Master Thesis

Method development for quantifying strontium-90 in bovine bones

by

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Abstract

In nuclear fission, fission products are produced, many of which are radioactive. The release of these radionuclides is viewed with concern by the public. In the course of the Fukushima Daiichi nuclear accident (2011), large activities of radionuclides were released into the environment. While other radionuclides (e.g., ^{134}Cs, ^{137}Cs, ^{131}I, etc.) are not only volatile but also allow straightforward detection and quantification, ^{90}Sr has been emitted in lower quantities, and is notorious for its laborious sample preparation. Data on ^{90}Sr from Fukushima are, therefore, scarce by comparison. With its chemical similarity to calcium and its relatively long half-life of 29 years, it poses a potential threat to human and environmental health.

Due to its chemical properties, radiostrontium is known to accumulate in bone tissue. Previous studies have shown that, if any, increased environmental levels of ^{90}Sr are to be found in close vicinity to the Fukushima Daiichi reactors. An interesting research opportunity arose when bones from a deceased cow that had lived approximately two kilometers from the reactor and was roaming freely for around one year after the accident were promised to the Applied Radiochemistry group at TU Wien.

In preparation for the arrival of this precious sample material, an analytical protocol for the extraction of ^{90}Sr had yet to be developed and is hence the topic of the present thesis. Since bone tissue is not generally regarded as a simple matrix, a complex sequence of sample preparation steps had to be developed. This included pyrolysis, acid digestion, solid phase strontium extraction, and analysis by liquid scintillation counting (LSC). The yield for this process was determined by spiking the sample with an internal, gamma-ray-emitting ^{85}Sr standard.

In this thesis, various bone samples were successfully prepared for strontium extraction. The extraction yield was demonstrated to be in the range of approximately 84 %. The present work provides a tool for the detection of radiostrontium in bone matrices, which may become valuable in the course of environmental monitoring following a nuclear release.

Zusammenfassung

Bei der Kernspaltung entstehen Spaltprodukte, von denen viele radioaktiv sind. Eine Freisetzung von dieser Radionuklide wird in der Öffentlichkeit mit Sorge betrachtet. Im Zuge des Atomunfalls im Kernkraftwerk Fukushima Daiichi (2011) wurden große Aktivitäten an Radionukliden in die Umwelt freigesetzt. Während andere Radionuklide (z. B. ^{134}Cs, ^{137}Cs, ^{131}I usw.) nicht nur flüchtig sind, sondern auch rasch und unkompliziert detektierbar sind, wurde ^{90}Sr in geringeren Mengen emittiert und ist bekanntermaßen aufwändig in der Probenvorbereitung. Die Datenlage zu ^{90}Sr aus Fukushima ist daher vergleichsweise spärlich. Aufgrund seiner chemischen Ähnlichkeit mit Calcium und seiner relativ langen Halbwertszeit von 29 Jahren ist ^{90}Sr besonders gesundheits- und umweltrelevant.

Aufgrund seiner chemischen Eigenschaften reichert sich Radiostrontium im Knochengewebe an. Frühere Studien haben gezeigt, dass erhöhte Umweltkonzentrationen von ^{90}Sr, wenn überhaupt, nur in unmittelbarer Nähe zu den Reaktoren des Kernkraftwerks Fukushima Daiichi zu finden sind. Eine interessante Forschungsmöglichkeit ergab sich, als der Gruppe für Angewandte Radiochemie der TU Wien die Knochen einer verstorbenen Kuh zugesagt wurden, die etwa zwei Kilometer vom Reaktorgelände entfernt gelebt hatte und sich nach dem Unfall etwa ein Jahr lang frei in dieser Gegend bewegen konnte.

In Vorbereitung auf das Eintreffen dieses wertvollen Probenmaterials musste noch ein Analyseprotokoll für die Extraktion von ^{90}Sr entwickelt werden, was daher Gegenstand der vorliegenden Arbeit ist. Da Knochengewebe im Allgemeinen nicht als einfache Matrix angesehen wird, musste eine komplexe Abfolge von Probenvorbereitungsschritten entwickelt werden. Dazu gehörten Pyrolyse, Säureaufschluss, Festphasenextraktion von Strontium und Analyse mittels Flüssigszintillationszählung (LSC). Die Ausbeute für diesen Prozess wurde durch Aufstockung der Probe mit einem internen, gammaemittierenden ^{85}Sr-Standard bestimmt.

In dieser Arbeit wurden verschiedene Knochenproben erfolgreich für die Strontiumextraktion vorbereitet. Die Ausbeute der Extraktion lag bei etwa 84 %. Die vorliegende Arbeit liefert ein Werkzeug für den Nachweis von Radiostrontium in Knochenmatrizen, das bei der Umweltüberwachung nach einer nuklearen Freisetzung von Bedeutung sein könnte.

Contents

1. Introduction

1.1 Radioactivity, radioactive decay and equilibrium systems

Radioactivity is a spontaneous process, in which an unstable nucleus transforms (decays) into a more stable nucleus. The energy difference between both nuclei is released in the form of highly energetic, ionizing radiation. The unit for activity (radioactivity) is a Becquerel (Bq, SI: Bq=s^{-1}) and it represents a radioactive decay per second. The activity can be defined by the equation (1.1.).

$$A = \frac{Decay}{Time} = \frac{-dN(t)}{dt} = \lambda \cdot N(t) \tag{1.1.}$$

The number of particles and the activity at a given time can be expressed through the first equation:

$$N(t) = N_0 \cdot e^{-\lambda t} \tag{1.2.}$$

Half-life is a unit of time in which the quantity of the substance reduces to its half, and it is correlated to the decay rate λ in the equation (1.3).

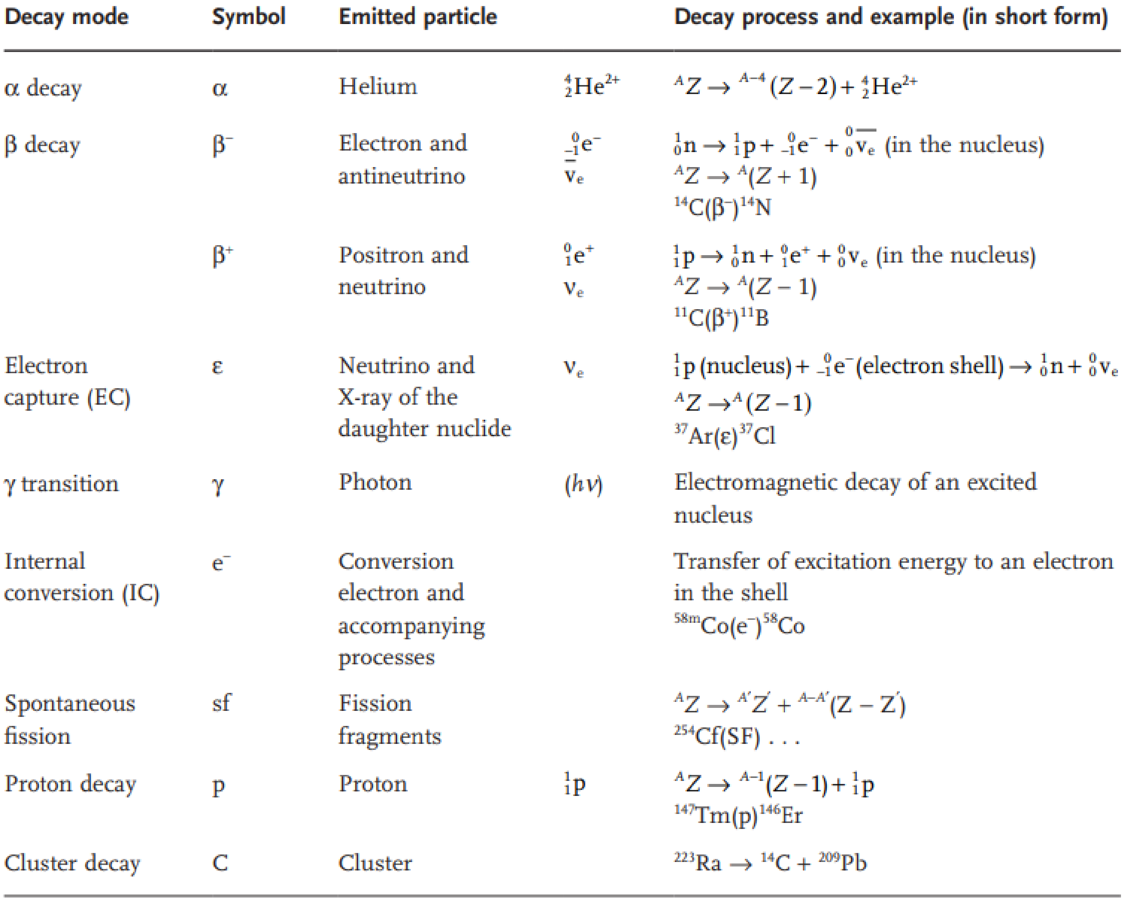
$$\lambda = \frac{\ln(2)}{t_{\frac{1}{2}}} \tag{1.3.}$$

The three most prominent decay types are alpha, beta, and gamma decay.

