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Activity concentration and transfer factors of natural and artificial radionuclides in the Swedish counties of Uppsala and Jämtland

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Abstract

The study was performed in two areas of Sweden respectively with high natural radioactivity background (County of Jämtland), and high ^{137}Cs deposition area deriving from Chernobyl fallout (County of Uppsala).
Aims of the study were the calculation of activity concentration and transfer factors for all 6 radionuclides in the two areas and analysis of influence of soil characteristics on radionuclides behavior. In both the locations 25 centimeters depth soil cores were collected together with grass samples. After analysis with HPGe detector results showed as expected high levels of deposition of ^{137}Cs in Uppsala area and high natural radionuclides activities in Jämtland. Nevertheless the highest activity value within all the radionuclides was found for ^{40}K in Skogsvallen, which is part of Uppsala County.
In the comparison between our results and those from previous studies, ^{232}Th and ^{40}K average activities appeared in the range of other countries, while differences were found for activity of ^{238}U, which appeared markedly higher if compared to literature. Transfer factors found in our study for all the natural radionuclides were generally higher in respect to literature, whereas the opposite values resulted from cesium TF values. Comparison between the main natural radionuclides suggested that natural radioactivity background seems to be mainly due to ^{238}U. From our results available phosphorus content in the soil seemed to have the lowest influence on the behavior of all the radionuclides, compared with other soil parameters. Furthermore Möjsjövik, one of Uppsala sampling sites, showed a different behavior from all the other sampling sites, probably due to differences in soil characteristics.

Contents

1. Introduction

Radioecology started to be considered a distinct natural science since early 20th century.
Within the different areas of radioecology, it is possible to enlist nuclear and atomic physics, radiochemistry, radiobiology and radiogeology. In early 20th century two scenarios were presented as a potential utilization of nuclear power by Vernadsky: destructive and constructive force, respectively for military purposes and benefit for human beings (Aleksakhin, 2010).
The sad events of the Second World War confirmed the fact that military purposes had prevailed on the potential positive ones in the early stages. In August 1945 Japan was shocked by US' nuclear attacks. Only the coming decades will benefit from nuclear power as a useful source for energy's growing demand. Another positive utilization of nuclear power is represented by nuclear medicine. In this practice small amounts of radioactive materials are used to treat different kinds of diseases. Typical applications are for example treatments for various types of cancer or heart diseases (General Nuclear Medicine, Web).
It is reasonable to consider the natural radiation background as the base of modern radioecological studies. "Natural environmental radioactivity is considered a basic parameter on which it increases due to the anthropogenic component (artificial radioactivity)" (Aleksakhin, 2010).
Three main events affected the growing interest in radioecological studies: nuclear tests of the second half of the twentieth century, the major nuclear accidents and the need to deal with environmental issues derived from nuclear energy industries (Aleksakhin et al., 2008).

1.1. Radioactivity

Radioactivity is part of everyday life. Depending on the typology of the source it is possible to divide it in natural and artificial radioactivity.
Natural sources are cosmic rays and the radioactive content of soil and rocks. Cosmic rays are a mixture of electrons, protons and traces of heavier nuclei. Through the interaction of cosmic rays with the upper atmosphere highly penetrating gamma rays and neutrons are produced. Cosmogenic radionuclides are permanently found on the earth. The most common are those

belonging to uranium and thorium decay series. Another important naturally occurring radionuclide is ^{40}K.

Artificial radioactivity derives from man activity and is generated in different applications, ranging from nuclear installations, nuclear accident or normal radionuclides applications used in industry, e.g. medical machinery and different kinds of electronic devices. The primary man-made products are ^{90}Sr and ^{137}Cs (Ahad, 2004).

1.1.1. Alpha decay

Alpha particles are composed by two protons and two neutrons. In the emission of an alpha particle therefore a nucleus loses this combination of protons and neutrons and this process changes the nucleus of an element to the nucleus of another element (Sutton, 1988).

In alpha decay the parent atom emits an alpha particle ($^4_2\alpha$) with the consequence of a daughter nucleus containing two protons and two neutrons less than the parent. It is a process mainly found in proton rich nuclides.

An example of α-decay is found in the formation of ^{222}Rn from the parent nuclide ^{226}Ra. The latter represents the main form in which Radium is present in nature. ^{226}Ra itself is a radioactive decay product in ^{238}U series. Its half-life is 1600 years and it decays emitting an alpha particle and results in the formation of ^{222}Rn (Peterson et al., 2007).

Alpha particles interact with matter giving up their energy. The energy range of Alpha particles is between 4 to 8 MeV. They travel short distances in which they deposit their energy, having the potential to cause a lot of damage. Protection from α-radiation can be provided by thin layers such as paper (Royal Society of Chemistry, Web).

1.1.2. Beta decay

Beta particles are charged particles, either electrons or positrons. They derive from nuclei having excess of neutrons leading to instability. Neutrons can turn into protons by emitting electrons in a process called beta decay. Another way for a nucleus to reach stability consists in the emission of a positively charged particle, called positron. In this process, called beta-plus decay, a proton is converted into a neutron, a positron and a neutrino:

$$p \rightarrow n + \beta^{+} + \upsilon$$

After the emission the positron gets strongly attracted by atomic electrons and the direct consequence of this annihilation is the emission of two photons. An example of β^{+} decay can be found in:

$$^{22}Na \rightarrow ^{22}Ne + \beta^{+} + \upsilon$$

Another typology of beta decay is the beta-minus decay (β^{-}). This decay happens when a nucleus emits a negative electron from an unstable radioactive nucleus. It is typical of nuclides with neutrons excess. Immediately after the emission of the beta particle, the daughter is positively charged having the same number of electrons as the parent atom. This positive charge is quickly lost through capturing by the daughter of an electron from the surrounding. Beta radiation is an external radiation hazard. An example of β^{-} decay is found in the decay of ^{14}C to ^{14}N:

$$^{14}C \rightarrow ^{14}N + \beta^{-} + \bar{\upsilon}$$

With this type of decay an electron is emitted together with an anti-neutrino.
The third type of beta decay is called Electron Capture. The process is similar to β^{+} decay, in fact the charge of the nucleus decreases. In this case the decay results in the ejection of a neutrino and the emission of an X-ray when the lack of the electron is filled by surrounding's electrons. An example is found in the decay from beryllium-7 to lithium-7.
For β-decay energy typically ranges from a few keV to a few MeV. It has a continuous spectrum of energies, between zero and a maximum value, in contrast to α-emission, in which energy spectrum is discrete.

1.1.3. Gamma decay

The third type of radiation leads to the formation of gamma rays. When alpha and beta decay lead to the formation of a new nucleus, the latter can be in the excited form, it has more ener-

gy than usual. This surplus energy is lost by the emission of a gamma ray. This radiation is characterized by a long range emission, being them neutral. Typical energies range from a few to 3000 keV. An example is the decay of ^{152}Dy to de-excited ^{152}Dy by emission of a photon:

$$^{152}Dy \rightarrow {}^{152}Dy_{de} + \gamma$$

These three different kinds of radiation represent a danger for human health in different ways, but they all cause damage primarily by losing energy passing through matter. The energy is lost by ionization, which can cause in biological tissue chemical reactions, leading to destruction of cells or changes in the functions. Ionization power highly depends on the speed at which particles are travelling (Sutton, 1988).

1.2. Natural radionuclides

Natural radioactivity, as already stated previously, comprises two different categories, primordial radionuclides and cosmogenic radionuclides. Those belonging to the first category were originated around $4{,}5\times10^{9}$ years ago with the formation of earth; cosmogenic ones are constantly produced as a consequence of cosmic rays bombing certain kind of nuclides. Radioactive chains are originated from a parent isotope that, through a series of decays, gets to a stable isotope. The principal natural chains are those originating respectively from ^{232}Th, ^{238}U and ^{235}U (Figures 1-2-3).

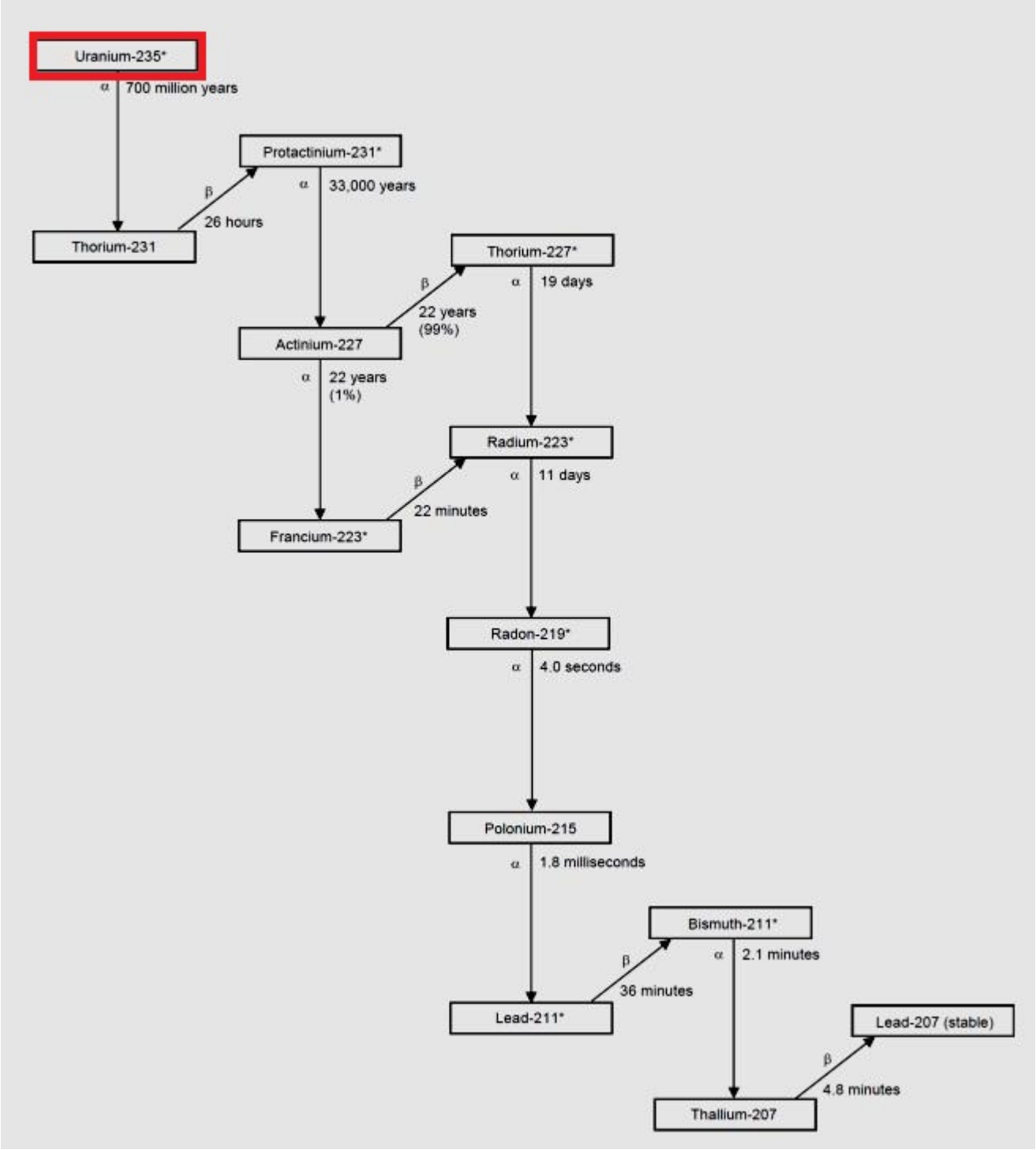

Figure 1. Decay series ^{235}U. The symbols α and β indicate alpha and beta decay and the times shown are half-lives. An asterisk indicates that the isotope is also a significant gamma emitter. Highlighted radionuclides are the ones our analysis was focused on.

