measurement. Instead, the activity concentration of ^{210}Po in K8 Soil was estimated by assuming equilibrium with ^{210}Pb, which was measured through gamma spectrometry. K9 Soil, K10 Soil and K10 Rödfyr was left unmeasured. The activity concentration for all samples except for water is given as Bq/kg of dry weight. The errors associated with the activity concentrations were calculated through the square-root of the sum of uncertainty components (K=1), including uncertainty in counts, measured weights and activity of the tracer.

To help reduce false positives in the detection of activity, a Minimum Detectable Activity (MDA) was determined for the four radionuclides, the MDA being a level of activity concentration practically achievable to detect by the measurement method. It includes both intrinsic parameters of the instruments such as background and efficiency, as well as the chemical yield, measuring time and amount of sample. It was calculated for a 95 % confidence level, with α-error = β-error = 0.05 (type I and type II error, respectively). In other words, the MDA is a beforehand determined activity concentration level that with 95 % probability can be detected by the measurement method. The MDA is calculated by

$$MDA = \frac{2.71 + 3.29\sqrt{CPS_{bg}}\sqrt{t_S}\sqrt{1 + \frac{t_S}{t_{bg}}}}{t_S \cdot \mathcal{E} \cdot Y \cdot S} \tag{4}$$

where CPS_{bg} is the count rate of the background (from a background measurement), t_S and t_{bg} the measurement time for the sample and the background and S the amount of the sample (12).

2.3. Assessment of Transfer Factors

To estimate the transfer of the U-isotopes and ^{210}Po in the soil-root-plant system at Kvarntorpshögen average soil-to-root (S-R), soil-to-plant (S-P) and root-to-plant (R-P) transfer factors (TF) were calculated. The $TF_{S\text{-}P}$ is defined as

$$TF_{S-P} = \frac{Activity\ Concentration\ in\ Plant\ [Bq/kg_{dw}]}{Activity\ Concentration\ in\ Soil\ [Bq/kg_{dw}]} \tag{5}$$

where *dw* is dry weight. The part of plants of concern is usually above ground, and for soil it is usually surface soil where the majority of the plant's roots are found (13). For the $TF_{S\text{-}R}$ the numerator is replaced by the activity concentration in the root, and for the $TF_{R\text{-}P}$ the numerator is replaced by the activity concentration in the plant and the denominator by the activity concentration in the root.

2.4. Mobile Detection System and Dose Rate Maps

Measurements on and around Kvarntorpshögen, for creating dose rate maps of the area, were carried out with a back-pack system including a NaI (Tl) detector (model 802-2x2, Canberra) connected to a digiDART high voltage supply and multichannel analyser (ORTEC, USA) and a GPS receiver (Trimble, USA). To create the dose maps the software Nugget and MapCreator (courtesy of Swedish Radiation Safety Authority) were used. The maps display Spectrum to Dose Index (SDI), a total photon dose rate determined by converting the number of detected pulses in the pulse height distribution spectrum with an energy dependent factor, and then by a calibration factor obtained by cross calibration with a handheld dose rate meter Automess 6150AD (Automation und Messtechnik, Germany, energy range 20 keV-7 MeV, dose rate range 50 nSv/h-100 µSv/h). The dose rates are shown by colour in the maps, from blue to red, where blue is relatively low, and red is relatively high. The dose rate and the corresponding colour is thus unique for each site. From the dose rate maps both maximum and mean ambient dose equivalent rate were acquired through input in MatLab version R2016a.

2.5. Radiological Risk Assessment for Biota

In this work, the ERICA Tool (Environmental Risk from Ionising Contaminants: Assessment Tool) Tier 1 and Tier 2 was used to perform the radiological risk assessment to biota (14). It aids in the estimation of absorbed doses and comparisons to background and "safe" exposures. The protection of biota against environmental exposure aims to keep deleterious radiation effects on a level where the impact on biodiversity, resilience and ecosystem function would be negligible. The most important biological endpoints therefore are those that could change the biota's structure and population size, such as early mortality, some forms of morbidity, impairment of reproductive capacity and induction of chromosomal damage.

Firstly, Tier 1 was used to screen out sites where the radiological risk to populations of biota regarding these endpoints is negligible. Tier 1 is a conservative assessment, requiring only inputs of the type of ecosystem studied (terrestrial, marine or fresh water), activity concentrations of the radionuclides of concern and selection of a Screening Dose Rate (SDR). The SDR serves as a Predicted No Effects Dose Rate and the ERICA Tool provides three options – either choosing 10 µGy/h for all ecosystems, or 40 µGy/h for terrestrial animals and 400 µGy/h for plants and aquatic organisms, or entering a custom value. The SDR 10 µGy/h is derived from the FREDERICA effects database, containing effects of ionising radiation in biota (15), while the SDR 40/400 µGy/h is derived from IAEA (16) and UNSCEAR (17) reports. In this work two Tier 1 assessments were carried out for each soil and water activity concentration, one for each predefined SDR provided by the ERICA Tool.

The results from the Tier 1 assessments are a Risk Quotient (RQ), a measure of how many times higher of a dose rate the most limiting organism is receiving from the input activity concentrations than from the SDR, based on a configuration giving the maximum exposure to that organism. The most limiting organism is the organism receiving the highest dose rate in an ecosystem, therefore making it the most limiting in regard to protection. The variety of biota, however, are immense, as well as size, shape, insides, occupancy, offspring, length of life, life stages and effects of radiation on them. To include all kinds of biota in a radiological risk assessment the diversities are simplified and a set of Reference Animals and Plants (RAPs) with relevant databases have thus been developed for each of the three environments. A RAP is defined by ICRP as “a hypothetical entity, with the assumed basic characteristics of a specific type of animal or plant, as described to the generality of the taxonomic level of Family, with defined anatomical, physiological, and life history properties, that can be used for the purposes of relating exposure to dose, and dose to effects, for that type of living organism” (18). Biota described to the generality of the taxonomic level of Family are assumed to have relatively constant biological features with respect to radiation effects. For the reference organisms used in the ERICA Tool, see Table 2.

Table 2 - Reference organisms used in the ERICA Tool.

Terrestrial	**Freshwater**
Amphibian	Amphibian
Annelid	Benthic fish
Arthropod – detritivorous	Bird
Bird	Crustacean
Flying Insects	Insect larvae
Grasses & Herbs	Mammal
Lichen & Bryophytes	Mollusc – bivalve
Mammal – large	Mollusc – gastropod
Mammal – small-burrowing	Pelagic fish
Mollusc – gastropod	Phytoplankton
Reptile	Reptile
Shrub	Vascular plant
Tree	Zooplankton

The most limiting organism in the results from Tier 1 is the most limiting of the RAPs for that ecosystem. A total RQ value >1 indicate on an exposure where deleterious effects can occur to the organism, while a value <1 can be interpreted as a site where the radiological risk to populations of biota is negligible. For sites where the total RQ was >1 for either one or both SDRs, the radiological risk assessment continued to the Tier 2 assessment.

Tier 2 estimates absorbed doses [µGy/h] from summed internal and external exposure and is a more detailed assessment concerning exposure conditions and transfer parameters. Inputs in addition to Tier 1 includes choice of organisms to investigate (reference and/or user-defined), possibility to add additional nuclides, review and edit of occupancy, transfer and radiation weighting factors, and dealing with multiple sites at once through spatial media activity concentrations. The inputs are related to absorbed dose rate through dose conversion coefficients [μGy h^{-1} Bq^{-1} kg]. The estimated doses are compared to the SDRs by RQ as in Tier 1, however, now by two RQs, one expected and one conservative (conservative in Tier 1). The expected RQ is calculated from the actual input media activity concentration, while the conservative is calculated by multiplying the expected RQ by an uncertainty factor. Assuming the distribution of the dose rate and RQ being exponential, and estimating the 95th percentile of the RQ, the uncertainty factor is 3. This gives the low probability of 5 % of the dose exceeding the SDR, while for the expected the probability is 50 %. The estimated doses are also compared to absorbed dose rates to the RAPs from natural background exposure. The Tier 2 assessment was carried out on Kvarntorpshögen, the Serpentine ponds, Surpölen and Norrtorpssjön, with both SDRs.

In both Tier 1 and Tier 2 the dose conversion coefficients (DCCs) for ^{238}U and ^{234}U does not include any progenies. Only for ^{235}U the daughter ^{231}Th is included. Therefore, progenies were manually added to make the assessments reflect the true radiation situation better. All progenies with half-lives approximately equal to or less than two months (i.e., assumed in equilibrium with their source), and if provided by the ERICA Tool with relevant databases, were included in the Tier 1 and Tier 2 assessments for soils, see Table 3. No progenies were included in assessments for water since the radionuclides behave differently in water.

Table 3 - Daughter nuclides manually included in the Tier 1 and Tier 2 assessments.