Alpha (α) decay releases an alpha particle, which is a ^{4}He nucleus/ $^4He^{2+}$ ion, with a very low penetrative ability. Its range depends on the energy of released particles, but it amounts to not more than a few centimeters in the air. The path of α particles is practically not influenced by their collision with electrons, and in the case of a collision with a nucleus it is either strongly deflected, or captured by the nucleus which induces a nuclear reaction – creating a new nucleus.

Beta (β) decay can occur as one of three different types of decay: β^-, β^+, or electron capture. During these decays, a neutron or a proton change into a proton or a neutron respectively, simultaneously emitting an electron or a positron together with an antineutrino or a neutrino. These released electrons are termed β^- particles. During electron capture, the nucleus captures an orbital electron, changing a proton into a neutron and creating a hole in the electron shell, which subsequently leads to the emission of X-rays or Auger electrons. The transformation of a proton into a neutron also leads to the emission of a neutrino. Beta particles typically carry less energy than alpha particles but have a higher penetrative ability. An α particle with 3 MeV of energy has a range of about 1.7 cm in air, whereas a β particle of the same energy travels a distance of about 10 m in air. Beta particles are, in contrast to α particles, markedly deflected by other electrons through collisions.

Gamma (γ) emission is a high energy photon released by a nucleus decaying from an excited state into a state of lower energy. Their absorption is in principle different from that of charged heavy particles (for example He nuclei) or β particles – they tend to give off their energy mostly in one process. Due to them not carrying a charge, their interaction with matter is small.



Decay mode	Symbol	Emitted particle		Decay process and example (in short form)
α decay	α	Helium	${}^{4}_{2}He^{2+}$	${}^{A}Z \rightarrow {}^{A-4}(Z-2) + {}^{4}_{2}He^{2+}$
β decay	β⁻	Electron and antineutrino	${}^{0}_{-1}e^{-}$ $\overline{\nu}_e$	${}^{1}_{0}n \rightarrow {}^{1}_{1}p + {}^{0}_{-1}e^{-} + {}^{0}_{0}\overline{\nu}_e$ (in the nucleus) ${}^{A}Z \rightarrow {}^{A}(Z+1)$ ${}^{14}C(\beta^{-}){}^{14}N$
	β⁺	Positron and neutrino	${}^{0}_{1}e^{+}$ ν_e	${}^{1}_{1}p \rightarrow {}^{1}_{0}n + {}^{0}_{1}e^{+} + {}^{0}_{0}\nu_e$ (in the nucleus) ${}^{A}Z \rightarrow {}^{A}(Z-1)$ ${}^{11}C(\beta^{+}){}^{11}B$
Electron capture (EC)	ε	Neutrino and X-ray of the daughter nuclide	ν_e	${}^{1}_{1}p$ (nucleus) + ${}^{0}_{-1}e^{-}$ (electron shell) $\rightarrow {}^{1}_{0}n + {}^{0}_{0}\nu_e$ ${}^{A}Z \rightarrow {}^{A}(Z-1)$ ${}^{37}Ar(\varepsilon){}^{37}Cl$
γ transition	γ	Photon	$(h\nu)$	Electromagnetic decay of an excited nucleus
Internal conversion (IC)	e⁻	Conversion electron and accompanying processes		Transfer of excitation energy to an electron in the shell ${}^{58m}Co(e^{-}){}^{58}Co$
Spontaneous fission	sf	Fission fragments		${}^{A}Z \rightarrow {}^{A'}Z' + {}^{A-A'}(Z-Z')$ ${}^{254}Cf(SF)$ . . .
Proton decay	p	Proton	${}^{1}_{1}p$	${}^{A}Z \rightarrow {}^{A-1}(Z-1) + {}^{1}_{1}p$ ${}^{147}Tm(p){}^{146}Er$
Cluster decay	C	Cluster		${}^{223}Ra \rightarrow {}^{14}C + {}^{209}Pb$

Figure 1: Radioactive decay types. Taken from (1). Reprinted with permission from Wiley-VCH © 2013

As radionuclides decay, they produce other, new radionuclides, which, in many cases will also decay (are unstable), creating a parent-daughter decay relationship in which the cumulative activity represents the realistic activity of the whole system. Depending on the ratio of the two half-lives, one of three different radioactive equilibrium is present: secular, transient or no equilibrium (1).

1.1.1. Secular equilibrium with $t_{1/2}(1) >> t_{1/2}(2)$

In the equation for secular equilibrium, the mothers decay rate is negligibly small, meaning it does not have to be considered:

$$N_2 = \frac{\lambda_1}{\lambda_2} \cdot N_1 (1 - e^{-\lambda_2 t_a}) \qquad (1.4.)$$

The daughter's activity reaches 99 % of the activity of the parent nuclide in a span of 8 half-lives, with the ingrowth speed being faster in the beginning and decreases with time as the daughter starts decaying at a bigger rate.

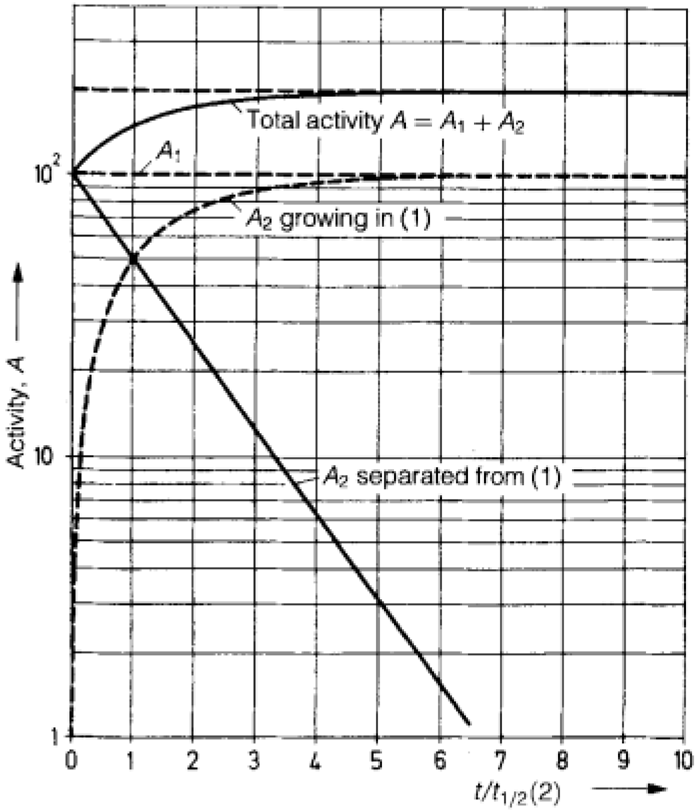


Figure 2: Secular equilibrium: activities of mother and daughter nuclides as a function of time with regards to half-life. Taken from (1). Reprinted with permission from Wiley-VCH © 2013

An example of a system eventually reaching secular equilibrium is ^{90}Sr and its daughter ^{90}Y.

1.1.2. Transient equilibrium with $t_{1/2}(1) > t_{1/2}(2)$

In this type of equilibrium, with daughter's half-life not being negligible in regard to parents' half-life, daughter's activity is always higher, and it is adjusted after various half-lives.

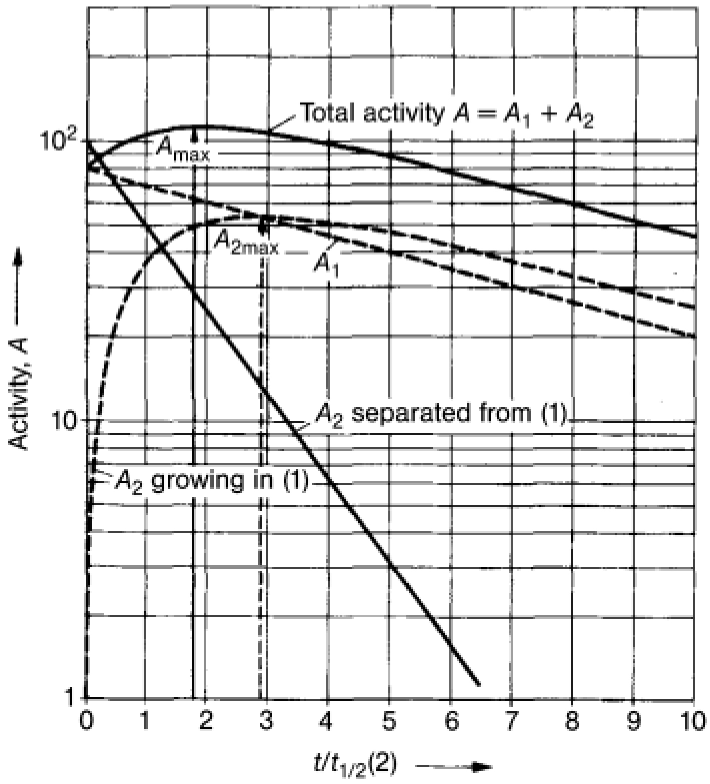


Figure 3: Transient equilibrium: activities of mother and daughter nuclides as a function of time with regards to half-life. Taken from (1). Reprinted with permission from Wiley-VCH © 2013

Example for transient equilibrium is ^{99}Mo and ^{99m}Tc with their respected half-lives being 66 h and 6 h.