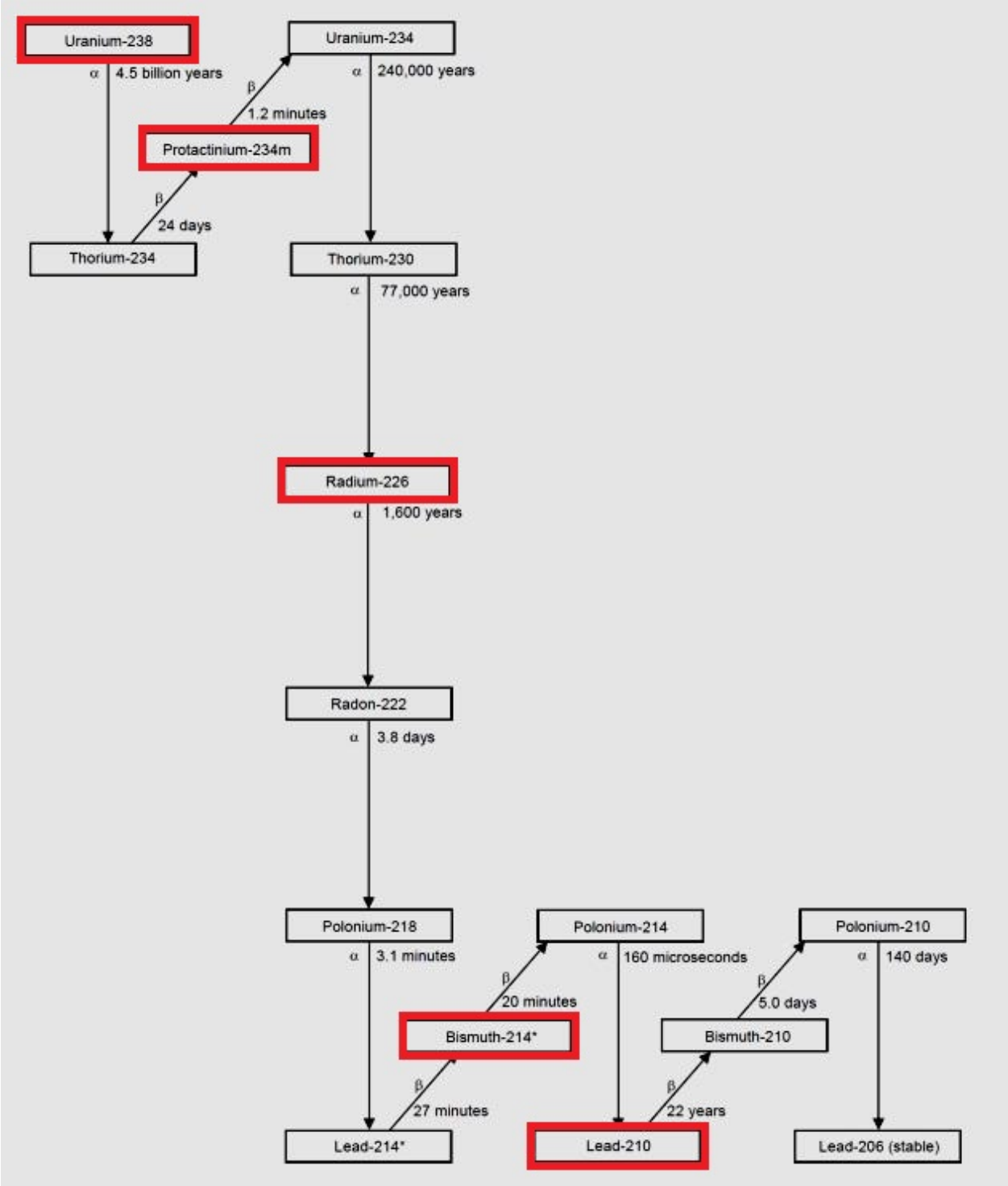

Figure 2. Decay series ^{238}U. The symbols α and β indicate alpha and beta decay and the times shown are half-lives. An asterisk indicates that the isotope is also a significant gamma emitter Highlighted radionuclides are the ones our analysis was focused on.

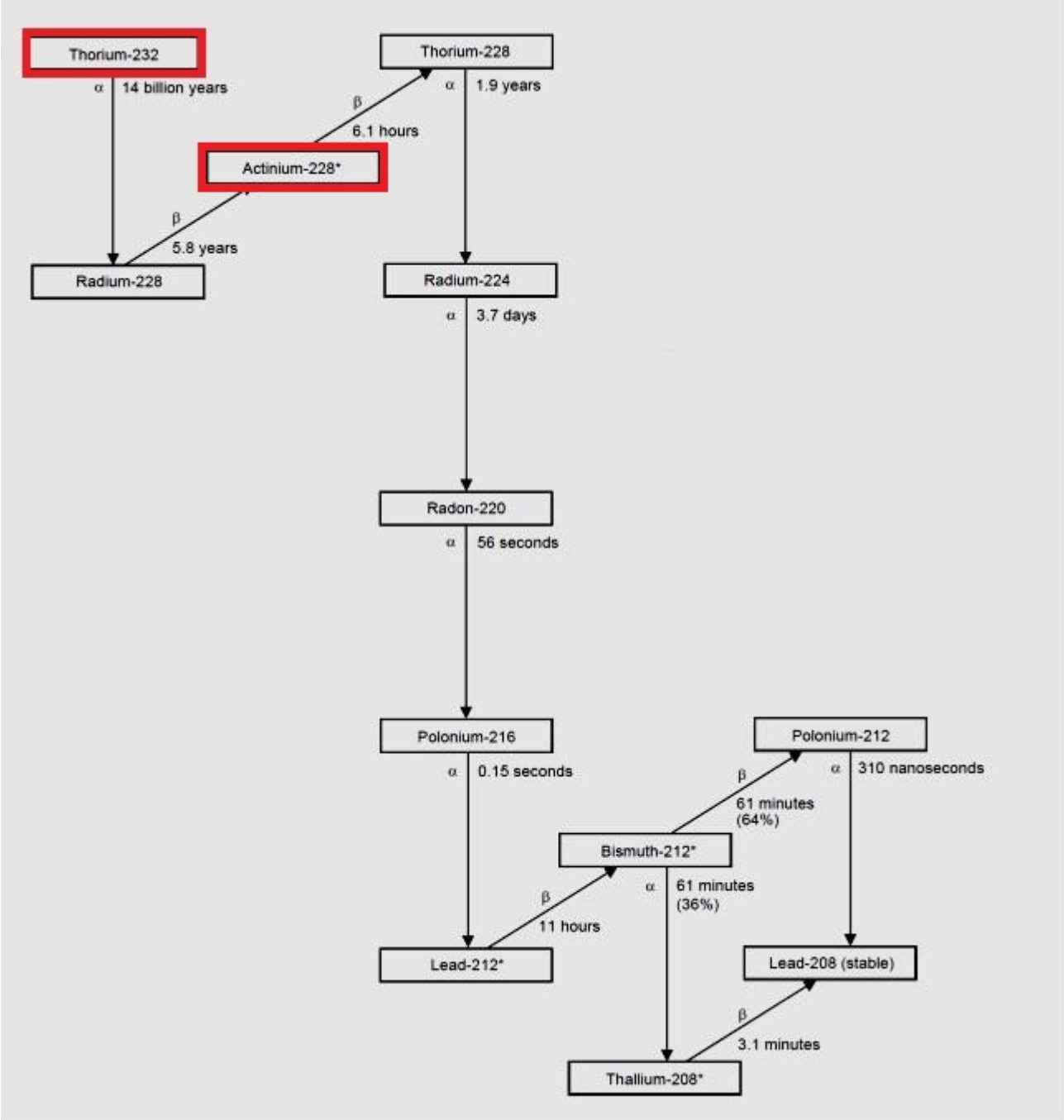

Figure 3. Decay series ^{232}Th. The symbols α and β indicate alpha and beta decay and the times shown are half-lives. An asterisk indicates that the isotope is also a significant gamma emitter. Highlighted radionuclides are the ones our analysis was focused on.

Radioactive series share a common characteristic. They all comprise radon, a radioactive gaseous isotope, typically alpha emitter. Radon isotopes have different half-life decay times, and usually when dealing with radon it is common to refer to ^{222}Rn, which is characterized by

longer half-life (3.82 days). This allows the isotope to diffuse through matter to air compartment leading to harmful effects on human health (Darby et al., 2006).
In facts Radon can be considered one of the main causes of lung cancer in non-smokers.
Entering human body through inhalation, radon decays in lung through different short living radioisotopes, until it reaches the steady state of ^{206}Pb. Once the decay products come in contact with the alveolus surface they emit alpha and beta particles, thus having physiological consequences. The main Radon contamination sources are soil, building materials and water (Andreotti, 2003).

^{232}Th is one of the natural thorium isotopes (together with e.g. ^{231}Th and ^{234}Th that are respectively daughters of ^{235}U and ^{238}U) with a half –life of $1{,}4\times10^{10}$ years. The final stable product of its decay chain is ^{208}Pb. Thorium is present at different concentrations in earth crust.
The main representatives of natural Uranium are the three isotopes 234, 235 and 238. The most abundant in earth crust is ^{238}U followed by a very weak presence of ^{235}U and ^{234}U (Eisenbud, 1997). Uranium is found in almost all soils and rocks at different concentrations depending on the nature and typology of soil (Andreotti, 2003).
As already mentioned for uranium, radium is also largely distributed in the earth crust. It's mainly present in mines and milling wastes and it is generally recognized as a waste product deriving from uranium activities. It is currently used in medical field for treatment of various kinds of cancer. It can enter the human body through either alimentation or respiration. The percentage of radium remaining in the blood stream reaches different vital parts of the body. Behaving similarly to calcium radium affects mainly bones and teeth. Both ^{226}Ra and ^{228}Ra decay to the gaseous product Radon, charged ions that attach directly to dust particles. Therefore the risk is directly connected with inhalation. Particles which remain unattached travel deep through the respiratory tract, till they reach lungs, where they stay with long residence time. In addition to internal α-radiation risk, there's an external γ exposure risk (Peterson, 2007).
Within natural radionuclides potassium isotopes ^{40}K is unstable. With a half-life of $1{,}227\text{x } 10^9$ years is one of the most important from bioenvironmental point of view. It decays by β^- decay to ^{40}Ca and β^+ or electron capture to ^{40}Ar (Ahad, 2004). ^{40}K is the radioactive isotope of Potassium, which is the eighth most abundant element in earth's crust. Its presence in the form of radioactive isotope, is accountable to 0.0117% in natural potassium (Choppin et al., 2002).
It has a branched decay chain, leading to the formation of ^{40}Ar and ^{40}Ca.

The soil-plant-man is the principal pathway being studied for the transfer of radionuclides to human beings (IAEA, 1982).

The plant uptake of the main natural radionuclides, uranium, thorium, potassium and radium, is naturally due to the plant's need of nutrients. Through the mineral uptake process the plant transfers natural radioactive substances, which accumulate in the plant vital portions, including the edible ones. Radionuclides are taken up by plants by some of the same mechanisms as plant nutrients due to their similar chemical and physical characteristics. In this way, radionuclides enter human food chain through consumption and lead to a long-term internal exposure to human body (Pulhani et al., 2004). The major concerns derive from ^{238}U, ^{232}Th series and ^{40}K.

Public opinion commonly tends to give more weight to man-made radiation, in terms of dangerousness. Analyzing the problem in terms of exposure, human beings are constantly exposed to natural radiation, since its origins can be found in different compartments: air, soil and even human body itself (Hafezi et al., 2005).

Cosmogenic radionuclides are constantly produced and brought to earth surface by precipitations. The causes of this process are found in the production of neutrons and protons by cosmic irradiation of the atmosphere. This radiation type is the main threat to frequent flyers, as for example air flights personnel. Cosmic radiation dose to travelers depends on different factors like altitude, latitude, duration of exposure and solar activity.

In the assessment of the risks deriving from radionuclides exposure it is important to know the dose limit of exposure and the background natural environmental radiation (UNSCEAR, 1993).

Usually radiation exposure assessment is done mainly because natural radionuclides get dissolved into water and can potentially lead to a contamination of foodstuff and introduction to the human food chain. Radioactive material can attach suspended particle (gaseous radionuclides) thus being incorporated through inhalation (Amaral et al., 2005)

1.3. Artificial radionuclides

Artificial radioactivity is present in the environment as a consequence of different anthropogenic sources. It is possible to enlist some of these, ranging from nuclear weapons to radioactive material leaching from nuclear power plants and waste products of general and medical industry. After the first denotation of nuclear bombs in New Mexico and Japan, other tests occurred mainly in the period between 1952 and 1963 (Avery, 1996). Radioactivity derived from nuclear testing has been subject to natural decay from then on. Another source of artificial radioactivity in the environment comes from controlled discharge waste resulting from nuclear reactors. This is the case for example of cooling waters for nuclear power plants. There are two basic waste products classes, high level and low level waste products. While the first ones must be stored, the latter are discharged into water basins, after proper monitoring. It's important to take into account the fact that releases from different kinds of power plants differ from case to case, for what concern radioactive content.

The degree of contamination deriving from ^{137}Cs waste products decreases with time. For example "in a spent reactor fuel with an initial ^{137}Cs activity comprising 33% of the total activity. ^{137}Cs will account for only approximately 6% after 300 years" (Oversby, 1987).

The last largest anthropogenic source of radionuclides in nature is attributed to accidental releases from nuclear power plants.

1.4. Nuclear accidents

As already mentioned nuclear power plants are one of the most discussed and up-to-date topics at present times.

Discussing nuclear accident, it comes natural to refer to one of the largest nuclear accident of all the time, the explosion in the fourth reactor of the Chernobyl power plant in 1986. As a consequence of the accident $7x10^{16}$ Bq ^{137}Cs were released (Avery, 1996). Radioactive material was mixed up with air particles in the atmosphere and reached altitudes of 1000 m. The spreading of contaminated material resulting from the fallout was driven by different factors among others comprising winds directions and rainfall patterns. Most likely rainfalls played a critical role in the degree of distribution of ^{137}Cs in soil profile (Avery, 1996).

Other examples of malfunctioning in nuclear power plants can be found in the Three Miles Islands accident (TMI). On March 28, 1979 the fuel core of reactor number two remained

uncovered due to a dropping in the level of the reactor coolant water. The result was the melting of one-third of the fuel (American Nuclear Society, Web). The main radionuclide found in the environment after this accident was xenon-133, a noble gas with a relatively short half-life (5.3 days) which represents a hazard from external exposure point of view, leading to production of gamma radiation (Dickinson College, Web).

Another accident involved the two air-cooled uranium metal reactors operating at Windscale, UK, in early 1950's. Around 20 kg of Uranium were released in the atmosphere. The reactor was seriously damaged and a release of volatile radioactivity, iodine and noble gases, took place. ^{131}I was spread to the surrounding areas due to varying weather conditions. Radioactive gases were transferred to animal breeding areas, with the consequent accumulation of radioactive iodine in milk. Estimates showed that the contamination involved an area of around 500 km^2 (U.S. Department of Energy, Web).