	Parents	**Daughters**
Tier 1	U-238	-
	U-234	-
	U-235	Th-227
Tier 2	U-238	Th-234
	U-234	Po-218, Pb-214, Po-214
	U-235	Th-227, Ra-223, Po-215, Po-211, Pb-211

2.6. Radiological Risk Assessment for Humans

The protection of humans against environmental exposure aims to prevent deterministic effects and minimize the risk of stochastic effects to individuals. The protection quantity relating human exposure to risk is the effective dose rate [Sv/h]. In this work the effective dose rate was conservatively estimated through the operational quantity ambient dose equivalent rate [Sv/h], which was measured at Kvarntorpshögen, Surpölen and Norrtorpssjön at 1 meter above ground. The measurements were carried out with both the handheld dose rate meter Automess and the back-pack system. The highest dose rate, as well as the average of the dose rates measured at the different sites, were used as the maximum and average effective dose rate estimate for an adult visiting the sites for an hour.

For both the maximum and average dose rate, the time before an adult receives 1 mSv at Kvarntorpshögen, Surpölen and Norrtorpssjön was calculated. The dose rates for both maximum and average measurements were then calculated for the most frequent visitor to knowledge – the kiosk worker, working 7 hours every day from May 30th to August 31st. Further, to demonstrate differences in dose rate between summer and winter, the dose rate measured in the ski slope at the pile was utilised. During the winter 20 cm of snow was assumed to cover the pile, which is estimated to attenuate 40 % of the exposure (19).

Furthermore, the difference between the estimated effective dose rate to adults to the effective dose rate to new-borns, 5-year-olds, 10-year-olds and 15-year-olds were estimated through comparison of ^{214}Bi-external dose rate coefficients for the different age groups from ICRP 144 (20). (^{214}Bi due to its several high energy gamma emissions). All the above estimated times before receiving 1 mSv were then also estimated for the four age groups of children, to demonstrate the differences in risk to children and adults.

The effective dose rate when swimming in Norrtorpssjön and Surpölen was also estimated, through multiplying measured water activity concentrations [Bq/kg] with external dose rate coefficients for immersion in water for adults from ICRP 144 [nSv h^{-1} Bq^{-1} m^{3}]. The radionuclides included in the estimate for swimming was ^{238}U, ^{234}U, ^{235}U and ^{210}Po. Again, the time before receiving 1 mSv for adults and children was estimated.

Then the collective dose from visiting Kvarntorpshögen during 2020 and 2019, both for maximum and average dose rates were estimated by multiplying the estimated effective doses for some representative visitors with the number of visitors during each year. The representative visitors were assumed to be two adults with two children (5-year-olds) coming once a year and staying for an hour.

Lastly, the effective doses were compared to yearly effective doses. The average yearly effective dose to non-smokers in Sweden, including all sources of exposure to the public, is shown in Figure 4. For the average non-smoker, the exposure from medical diagnostics is 0.9 mSv/y, from ^{137}Cs 0.01 mSv/y, from naturally occurring radionuclides in food 0.2 mSv/y, from radon in indoor air 0.2 mSv/y, from potassium in the body 0.2 mSv/y, from (ground and) building materials 0.6 mSv/y, and from cosmic radiation 0.3 mSv/y. For the groups under special conditions, it can be deduced from the figure their respective dose from the various exposures. For example, a frequent (one long-distance flight per week) air passenger increases its yearly effective dose by 1 mSv. This means that a long-distance flight increases the average individual's yearly dose by 0.019 mSv.

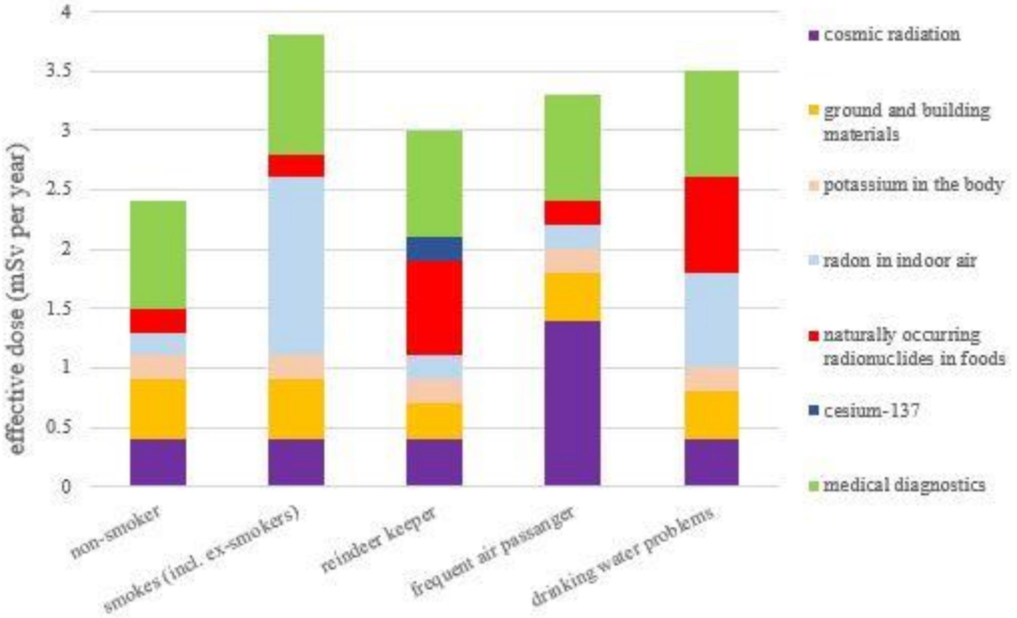


Figure 4 – Average yearly effective dose to the public of non-smokers, smokers and other groups under special conditions. Modified after Andersson *et al.* Strålmiljön i Sverige (21).

3. Results

3.1. Dose Rate Maps

Figures 5-7 shows the dose rate maps of Kvarntorpshögen, Surpölen and Norrtorpssjön. The maximum ambient dose equivalent rates were 1.8, 2.1 and 1.7 µSv/h, respectively, while the mean dose rates were 0.7, 0.9 and 0.4 µSv/h, respectively. However, at Surpölen and Norrtorpssjön measurements of up to 3 and 4.3 µSv/h were carried out with the Automess right next to exposed alum shale walls around the lakes, thus being the maximum dose rates at the sites.

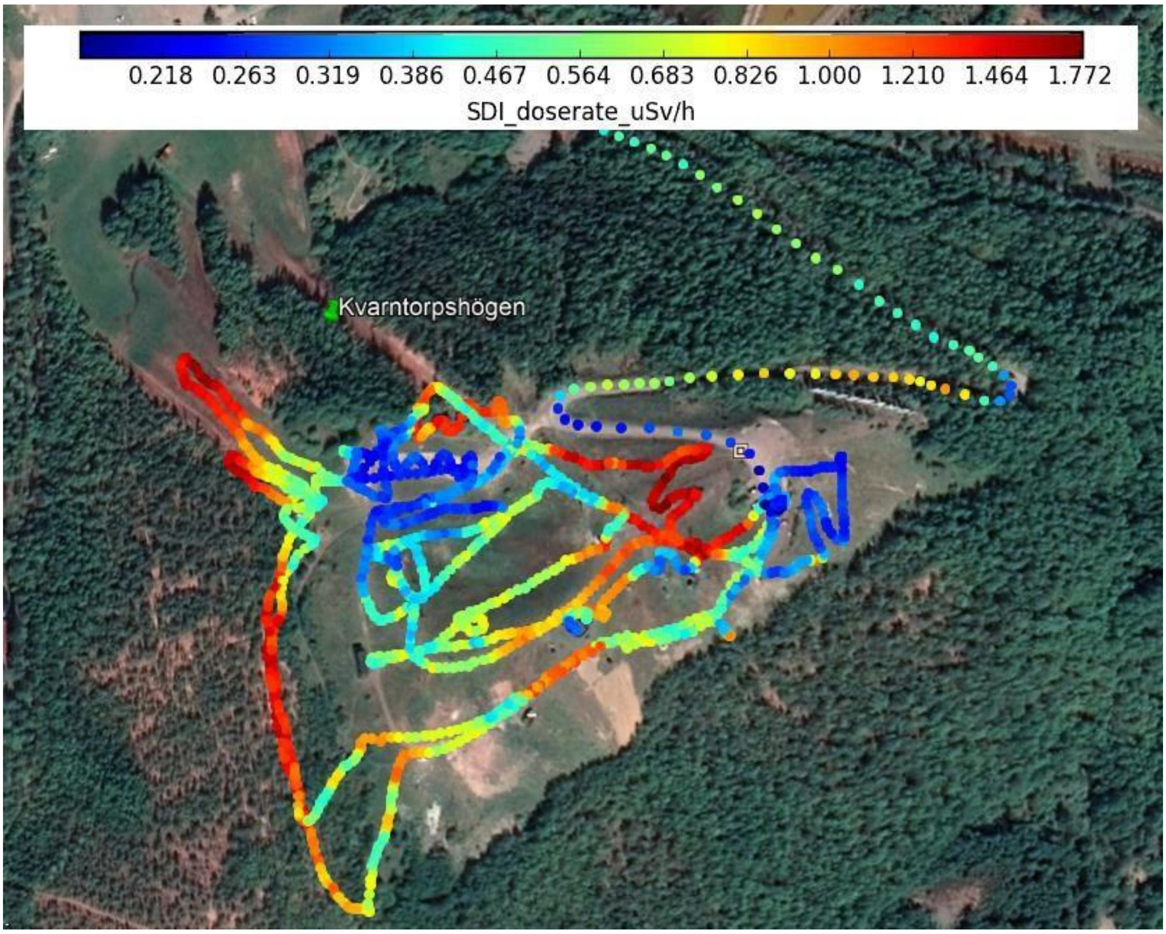


Figure 5 - Dose rate map of Kvarntorpshögen showing the dose rate in colour from blue to red, where blue is relatively lower dose rates and red is relatively higher at the site. Map data ©2020 Google.