1.1.3. No equilibrium $t_{1/2}(1)<t_{1/2}(2)$

With the daughter's half-life being longer than its parents' half-life, the activity ratio never reaches a state of equilibrium and keeps changing until the parent is fully decayed.

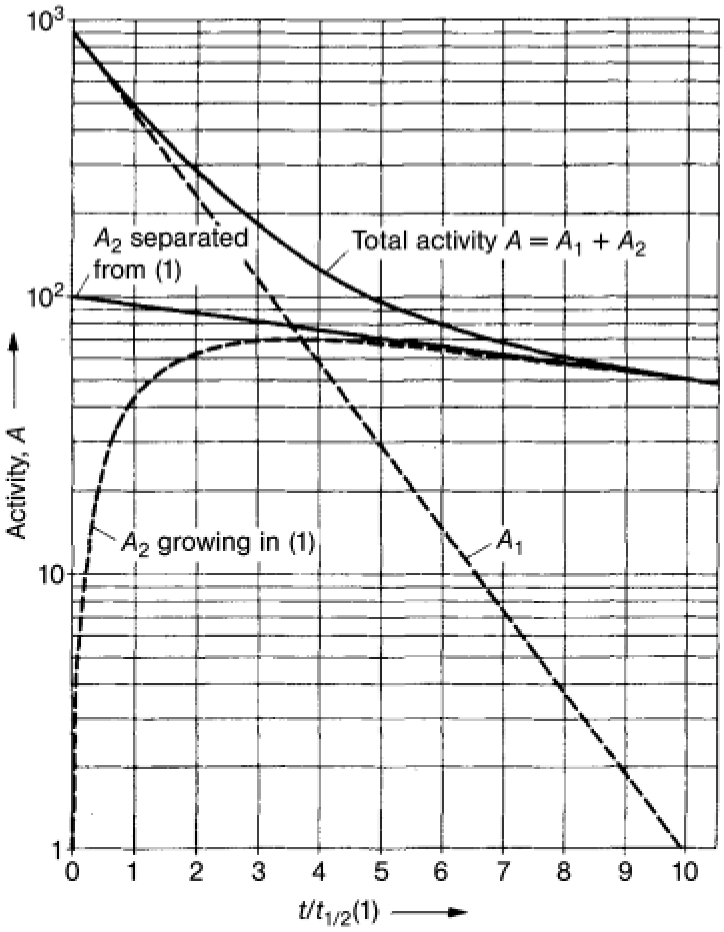


Figure 4: No radioactive equilibrium. Taken from (1). Reprinted with permission from Wiley-VCH © 2013

1.2 Nuclear reactions, nuclear fission, and nuclear power

A nuclear reaction occurs when a nucleus interacts with a proton, a neutron, another nucleus, an elementary particle, or a photon, resulting in the formation of one or more new nuclei and potentially other particles. The probability of a nucleus absorbing a particle is expressed through the isotope-specific cross section σ, with its unit being barn [$b=10^{-28}$ m^2], together with the type of projectile particle. These reactions are, like chemical reactions, always accompanied by a release or absorption of energy.

In 1938, in a series of studies, Hahn and Strassmann discovered what they thought was a radium isotope, formed by bombarding uranium with slow neutrons – allegedly – inducing two α-decays from the nucleus. Meitner and Frisch realized that what they discovered was not radium, but chemically similar radioactive barium – formed by splitting of the uranium nucleus, a process we now know as nuclear fission (2) (3).

During fission, a nucleus splits into two smaller fragments/nuclei, roughly each having a half of the initial nucleus's mass. The energy range accessible through nuclear reactions is much higher than that of mentioned radioactive decay – hence there are other usages besides analytical procedures – such as energy production in nuclear power plants.

Neutron-induced fission is used in nuclear reactors, in which commonly ^{235}U absorbs a neutron – becoming highly-excited ^{236m}U – prompting fission, splitting into two smaller nuclei with kinetic energy, releasing 2-5 neutrons, gamma radiation, and antineutrinos in the process.

The decisive features for production of energy by nuclear fissions are the thermal energy released (which is very high for ^{235}U-^{236}U), and that at least one of several released neutrons induces another fission reaction – making a chain reaction possible.

Due to thermal (slow, <0.1 eV) neutrons having much higher probability of inducing a nuclear reaction, the neutrons released during the fission are slowed down, creating a sustained and controlled chain reaction. The produced atoms mass distribution of the fission yield can be seen in Figure 5 (4) (1).

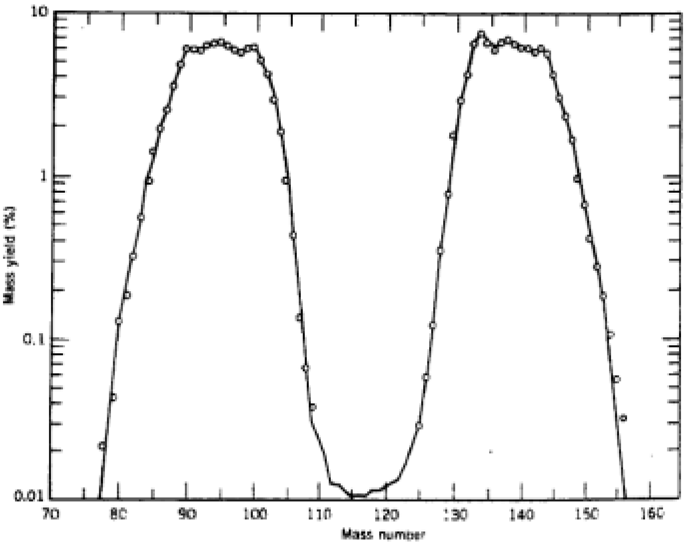


Figure 5: Uranium-235 fission products mass yield. Taken from (5)

1.3 The Fukushima nuclear accident

On 11th of March 2011, the Tohoku Earthquake with a maximum force of 9.0 Mw struck near Japan, 130 km east of Sendai and 163 km northeast the Fukushima, where multiple nuclear power plants (NPP) were positioned (Figure 6). The Fukushima Daiichi NPP (FDNPP) consisted of six units of boiling water reactors, with a combined power capability of around 4500 MWe (6). Due to the earthquake-induced shutdown, the east coast of Honshu experienced an almost complete electrical blackout, with FDNPP being amongst the affected infrastructure – activating the power plant's emergency cooling system powered by generators running on diesel fuel. The earthquake also triggered a tsunami, which destroyed the large parts of the coastal area in the vicinity. FDNPP was designed to be protected from a tsunami of up 5.7 m, with an additional safety margin of 4.3 m due to the plants grade level, reaching the tsunami preparedness of 10 m (Figure 7). With waves height reaching as high as 14 m at the plant site, ocean water flooded the diesel tanks fueling the generators, leaving the power plant's emergency cooling system inoperable (7). This resulted in a partial core meltdown in reactor Units 1, 2 and 3 in the following days, allowing hydrogen to build up in these three reactor buildings which subsequently caused explosions in the reactor buildings. Based on the best available information, these explosions affected only the reactor buildings and not the pressure vessels or the reactors themselves (7) (6).

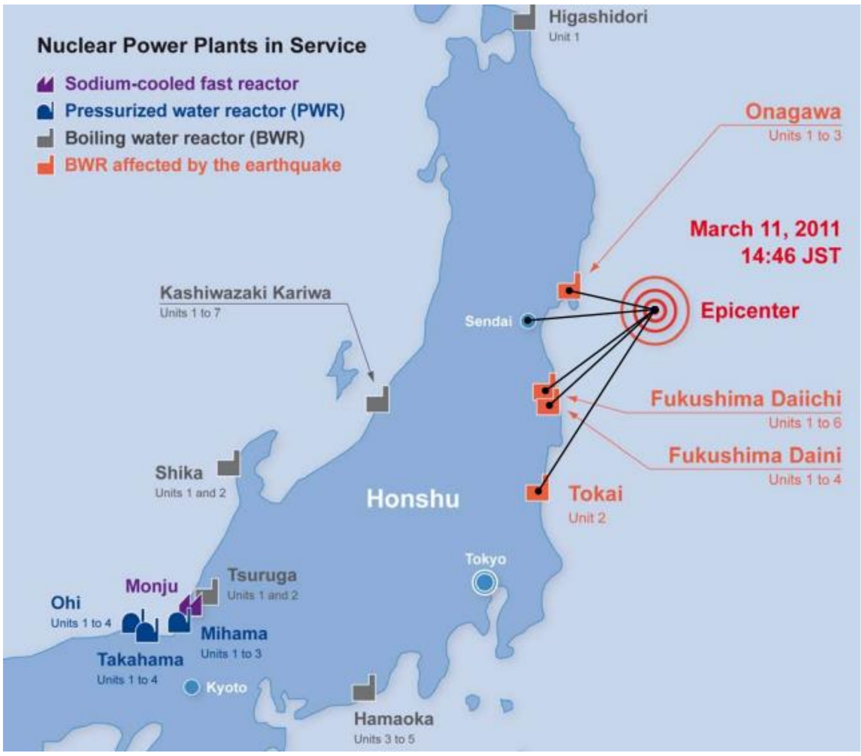


Figure 6: Epicenter of the earthquake and its position in regard to NPPs in Japan. Modified after [2]