The most recent nuclear accident is the one that took place in Japan in 2011. On the 11th of March early morning local time, an earthquake took place on the coasts of the region of Tohoku. The earthquake (with a magnitude of 9.0 in Richter scale) had as an immediate consequence the formation of a giant tsunami. This caused, besides physical direct damages to the eastern part of Japan, a serious accident at the Fukushima Nuclear Power Plant (NPP) of Tokyo Electric Power Co. (TEPCO), in particular reactor n.1.

Emergency generator stopped working as a consequence of two different subsequent tsunami waves that caused the flooding of buildings containing the nuclear reactors. Cooling systems were at that time not available anymore. The day after levels of cooling waters of reactors number 1 and 3 were lowered with partial melting of fuel rods (Mimura et al., 2011). Estimates by the Incorporated Administrative Agency Japan Nuclear Energy Safety Organization (JNES) mention that the discharge of radioactive material from the reactors were approximately $1.6x10^{17}$ Bq for ^{131}I and $1.5x10^{16}$ Bq for ^{137}Cs. For what concerns contaminated water outflow results via sampling showed a discharge of about 4.7×10^{15} Bq in the sea. (NERH Government of Japan, 2011).

1.5. Detection of ionizing radiation

Detectors' functioning is based on the interaction between a particle like for example α–particles, or a photon and the detector material.
The energy contained in the radiation is transmitted to the detector and subsequently converted to an electrical signal, readable by a proper device. Detectors give an outcome which is translated into electrical signals. If necessary these signals can be filtrated, amplified and eventually converted into digital data (Andreotti, 2003). The interaction of radiation with matter is carried out mainly due to three effects:

- *Photoelectric effect*: photons deriving from γ-rays interact with the atoms of the detection material and their energy is entirely transferred to those atoms. An electron will be emitted from the orbital with an energy E equal to the difference between the energy of the incident photon and the linking one:

  $$E = h\upsilon - E_b$$

  The lack of an electron will cause the ionized atom to capture a free one, re-gaining the electronic equilibrium with the emission of an X-ray. This process will be revealed in the machine, represented as a full energy peak. This kind of interaction is typical for low energy γ-rays.

- *Compton effect:* this is another case of interaction between a photon and an electron of the detection material. The incident γ-ray will be deflected of an angle θ compared to the initial direction inversely proportional to the energy. Therefore the main difference with photoelectric effect lies in the fact that part of the energy of the photon is transferred to the electron and the rest of the energy is conserved by the photon to keep its movement.
- *Pairs production*: this interaction takes place in case the energy of the photon doubles the electron energy in the stable state:

  $$h\upsilon \geq 2m_0c^2 = 1.02MeV$$

In this case there's the formation of electron-positron pairs with total energy:

$$E_{e^-} + E_{e^+} = h\upsilon - 2m_0c^2$$

The energy contained in the pair is released in the target material.

1.6. Detectors of ionizing radiation

Depending on the radiation analysis needed it is possible to have recourse to different techniques and measurements. All the track measurements are based on the concept that nuclear particles in cloud chambers, in solids and photographic emulsions form tracks that can be observed, thus proving the existence of a nuclear reactions and radioactive decay processes.
Detection by a device can be performed in the following way: when a nuclear particle enters a device it produces either excitation or ionization. If the excitation is followed by fluorescent de-excitation, the light emitted can be recognized by proper detector as photomultipliers (PMT) and subsequently transformed into in electric signal. These kinds of devices are called Scintillation detectors. Depending on the material of the scintillation material it is possible to find solid, liquid or gas scintillator detectors.
The second type of detectors is based on the production of charge carriers between the charged electrodes contained in the detector itself. This charge creates also in this case a current signal. Devices based on this process can be solid (semiconductor crystals) or gas filled, usually divided into ion chambers, proportional and Geiger-Müller tubes.

1.7. High purity Germanium detector (HPGe).

In our analysis, the presence of radionuclides in the samples collected has been assessed trough γ-spectrometry using a High-Purity Germanium detector (HPGe).
A Germanium detector is a semiconductor diode in which the intrinsic region is sensitive to ionizing radiation. Photons interact with material contained in the volume of the detector with a consequent production of charge carriers that are moved by the electric field towards the electrodes. This charge, proportional to the energy in analysis brought by the photons, will be converted into a voltage pulse by a preamplifier. HPGe provides the best energy resolution for

detection of radiation but low efficiency. Depending on the impurity dopant it is possible to find three different kinds of detectors:

- N-type: germanium crystals with donor (N-type) impurities.
- P-type: germanium crystals with acceptor (P-type) impurities.
- P-PC: A point contact is used instead of the inner core of a coaxial crystal.

A γ-detector is mainly characterized by three components: a detector system, electronic chain and a recording and identification system.

- *Detector system* is particularly important for background rumor filtering. This component is useful to reduce the background noise due to the presence of natural radioactivity in the surroundings.

- *Electronic chain* is the system through which the signals are analyzed. It can be divided into:
    - Preamplifier: the first element of the electronic chain which causes a response proportional to the quantity of charges produced by the incident ray.
    - Amplifier: the main function of the second element of the electronic chain is creating and amplifying the signal. Pulses are properly converted into signals in a way that the ratio between signal and background noise is maximized. Within the different phases, this is the step in which operational decision has to be taken, user's discretion, like the time unit or gain.
    - ADC/MCA: the analog digital converter (ADC) rounds up the analogic information and records the pulses height distribution. This information is subsequently collected by a multichannel analyzer (MCA), which combines the number of signals with widths included in the same range in one single channel.

- The final step of the process is the *recording and identification system.* Data are stored and transferred to a calculator, commonly capable to render the results in a user-friendly interface.

Based on the analysis of radionuclides different characteristics of the detector have to be taken into account. The choice of a particular kind of detector is driven by the specific required range of gamma ray object of the study. Resolution and efficiency are directly linked with a good result in the identification of a specific radionuclide. Resolution is the capacity of a detector to separate two different peaks close to each other in energy range. Resolution increases with narrowing of peaks. Efficiency is the ratio between the number of counts produced by γ-decay and the activity, expressed in disintegration per second.
Characteristics of the HPGe used for our analysis are presented in Table 1.

Table 1. Specific characteristics of HPGe detector used for the measurements

Dimensions	
Detector Diameter	66.5 mm
Detector Lenght	68.8 mm
Absorbing Layers	
Aluminum	1.00 mm
Inactive Germanium	0.5 μm
Performance	
Resolution (FWHM[a]) at 1.33 MeV, ^{60}Co	1.90 keV
Relative Efficiency at 1.33 MeV, ^{60}Co	0.90 keV

[a] Full With Half Maximum

1.8.1 Energy and Efficiency calibration

Before the measurement session, the equipment was first calibrated, that means that standard values were set in the device in order to obtain reliable results. The machine was calibrated using a gamma emitter sample of know energy that allows determining a linear equation relating gamma energy to channel number:

$$E = mx + c$$

Where E corresponds to the gamma energy, m and c are constants, and x is the relative channel.

Once energy calibration was performed, the machine was calibrated concerning efficiency. Efficiency depends on gamma ray energy, geometry and density of the sample. The different parameters for the main radionuclides, provided in the standard sample documents, were entered in the machine settings through the detector software. The standard sample was then analyzed in the detector for a cycle from which an efficiency curve was obtained. The curve relates different levels of energies (keV) to a specific efficiency, expressed as a percentage, within a given range. The wider the range in energy, the larger is the number of radionuclides whose concentration can be determined. "The wider the range in energy, the larger the number of radionuclides whose concentration can be determined. To measure the main natural γ-ray emitters, the efficiency should be known at least from 46-54 keV (^{210}Pb) to 1836 keV (^{88}Y)" (Harb et al., 2008).

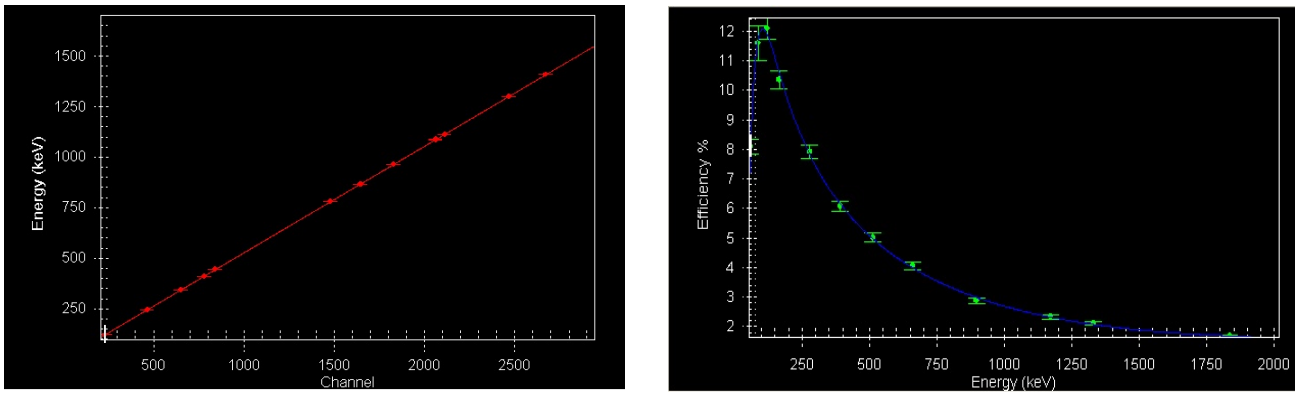

Figure 4. Energy calibration, linear plot and efficiency calibration curve of the HPGe detector used in our analysis.

1.9. Aim of the study

The aim of the study was to:

- Calculate activities concentration of different natural radionuclides and ^{137}Cs in two different Swedish Provinces with high and low levels of natural and artificial radiation.
- Estimate the transfer factor of these radionuclides from soil to plant
- Analyze correlations between soil characteristics and radionuclides concentration

2. Materials and Methods

2.1. Study area

Two areas of Sweden were selected for the study attending to the natural background. First, a high natural background area located in the Jämtland County was chosen (Fig.5). The samples from this area were taken in 4 locations: Hallen, Myrviken, Backfors and Vikdrolet. The second area in our study corresponds to a low natural background area in the surroundings of Uppsala (Fig.5). However in this area the deposition of ^{137}Cs due to the Chernobyl fallout was particularly high. The samples from this area were taken in the locations of Lövstalot, Möjsjövik and Skogsvallen. The sampling was performed to assess the radionuclides content and activity in each soil and grass sample.

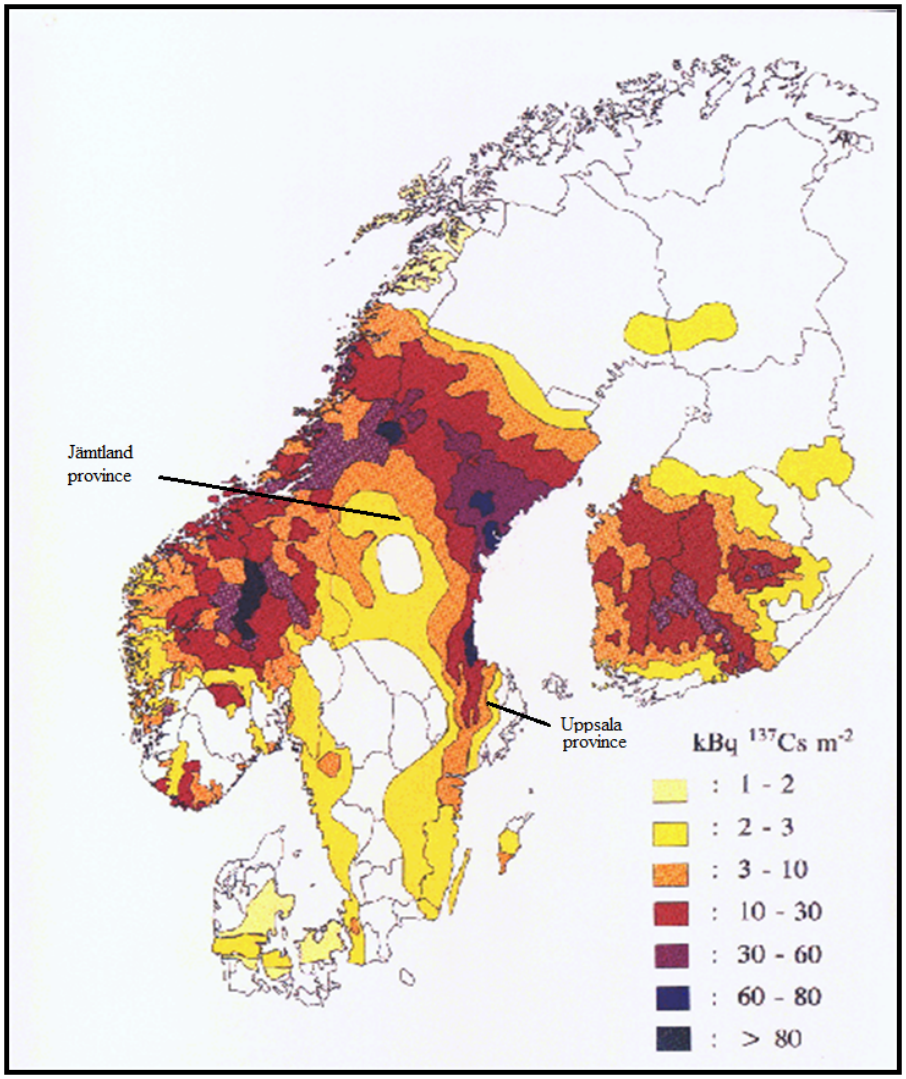

Figure 5. Maps of deposition of ^{137}Cs in Sweden with indication of the two provinces

2.1.2 Description of the study locations

Möjsjövik is located 25 km west of Uppsala in Uppsala County. The soil is characterized by organic matter and used mainly for cattle and sheep grazing (Rosèn et al., 1999). See Appendix 4 for the samples site geographical coordinates for the three sub-locations A,B and C.