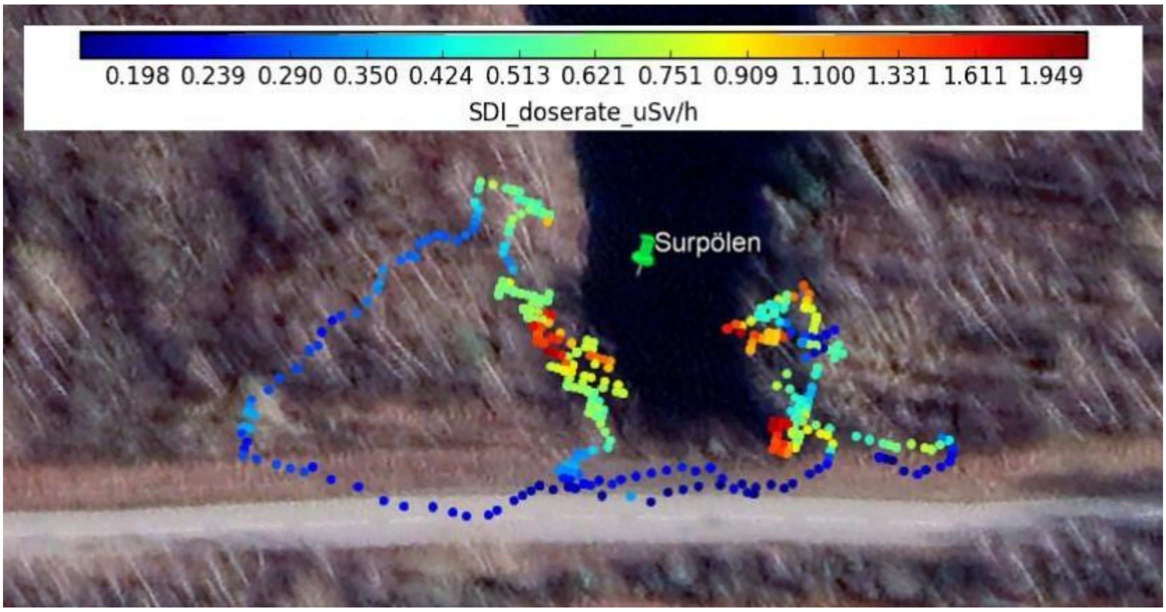


Figure 6 - Dose rate map of Surpölen showing the dose rate in colour from blue to red, where blue is relatively lower dose rates and red is relatively higher at the site. Map data ©2020 Google.

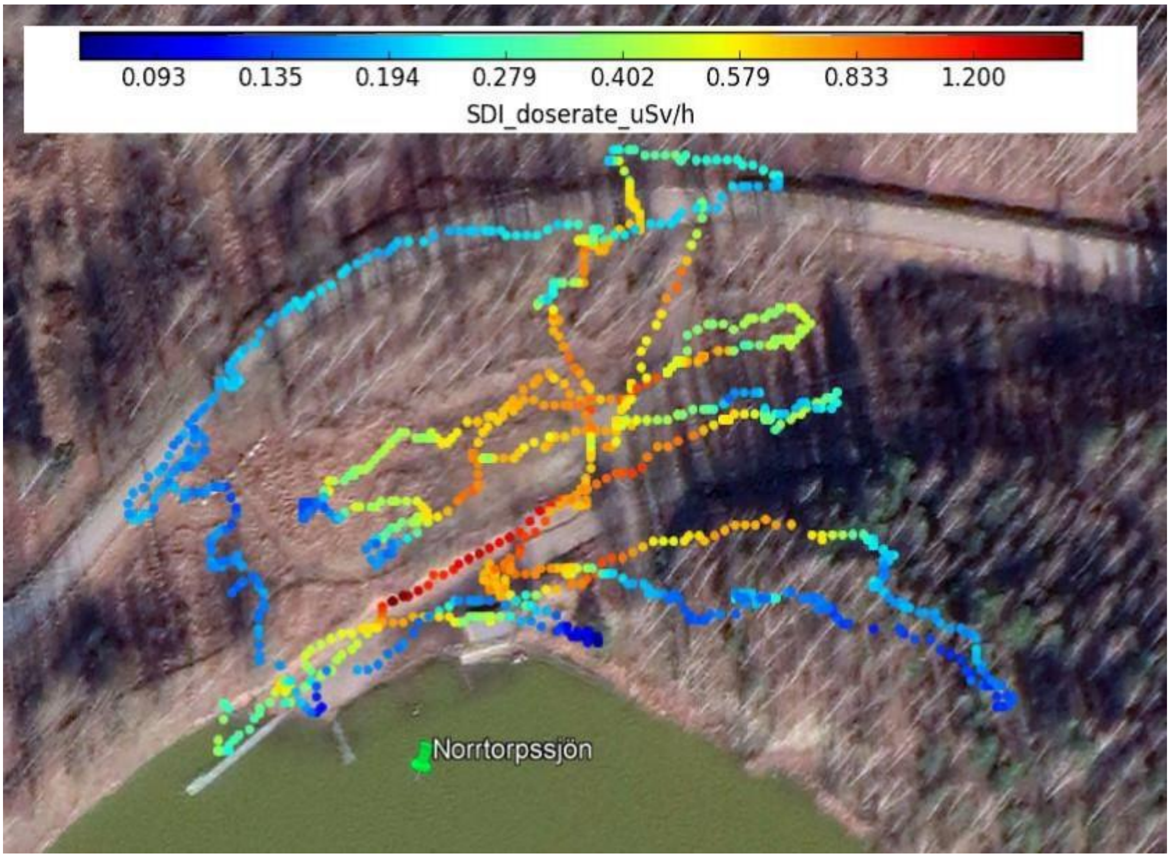


Figure 7 - Dose rate map of Norrtorpssjön showing the dose rate in colour from blue to red, where blue is relatively lower dose rates and red is relatively higher at the site. Map data ©2020 Google.

3.2. Activity Concentration of Alpha Emitters in Kvarntorp

Table 4 shows the average activity concentrations in Bq/kg *dw* with uncertainties (K=1) of ^{238}U, ^{235}U, ^{234}U and ^{210}Po in shale ash, soil, roots and plants at Kvarntorpshögen and the Serpentine ponds. The activity concentration was highest in the shale ash, thereafter decreasing from soil to root to plant at Kvarntorpshögen, to plants at the Serpentine ponds. The radionuclides ^{238}U, ^{234}U and ^{210}Po had approximately the same activity concentrations in each type of sample, while ^{235}U was lower.

Table 4 - Average activity concentrations in Bq/kg dry weight with uncertainties (K=1) of ^{238}U, ^{235}U, ^{234}U and ^{210}Po in shale ash, soil, root and plant samples at Kvarntorpshögen (K) and the Serpentine Ponds (SP). The number of samples used for the average is also displayed.

Sample	**^{238}U**	**^{235}U**	**^{234}U**	**^{210}Po**	**# samples**
Shale ash (K)	1722 ± 124	114 ± 11	1866 ± 127	1745 ± 95*	3
Soil (K)	698 ± 43	35 ± 4	650 ± 40	635 ± 141*	9
Roots (K)	49.0 ± 1.7	2.8 ± 0.2	50.2 ± 1.8	49.4 ± 4.9	6
Plants (K)	9.4 ± 0.8	1.1 ± 0.2**	10.5 ± 0.9	15.9 ± 2.1	16
Plants (SP)	6.6 ± 1.0	0.6 ± 0.1**	7.2 ± 1.0	5.6 ± 0.7**	14

*= Radionuclide not measured in all samples

**= Radionuclide below MDA and not included in the average

Table 5 shows the activity concentrations in mBq/kg with uncertainties (K=1) of ^{238}U, ^{235}U, ^{234}U and ^{210}Po in water from the Serpentine ponds, Surpölen and Norrtorpssjön. The activity concentrations of the U-isotopes were highest in Surpölen and Norrtorpssjön, while ^{210}Po was highest in SP1 and Surpölen. The radionuclides ^{238}U and ^{234}U had approximately the same activity concentrations in each sample (except for Surpölen), with ^{235}U being lower.