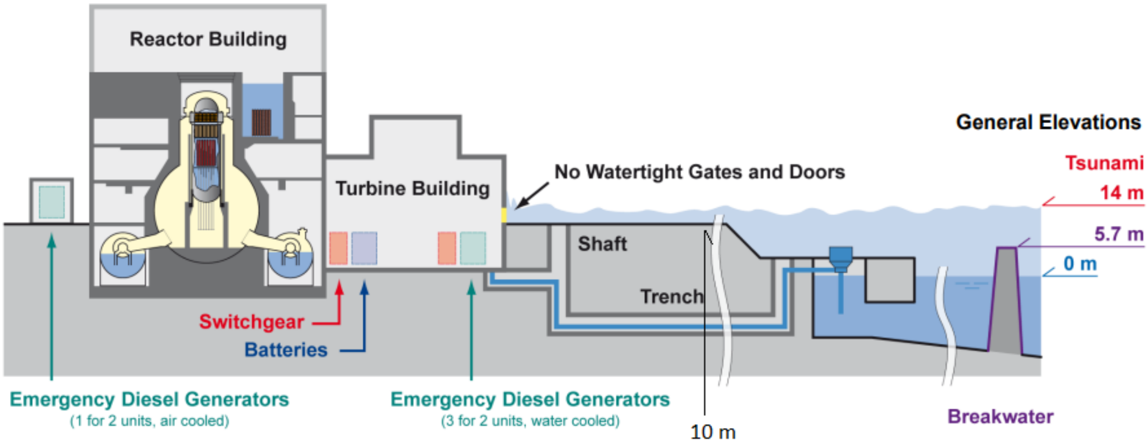


Figure 7: Daiichi NPP setup and general elevation. Modified after [2]

Following the nuclear accident at Fukushima Daiichi Nuclear Power Plant, a fraction of the fuel's activity was released into the environment – approximately 520 PBq (excluding noble gases) (7). Volatile compounds such as gamma-ray-emitting ^{134}Cs, ^{137}Cs and ^{131}I were easily and constantly monitored in the area. On the other hand, very little information on the less volatile ^{89}Sr and ^{90}Sr was at hand because of the laborious matrix separation process for an appropriate measurement of the beta radiation with Liquid Scintillation Counting (LSC) or gas ionization detectors. The similarity of strontium to calcium makes radiostrontium a notorious bone-seeker, increasing the chances for leukemia or bone cancer. Previous studies indicated that radiostrontium was released in low amount, and due to its low volatility, it is most likely to be found in close proximity to FDNPP (7) (8).

After the discovery of bovine bones 2 km away from the FDNPP, an opportunity arose to address the research gap on the understudied ^{90}Sr released from the reactors. After further investigation, it was found out that the cows these bones were stemming from, were set free from the barn as the evacuation began after the accident. Due to this environmental sample being promised to the Applied Radiochemistry group at the TU Wien, this thesis with the purpose of establishing and evaluating an analytical protocol for the extraction and measurement of ^{90}Sr was carried out.

1.4 Strontium

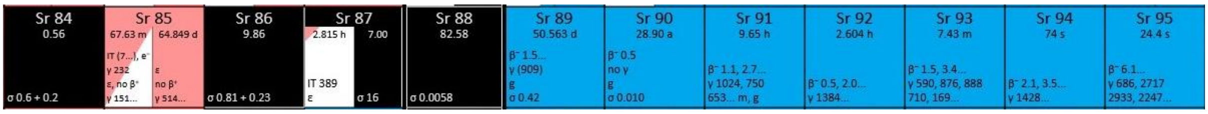
Strontium is an alkaline earth metal with the atomic number 38, belonging in the second group of the periodic table. In the nature it is mostly found in the form of carbonates (strontianite) and sulfates (celestite), with its only environmentally relevant oxidation state being +2, like all other alkaline earth metals. Strontium is not biologically significant for humans, but since it is a calcium-analogue (due to its ionic radius and oxidation state) it is actively incorporated by the organism as strontium-apatite $(Ca,Sr_3(PO_4)_2$ x $Ca,Sr(F,Cl,OH)_2$ in teeth and bone. The biological half-life of strontium is between 3 and 20 years, and factors influencing it are age, sex, height, food incorporation, etc. (9) (10).

Strontium has 4 stable (^{84}Sr, ^{86}Sr, ^{87}Sr, ^{88}Sr) and 26 known radioactive isotopes, with ^{90}Sr being the environmentally most relevant radionuclide since it is produced in nuclear reactors and during nuclear weapon explosions. ^{90}Strontium does not occur naturally (10).

^{90}Strontium is a pure β^-- emitter, decaying into ^{90}Y with the endpoint energy of 546 keV and a half-life of 29 years. ^{90}Y has a relatively short half-life of 64 h, thus reaching a state of secular equilibrium with ^{90}Sr with 97 % of initial activity of ^{90}Sr in approximately 14 days. This rapid ingrowth of ^{90}Y has radioanalytical implications and can be used for internal quality control. Mass spectrometric determination of ^{90}Sr can be challenging due to isobaric interferences, most prominently with ^{90}Zr (11) (12).

Since the laborious separation of ^{90}Sr from other beta-emitters (most importantly the fission product ^{137}Cs) is not easily followed by the radiation of the target nucleus, gamma-ray-emitting ^{85}Sr is often used as a chemically identical radiotracer. This isotope is oftentimes produced by neutron capture in natural ^{84}Sr or via ^{85}Rb(p,n)^{85}Sr, It decays through an electron capture process, releasing a 514 keV gamma photon emission (96 % probability), with a half-life of 64.8 days. Due to its relatively short half-life, gamma emission, and production simplicity, it is the ideal radiotracer for the determination of the extraction yield of Sr (9) (12).

The fission product ^{89}Sr is a relevant fission product to be considered directly after a nuclear accident or a nuclear weapon test fallout due to its relatively short half-life of 50.5 days. This radioisotope decays through a beta-decay with an endpoint energy of 1502 keV into stable ^{89}Y (12).



Sr 84	Sr 85	Sr 86	Sr 87	Sr 88	Sr 89	Sr 90	Sr 91	Sr 92	Sr 93	Sr 94	Sr 95
0.56 σ 0.6 + 0.2	67.63 m IT (7...), e⁻ γ 232 ε, no β⁺ γ 151... 64.849 d ε no β⁺ γ 514...	9.86 σ 0.81 + 0.23	2.815 h IT 389 ε 7.00 σ 16	82.58 σ 0.0058	50.563 d β⁻ 1.5... γ (909) g σ 0.42	28.90 a β⁻ 0.5 no γ g σ 0.010	9.65 h β⁻ 1.1, 2.7... γ 1024, 750 653... m, g	2.604 h β⁻ 0.5, 2.0... γ 1384...	7.43 m β⁻ 1.5, 3.4... γ 590, 876, 888 710, 169...	74 s β⁻ 2.1, 3.5... γ 1428...	24.4 s β⁻ 6.1... γ 686, 2717 2933, 2247...

Figure 8: Strontium isotopes (13)

2. Materials and methods

In order to address interferences in the measurement, ^{90}Sr had to be separated before counting the beta radiation. Therefore, a solid phase extraction method with Triskem® Sr-specific resin was used. In the first step, ashed bones were dissolved in 8 mol/L HNO_3 solution before loading the sample onto the cartridge to specifically bind strontium to the crown-ether material of the resin (Figure 9). In this process, other elements either passed through during the loading process, were washed away before eluting strontium, or stayed behind in the cartridge when strontium was eluted. Following the separation procedure, the end fraction containing strontium was measured and analyzed for the ^{90}Sr content in bovine bones. Yield for the separation process was determined with a ^{85}Sr tracer produced in the TRIGA Mark II reactor at the TRIGA Center Atominstitut of TU Wien.

2.1. NAA

The concept of neutron-induced nuclear reactions and measuring the decay process of created new radioisotopes is regularly used in analytical chemistry – in the Neutron Activation Analysis (NAA) procedure.

If parameters such as provided neutron flux, the elements half-life, cross section, and the number of atoms is known, the activity produced by neutron capture can be calculated by the equation (2.1.1.):

$$A = \sigma \cdot \Phi \cdot N_A \cdot (1 - e^{-\lambda t_a}) \qquad (2.1.1.)$$

To calculate the activity after activation with neutrons at a specific time regarding the decay of produced radioisotopes, the previous equation is transformed into:

$$A = \sigma \cdot \Phi \cdot N_A \cdot (1 - e^{-\lambda t_a}) \cdot e^{-\lambda t_w} \qquad (2.1.2.)$$

2.2. Bone matrix

Bovine bones are primarily composed of inorganic substances – 70 % (mineral phase: mainly hydroxyapatite: $Ca_{10}(PO_4)_6(OH)_2$), and with the rest 30 % being organic substances (mainly collagen) (14).

Bone matrix is a two-phase system, in which the stiffness is provided by the mineral phase and collagen providing the ability to absorb energy – making it somewhat elastic. Mechanical properties of bones can therefore be altered by altering the collagen, making bones more susceptible to fractures (15).