The second site, Lövstalöt, is an area 12 km north of Uppsala. Area used for pasture since the last centuries, its soil composition is mainly characterized by sandier soil up-slope and clay soil down-slope. The main land cover is grass with forbs. Soil characteristics are pH around 5.5 and 3.6% of organic carbon and 0.3% of nitrogen. (Herman, 1997). Geographical coordinates in Appendix 4.

The third location in Uppsala area is Skogsvallen. Located north-west of Uppsala it is an area characterized by mainly silty clay soil and used mainly for permanent pasture. No animal grazing is present. Farming is practiced extensively and the soil has not been ploughed since 1986. The samples were collected in a narrow field strip enclosed in between an esker and a moraine hill (Persson, 2008).

The second location object of the study is located in the province of Jämtland, in the area surrounding the lake Storsjön. Situated in the north-western part of Sweden, Jämtland is the second largest province in the country, covering a surface of about 34000 square kilometers. Most of the province is a highland region. Since last ice age ended materials like stones and boulders were left behind and therefore the great part of the soil compound in the province is till. The majority of the population in the province has always been living in the area surrounding the lake Storsjön. This was due to the fertility of the soil composition that makes it particularly suitable for farming and general agricultural activities. See Appendix 4 for GPS coordinates.

2.2. Soil and Grass sampling

In all the seven sampling locations of the two Counties, three sub-sites of circular shape called A, B and C were designed with a radius of 3 m each (28.27 m^2), with a distance of 20 m referring to the center of each sub-site (Fig. 7). Thus the total sampling area accounted for 84.82 m^2 in each location. In each sub-site twelve 25 cm depth soil cores were collected with a soil sampler together with four grass samples (a surface of 1 m^2 each). The result was that in both locations a total of 36 bulk soil cores and 12 m^2 of grass sample were randomly collected. See figure 7 for a description of the sampling scheme. The soil sampler's diameter is 4.6 cm and the depth of sampling was set by adjusting the sampler length to 25 cm (Figure 6).

The grass samples were collected by cutting with grass shears five centimeters above ground level to avoid any contamination with soil, within the borders of a one square meter metal shape in each quarter of each sub-site. Samples were collected in plastic bags during field work and then transferred to the lab for treatment and analysis.

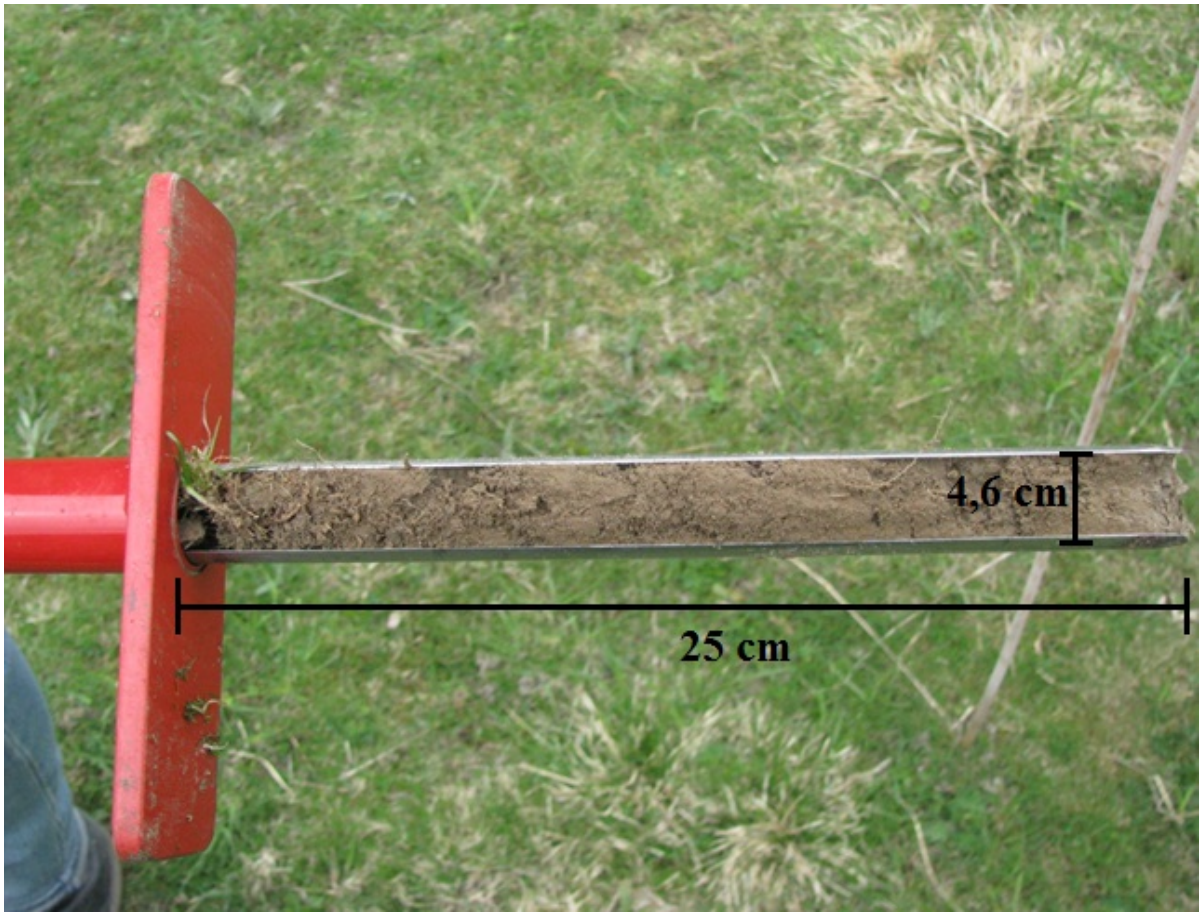

Figure 6. Soil bulk sampler

After collection single grass and soil samples collected in the same sub-site were mixed together in order to select a random aliquot of the area of study. This process is necessary to avoid sampling bias.

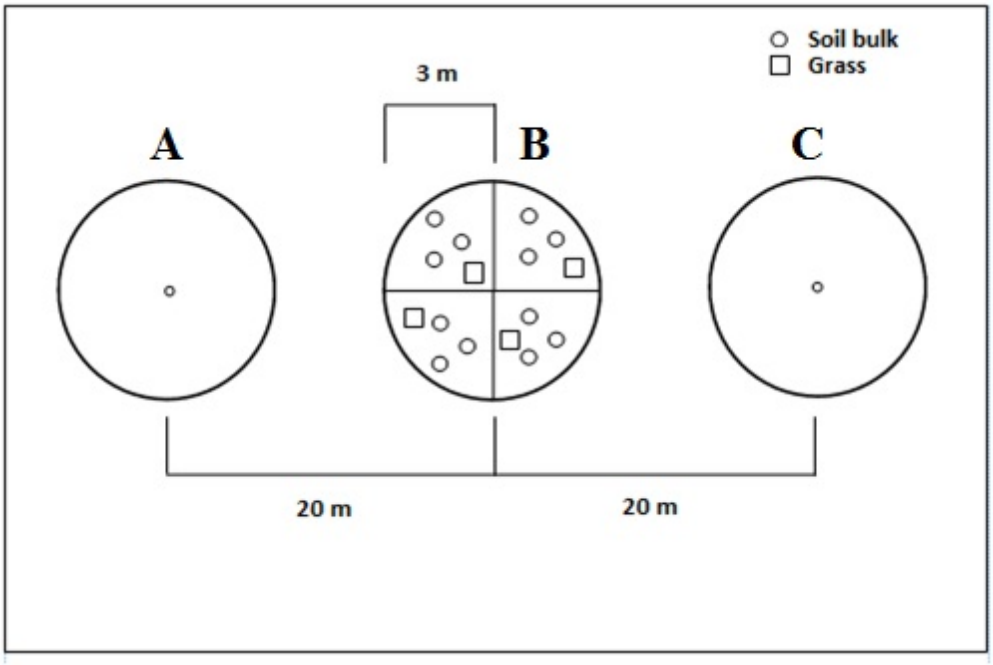

Figure 7. Sampling work procedure scheme

2.3. Samples preparation

The soil and grass samples were dried at a temperature of around 60°C for one week. The samples' weight was registered both before and after drying, in order to have an estimate of the water content. After drying bulk soil was sieved and then shredded with a pestle in order to reduce soil particle's size (the size of the sieve's meshes was 2 mm). For the same reason grass samples were milled with a milling machine after drying.

After homogenization, a representative amount from all samples was stored in 60 ml plastic petri dishes, closed with parafilm, to prevent any radon leakage. These containers were put in HPGe detector for the analysis. Each soil sample needed a 24 hours analysis cycle for proper nuclides detection, while grass samples run 72 hours cycles. This difference in analysis duration was due to difference in the detection limits. The activities registered were expressed in Bq/kg dry weight.

For measurements of activity concentration of ^{226}Ra the short living daughters were used: ^{214}Pb and ^{214}Bi with respective half-lives of 27 and 20 minutes. It is possible to achieve secular equilibrium between ^{226}Ra and ^{222}Rn due to the large difference in their half-lives, which are 3.8 days and 1600 years, respectively. In order to reach this equilibrium we had to wait around 30 days. Once this time is elapsed, it is possible to use ^{222}Rn gamma daughters to determine radon's activity and as a consequence activity of ^{226}Ra in the sample. This is possible

because, as previously mentioned, samples are sealed off and therefore all radon generated in the sample is due to ^{226}Ra content.

The energy used for calculation of the activities for the different radionuclides are displayed in table 2.

Table 2. Radionuclides of interest in our work and daughters used to estimate parent's activities with respective energy levels.

Radionuclide	Daughter (energy level)
^{238}U	^{234m}Pa (1001 keV)
^{232}Th	^{228}Ac (911.6 keV)
^{226}Ra	^{214}Bi (1764.49 keV)
^{210}Pb	^{210}Pb (46.54 keV)
^{40}K	^{40}K (1460 keV)
^{137}Cs	^{137}Cs (661 keV)

2.4. Data treatment

The transfer factor is a tool in the form of a mathematical equation that is used to express the uptake of radionuclides from soil by the plants. For ^{137}Cs, it is obtained by the activity of plant dry matter divided by the activity of deposition on the ground (Rosén et al., 1999).

$$TF_g = \frac{\text{Activity concentration plant dry matter}}{\text{Activity deposited on ground}} \left[\frac{Bq_{plant}\ Kg_{dw}^{-1}}{Bq_{soil}\ m^{-2}} = \frac{m^2}{Kg_{dw}} \right] \qquad (1)$$

For natural radionuclides instead of considering deposition on the ground, the activity concentration in plant matter is divided directly by the activity in dry soil matter.

$$TF_g = \frac{\text{Activity concentration plant dry matter}}{\text{Activity in dry soil matter}} \left[\frac{Bq_{plant}\ Kg_{dw}^{-1})}{Bq_{soil}\ Kg_{dw}^{-1}} \right] \qquad (2)$$

Levels of ground deposition for ^{137}Cs were calculated based on activity measurements and the characteristics of the single bulk sample. In fact deposition was calculated as activity concentration of radiocaesium in the sample, multiplied by the weight of the petri dish and divided by the diameter of the soil sampler:

$$\text{Ground depositon} = \frac{Activity_{^{137}Cs} \cdot Weight}{d} \left[\frac{\mathrm{Bq_{soil}\ Kg_{dw}}^{-1}\mathrm{Kg_{dw}})}{\mathrm{m^2}} = \frac{\mathrm{Bq_{soil}}}{\mathrm{m^2}} \right] \qquad (3)$$

The data obtained from the measurements were compared and analyzed through linear regression trend lines, with focus on R^2 values and the respective correlation coefficients between the different data sets.

3. Results

In the analysis, activity concentration was measured through the HPGe for both natural radionuclides and ^{137}Cs. Appendix 1 shows the activity concentrations measured in samples collected in Jämtland province, expressed in Bq kg^{-1}. Activity concentrations for the Uppsala area are displayed in Appendix 2.

Data lack is due to the minimum detectable activity (MDA) overcoming the effective activity measured for the single radionuclide. The uncertainties correspond to the standard deviations of the activity concentrations related to the three different subsites (A, B and C).

3.1. Jämtland province

3.1.1. Soil

In table 3 the averaged activity concentrations of the three different subsites (A, B and C) for each sampling site are displayed. The highest values in the whole province are registered for ^{40}K, followed by ^{238}U, ^{226}Ra and ^{210}Pb. The lowest activities are those measured for ^{137}Cs and ^{232}Th.

In Hallen sampling location, all the natural radionuclides show a low activity concentration, compared to the other sites except for ^{40}K.