Table 5 - Activity concentrations in mBq/kg with uncertainties (K=1) of ^{238}U, ^{235}U, ^{234}U and ^{210}Po in water samples from the serpentine ponds (SP1-SP6), Surpölen (S) and Norrtorpssjön (N).

Sample	**^{238}U**	**^{235}U**	**^{234}U**	**^{210}Po**
SP1	98.8 ± 7.8	7.8 ± 1.5	98.8 ± 7.8	13.8 ± 3.4
SP2	186.0 ± 12.3	7.3 ± 1.5	199.6 ± 13.0	5.6 ± 1.8
SP3	127.8 ± 6.9	7.5 ± 1.2	129.4 ± 7.0	4.8 ± 1.3
SP4	121.6 ± 7.6	6.0 ± 1.3	110.5 ± 7.1	< MDA
SP5	180.7 ± 13.7	6.7 ± 1.8	181.3 ± 13.8	3.7 ± 0.7
SP6	68.3 ± 4.8	4.3 ± 0.9	66.9 ± 4.7	< MDA
S	416.9 ± 22.6	42.8 ± 3.8	496.4 ± 26.4	9.6 ± 2.3
N	359.6 ± 19.3	17.0 ± 1.9	369.7 ± 19.7	< MDA

3.3. Transfer Factors

Table 6 shows the average soil-to-root ($TF_{S\text{-}R}$), soil-to-plant ($TF_{S\text{-}P}$) and root-to-plant ($TF_{R\text{-}P}$) transfer factors with uncertainties (K=1) of ^{238}U, ^{235}U, ^{234}U and ^{210}Po at Kvarntorpshögen. The $TF_{S\text{-}R}$ was quite similar for the four radionuclides, ranging between 7-8 %. The $TF_{S\text{-}P}$ was lower than $TF_{S\text{-}R}$, however, not similar for the four radionuclides. The transfer of ^{238}U and ^{234}U were about 1.4 and 1.6 %, respectively, while ^{235}U and ^{210}Po were about 3.1 and 2.5 %, respectively. The $TF_{R\text{-}P}$ was higher than the $TF_{S\text{-}R}$ and again not similar for the four radionuclides. The transfer of ^{238}U and ^{234}U were around 20 % while ^{235}U and ^{210}Po were about 39 and 32 %.

Table 6 – Average soil-to-root ($TF_{S\text{-}R}$), soil-to-plant ($TF_{S\text{-}P}$) and root-to-plant ($TF_{R\text{-}P}$) transfer factors with uncertainties (K=1) of ^{238}U, ^{235}U, ^{234}U and ^{210}Po at Kvarntorpshögen.

Transfer Factor	**^{238}U**	**^{235}U**	**^{234}U**	**^{210}Po**
$TF_{S\text{-}R}$	0.0702 ± 0.0050	0.0800 ± 0.0108	0.0772 ± 0.0188	0.0778 ± 0.0189
$TF_{S\text{-}P}$	0.0135 ± 0.0014	0.0314 ± 0.0067	0.0162 ± 0.0017	0.0250 ± 0.0065
$TF_{R\text{-}P}$	0.1918 ± 0.0176	0.3928 ± 0.0767	0.2092 ± 0.0194	0.3219 ± 0.0532

3.4. Radiological Risk Analysis for Biota

Table 7 shows the results from the ERICA Tool assessment Tier 1 where all soil and water samples except for K9 Soil and K10 Soil were included (since ^{210}Po was not measured in these). The total risk quotients for both of the default SDRs are displayed, for each of the soil and water sampling sites. The RQs assessed with the SDR 10 µGy/h was above 1 for all sites except for SP4 and SP6, while the RQs assessed with the SDR 40/400 µGy/h only was above 1 for one soil site.

The radionuclide that contributed the most to the total RQ was ^{210}Po for the soil and shale ash sites on Kvarntorpshögen, with ^{238}U and ^{234}U following being approximately equal to each other, and ^{235}U and ^{227}Th contributing the least. For the water sites, ^{210}Po contributed most to the total RQ (not for SP4, SP6 and N since ^{210}Po < MDA and therefore not included), followed by ^{238}U and ^{234}U still being approximately equal to each other, and ^{235}U contributing the least. The most limiting RAP, for which the RQs were calculated, was Lichen & Bryophytes for all the included radionuclides on terrestrial sites. For freshwater sites, the most limiting RAP was Amphibian for the U-isotopes and Insect larvae for ^{210}Po.

All soil samples had a total RQ greater than 1 for one or both of the SDRs. Therefore, none of the studied sites at Kvarntorpshögen were screened out as being a site where the radiological impact on biota is negligible. The assessment continued to Tier 2 for the maximum (K1 Soil), average (K4 Soil) and minimum (K5 Soil) risk sites.

All water samples except for SP4 and SP6 had a total RQ greater than 1 for the SDR 10 µGy/h. Therefore, at this stage of the assessment, SP4 and SP6 were screened out as being sites where the radiological impact on biota is negligible. The assessment continued to Tier 2 for S, N and the maximum (SP1) and minimum (SP5) risk sites at the Serpentine ponds, as well as for SP3 being situated at the centre of the ponds.

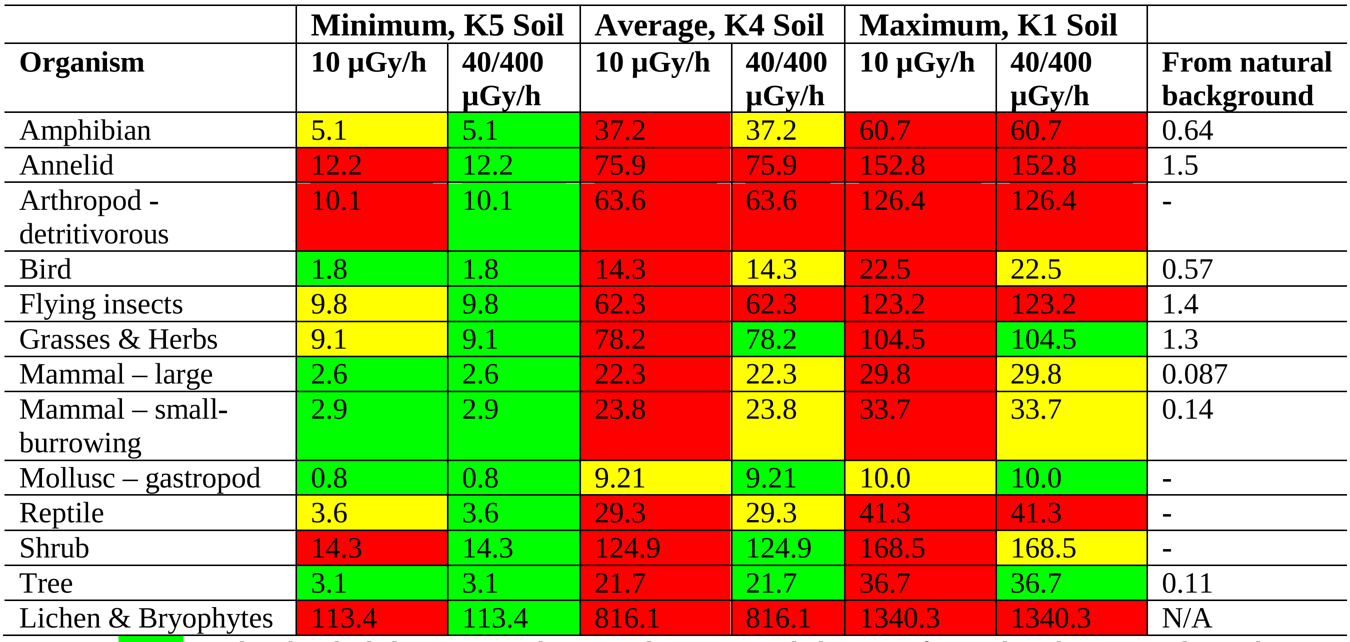
Table 7 - The total risk quotient (RQ) for the two default SDRs for soil and water sampling sites, obtained from the ERICA Tool assessment Tier 1. Red colour are values > 1 and green colour are values < 1.