Decomposition of the organic components in bones occurs between 360 and 525°C – including burning off almost all of the organic material and therefore removing bones' elasticity (16).

2.3. SPE – Sr resin/chromatography

In order to separate strontium from the matrix, solid phase extraction procedure with commercially available Sr-resin from the company Triskem® was used. The resin was packed into cartridges, with 2 mL of 50-100 µm particle size material. The part binding to strontium is the crown ether (4,4’(5’)-di-t-butylcyclohexano-18-crown-6) (Figures 9,10), into which a strontium ion coordinated by two nitrate anions fits perfectly. The strontium-specific agent is diluted in 1-octanol (1 % solution) and embedded on an inert stationary phase – 1 mol/L of resin. The capacity of the Sr-resin is reported to be 8 mg/mL of resin. Even though calcium shows very little affinity for this specific Sr-resin ($k'_{Ca,max}<0.3$, a study conducted at BNFL (UK) showed that chemical recovery of strontium decreases when the amount of calcium loaded onto a 2 mL cartridge is greater than 300 mg (17).

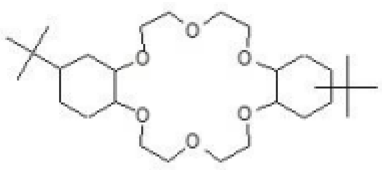


Figure 9: Crown ether from the Sr-resin (17)

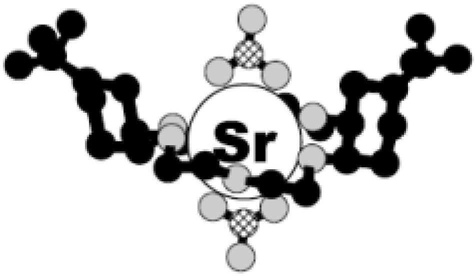


Figure 10: Crown ether from Sr-resin binding to strontium (17)

Strontium’s affinity toward the crown ether increases with nitrate ion concentration, reaching its maximum at k_{eq}~90 at 3-8 M HNO_3 and its reported minimum at k_{eq}~0.2 at 0.02 M HNO_3 (17). Existing literature shows that there is no noticeable difference in elution efficiencies between the 0.05 and 0.01 mol/L HNO_3 solutions, and that both are sufficient enough to separate Pb from Sr, with Pb being retained by the cartridge at these concentrations of nitric acid (18).

2.4. Gamma radiation and spectrometry

If a nucleus of a higher energy state decays into a nucleus of a lower energy state, a γ-emission (photon) is emitted. These photons are electromagnetic waves of a low (γ) wavelength and high energy (20 keV up to 10 MeV), and because of this reason they can travel great lengths without interacting with matter - making them highly penetrative. They have three mechanisms of interaction with matter, which depend primarily on the energy of the γ-ray: photoelectric effect (<100 keV), Compton effect (between 100 and 5000 keV) and pair production (>5 MeV). The wavelengths produced in this decay are isotope-specific and can be measured with the goal of qualifying or quantifying certain elements.

Most prevalent γ-detectors are semiconductor and scintillation detectors.

High Purity Germanium (HPGe) gamma spectroscopy (semiconductor detector) is seen as the high energy resolution but low counting efficiency detector, often requiring longer counting times. This detector consists of a high purity germanium crystal which is cooled by liquid nitrogen, shielding, electronics and a data processor. The HPGe crystal consists of two crystals (electrodes) joined together: one with electron excess (n-doped) and the other one with hole excess (p-doped). Through ionizing radiation, free electrons and holes are produced in the crystals, with their number being dependent on the energy transmitted through the radiation. Both free electrons and holes will all travel to the electrode junctions – giving rise to a pulse that can be measured in an outer circuit – allowing for calculation of energy transmitted through the γ-ray (1).

Scintillation detector consists of a crystal (for example NaI doped with small quantities of Tl), which after absorbing radiation emits photons. These photons then release electrons at the photocathode or photomultiplier tube, which get multiplied by the dynodes of the multiplier, giving pulses of several millivolts. These detectors usually have lower energy resolution but higher counting efficiency (1).

2.5. Liquid Scintillation Counting – LSC

Since beta particles have low penetrative ability, they are difficult to measure directly, and one of the ways of measuring them is transforming them into easier measurable photons. In this process, the liquid sample (β^--emitter) is mixed with a liquid scintillation cocktail (scintillator) – which produces photons from interactions with beta radiation. These photons can be detected with the classical scintillation detector. Main advantage of this method is its high counting efficiency of ~100 %.

The sample for LSC should be as pure as possible to minimize self-absorption, and in little water as possible in which the analyte is dissolved after separation, since foreign substances often reduce the light output and counting efficiency – an effect known as quenching. The scintillation liquid (solution containing the radioactive sample and scintillator) can also consist of multiple scintillators if necessary. The sample container must be resistant to the solvent, and transparent at the wavelength produced by the scintillator (1).

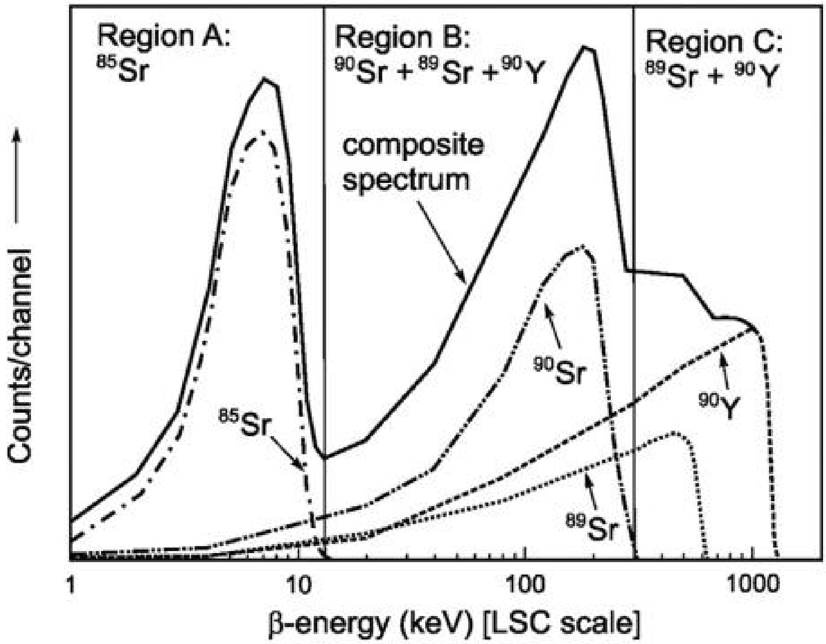


Figure 11: ^{90}Strontium LSC spectrum (19)

2.6. Internal std/tracer system

When performing solid phase extraction, it is important to determine the extraction yield since losses are to be expected. This is done through an internal standard – a radiotracer in many radioanalytical techniques. ^{85}Sr can be used as an internal standard for the ^{90}Sr extraction, since it's a gamma-emitter – being simpler to measure. Due to them being different isotopes of the same element, they exhibit the same chemical properties and the same behavior during the extraction process.

The yield is calculated through the equation:

$$Recovery\ yield\ [\ \%] = \frac{Meassured\ {}^{85}Sr\ activity}{Added\ {}^{85}Sr\ activity} \cdot 100\ \% \qquad (2.6.1)$$

2.7. Chemicals and materials

Bovine bones bought in supermarket Spar in Vienna, Austria

Bovine bones found in a forest in district Melk, Lower Austria

Hydrogen Peroxide 30 % by Carl Roth; Art.# 8070.4

Nitric acid 65 % by ROTH; Art.# X898.2

Oxalic acid dihydrate ≥99 % by ROTH; Art.# 8879.1

Strontium nitrate extra pure by Riedel-de Haen

^{90}Sr stock solution with an unknown concentration

Sr Resin (particle size: 50-100 µm, 2mL) from Triskem®

LSC glass vials

Ultima Gold™ scintillation cocktail

2.8. Instrumentation and software

HIDEX NaI gamma counter, this instrument is referred to as "gamma counter" in this thesis

222 cm^3 HPGe gamma spectrometer, this instrument is referred to as "gamma spectrometer" in this thesis

PerkinAlmer Tri-Carb 2910 TR Liquid Scintillation Analyzer, this instrument is referred to as "beta counter" in this thesis

CEM Mars6 Microwave

TRIGA Mark II research reactor

Thermo Scientific Heraeus furnace

Efco 180 muffle furnace

2.9. Experiments

2.9.1. Sample preparation

The first bone (reference) sample was a commercially available bone at a local supermarket and was stored at approximately 5°C in a refrigerator. However, since the anticipated samples from Fukushima are 12 years old bones with minimal organic matter, the constitution differed from our obtained(reference) samples. Therefore, before the sample was ashed, organic matter removal steps were performed. Bones were immersed into a 15 % H_2O_2 solution overnight (~16h), after which the organic parts (primarily bone marrow) were softened and removed by hand. Bones were subsequently ashed for 1 h at 500°C and were under an open flame for another 1 h due to the extreme smoke build up – pyrolyzing the sample completely as seen in Figure 8. This sample was pulverized with a pestle and mortar producing bone ash as seen in Figure 9.