Table 3. The average activity concentrations in soil bulk samples in Jämtland area (Bq kg^{-1}) with relative uncertainties registered. ^{137}Cs deposition unit is expressed in Bq m^{-2}

	Myrviken	Hallen	Backfors	Vikdrolet
Pb-210	198±28	65±7	194±18	425±55
Th-232	31±2	31±3	37±2	34±3
U-238	426±102	-	478±19	631±107
Ra-226	259±9	49±4	249±8	411±13
K-40	697±20	681±21	845±22	872±24
Cs-137	9±0.4	5±1	5±0.3	9±0.4
Cs-137 (Deposition)	326±29	146±9	199±42	316±53

Figure 8 shows a graphical presentation of the results again it is clear the difference between ^{40}K and the other radionuclides in all the four sampling locations. Within natural radionuclides, ^{238}U shows slightly higher values compared to ^{210}Pb and ^{226}Ra. ^{137}Cs activity levels appear to be very low.

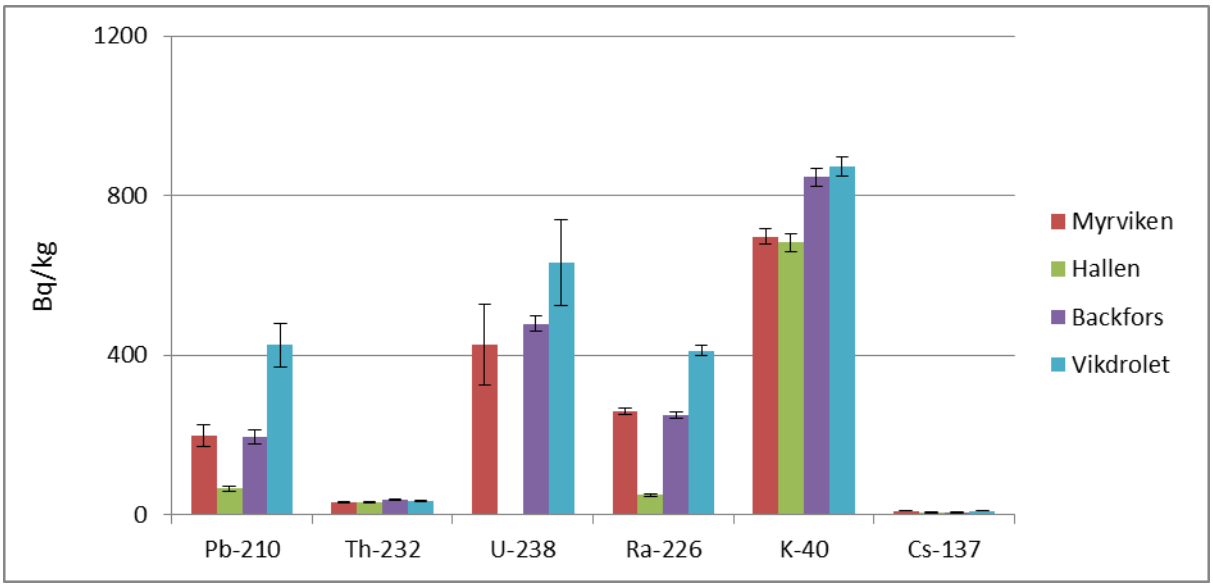

Figure 8. Comparison between the activity concentrations in soil samples for Jämtland sampling sites

3.1.2. Grass

Grass samples collected in northern part of Sweden show, as for soil samples, a marked natural radioactivity component. The highest values of activity concentration are represented by ^{40}K, with a peak in the location of Myrviken. ^{232}Th appears to be lower in activity compared to the other available natural radionuclide values and artificial radioactivity represents the lowest value.

The graph (Fig.9) brings the attention on ^{40}K in particular, since it is the only radionuclide to be present in all the locations and with highest values. The only exception data is found in Backfors, in which the highest activity concentration corresponds to ^{238}U with 664 Bq kg^{-1}. Still the trend is the one found in soil bulk measurements, according to which natural radionuclides activity overcomes distinctly the one obtained for ^{137}Cs.

^{210}Pb activities are not displayed in the analysis, exemption done for Myrviken location, due to MDA values being higher than activities detected.

Table 4. The average activity concentrations in grass samples in Jämtland area (Bq kg^{-1})

	Myrviken	Hallen	Backfors	Vikdrolet
Pb-210	213±2	-	-	-
Th-232	27	-	24±2	27±1
U-238	829±55	-	664±85	385±76
Ra-226	178±4	-	172	146±7
K-40	1141±38	856±35	499±30	560±31
Cs-137	-	-	-	3

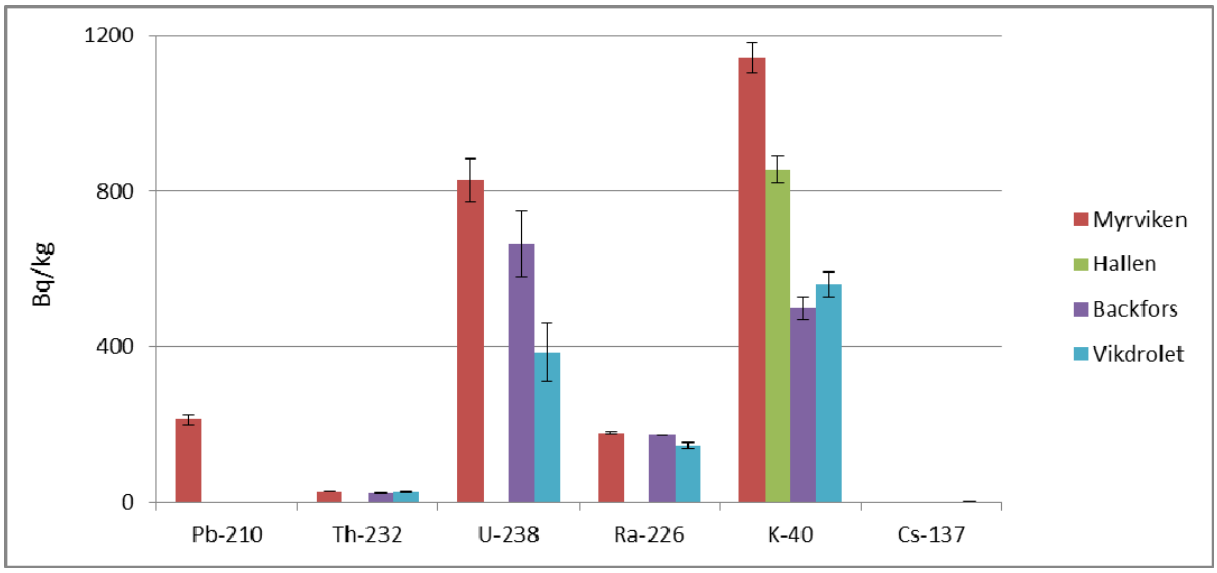

Figure 9. Comparison between the activity concentrations in grass samples for Jämtland sampling sites

3.1.3. Transfer factor

The transfer factor calculated with equation (1) for the 5 natural radionuclides object of the study are displayed in Table 5.

The highest values are registered for ^{238}U in Myrviken site and for ^{40}K in both Myrviken and Hallen. Backfors and Vikdrolet show an average transfer factor of roughly 0.6 for all the radionuclides analyzed.

Table 5. Transfer factor for natural radionuclides in Jämtland province.

	Myrviken	Hallen	Backfors	Vikdrolet
Pb-210	1.1	-	-	-
Th-232	0.9	-	0.6	0.8
U-238	1.7	-	0.8	0.8
Ra-226	0.7	-	0.5	0.5
K-40	1.6	1.3	0.6	0.6

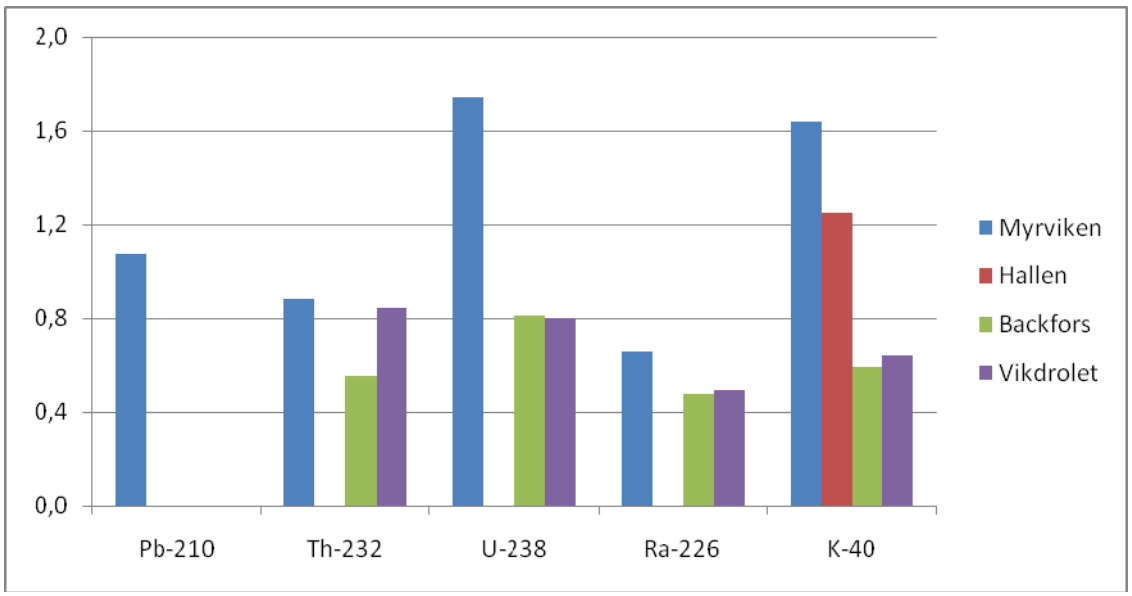

Figure 10. Comparison between transfer factors for Jämtland sampling sites

3.2. Uppsala province

3.2.1. Soil

Table 6 shows the results from the lab analysis obtained for Uppsala area for what concerns the averaged activities in each site.

In the analysis, two different datasets for Möjsjövik location were used. The measurements differ in terms of sampling date and show significant variances. In the comparison between Möjsjövik 2009 and Möjsjövik 2011 a noticeable decrease in activity concentration for most of the radionuclides from 2009 and 2011 appears clear in Table 6. In details ^{232}Th, ^{40}K and

^{137}Cs activities decreased roughly of a 30% of the initial values, while ^{226}Ra decreased to 50% of the activity registered in 2009. In general activity concentrations for ^{137}Cs are higher if compared to the values registered in Jämtland, both in Lövstalöt and Skogsvallen, with peaks in both Möjsjövik measurements, in which activity of the artificial nuclide is in 2011 equal to 10 times the activities found in the other two sites. To notice that ^{40}K in Skogsvallen and Lövstalöt registered similar activities values (1040 and 849 Bq kg^{-1} respectively) if compared to those found for ^{137}Cs in the County.

Thorium and radium display lower values in all the sampling sites.

From available data, uranium and lead isotopes show high values both in Möjsjövik (2011) and Lövstalöt.

Table 6. The average activity concentrations in soil bulk samples in Uppsala area (Bq kg^{-1}). ^{137}Cs deposition unit is expressed in Bq m^{-2}

	Möjsjövik[a]	Möjsjövik (2011)[b]	Lövstalöt	Skogsvallen
Pb-210	-	719±90	356±45	-
Th-232	47±3	32±3	50±2	63±2
U-238	-	593±115	307±79	-
Ra-226	62±7	33±6	49±4	65±5
K-40	110±11	76±15	849±22	1040±37
Cs-137	1086±24	784±14	70±2	75±2
Cs-137 (Deposition)	24219±3530	17372±2833	2960±448	4067±2872

a. Activities concentration values obtained from field work in 2009 in the area of Möjsjövik

b. Activities concentration values obtained from field work in 2011 in the area of Möjsjövik

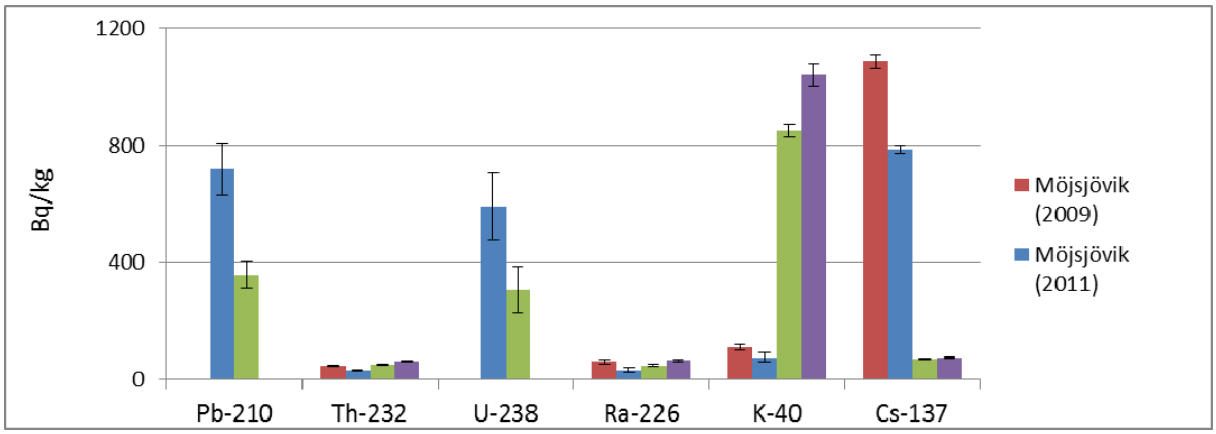

Figure 11. Comparison between the activity concentrations in soil samples for Uppsala sampling sites

3.2.2. Grass

From Fig.12 is clear the difference between ^{40}K activity and the rest of the radionuclides. The analysis outcome shows the highest activity concentrations for ^{40}K in the three locations (Table 7). With available data, high activity was also registered for ^{238}U.