Soil Samples			**Water Samples**		
Sample	**RQ, 10 µGy/h**	**RQ, 40/400 µGy/h**	**Sample**	**RQ, 10 µGy/h**	**RQ, 40/400 µGy/h**
K1 Soil	59.5	1.4	SP1	10.3	0.3
K2 Soil	9.9	0.2	SP2	5.3	0.2
K3 Soil	16.7	0.4	SP3	4.3	0.1
K4 Soil	32.5	0.8	SP4	0.9	< 0.1
K5 Soil	6.7	0.2	SP5	3.9	0.1
K7 Soil	7.6	0.2	SP6	0.5	< 0.1
K8 Soil	37.8	0.9	S	10.1	0.3
			N	2.7	0.1

Table 8 shows minimum, average and maximum absorbed dose rates to RAPs at Kvarntorpshögen. The minimum ranged from about 1–113 µGy/h for the different RAPs, the average from about 9-816 µGy/h and the maximum from about 10.0 – 1340 µGy/h. The most limiting RAP for both minimum, average and maximum absorbed dose rate was Lichen & Bryophytes. In comparison to the absorbed dose rate to the RAPs from natural background (available in the ERICA Tool for some of the RAPs), both the minimum, average and maximum absorbed dose rates to the RAPs at Kvarntorpshögen was higher, although not much when considering the minimum absorbed dose rates.

Table 8 also shows both the expected and conservative RQs via colouring for both SDRs. Green indicates that the absorbed dose rate to the RAP is less than the SDR for both the expected and conservative RQ for that SDR. Yellow indicates that the absorbed dose rate to the RAP is less than the SDR for the expected RQ but not for the conservative RQ for that SDR. Red indicates that the absorbed dose rate to the RAP is greater than the SDR for both the expected and conservative RQ for that SDR. Thus, for minimum absorbed dose rate at Kvarntorpshögen four RAPs exceeded the 10 µGy/h SDR for both the expected and conservative RQs, while four RAPs only exceeded it for the conservative RQ. Five RAPs did not exceed the SDR for neither the expected nor the conservative RQ. For the 40/400 µGy/h SDR, all RAPs for the minimum absorbed dose rate at Kvarntorpshögen did not exceed it for neither the expected nor the conservative RQs. For the maximum absorbed dose rate at Kvarntorpshögen, however, all RAPs but one exceeded the 10 µGy/h SDR for both the expected and conservative RQs. For the 40/400 µGy/h SDR six RAPs exceeded it for both the expected and the conservative RQs, while four only exceeded it for the conservative RQs and three did not exceed it for neither the expected nor the conservative RQs. For the average absorbed dose rate, the results were the same as for the maximum, except for two dose rates (Amphibian and Reptile) only exceeding the SDR 40/400 µGy/h for the conservative RQ, instead of both expected and conservative. The radionuclide that contributed the most to both the internal and external absorbed dose rate to all terrestrial RAPs on Kvarntorpshögen was ^{214}Pb, and the internal exposure made up most part of the total exposure.

Table 8 - Absorbed dose rate in µGy/h to RAPs at Kvarntorpshögen. Expected and conservative RQs included via colouring for both SDRs. Absorbed dose rate to RAPs from natural background also included as comparison. Obtained from ERICA Tool assessment Tier 2.



	Minimum, K5 Soil		**Average, K4 Soil**		**Maximum, K1 Soil**		
Organism	**10 µGy/h**	**40/400 µGy/h**	**10 µGy/h**	**40/400 µGy/h**	**10 µGy/h**	**40/400 µGy/h**	**From natural background**
Amphibian	5.1	5.1	37.2	37.2	60.7	60.7	0.64
Annelid	12.2	12.2	75.9	75.9	152.8	152.8	1.5
Arthropod - detritivorous	10.1	10.1	63.6	63.6	126.4	126.4	-
Bird	1.8	1.8	14.3	14.3	22.5	22.5	0.57
Flying insects	9.8	9.8	62.3	62.3	123.2	123.2	1.4
Grasses & Herbs	9.1	9.1	78.2	78.2	104.5	104.5	1.3
Mammal – large	2.6	2.6	22.3	22.3	29.8	29.8	0.087
Mammal – small-burrowing	2.9	2.9	23.8	23.8	33.7	33.7	0.14
Mollusc – gastropod	0.8	0.8	9.21	9.21	10.0	10.0	-
Reptile	3.6	3.6	29.3	29.3	41.3	41.3	-
Shrub	14.3	14.3	124.9	124.9	168.5	168.5	-
Tree	3.1	3.1	21.7	21.7	36.7	36.7	0.11
Lichen & Bryophytes	113.4	113.4	816.1	816.1	1340.3	1340.3	N/A

Green = The absorbed dose rate to the RAP do not exceed the SDR for neither the expected nor the conservative RQ for that SDR.
Yellow = The absorbed dose rate to the RAP do not exceed the SDR for the expected RQ but do for the conservative RQ.
Red = The absorbed dose rate to the RAP exceeds the SDR both for the expected and conservative RQ for that SDR.

Table 9 shows minimum, centre and maximum absorbed dose rates to RAPs at the Serpentine ponds. The minimum and centre ranged from below 1 to about 20 µGy/h for the different RAPs, and the maximum ranged from about 1–56 µGy/h. In comparison to the absorbed dose rate to the RAPs from natural background, both the minimum, centre and maximum absorbed dose rates to the RAPs at the serpentine ponds are lower or quite similar, with the maximum for Mollusc – bivalve being the highest away from it.

Table 9 also shows both the expected and conservative RQs via colouring for both SDRs. For both the minimum, centre and maximum absorbed dose rate, no RAPs exceeded the SDR 40/400 µGy/h for neither the expected nor the conservative RQ. Both the minimum and maximum did not exceed the SDR 10 µGy/h for neither the expected nor the conservative RQ for eight RAPs, while the centre did not exceed it for nine RAPs. Both the minimum and maximum exceeded the 10 µGy/h for only the conservative RQ, for one RAP.

The radionuclide that contributed the most to minimum, centre and maximum external and internal exposure to most of the RAPs at the Serpentine ponds are ^{235}U and ^{210}Po, respectively. The internal exposure made up most part of the total exposure.

Table 9 - Absorbed dose rate in µGy/h to RAPs at the Serpentine Ponds. Expected and conservative RQs included via colouring for both SDRs. Absorbed dose rate to RAPs from natural background also included as comparison. Obtained from ERICA Tool assessment Tier 2.

	Minimum, SP5		**Centre, SP3**		**Maximum, SP1**		
Organism	**10 µGy/h**	**40/400 µGy/h**	**10 µGy/h**	**40/400 µGy/h**	**10 µGy/h**	**40/400 µGy/h**	**From natural background**
Amphibian	🟩 1.3	🟩 1.3	🟩 1.1	🟩 1.1	🟩 1.5	🟩 1.5	1.1
Benthic fish	🟩 0.9	🟩 0.9	🟩 0.8	🟩 0.8	🟩 1.3	🟩 1.3	31
Bird	🟩 0.7	🟩 0.7	🟩 0.7	🟩 0.7	🟩 1.2	🟩 1.2	0.92
Mammal	🟩 1.4	🟩 1.4	🟩 1.1	🟩 1.1	🟩 1.5	🟩 1.5	
Pelagic fish	🟩 0.9	🟩 0.9	🟩 0.8	🟩 0.8	🟩 1.3	🟩 1.3	1.1
Phytoplankton	🟩 0.9	🟩 0.9	🟩 0.7	🟩 0.7	🟩 1.1	🟩 1.1	30
Reptile	🟩 1.5	🟩 1.5	🟩 1.3	🟩 1.3	🟩 2.2	🟩 2.2	
Crustacean	🟩 2.9	🟩 2.9	🟩 2.6	🟩 2.6	🟨 4.6	🟩 4.6	
Insect larvae	🟥 16.1	🟩 16.1	🟥 19.8	🟩 19.8	🟥 54.1	🟩 54.1	
Mollusc – bivalve	🟥 19.5	🟩 19.5	🟥 22.3	🟩 22.3	🟥 56.0	🟩 56.0	25
Mollusc – gastropod	🟥 19.5	🟩 19.5	🟥 22.3	🟩 22.3	🟥 56.0	🟩 56.0	
Vascular plant	🟨 3.7	🟩 3.7	🟩 2.79	🟩 2.79	🟩 2.7	🟩 2.7	4.2
Zooplankton	🟥 16.1	🟩 16.1	🟥 19.8	🟩 19.8	🟥 54.1	🟩 54.1	

Green = The absorbed dose rate to the RAP do not exceed the SDR for neither the expected nor the conservative RQ for that SDR.

Yellow = The absorbed dose rate to the RAP do not exceed the SDR for the expected RQ but do for the conservative RQ.

Red = The absorbed dose rate to the RAP exceeds the SDR both for the expected and conservative RQ for that SDR.