Figure 12: Pyrolyzed bone #1

Figure 13: Pyrolyzed bone (ash) #1 ground

A new sample resembling the Fukushima bones with its minimal organic content was collected in a forest in Lower Austria, which was cut with an iron saw into smaller pieces (Figure 10) and ashed in a ceramic-muffle furnace (Figure 11). The end-temperature of 550°C was reached in approximately an hour, with 5°C per minute heating ramp, and held for 3 hours, before cooling the sample down to room temperature.

Figure 14: Cut bone samples #2

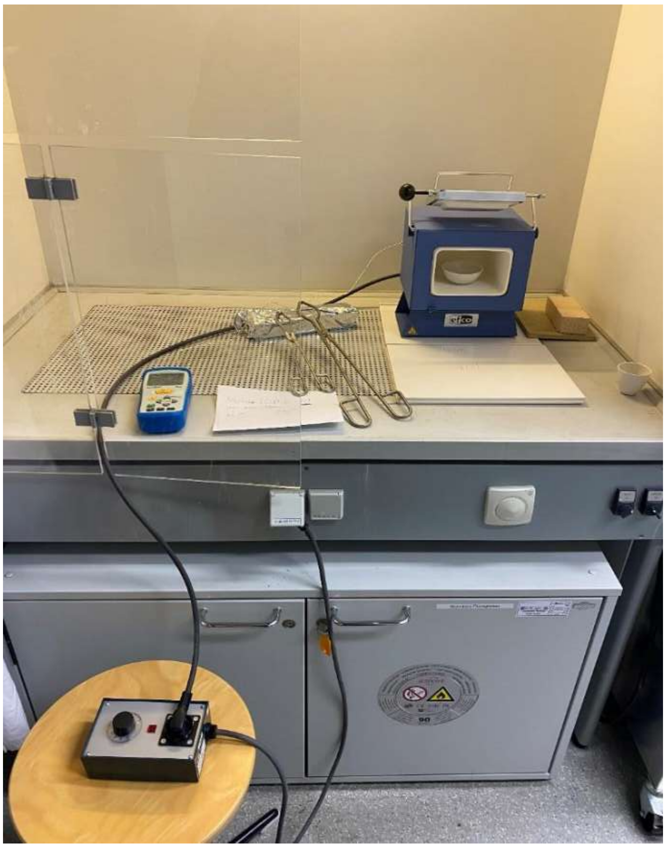


Figure 15: Ceramic muffle oven setup for bone samples #2

2.9.2. Ash dissolution

Pulverized and homogenized samples were weighed and prepared for the dissolution. The first try was performed with various amounts of ash as presented in Table 1 in 10 mL 8 mol/L HNO_3 over night at 50°C, which was unsuccessful. The second try was performed with 4 times 0.5 g of bone ash in the CEM Mars6 microwave at 200°C for 30 min – method for bone-ash dissolution from the microwave manual (Figure 12). This procedure was successful in dissolving the ash in 8 mol/L HNO_3 producing a clear golden liquid.

Table 1: Ash dissolution, first try

Sample	Ash mass [g]
1	0.1
2	0.2
3	0.5
4	1

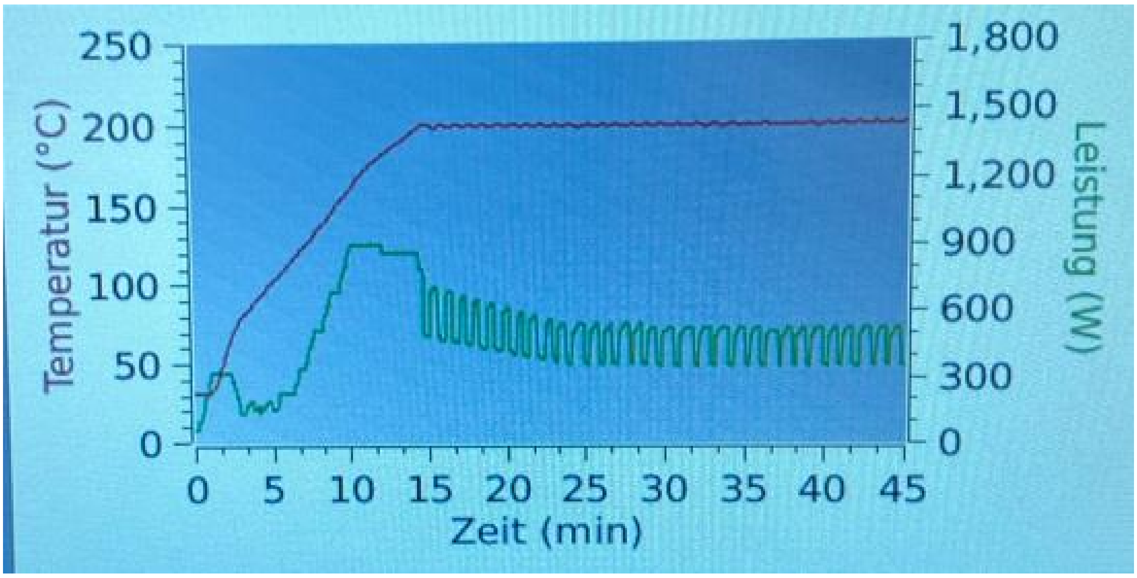


Figure 16: Microwave program for bones dissolution in HNO_3

2.9.3. Strontium extraction

For the extraction of strontium, 2 mL Sr-resin cartridges with particle size: 50-100 µm from the company Triskem® were used. The resin was conditioned once with 5 mL 8 mol/L HNO_3 1 h before using them and once with 5 mL 8 mol/L HNO_3 directly before loading for the total conditioning volume of 10 mL. After the conditioning step, the resin was loaded with 10 mL sample solution in 8 mol/L HNO_3 over the course of approximately 2 min (5 mL/min). The sample-holding container (microwave Teflon tubes) was washed with additional 5 mL 8 mol/L HNO_3 which were also loaded onto the cartridge, together with wash fractions of 5 mL 8 mol/L HNO_3 and 10 mL 3 mol/L HNO_3. For the ^{90}Sr samples, the 3 M HNO_3 wash solution also contained 0.25 mol/L of oxalic acid. Strontium was eluted with 2x 5 mL 0.05 mol/L HNO_3 solution, or 0.01 mol/L HNO_3 solution for ^{90}Sr samples.

2.9.4. Gamma spectrometer analysis of bone ash

The produced bone ash, with the sample mass of 100 mg, was activated in the reactor under the parameter presented in Table 2. After a cooling time of 14 days, the sample was measured with the gamma spectrometer for 24 h.

Table 2: Activation parameters

Neutron Flux	^{84}Sr abundance	^{84}Sr cross section [b=10^{-24} cm^2]	Irradiation time [min]	^{85}Sr half-life [d]	Decay constant [min]
10^{12}	0.00562	0.5	1260	64.849	7.405^{-6}

2.9.5. Gamma counter analysis of separated strontium

The 10 mL 0.05 mol/L HNO_3 solutions with eluted strontium were evaporated to dryness in a 25 mL crystalizing dish over night at 40°C. Masses of empty dishes and dishes with the solids after evaporation of extracted Sr samples are summed in Table 3. The remaining solids from the bottom of the dishes were redissolved in 1 mL 0.02 mol/L HNO_3 and transferred to 1.5 mL Eppendorf tubes together with 1 mL 0.02 mol/L HNO_3 blank sample. All samples were evaporated to dry in the 1.5 mL Eppendorf tubes and redissolved in 0.5 mL 0.02 ml/L HNO_3. The 0.5 mL solutions were split into 0.3 mL and 0.2 mL fractions, from which the 0.3 mL fractions were activated for 3 days under the circumstances presented in Table 2 and counted with the NaI-well gamma counter for 3 h.

Table 3: Beakers masses before and after eluted Sr-fraction drying

Sample	Empty crystalizing dish [g]	Full crystalizing dish [g]	Δ [mg]
1	18.64487	18.64527	0.40
2	19.54977	19.54994	0.17
3	19.55155	19.55171	0.16
4 – first half	22.70206	22.70236	0.30
4 – second half	18.54839	18.54804	-0.40

2.9.8. Calibration curve: activated $Sr(NO_3)_2$

A known amount of $Sr(NO_3)_2$ (80 mg) was activated in the TRIGA Mark II reactor under the parameters presented in Table 2, producing the radioisotope ^{85}Sr. Out of the 80 mg activated, 60 mg of $Sr(NO_3)_2$ were dissolved in 50 mL 8 mol/L HNO_3 creating a stock solution with ~0.5 mg of Sr^{2+}/mL, which was used as a radiotracer for the future extraction experiments. Standards prepared (5 non-extracted standards and 1 blank) were subsequently measured with the gamma counter (Table 4).