As for ^{137}Cs, activity values remain noticeably high in Möjsjövik, opposed to Lövstalöt and Skogsvallen, in which evidence shows both markedly low activities.

Table 7. The average activity concentrations in grass samples in Uppsala area (Bq kg^{-1})

	Möjsjövik (2009)	Lövstalöt	Skogsvallen
Pb-210	130	158	-
Th-232	24±5	20	-
U-238	-	431±35	-
Ra-226	156	-	51±4
K-40	435±28	770±17	543±32
Cs-137	228±5	3±0.1	22±1

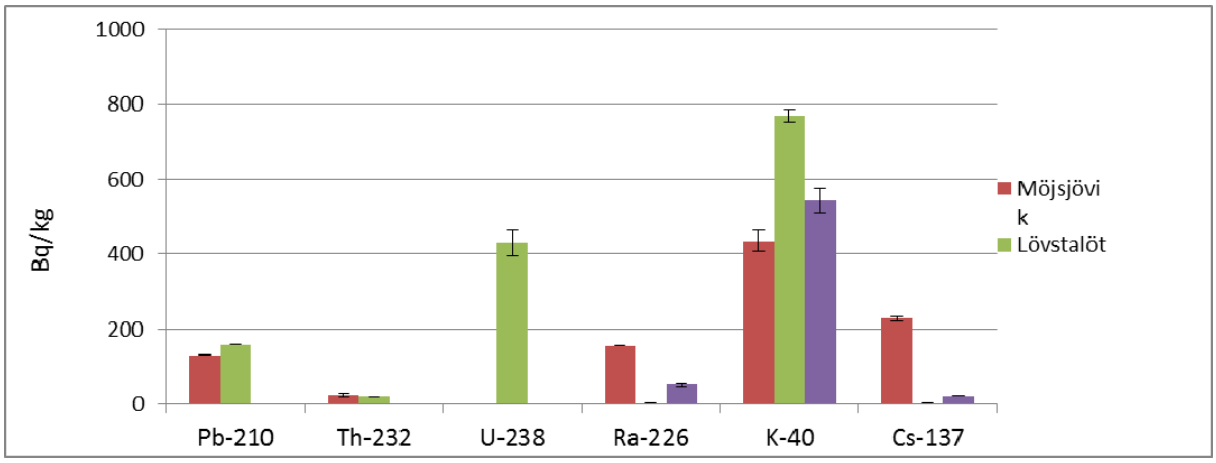

Figure 6. Comparison between the activity concentrations in grass samples for Uppsala sampling sites

3.2.3. Transfer factor

In the province of Uppsala the highest TF values were registered in Möjsjövik, as it is possible to see for example for ^{40}K. To notice a value of 1.6 in Lövstalöt, which is the second highest obtained between the three sites.

Table 8. Transfer factor for natural radionuclides in Uppsala province.

	Möjsjövik (2009)	Lövstalöt	Skogsvallen
Pb-210	-	0.5	-
Th-232	0.3	0.4	-
U-238	-	1.6	-
Ra-226	1.3	-	1.0
K-40	4.3	0.9	0.5

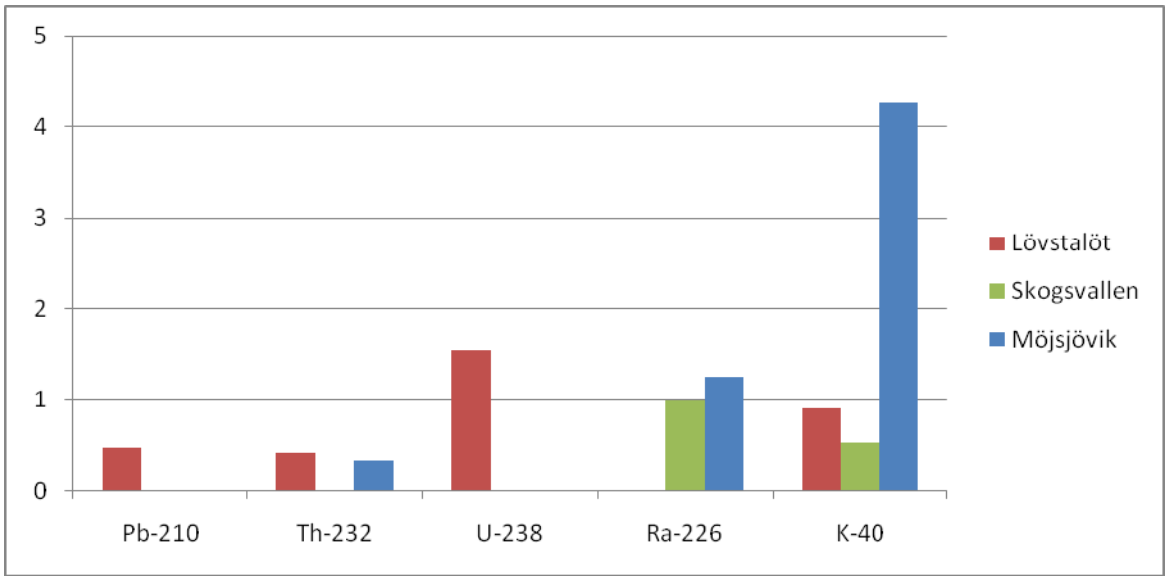

Figure 7. Comparison between transfer factors for Uppsala sampling sites

3.3. Transfer factor ^{137}Cs

For radiocesium transfer factors have been calculated with equation (2). Taking into account the difference in the levels of activity for this radionuclide in the two different locations in Sweden, the highest rate of uptake is found in Möjsjövik and Vikdrolet, although Möjsjövik activities are markedly higher compared to the low values registered in Vikdrolet. The smallest value is found in Lövstalöt (Table 9).

Table 9. Transfer factor for ^{137}Cs (m^2 kg dw $^{-1}$).

	Myrviken	Hallen	Backfors	Vikdrolet	Möjsjövik (2009)	Lövstalöt	Skogsvallen
Cs-137	-	-	0.00499	0.00934	0.00997	0.00038	0.00860

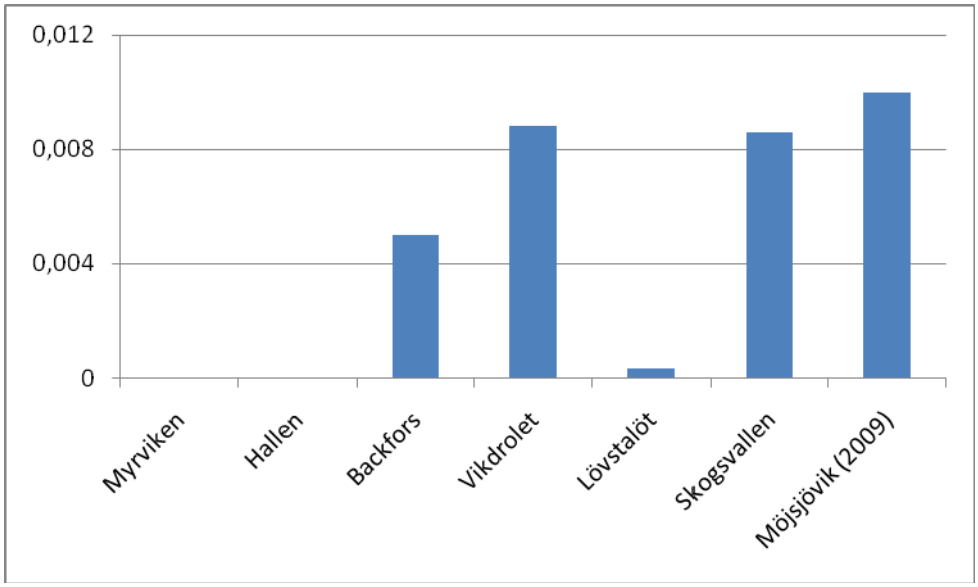

Figure 8. Comparison between transfer factors of ^{137}Cs for all the sampling sites

4. Discussion

4.1. Activity Concentration comparison

Table 10 summarizes the maximum, minimum, and mean values of activities concentration in soil bulk for each radionuclide in each of the eight locations object of our study. What appears immediately clear is that the lowest ranges of values for what concerns artificial radioactivity are found at the first four locations. In fact for ^{137}Cs activities ranges, for the first four sites, show a minimum value of 4 Bq kg^{-1} (the lowest value registered) in Hallen and Backfors and a maximum of 12 Bq kg^{-1} in Myrviken. This is consistent with what stated previously with respect to the general background of the area (see § 2.1). In fact, Hallen, Backfors and Myrviken are located in Jämtland Province, which is characterized by a high natural radioactivity background and a low artificial component. This is further on confirmed by generally high activity concentration data obtained in Jämtland sampling sites, exception done for ^{232}Th, whose low activity values resulted homogeneous in both the provinces.

In Uppsala there was a low natural background with low ^{226}Ra concentration and as already mentioned low ^{232}Th concentrations. The other natural radionuclides gave high activity values

in almost all the locations. The highest activity concentration in both the provinces, within natural radionuclides, is found for ^{40}K in Skogsvallen with an average of 1040 Bq kg^{-1}. Ramasamy et al. (2009) in similar study showed that high potassium activity concentration might be related to an increase amount of clay minerals. The highest value of activity for ^{137}Cs was found in the area of Uppsala, in particular in Möjsjövik during the year 2009.

The same comparison could not be performed with activities obtained in grass samples, for a lack of data for what concern ^{137}Cs activity in Jämtland province. Activity values for potassium, radium and uranium are as for soil samples, rather high. Available data suggest that trend in activity in grass is similar to that of soil. Comparing the results obtained in our study with literature from different studies (Table 12), ^{232}Th together with ^{40}K average activities appear in the range of other countries available data. A markedly difference with other studies is found for ^{238}U. Activity concentrations resulted from our study show around ten times the values obtained from previous studies in Sweden (UNSCEAR, 2000) and in other countries. This outrange value is most likely due to measurements issues. ^{234m}Pa is part of uranium decay chain and its half-life is markedly shorter compared to ^{238}U. The two radioelements are considered in secular equilibrium once ^{234m}Pa half-life is reached and therefore the activity of ^{238}U is similar to the activity of ^{234m}Pa. In our study ^{238}U activity was obtained through this method which can sometimes lead to experience some reliability uncertainties. Radium activity is another value that gave high average activity results compared to literature, although range values are in line with previous studies.

Table 10. Average activity concentrations and relative maximum and minimum values in soil bulks samples for all the radionuclides in all sampling locations (Bq Kg^{-1}). ^{137}Cs deposition unit is expressed in Bq m^{-2}.

	Pb-210		Th-232		U-238		Ra-226		K-40		Cs-137		Cs-137 (Deposition)	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Myrviken	198	180-228	31	29-33	426	328-475	259	223-303	697	685-706	9	9-12	326	294-350
Hallen	65	0-65	31	29-33	-	-	49	46-51	681	641-718	5	4-6	146	136-152
Backfors	194	152-217	37	34-40	478	415-515	249	212-283	845	820-879	5	4-6	199	157-241
Vikdrolet	425	168-899	34	28-40	631	428-985	411	270-658	872	760-980	9	7-10	316	258-362
Möjsjövik[a]	-	-	47	39-58	-	-	62	42-82	110	83-128	1086	897-1300	24219	20455-27455
Möjsjövik[b]	719	701-734	32	25-36	593	457-848	33	27-43	76	76-77	784	657-981	17372	15298-20600
Lövstalöt	356	312-384	50	48-53	307	269-345	49	41-55	849	842-855	70	61-83	2960	2609-3464
Skogsvallen	-	-	63	61-66	-	-	65	51-77	1040	1010-1060	75	24-128	4067	1271-7008

a. Activities concentration values obtained from field work in 2009 in the area of Möjsjövik

b. Activities concentration values obtained from field work in 2011 in the area of Möjsjövik

Table 11. Average activity concentrations and relative maximum and minimum values in grass samples for all the radionuclides in all sampling locations (Bq kg^{-1}).