Table 10 shows the absorbed dose rates to RAPs at Surpölen and Norrtorpssjön, ranging from about 2-51 and 1-11 µGy/h for the different RAPs, respectively. In comparison to the absorbed dose rate to the RAPs from natural background, both the dose rates at Surpölen and Norrtorpssjön was lower or quite similar, with Mollusc – bivalve from Surpölen being the highest away from it.

Table 10 also shows both the expected and conservative RQs via colouring for both SDRs. At both Surpölen and Norrtorpssjön no absorbed dose rates exceeded the SDR 40/400 µGy/h for neither the expected nor the conservative RQ. At Surpölen five RAPs exceeded the SDR 10 µGy/h for the conservative RQ, while four did not exceed for neither the expected nor the conservative RQ, and four exceeded it for both the expected and conservative RQ. At Norrtorpssjön two RAPs exceeded the SDR 10 µGy/h for the expected RQ, while seven did not exceed for neither the expected nor the conservative RQ, and four exceeded it only for the conservative RQ.

The radionuclide that contributed the most at Surpölen to external and internal exposure to most of the RAPs was ^{235}U and ^{210}Po, respectively. The radionuclide that contributed the most at Norrtorpssjön to external and internal exposure to most of the RAPs was ^{235}U and ^{234}U (closely followed by ^{238}U), respectively. The internal exposure made up most part of the total exposure, at both Surpölen and Norrtorpssjön.

Table 10 - Absorbed dose rate in µGy/h to RAPs at Surpölen and Norrtorpssjön. Expected and conservative RQs included via colouring for both SDRs. Absorbed dose rate to RAPs from natural background also included as comparison. Obtained from ERICA Tool assessment Tier 2.

	Surpölen		**Norrtorpssjön**		
Organism	**10 µGy/h**	**40/400 µGy/h**	**10 µGy/h**	**40/400 µGy/h**	**From natural background**
Amphibian	🟨 3.5	🟩 3.5	🟩 2.3	🟩 2.3	1.1
Benthic fish	🟩 2.4	🟩 2.4	🟩 1.4	🟩 1.4	31
Bird	🟩 1.9	🟩 1.9	🟩 1.0	🟩 1.0	0.92
Mammal	🟨 3.5	🟩 3.5	🟩 2.3	🟩 2.3	
Pelagic fish	🟩 2.4	🟩 2.4	🟩 1.4	🟩 1.4	1.1
Phytoplankton	🟩 2.2	🟩 2.2	🟩 1.4	🟩 1.4	30
Reptile	🟨 4.0	🟩 4.0	🟩 2.2	🟩 2.2	
Crustacean	🟨 7.5	🟩 7.5	🟨 3.9	🟩 3.9	
Insect larvae	🟥 41.7	🟩 41.7	🟨 3.9	🟩 3.9	
Mollusc – bivalve	🟥 50.7	🟩 50.7	🟥 10.8	🟩 10.8	25
Mollusc – gastropod	🟥 50.7	🟩 50.7	🟥 10.8	🟩 10.8	
Vascular plant	🟨 9.8	🟩 9.8	🟨 7.2	🟩 7.2	4.2
Zooplankton	🟥 41.7	🟩 41.7	🟨 3.88	🟩 3.88	

🟩 Green = The absorbed dose rate to the RAP do not exceed the SDR for neither the expected nor the conservative RQ for that SDR.

🟨 Yellow = The absorbed dose rate to the RAP do not exceed the SDR for the expected RQ but do for the conservative RQ.

🟥 Red = The absorbed dose rate to the RAP exceeds the SDR both for the expected and conservative RQ for that SDR.

3.5. Radiological Risk Analysis for Humans

Maximum dose rate and average dose rate for an adult at Kvarntorpshögen was 1.8 and 0.7 µSv/h, respectively. At Surpölen worst case and average case was 3 and 0.9 µSv/h, and at Norrtorpssjön 4.3 and 0.4 µSv/h, respectively. For visits longer than an hour, the dose accumulates linearly with time. For example, a two-hour visit (or a one hour visit twice a year) at Kvarntorpshögen would double the dose to 3.6 and 1.4 µSv. To acquire the dose for several visits during a year, the dose at the visited site is multiplied with the number of visiting hours.

The differences between the estimated effective dose rate to adults to the effective dose rate to 15-year-olds, 10-year-olds, 5-year-olds and new-borns was 3.4 %, 8.9 %, 16.8 % and 33.1 % higher, respectively. The times before adults and children of different age groups receive 1 mSv at Kvarntorpshögen, Surpölen and Norrtorpssjön are shown in Table 11, for both maximum and average dose rates. The times directly correlates to the dose rates, with higher dose rates giving less time before receiving 1 mSv, and lower dose rates giving more time before receiving 1 mSv. For the different age groups, it was shown by the differences in percent as well as in the table that smaller individuals receive higher doses faster than larger individuals.

The dose received by the most frequent visitor on the pile, the adult kiosk worker working a total of 27 whole days, was 1.2 mSv/y for maximum dose rate and 0.5 mSv/y for average dose rate. The kiosk worker thus exceeded the time for receiving 1 mSv at Kvarntorpshögen for maximum dose rate, but not for average.

The dose rate to an adult skiing in a 20 cm snow-covered ski slope at Kvarntorpshögen was 0.4 µSv/h (0.7 µSv/h without snow-cover). The time before receiving 1 mSv from skiing thus was 104 days, assuming the snow-cover is present that long (compared to 60 days needed in the slope without a snow-cover to receive 1 mSv). For the four age groups of children the time before receiving 1 mSv from skiing was 101, 96, 89 and 78 days, respectively. This is the same number of days required at an un-covered Norrtorpssjön.

The effective dose rate for immersion in water at Surpölen and Norrtorpssjön for an adult was $8 \cdot 10^{-10}$ µSv/h and $6 \cdot 10^{-10}$ µSv/h, respectively. The time before receiving 1 mSv from immersion in the waters was 135 and 180 *years*, respectively. For the four age groups of children the time before receiving 1 mSv was all above a 100 years. Compared to the times needed to receive the same external dose rate on land, the external dose rate from immersion in water was very low.

The effective dose to the representative visitors at Kvarntorpshögen (two adults and two 5-year-olds, average dose rate without snow-cover) was 3 µSv. The number of visitors during 2019 and 2020 were approximately 27 000 and 40 000. The collective dose from visiting Kvarntorpshögen during 2019 and 2020 therefore was 0.08 and 0.12 manSv, respectively.

Table 11 – Days (24 hours) before adults and children of different age groups receive 1 mSv at Kvarntorpshögen, Surpölen and Norrtorpssjön, for both maximum and average dose rate.

	Adults	**15-year-olds**	**10-year-olds**	**5-year-olds**	**New-borns**
Kvarntorpshögen					
Maximum	23	22	21	20	17
Average	60	58	55	51	45
Surpölen					
Maximum	14	13	13	12	10
Average	46	45	43	40	35
Norrtorpssjön					
Maximum	10	9	9	8	7
Average	104	101	96	89	78

4. Discussion

4.1. Activity Concentrations on Land

The activity concentration of uranium in shale ash has been seen to vary between 2500 – 6000 Bq/kg (1). The measured activity concentration of uranium at Kvarntorpshögen was on average 3700 ± 180 Bq/kg and therefore within this range. The activity concentration of ^{238}U alone in the shale ash was on average 1722 ± 124 Bq/kg. The average of ^{234}U was 1866 ± 127 Bq/kg, and therefore in equilibrium with ^{238}U in the shale ash at Kvarntorpshögen. The average of ^{235}U was 114 ± 11 Bq/kg. This is 6.6 % of the average ^{238}U contents in the shale ash at Kvarntorpshögen, which is slightly higher than the 4.6 % seen in natural uranium. The slight increase could be due to an overestimation of the net counts of ^{235}U caused by in-scattering from the ^{234}U-peak, which is more prevalent in samples with high concentration of U. The average of ^{210}Po was 1745 ± 95 Bq/kg, therefore also in equilibrium with ^{238}U and ^{234}U.

In Swedish soil from 0-25 cm depth, the average contents of ^{238}U was about 70 ± 60 Bq/kg (22), while at Kvarntorpshögen soil from 0-10 cm depth had the average contents of 698 ± 43 Bq/kg. This is a factor ten higher activity concentration, showing that Kvarntorpshögen have technologically enhanced NORM levels. The average of ^{234}U was 650 ± 40 Bq/kg, therefore in equilibrium with ^{238}U in the soil at Kvarntorpshögen. The average of ^{235}U was 35 ± 4 Bq/kg. This is about 5 % of the ^{238}U contents in the soil at Kvarntorpshögen, therefore in equilibrium with ^{238}U. The average of ^{210}Po was 635 ± 141 Bq/kg, therefore also in equilibrium with ^{238}U and ^{234}U.