Table 4: Activated Sr-nitrate non-extracted standards

Sample	Volume [mL]	Amount (Sr^{2+}) [mg]
1	0 (blank)	0 (blank)
2	0.5	0.25
3	1	0.5
4	2	1
5	5	2.5
6	10	5

2.9.7. Strontium-separation extraction yield determination

Bone ash samples (6 times 0.5 g) were weighed and dissolved in HNO_3 as explained in the section 2.9.2.. After acid digestion, the 10 mL samples were spiked with known amounts of ^{85}Sr tracer presented in Table 5 – same amounts used for standards in the previous section: 2.9.8.. The strontium has been subsequently extracted as explained in the 2.9.3. section followed by measuring with the gamma counter.

Table 5: Samples for extraction yield determination

Sample	Ash [g]	Spike [mL]	Spike (Sr) [mg]
1	0.5	0 (blank)	0 (blank)
2	0.5	0.5	0.25
3	0.5	1	0.5
4	0.5	2	1
5	0.5	5	2.5
6	0.5	10	5

2.9.8. ^{90}Strontium extraction and measurement – LSC and solubility test

The first samples with ^{90}Sr were 2 times 10 mL 8 mol/L HNO_3 solutions spiked with 100 µL of a ^{90}Sr stock solution with unknown activity or concentration. Solid phase extraction was performed on the samples as described in section 2.9.3. Strontium extraction After the elution, sample 1 was evaporated to 50 % of its initial volume (10 to 5 mL) and sample 2 was evaporated to dryness and re-dissolved in 1 mL of deionized water, before being transferred to the LSC glass vials. The beakers used for evaporation were washed with an additional 1 mL of deionized water which was then combined with the aqueous samples in LSC glass vials. Both samples were mixed with 9 mL of Ultima Gold scintillation cocktail and measured by the beta counter for 30 min. Sample 1 did not mix fully – creating a cloudy liquid, whereas the sample 2 created a clear liquid when mixed with the cocktail, as seen in Figure 17.

Figure 17: Sample 1(left), sample 2 (right)

2.9.9. ^{90}Strontium spike in bone ash – LSC and miscibility test

For the next experiment, 3 times 0.5 g of bone ash samples were dissolved in 10 mL 8 mol/L HNO_3 and spiked with 100 µL of the same stock-solution from experiment 2.9.8.. Solid phase extraction was subsequently performed as described in 2.9.3. section. Eluted samples (10 mL) were evaporated multiple times to dryness before being redissolved in 10 mL deionized water under heating, with the number of evaporations being presented in Table 6. After the specified number of evaporations, samples 3 and 4 were evaporated to approximately 1 mL before being transferred to the LSC glass vials. Their evaporation containers (crystallization dishes) were washed with an additional 1 mL of water, which was then combined with the samples in the LSC glass vials – making the end-volume approximately 2 mL. Sample 5 was evaporated to approximately 4 mL before being transferred to LSC vial, and again combined with 1 mL of water that was used to wash the container it was evaporated in – making the end volume approximately 5 mL.

All samples were then combined with 9 mL of Ultima Gold scintillation cocktail directly in the 20 mL LSC glass vials before being measured by the beta counter for 30 minutes.

Samples 3 and 4 demonstrated good miscibility with the cocktail as seen in Figure 18 (samples marked as 1 and 2), whereas the sample 5 created a cloudy liquid upon mixing with the cocktail.

Figure 18: Samples 3,4, and 5 (marked as 1,2, and 3)

Table 6: ^{90}Strontium spike in bones matrix procedure variables

Sample	Number of evaporations until dryness	End-volume of sample$_{aq}$ [mL]	Volume of Ultima Gold scintillation cocktail [mL]
3	2	2	9
4	3	2	9
5	5	5	9

3. Results and discussion:

3.1. Bovine bones ashing and dissolving

Since the 150 g of bones used for the initial ashing experiment were still wet, ashing led to excessive smoke build-up, and the muffle furnace had to be turned off after one hour. The bones were allowed to burn after shutdown for another hour, which subsequently ashed them. This process charred them and made them brittle enough to be pulverized with a pestle and mortar.

The next ashing experiment was conducted with a smaller sample size of approximately 6 g and a temperature ramp as described in section 2.9.1.. The new oven was smaller and fit in the digestor, ensuring better ventilation. This time, no noticeable amount of smoke was released – this was most likely due to the smaller sample size and its dryness. After this procedure, the bones lost approximately 20 % of their mass and turned gray. Their brittleness once again allowed for them to be pulverized with a pestle and mortar.

As mentioned in section 2.9.2., an earlier attempt to dissolve the bones by heating them up to 50°C and stirring them was unsuccessful. Dissolving the ash in 8 M nitric acid at 200°C for 30 min in a microwave, however, turned out to be successful and was therefore chosen as the method for ash dissolvement for our experiment due to its simplicity, effectiveness, and reproducibility.

3.2. Bovine bones ash NAA

NAA was performed on the initially produced bone ash as described in section 2.9.1. to demonstrate strontium's natural occurrence in bovine bones. The background was too high even after 14 days of cooling, and with strontium's 514 keV peak being a shoulder of the annihilation peak at 511 keV peak, only qualitative determination was possible.

After this attempt, it was concluded that a strontium separation was required before its quantitative determination.

3.3. Determination of Sr in bovine bones - SPE + gamma counting

The bovine bone samples were prepared as described in sections 2.9.1, 2.9.2., 2.9.3., and counted by using the gamma counter as described in section 2.9.5.. The results can be seen in Table 7.

Table 7: Counted extracted strontium from bovine bones and Sr mass corrected with the 0.6 fraction separation factor

Sample	^{85}Sr CPM [counts/minute]	^{85}Sr activity [Bq]	Sr mass with 0.6 correction factor [mg]	% of bone ash mass
1	692	12	0.064	0.013 %
2	2060	34	0.182	0.036 %
3	652	11	0.059	0.012 %
4_1	670	11	0.059	0.012 %
4_2	-	-	-	-
Blank	-	-	-	-

The strontium mass was calculated from the activity using the activation equation (2.1.1.) and activation parameters from Table 2, with the activity in Becquerel calculated by the gamma counter software. Due to the splitting of the fractions (0.3 mL activated and counted out of the 0.5 mL samples), all results will be corrected with a 60 % correction factor. In the following sections, the Sr mass will be recalculated using a standard calibration curve and the extraction yield factor.

As shown in Figure 19, ash sample 2 has a higher background than other samples, demonstrating a higher activity. Sample 4_2, being the second 5 mL elution fraction of sample 4, showed no ^{85}Sr as seen in Figure 20.