	Pb-210		Th-232		U-238		Ra-226		K-40		Cs-137	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Myrviken	-	-	27	24-31	829	657-1000	178	42-251	1141	933-1260	-	-
Hallen	-	-	-	-	-	-	-	-	856	738-974	-	-
Backfors	-	-	24	21-28	664	408-1020	172	25-251	499	488-506	-	-
Vikdrolet	-	-	27	21-33	385	-	146	43-249	560	478-690	3	-
Möjsjövik[a]	130	-	24	17-32	-	-	156	-	435	355-562	228	168-348
Möjsjövik[b]	-	-	-	-	-	-	-	-	-	-	-	-
Lövstalöt	158	152-164	20	14-27	431	417-444	-	-	770	762-778	3	-
Skogsvallen	-	-	-	-	-	-	51	-	543	462-662	22	9-38

a. Activities concentration values obtained from field work in 2009 in the area of Möjsjövik

b. Activities concentration values obtained from field work in 2011 in the area of Möjsjövik

Table 12. Comparison of mean activity concentrations in soil samples (Bq kg^{-1}) of the main natural radionuclides with published results from other countries

	Th-232		U-238		Ra-226		K-40	
	mean	range	mean	range	mean	range	mean	range
Our study	***41***	***25-66***	***487***	***269-985***	***147***	***27-658***	***646***	***76-1060***
Sweden (UNSCEAR, 2000)	42	14-94	-	-	42	12-170	780	560-1150
Denmark (UNSCEAR, 2000)	19	8-30	-	-	17	9-29	460	240-610
Svalbard, Artic Region (Dowdall et al.)	24	9-43	32	17-49	36	15-62	362	115-614
China (Dai et al.)	50	38-66	-	-	32	24-40	720	498-858
Brazil (Flues et al.)	-	9-282		9-82	-	-	-	59-412
Hungary (Papp et al.)	24	15-41	41	0-1346	43	21-1256	330	176-567
Greece (rouni et al)	-	12-43	-	-	-	23-337	-	154-631
Greece (papastefanou et al.)	-	-	48	17-360	200	50-941	-	-
Germany (Bunzl et al.)	-	22-48	-	-	-	13-58	-	259-703
Germany (Rosner et al.)	27	11-48	31	15-67	35	11-59	362	244-703

4.2. Transfer Factors comparison

Transfer factors found in our study for all the natural radionuclides are generally higher in respect to those provided by IAEA (2010). The opposite condition instead results from cesium TF values. Literature shows higher TF than almost all the measurements performed for all the locations. Backfors, Lövstalöt and Skogsvallen show TF minor than 0.01. These could be related to the nature of soil texture. These sites share a common characteristic represented by a strong fine particle component.

A minimum is found in Lövstalöt, which is characterized by only 4% of sand and around 60% silt-clay. At the same time, Skogsvallen shows a TF value closer to 0.009, probably for its higher nutrients content compared to the other two sites (Tab.13). Similar value was registered in Vikdrolet, but in this case, no reasonable explanation could be provided, from the moment that no significant links to soil characteristics is noticed in the area. The only exception, for what concern ^{137}Cs TF, is in Möjsjövik, where values are similar to IAEA. This slight difference with other sites can probably be attributed to the nature of the soil composition, even though low activity concentrations of radiocaesium obtained for grass samples still evidence a low root uptake by plants. Möjsjövik soil is characterized by a high organic content. Fresquez et al. (1998) proved that organic soils act as binding agent and therefore tend to prevent plant uptake of radionuclides.

Within natural radionuclides the highest TF values are those found in Myrviken. This location is part of Jämtland province and it is the sampling site with the highest organic matter within the same province. In addition, even if soil texture is rather equally distributed between fine and gross particle, there is a slight preponderance of sand. In sandy soil coarse sized particles constitute the main component of the texture. Compared to clays these soils have larger pores are present and therefore mobility of radionuclides is enhanced, depending also on the nuclide's characteristics (Blanco Rodriguez et al., 2008).

The high values found for ^{40}K in Möjsjövik can be due to disturbances to grass samples during sampling procedure. Furthermore, to notice from available data that Backfors and Vikdrolet returned similar TF. This can be explained by similar soil composition. Available nutrients are rather similar in concentrations even if Backfors soil texture shows a slightly higher silty-clay fraction.

Table 13. Comparison between TF in all the location and UNSCEAR values (2010)

	Pb-210	Th-232	U-238	Ra-226	K-40	Cs-137 (m^2 kg_{dw}^{-1})
Myrviken	1.1	0.9	1.7	0.7	1.6	-
Hallen	-	-	-	-	1.3	-
Backfors	-	0.6	0.8	0.5	0.6	0.0050
Vikdrolet	-	0.8	0.8	0.5	0.6	0.0088
Möjsjövik (2009)	-	0.3	-	1.3	4.3	0.0100
Lövstalöt	0.5	0.4	1.6	-	0.9	0.0004
Skogsvallen	-	-	-	1.0	0.5	0.0086
UNSCEAR	0.07	0.04	0.02	0.04	-	0.015

4.3. Thorium-Uranium and Radium-Uranium ratios

Comparing the activity ratios of the two parent isotopes of the natural decay chain no substantial difference between the two areas could be found. As it is possible to see from Table 14, Mösjövik shows similar average values as Jämtland. The only exception is Lövstalöt site, which is the only sampling point in which $^{232}Th/^{238}U$ ratio appears slightly higher than the rest of the sites. Nonetheless uranium generally shows a predominant activity if compared to thorium. The comparison of activities ratios between ^{226}Ra and ^{238}U indicates different situation in the two provinces, in facts a clearer difference between the two provinces is instead noticed for $^{226}Ra/^{238}U$ activities. Table 14 shows that in all the four Jämtland sampling sites, even if with slightly different activities, all ratios lie around 0.6, value that markedly differs from the ratios found in Uppsala province. Nevertheless, within Uppsala area, no significant homogeneity in the results was found, with values of 0.06 in Möjsjövik and 0.16 in Lövstalöt. Previous studies (P. Blanco Rodriguez, 2008) showed that breaking of radioactive equilibrium between members of the same radioactive chain gives indication about an increased mobility of the single radioisotope. In both provinces, data suggest that radioactive equilibrium between radium and uranium is not reached. While in Uppsala area we see a markedly stronger uranium component compared to radium, radium activities in Jämtland appear to be around 60% of uranium and therefore closer to the equilibrium.

Mobility of radium in soil is connected among other parameters also to soil nutrients content. The extremely weak presence of ^{226}Ra in Möjsjövik can be explained by the high primary elements concentration in the site's soil. Möjsjövik as expected shows the lowest radium concentration and at the same time the highest Mg percentage within all the sites. As proved in previous studies (P. Blanco Rodriguez, 2008), the explanation can be magnesium disposition to displace Ra in soil, thus filling up available sites on soil particles and enhancing radium availability.

Higher ^{226}Ra activity in Lövstalöt compared to Möjsjövik could be due to high clay content and therefore enhanced interaction between the nuclide and the soil particles due to a weak nutrient content (the lowest registered in both provinces). According to previous studies (El-Arabi et al., 2006) the reason for high radionuclides activity in clays is due to the fact that clay minerals are mainly composed by aluminium silicates and they are characterized by small sized grain and negative charged surface. In this way clay particles easily absorb cations on their surface.

While low radium activity in Hallen can be explained by the predominance in soil texture of a sandy fraction, Myrviken's high value of ^{226}Ra seems to be rather contradictory, since both high organic matter content and nutrients would suggest a high mobility for radionuclides and therefore less concentration in the topsoil.

Table 14. Activity ratios between radium-uranium and thorium-uranium in all the locations

Site	Ra-226/U-238	Th-232/U-238
Jämtland		
Myrviken	0.61	0.07
Hallen	-	-
Backfors	0.52	0.08
Vikdrolet	0.65	0.05
Uppsala		
Möjsjövik (2011)	0.06	0.05
Lövstalöt	0.16	0.16
Skogsvallen	-	-

4.4. Comparison of activities of ^{238}U, ^{232}Th and ^{40}K

Figure 15 displays the plots obtained from the comparison between the activities registered for the parent nuclides of the two main natural decay series. In the comparison between potassium and the other two parent nuclides concentrations registered for Möjsjövik influence significantly regression analysis since they differ markedly from the other sites trends.

Uranium and potassium showed no significant correlation, independently from the geographical locations. Considering both the provinces together the correlation coefficient appears rather low (-0.39) with a R^2=0.15 and there is no visible link between the values in the plot.

Correlation coefficient between thorium and potassium appears as well rather low (0.33) with a R^2= 0.11. Again, these values suggest weak interactions between activities of the two radionuclides. Interesting to notice that in this correlation, not taking into account the extreme values registered in Möjsjövik, we obtain a significantly high correlation coefficient of 0.86 and R^2=0.75 (Figure 15 A-D).

Uranium and thorium have a good negative correlation (-0.72) with R^2=0.51, in fact from plot C in Fig.12 it is possible to notice how to an increase in uranium concentration corresponds a slight decrease in thorium activity. Although other studies showed opposite trends to those found in our work (Hussain et al., 2010), Ababneh et al. (2009) found similar negative correlation and trend in another study. Interactions between primordial radionuclides could be due to the fact that these elements are present in soil since the formation of earth. Once earth's crust solidified these radionuclides remained trapped in rocks.

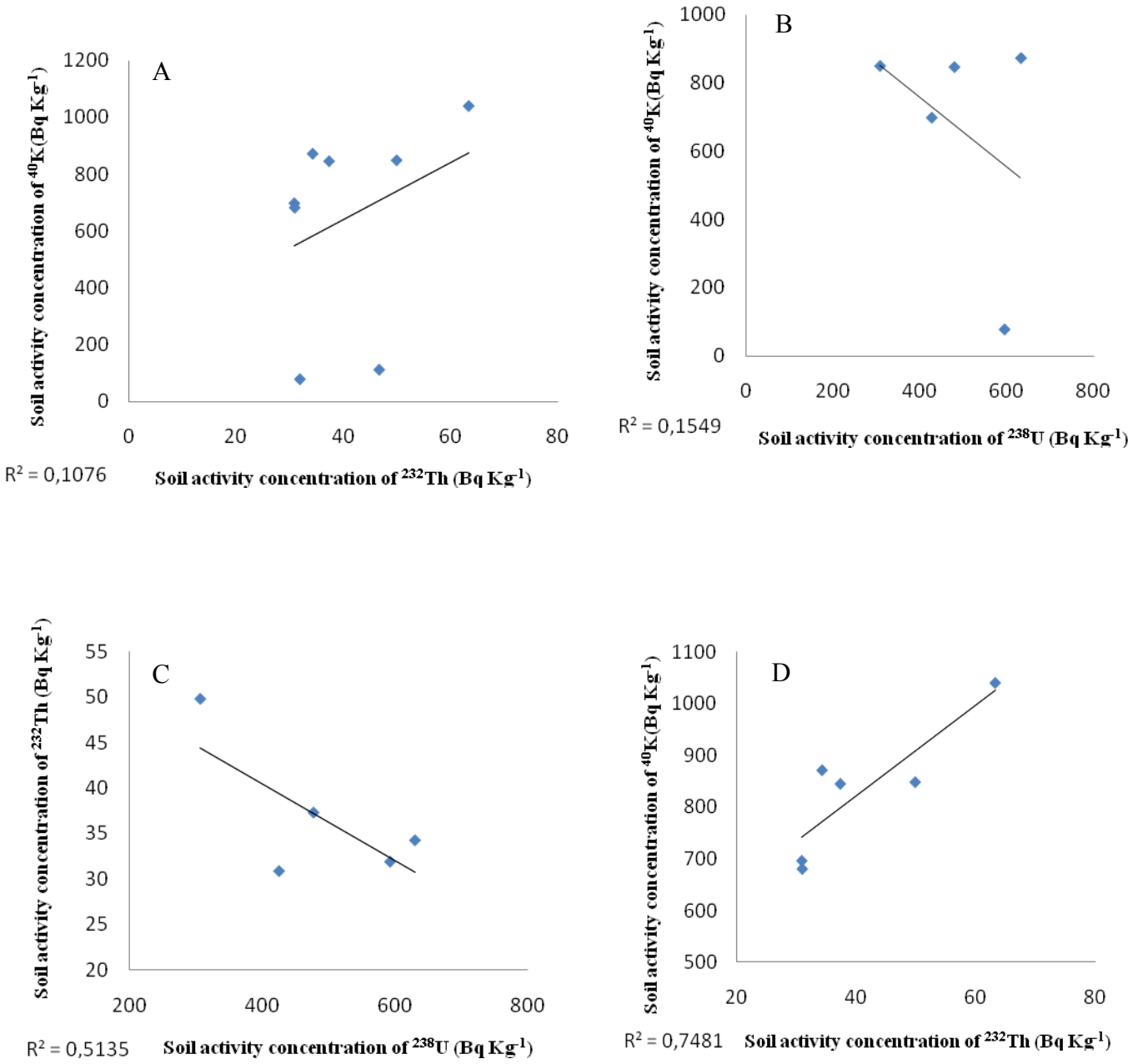

Figure 9. Plot of the correlation between the activities concentrations of the main parent nuclides and ^{40}K for soil samples. Plots A,B and C display comparison between activity values obtained in all the locations. Plot D displays activity concentrations of potassium and thorium in all the locations, not taking into account those found in Möjsjövik.

4.5. Comparison of activity concentrations and soil parameters

The comparison between activity concentrations and the chemical parameters that characterize the study locations was performed for all the radionuclides object of our study. The correlation coefficients obtained from these comparisons can be found in Appendix 6. From the graph in Figure 16 it is possible to notice consistent differences in the way the activities of the radionuclides are influenced by the different parameters.

What appears clear is that both radiocaesium and ^{210}Pb are positively correlated with most of the parameters exception done for pH, which seems to be in negative correlation with both the nuclides. At the same time, a marked negative correlation exists between ^{40}K and most of the parameters.

For what concern the rest of the radioisotopes, ^{226}Ra, ^{238}U and ^{232}Th, our results showed low correlations between activities and chemical parameters. The highest correlation was found between pH and ^{232}Th activity, although with a rather low value (-0.63).

Within all the soil parameters must be mentioned that phosphorus available content (P-AL) seems to have the lowest influence on the behavior in soil of all the radionuclides.

Interesting to notice that, as already found, the correlation coefficients for most of the radionuclides vary depending on the presence or not of Möjsjövik data. Figure 17 shows some examples of how marked the variation between the coefficient can be in different cases.