In roots at Kvarntorpshögen ^{238}U, ^{234}U and ^{210}Po were in equilibrium, while the ^{235}U was about 5.7 % of the ^{238}U contents. In plants at Kvarntorpshögen only ^{238}U and ^{234}U was in equilibrium, while ^{210}Po had a higher activity concentration. This slightly higher value could be due to atmospheric deposition of ^{210}Po, originating from ^{222}Rn emerging from the ground to the leaves of the plant, thus increasing the activity concentration of ^{210}Po in a way not available for the U-isotopes. The ^{235}U was about 11.7 % of the ^{238}U contents, because of the high uncertainties of activity concentrations when the detection is near the MDA.

In plants at the Serpentine ponds the activity concentration levels of the nuclides were lower than in the plants at Kvarntorpshögen. It could be because of the distance from the pile and the land around the ponds consisting of much less shale ash than the pile. However, to conclude anything with certainty, measurements of soil at the Serpentine ponds is needed. The ^{235}U was about 9.1 % of the ^{238}U contents, again because of the high uncertainties when the activity concentration is near the MDA.

4.2. Transfer Factors

For ^{238}U, the average transfer from soil to root was about 7 %, while 1.4 % was transferred from the soil to the plant, meaning a 19.2 % transfer from root to plant. For ^{234}U the average transfer in the soil-root-plant system was very similar to ^{238}U, which is expected since they belong to the same chemical group with similar chemical properties, thus behaving similarly in nature. For ^{210}Po, the average transfer from soil to root was also similar to that of ^{238}U, however, the transfer from root to plant was about 32 %. Despite ^{238}U and ^{210}Po belonging to different chemical groups with different chemical behaviour, their transfer was quite similar, with the slight increase of transfer for ^{210}Po probably explained by the ^{222}Rn emerging from the ground and adsorbing onto the plant. For ^{235}U, belonging to the same chemical group as ^{238}U, the average transfer was also similar to that of ^{238}U, with the slight increase being explained by the high uncertainties associated with the activity concentrations near the MDA.

4.3. Activity Concentrations in Water

In the Serpentine ponds the activity concentration of ^{238}U and ^{234}U varied between about 70-200 mBq/kg. The lowest was measured for SP6 in the end of the treatment dams, having decreased from the beginning of the dams at SP1 with about 30 mBq/kg. The activity concentrations of ^{235}U and ^{210}Po, taking values of less than about 10 and 15 mBq/kg, respectively, also decreases from the beginning to the end of the dams. The total activity concentration was highest at SP2 and SP5 and second highest at SP3 and SP4.

At Surpölen and Norrtorpssjön the activity concentrations of the U-isotopes were higher than at the Serpentine ponds. They are pit lakes, and radionuclides will through weathering and leaching move from the shale walls of the lakes into the water, therefore continuously contaminating the water. The higher activity concentrations at Surpölen than at Norrtorpssjön is most probably because of Surpölen being acidic (pH around 3). Lower pH increases the mobility of most metals, resulting in higher amounts being in the sampled surface water than at the bottom. The activity concentration of ^{210}Po in all water samples was very low (some even less than MDA), especially compared to it being similar to ^{238}U and ^{234}U in shale ash, soil and plants. This can be explained by polonium and uranium belonging to different chemical groups, thus behaving differently in water. ^{210}Po in standing water is taken up by particulates and settled to the bottom, thus being removed from the water. It can also become volatile through action of microorganisms (23). For comparison, from 2875 points of groundwater measured of ^{238}U by the SGU across Sweden, about half were below 60 mBq/kg, while 15 % were equal to or higher than 370 mBq/kg (24). The water in Kvarntorp thus was in the half with activity concentrations above 60 mBq/kg being quite average, while Surpölen and Norrtorpssjön was in the 15 % equal to or higher than 370 mBq/kg being more unusual. However, more samples along different points of the lake should be taken to obtain an average for the activity concentration in surface water.

4.4. Radiological Risk to Biota

At **Kvarntorpshögen** the Tier 2 assessments showed quite different results for the minimum and maximum absorbed dose rate to the RAPs. For the risk assessment to terrestrial RAPs at the pile, the average absorbed dose rate therefore was used, to represent the whole pile instead of only a few square meters on it (however the average being quite similar to the maximum in terms of the expected and conservative RQs).

The absorbed dose rates estimated to the RAPs included natural background levels of the nuclides being investigated. For the average dose rates, none was similar to the upper range of the normal natural background stated in the ERICA Tool. The absorbed dose rates to biota at Kvarntorpshögen thus are enhanced and there might be cause for concern.

For the SDR 10 µGy/h, all RAPs except the "Mollusc – gastropod" had an expected RQ above 1, however the absorbed dose rate to the Mollusc – gastropod being very close to the SDR (probability higher than 5 % of exceeding it). For the SDR 40/400 µGy/h, the four RAPs Annelid, Arthropod – detritivorous, Flying insects and Lichen & Bryophytes also had an expected RQ above 1. However, for Lichen & Bryophytes the absorbed dose rate most probably was an overestimate since the ERICA Tool utilises soil to biota concentration ratios which may not always be applicable for Lichen & Bryophytes since lacking roots. They instead get their nutrients (and pollutants) from atmospheric deposition. The results that the internal exposure makes up most part of the exposure to RAPs at Kvarntorpshögen thus do not apply for Lichen & Bryophytes, since the external was less than 1 µGy/h, while the total was about 800 µGy/h. In addition, UNSCEAR suggest Lichen & Bryophytes being one of the least radiosensitive organisms (17).

The RAPs Amphibian, Bird, Mammal – large, Mammal – small-burrowing and Reptile had a conservative RQ above 1 for the SDR 40/400 µGy/h. The probability of exceeding the SDR thus was above 5 %, and the risk cannot be neglected. The RAPs Grasses & Herbs, Mollusc – gastropod, Shrub and Tree had a conservative RQ below 1 for the SDR 40/400 µGy/h. However, without information about which screening dose rate is the "Predicted No Effects Dose Rate" for these RAPs, a statement cannot be made that no deleterious effects are present, since their expected RQ was above 1 for the SDR 10 µGy/h.

At Tier 1 the most limiting organism was Lichen & Bryophytes. However, as explained above, Lichen & Bryophytes most probably are not the most limiting organism, meaning that the RQ at Tier 1 for the terrestrial sites was overestimated. At Tier 2 it was nonetheless confirmed that a radiological risk to biota at Kvarntorpshögen cannot be neglected.

The radionuclide that contributed the most to both internal and external exposure at Kvarntorpshögen was ^{214}Pb. ^{214}Pb is however, only a short-lived progeny of ^{226}Ra (and of course of ^{238}U, and ^{234}U which it was assumed to be in equilibrium with), continuously supplying more through its own decay in the soil. The internal exposure was greatest since many of the radionuclides in the naturally occurring decay chains primarily decay by emitting alpha particles, whose range is about 10 cm in air. The number of nuclides contributing to the internal exposure was also greater than the number of nuclides contributing to the external exposure (mostly only ^{214}Pb, ^{238}U and ^{210}Po), with the root uptake for plants and ingestion by animals probably playing a big roll.

At the **Serpentine Ponds**, the Tier 2 assessment showed quite similar results for the minimum, centre and maximum absorbed dose rate to the RAPs. For the risk assessment to freshwater RAPs at the ponds, the maximum absorbed dose rate was therefore used, to be conservative while still representing the whole water treatment system.

For the maximum absorbed dose rates, all but one was lower or quite similar to the upper range of the normal background stated in the ERICA Tool. For all RAPs with either expected and/or conservative RQs less than 1 for both SDR, it can be concluded that there is negligible cause for concern. This leaves Insect larvae, Mollusc – bivalve, Mollusc gastropod and Zooplankton, all of which had an expected RQ above 1 for the SDR 10 µGy/h. The minimum and centre absorbed dose rates to these RAPs was in the upper range of the natural background, while the maximum was approximately twice as big. This is because of the slightly higher activity concentration of ^{210}Po in the water at SP1 than in the rest of the Serpentine ponds. Without information about which screening dose rate is the "Predicted No Effects Dose Rate" for these four RAPs, a statement cannot be made that no deleterious effects are present for biota at SP1.

At tier 1 the most limiting organism was Amphibian for the U-isotopes and Insect larvae for ^{210}Po, with ^{210}Po making the Insect larvae the most limiting organism for the total assessment. At Tier 2 it was shown that the risk to Mollusc – bivalve, Mollusc – gastropod and Zooplankton was as limiting as Insect larvae in regard to ^{210}Po. ^{210}Po posed the greatest risk as internal exposure, which is why, for example Bird, Mammal or Pelagic fish do not have as high internal exposure due to ^{210}Po, since ^{210}Po is sparingly soluble in water and mainly found in the bottom sediments. The ERICA Tool estimates the activity concentration of a nuclide in the sediment from the activity concentration in water through a distribution coefficient, where it is the highest for ^{210}Po, similar for both ^{238}U and ^{234}U, and lowest for ^{235}U.