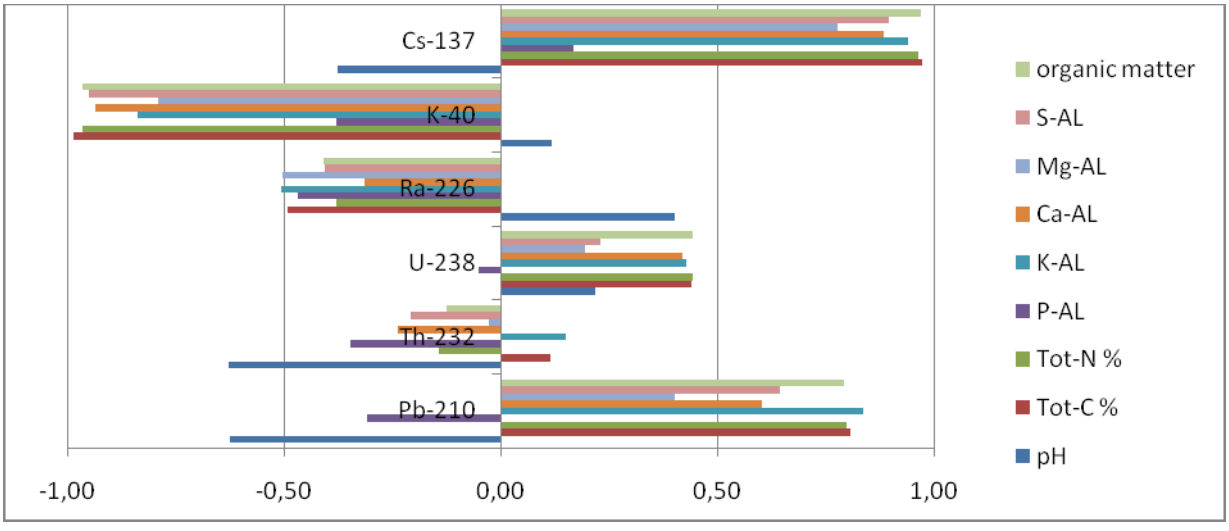

Figure 16. Degree of correlation between the activities obtained in our study for all the radionuclides and the different soil parameters

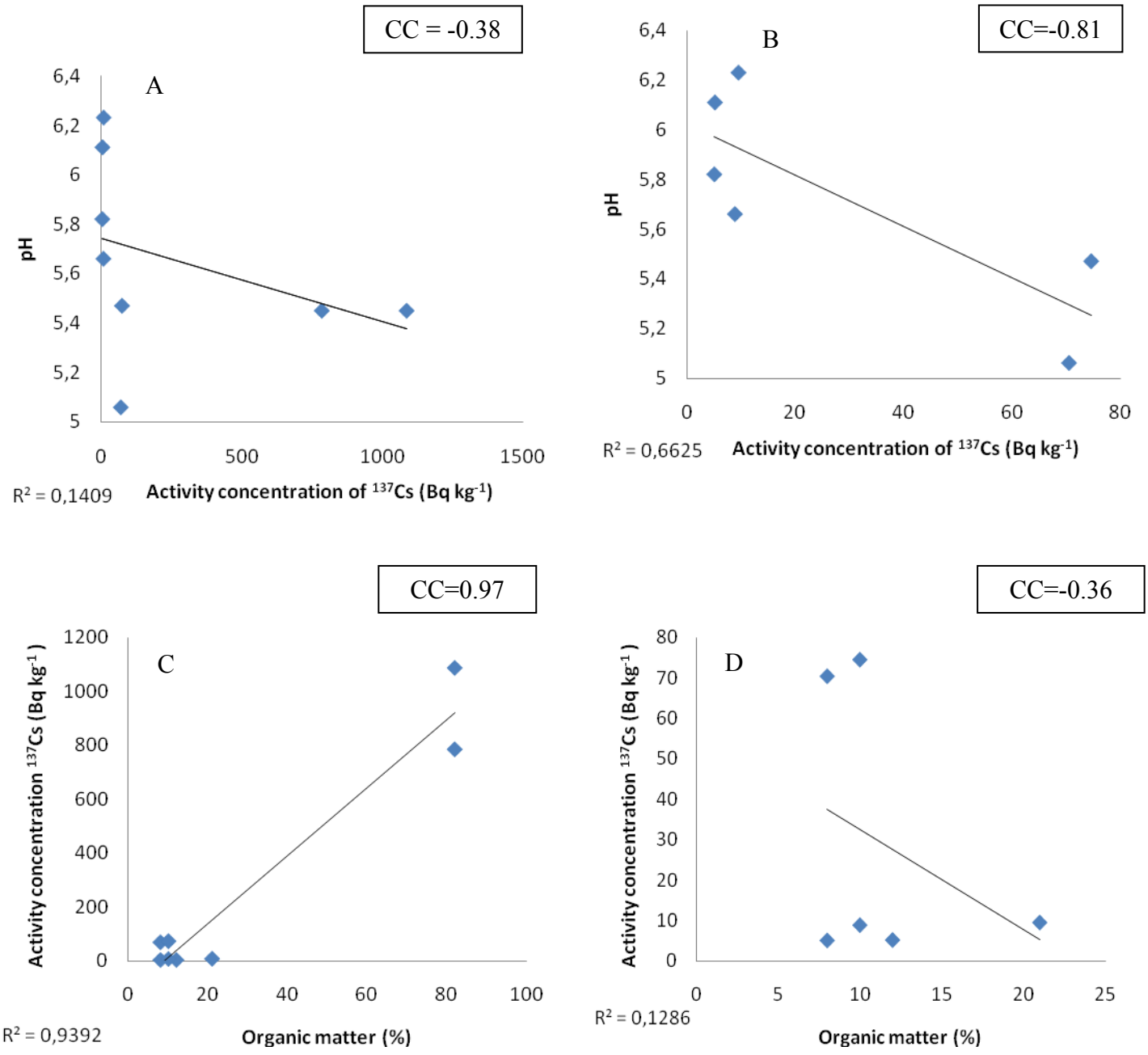

Figure 10. Example of comparison between correlation coefficients (CC) obtained for radiocaesium activity against respectively pH and organic matter in the soil. Plot A and C are obtained considering all the sampling locations. Plots B and D are obtained not taking into account activities registered in Möjsjövik.

5. Conclusions

- The results found in our study confirm the expected difference between the two provinces for what concern radioactivity background. Jämtland province shows a high natural background, while Uppsala province shows lower values. At the same time the latter shows high deposition rates of radioacaesium while Jämtland confirms a low trend.
- The differences found in the activities of the parent nuclides of the two main natural decay chains, ^{238}U and ^{232}Th, suggest that the natural radioactive background is mostly due to ^{238}U and the members of the same chain.
- Möjsjövik if compared to the other study locations seems to have a totally different behavior. This might be attributable to the peculiar nature of the soil texture and composition.

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Appendix

Appendix 1

Activity concentration in soil bulk and grass samples for each subsite in the Jämtland area (Bq kg^{-1}).

	Myrviken			Hallen			Backfors			Vikdrolet		
	SOIL											
	A	B	C	A	B	C	A	B	C	A	B	C
Pb-210	187±26	228±31	180±28	-	-	65±20	152±21	217	214±31	168±28	208±31	899±107
Th-232	33±2	31±2	29±3	30±3	33±3	29±3	38±2	34±3	40±2	36±2	28±2	40±3
U-238	328±82	475±101	474±122	-	-	-	415±57	503	515	428±115	479±84	985±122
Ra-226	252±8	303±10	223±9	51±6	46±5	-	253±7	212±8	283±9	270±9	306±10	658±19
K-40	706±20	700±20	685±21	685±21	718±21	641±21	837±20	820±22	879±24	875±23	760±21	980±26
Cs-137	9±30	8±0.3	12±1	6±1	5	6±1	6±0.2	4	5	7±0.3	10	10
	GRASS											
	A	B	C	A	B	C	A	B	C	A	B	C
Pb-210	175	243	221±36	-	-	-	-	-	-	-	-	43
Th-232	26	24	31	-	-	-	-	28	21	33	21	-
U-238	-	1000	657	-	-	-	1020	408±113	564±142	-	385±118	118
Ra-226	251	240	42±11	-	-	-	25	251	241	249	-	1143
K-40	1260±40	933±34	1230±40	855±38	974±36	738±32	488±32	506±30	503±28	478±33	513±31	31
Cs-137	-	-	-	-	-	-	-	-	-	-	-	3

Appendix 2

Activity concentration in soil bulk and grass samples for each subsite in the Uppsala area (Bq kg^{-1}).

	Möjsjövik (2009)			Möjsjövik (2011)			Lövstalöt			Skogsvallen		
	SOIL											
	A	B	C	A	B	C	A	B	C	A	B	C
Pb-210	-	-	-	701±87	734±92	721±91	312±40	371±47	384±48	-	-	-
Th-232	44±2	58±2	39±3	36±3	34±3	25±3	48±2	48±2	53±2	66±2	63±2	61±2
U-238	-	-	-	474±98	848±169	457±79	-	269±68	345±88	-	-	-
Ra-226	82±13	-	42±7	27±6	31±6	43±6	41±4	55±5	50±4	68±5	77±5	51±4
K-40	119±10	128±9	83±15	77±14	76±16	-	842±22	855±22	849±22	1050±37	1060±38	1010±36
Cs-137	1300±30	1060±25	897±17	715±13	981±18	657±12	61±1	83±2	67±1	24±1	72±2	128±3
	GRASS											
	A	B	C	A	B	C	A	B	C	A	B	C
Pb-210	-	-	130	-	-	-	164	152	-	-	-	-
Th-232	-	32±15	17	-	-	-	27	14	-	-	-	-
U-238	-	-	-	-	-	-	444±104	417	-	-	-	-
Ra-226	-	-	156	-	-	-	-	-	-	-	-	51±13
K-40	355±24	389±36	562±23	-	-	-	778±26	762±25	-	462±35	662±31	505±32
Cs-137	168±4	168±5	348±6	-	-	-	3	-	-	19±1	38±2	10±1

Appendix 3

Soil characteristics for the different locations object of our study

	%								mg/100g				
	clay	silt	loam	sand	organic matter	Tot-C	Tot-N	pH	P-AL	K-AL	Ca-AL	Mg-AL	S-AL
Jämtland													
Myrviken	25	27	20	29	21	9.5	0.9	6.23	4.6	10.9	591.8	25.5	5.0
Hallen	18	20	17	44	12	4.3	0.4	6.11	6.9	8.0	312.2	17.5	2.3
Backfors	28	38	26	8	8	3.1	0.3	5.82	1.1	4.2	196.9	9.3	0.7
Vikdrolet	23	23	25	29	10	4.1	0.4	5.66	1.5	8.8	239.1	7.6	1.2
Uppsala													
Möjsvjövik	-	-	-	-	82	44.4	3.0	5.45	3.9	36.2	959.6	32.4	8.6
Lövstalöt	31	30	35	4	8	3.3	0.3	5.06	2.1	8.5	119.2	9.7	2.1
Skogsvallen	47	39	9	6	10	3.7	0.3	5.47	2.7	19.1	260.9	21.0	1.3

Appendix 4

Characteristics of sampling locations in both the provinces

	Uppsala			Jämtland			
	Skogsvallen	Möjsjövik	Lövstalöt	Myrviken	Backfors	Vikdrolet	Hallen
GPS coordinates	60° 10' 1" N 17° 11' 19" E	59° 56' 60" N 17° 13' 60" E	59° 57' 10" N 17° 35' 0" E	62º 59’ 11.0’’ N 14º 24’ 26’’ E	63º 01’ 10.3’’ N 14º 21’ 50’’ E	63º 00’ 55.5’’ N 14º 23’ 02.3’’ E	63º 10' 5" N 14º 7' 13" E
Grazing area (ha)	-	3	50	>40	-	-	>40
Grazing period	-	15/05 – 15/09	15/05 – 15/09	01/06 - 01/09	-	01/06 - 01/09	01/06 - 01/09
Number of animals grazing	-	10	140	100	-	<20	120
Type of breed	-	Horses	Swedish lowland livestock, (SRB), Cow	Swedish lowland livestock, (SLB), Cow	-	Sheep	Swedish lowland livestock, (SLB), Cow
Type of farm	extensive	Extensive	intensive	intensive	extensive	extensive	intensive

Appendix 5

Average MDA for Jämtland and Uppsala provinces (Bq kg^{-1})

	Jämtland	Uppsala
Pb-210	81	71
Th-232	10	8
U-238	403	340
Ra-226	21	18
K-40	59	47
Cs-137	2	3

Appendix 6

Correlation coefficients between activity concentrations and soil chemical parameter

Unit		Pb-210	Th-232	U-238	Ra-226	K-40	Cs-137
	pH	-0.62	-0.63	0.22	0.40	0.12	-0.38
%	Tot-C	0.81	0.12	0.44	-0.49	-0.99	0.97
	Tot-N	0.80	-0.14	0.44	-0.38	-0.96	0.96
	Organic matter	0.79	-0.12	0.44	-0.41	-0.97	0.97
Mg/100g	P-AL	-0.31	-0.35	-0.05	-0.47	-0.38	0.17
	K-AL	0.84	0.15	0.43	-0.51	-0.84	0.94
	Ca-AL	0.60	-0.24	0.42	-0.31	-0.94	0.88
	Mg-AL	0.40	-0.03	0.19	-0.50	-0.79	0.78
	S-AL	0.64	-0.21	0.23	-0.41	-0.95	0.90