At **Surpölen,** the Tier 2 assessment showed quite similar results to the Tier 2 assessment at SP1, with the difference of Surpölen having just slightly higher activity concentrations of ^{238}U (giving a few RAPs with dose rates exceeding the SDR for the conservative RQ, where SP1 do not) and slightly lower activity concentrations of ^{210}Po (giving just slightly lower dose rates to Insect larvae, Mollusc – bivalve, Mollusc – gastropod and Zooplankton).

For the absorbed dose rates, all but one was lower or quite similar to the upper range of the normal background stated in the ERICA Tool. For all RAPs with either expected and/or conservative RQs less than 1 for both SDR, it can be concluded that there is negligible cause for concern. This leaves Insect larvae, Mollusc – bivalve, Mollusc gastropod and Zooplankton, all of which had an expected RQ above 1 for the SDR 10 µGy/h, and a dose rate approximately twice the background, caused by the ^{210}Po. Without information about which screening dose rate is the "Predicted No Effects Dose Rate" for these four RAPs, a statement cannot be made that no deleterious effects are present for biota at Surpölen. Contribution of radionuclides to internal, external and total exposure was the same as at the Serpentine ponds.

At **Norrtorpssjön** the Tier 2 assessment showed a slightly lower risk than at Surpölen and SP1, which can be explained by ^{210}Po not being included in the assessment since being below the MDA. For the absorbed dose rates, all was lower or quite similar to the upper range of the normal background stated in the ERICA Tool. For all RAPs, independent of expected or conservative RQs, it can thus be concluded that there is negligible cause for concern.

However, since only the measured radionuclides were included in the water assessment, the final impact could differ if all radionuclides were measured (e.g., ^{226}Ra). For example, Th-isotopes were not included (as it was in the soil assessment), since the thorium/uranium ratio is observed to be very low in natural waters. However, the activity concentration of Th-isotopes is higher in the sediment since it precipitates in water (25). The risk assessment to freshwater biota therefore could be an underestimate and the statement above that there is no cause for concern for most RAPs, thus applies in regard to ^{238}U, ^{235}U, ^{234}U and ^{210}Po only (not ^{210}Po at Norrtorpssjön). There might be a radiological risk associated with other nuclides not included in the assessment. Furthermore, the ERICA Tool only considers the radiological exposure of nuclides. In some cases, however, the chemical toxicity of U- and Th-isotopes will dominate the risk to organisms. The total risk to biota can thus be much greater than what the results from a radiological risk assessment states.

The information on the radiological risks to biota in Kvarntorp, generated in this work, is limited. However, the site is still under investigation, so the risks will be established in the future.

4.5. Radiological Risk to Humans

The probability of inducing cancer (both fatal and non-fatal) for the whole population is approximately 0.017 % per mSv, or 0.0055 % per mSv for fatal cancer only (26). An average non-smoker in Sweden thus increases its probability of inducing cancer by 0.041 % each year, or 0.013 % for inducing fatal cancer, just by living life in modern society. The probability of inducing cancer increases the same way for all exposures, whether it be a long-distance flight, medical diagnostics, or a trip to Kvarntorpshögen.

For an adult to receive the same exposure at Kvarntorpshögen (maximum dose rate) as during a long-distance flight, a visiting time of 10.7 hours is required. For a 15-year-old, a 10-year-old, a 5-year-old and a new-born, a visiting time of 10.3, 9.8, 9.1 and 8 hours is required, respectively. Because the radiological risk to humans is not of concern for long-distance flights once in a while, then it is not of concern for sporadic summer and winter visits at Kvarntorpshögen either, independent of age. Neither any risk to foetuses is of concern, since the exposure time is limited (27). The greater dose rate to children of younger ages compared to adults is a result of being closer to the contamination in the ground and having less overlying tissue shielding radiosensitive organs.

For an adult to receive the same exposure at Norrtorpssjön (maximum dose rate) as during a long-distance flight, a visiting time of 4.5 hours is required. For a 15-year-olds, a 10-year-old, a 5-year-old and a new-born, a visiting time of 4.3, 4.1, 3.8 and 3.4 hours is required, respectively. However, for this the person needs to stand right next to the exposed alum shale walls, and the maximum dose rate measured walking around with the back-pack was only 1.7 µSv/h (instead of 4.3 µSv/h) which is lower than the worst case at Kvarntorpshögen (1.8 µSv/h). At Surpölen, the maximum measured with the back-pack was 2.1 µSv/h (instead of 3 µSv/h), which is only slightly higher than at Kvarntorpshögen. Thus, the radiological risk to humans is not of concern at Surpölen or Norrtorpssjön either, neither on land nor in the water.

However, a fisher might stand right next to the exposed alum shale walls for hours at a time. For an adult to receive 1 mSv at worst case at Norrtorpssjön, 10 days is required. This corresponds to for example fishing 6 hours once a week for 40 out of the 52 weeks of the year. This radiological risk would also be of negligible concern since only increasing the risk of inducing cancer by approximately 0.017 % (fatal by 0.0055 %). A 10-year-old child fishing in this scenario would give a dose of 1.1 mSv, or a new-born 1.3 mSv, thus increasing the total cancer risk by 0.019 % and 0.006 %, respectively.

To set this into perspective, the number of people that would get a fatal cancer form from fishing as above would be approximately 22 people, assuming 10 000 visitors a year, with a yearly dose of 1.3 mSv per year for 30 years, with the risk of inducing cancer being 0.0055 % per mSv ($1.3 \cdot 30 \cdot 0.00005 \cdot 10000 \approx 22$). With a 10-year survival of 58 % for all cancer forms (27), this would correspond to approximately 10 deaths caused by radiation induced cancer. To set that into perspective, 30 % of all deaths in Sweden in caused by cancer (27). For the 10 000 mentioned above, 3000 people ($10000 \cdot 0.3 = 3000$) is expected to die of cancer, compared to the 10 from the radiation exposure at maximum dose rate in the Kvarntorp area. Of course, there are a lot of uncertainties with a calculation like this, but it puts the risks into perspective.

Further, it can then be concluded that the radiological risk to the kiosk worker at Kvarntorpshögen, also is of limited concern, since for maximum dose rate receiving an additional dose rate of 1.2 mSv/year and for average dose rate only 0.5 mSv/year. From this it can also be concluded that there are no risks of deterministic effects from external exposure in the Kvarntorp area, since requiring > 250 mSv (28).

The yearly effective doses to the representative visitors at Kvarntorpshögen is 9.6 mSv, assuming yearly doses to adults and children being the same. The collective dose from yearly doses during 2019 and 2020 for the 27 000 and 40 000 visitors therefore are approximately 260 and 380 manSv.

The collective doses from visiting Kvarntorpshögen (0.08 and 0.12 manSv) thus are negligible in comparison.

The effective dose rate in this work was also a conservative estimate, being the measured ambient dose equivalent rate, defined as the dose rate one meter above ground, absorbed at 1 cm depth in tissue equivalent material. Not all organs and tissues are placed at that height and depths in an adult body. However, since the risk was of little concern even for maximum dose rate at Norrtorpssjön, the ambient dose equivalent can be concluded to have been a good enough estimate for this assessment.

5. Conclusions

External dose rates in the Kvarntorp area are highest by exposed alum shale walls at the pit lakes and by shale ash at Kvarntorpshögen. The average activity concentrations of ^{238}U, ^{235}U, ^{234}U and ^{210}Po decreases in the soil-root-plant system at the pile. ^{238}U, ^{235}U, ^{234}U and ^{210}Po are in equilibrium in soil and roots but not in plants, probably due to atmospheric deposition of ^{210}Po. The average activity concentration of ^{238}U, ^{235}U, ^{234}U and ^{210}Po in plants at the Serpentine ponds were seen to be slightly lower than in plants at Kvarntorpshögen.

For water in the Kvarntorp area, the highest activity concentrations were found mainly in Surpölen because of its acidity, second highest in Norrtorpssjön and lowest in the Serpentine ponds. In the water, ^{210}Po was not in equilibrium with the U-isotopes due to differences in solubility. In the Serpentine ponds a decrease of activity concentration was seen the further away the ponds extend from the pile, showing that the treatment dams are working to reduce the concentration of radionuclides in the water.

The radiological risk to biota at Kvarntorpshögen, the Serpentine Ponds, Surpölen and Norrtorpssjön cannot be concluded as being of negligible concern. Further assessments are necessary to establish the risk. However, the radiological risk to humans at any site in the Kvarntorp area is of limited concern compared to average yearly effective doses to the Swedish population.

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