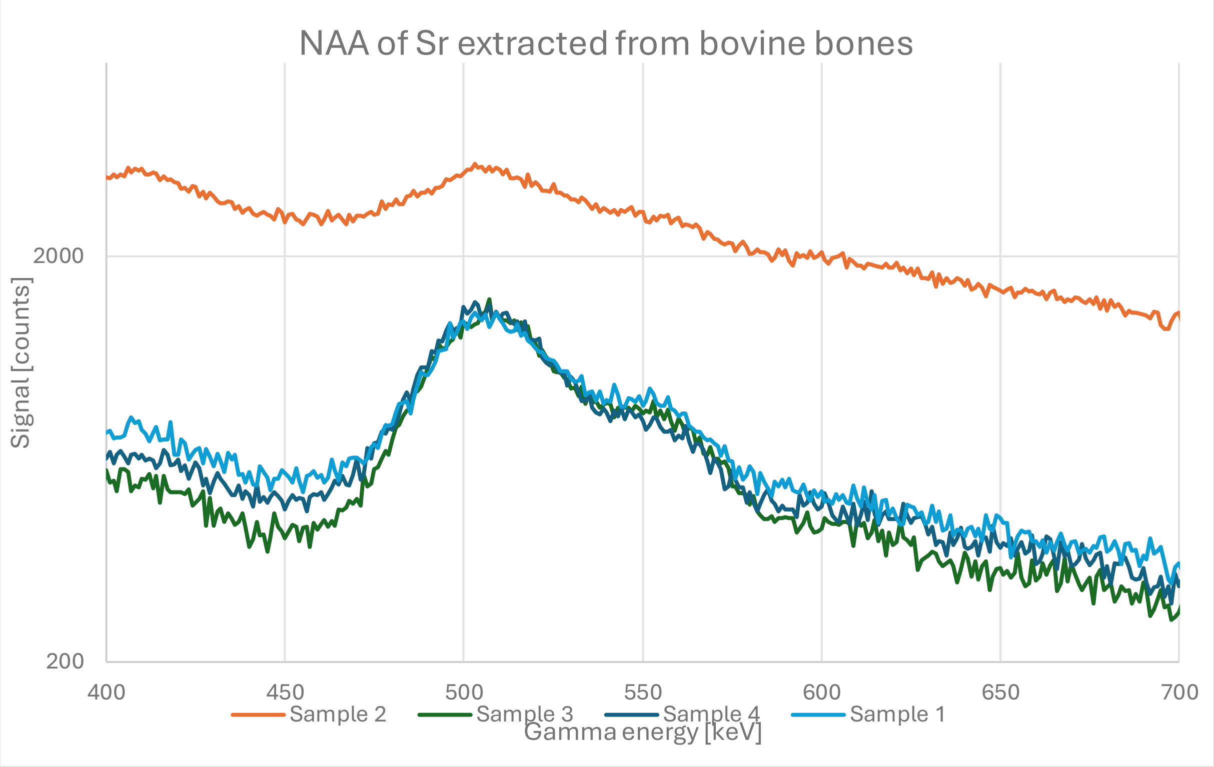


Figure 19: NAA of Sr extracted from bovine bones – logarithmic scale

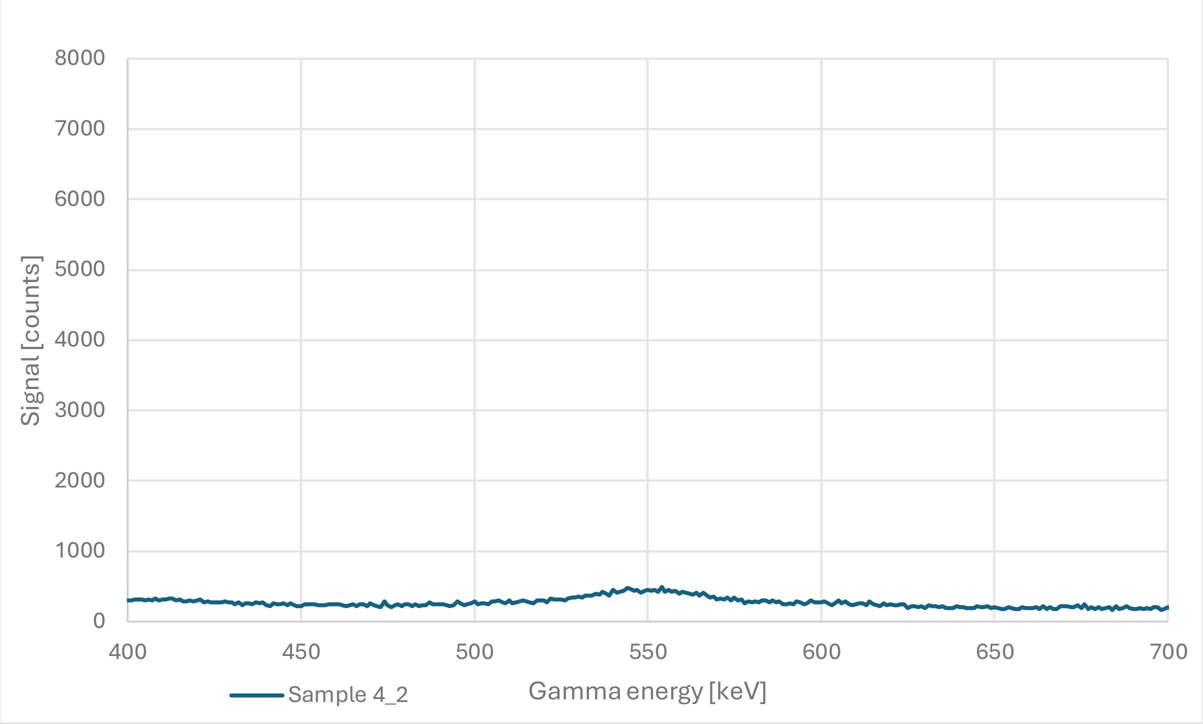


Figure 20: NAA of Sr extracted from bovine bones sample 4_2

The blank sample demonstrated a peak at approximately 555 keV, which could be a contamination in either this specific Eppendorf tube or a different glass piece since it is only seen in the blank sample (Figure 21).

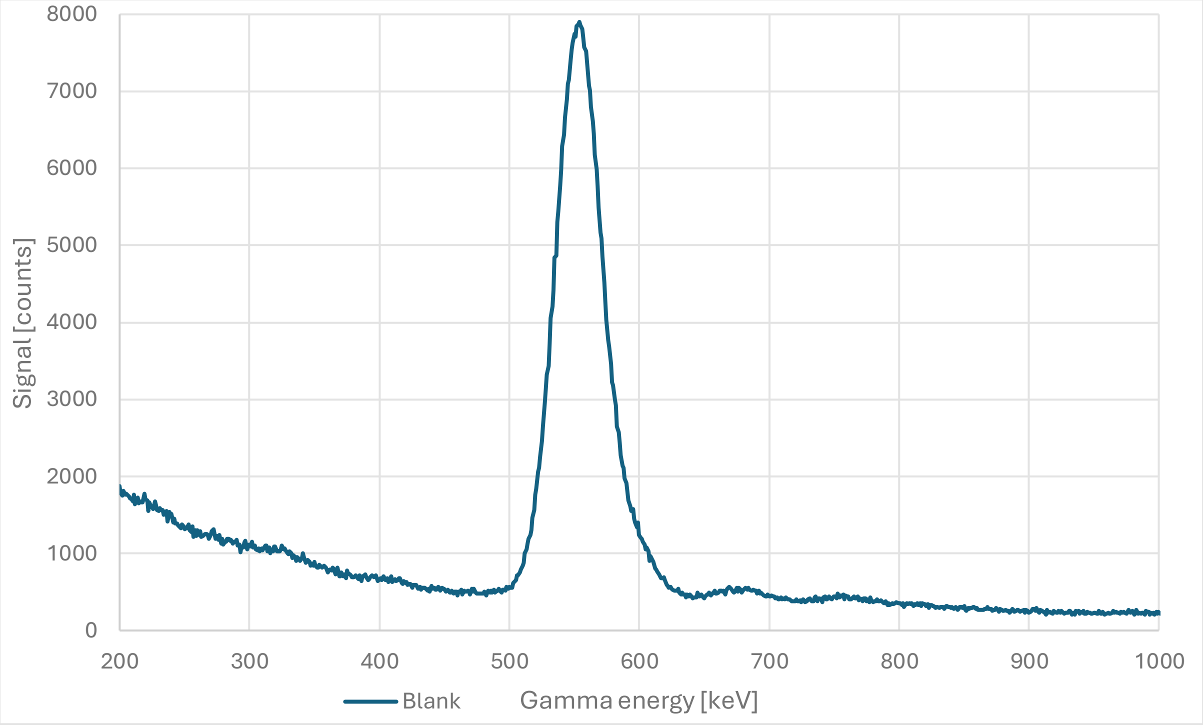


Figure 21: Blank sample (1 mL 0.02 mol/L HNO_3 evaporated to dry in 1.5 mL Eppendorf tube and tread like other samples further on)

3.4. Extraction yield determination and strontium amount in bone ash recalculation

In order to determine the extraction losses of Sr during the SPE procedure, the gamma-emitting isotope ^{85}Sr was used as an internal standard. The extraction protocol described in section 2.9.3. was performed on 5 spiked and one blank ash sample. All fractions were gamma-counted together with non-extracted standards to determine the extraction yield. The strontium resin manufacturer reported an extraction yield above 85 %, but also a decrease of the chemical recovery of strontium if the Ca^{2+} amount is more than 300 mg when using the 2 mL cartridge. According to NIST's literature value of bone ash being ~38 % calcium, our 0.5 g samples are supposed to contain less than 300 mg of calcium if using that literature value for calcium amount approximation (20). The reported strontium limit of the cartridge is at 8 mg of Sr^{2+}, which is only relevant for the added carrier in the case of our samples (17). As presented in Table , losses occurred in both the load and the wash fractions, together with the additional losses of Sr remaining in the resin.

All the calculations from counts/minute to becquerel were done by the Hidex software with the instrument's ^{85}Sr efficiency metric. The gamma-counter measurement of the non-extracted ^{85}Sr standard from Section 2.9.8. was used to produce a calibration curve (Figure 23) and represent 100 % of activity in the Sr extraction calculations presented in Table 8.

Although the ^{85}Sr isotope offers reliable and straightforward counting/yield determination as a tracer/internal standard, it could hinder the LSC measurement of ^{90}Sr. Therefore, it was not used as a tracer for ^{90}Sr determination.

Table 8: Strontium extraction recoveries

sample	Load [Bq]	1st wash [Bq]	2nd wash [Bq]	**Eluate** [Bq]	**Yield [%]**	Cartridge [Bq]	**Non-extracted standard** [Bq]	traced [%]
Blank	1	1	1	1	-	1	1	-
0.25 mg	1	1	1	**20**	**83**	2	**24**	104
0.5 mg spike	1	1	1	**41**	**87**	3	**47**	100
1 mg spike	1	2	2	**80**	**81**	4	**99**	90
2.5 mg spike	5	5	6	**187**	**86**	9	**216**	98
5 mg spike	10	11	14	**356**	**83**	13	**430**	94

The yield for Sr extraction with Sr-resin by Triskem® from bone ash matrix is demonstrated to be **84±2.5 %.**

The reason for tracing ≥100 % of ^{85}Sr is most likely the background which was rounded to 1 Bq in the lower concentration samples, which should not hinder the extraction yield calculation.

The values from the previous experiment (Section 3.3.) corrected with the extraction yield are presented in Table 9.

Table 9: Counted extracted strontium from bovine bones and Sr mass corrected by the extraction yield factor and the 0.6 fraction separation factor

Sample	^{85}Sr CPM [counts/minute]	^{85}Sr activity [Bq]	Sr mass **corrected** [mg]	% of bone ash mass **corrected**
1	692	12	0.127	0.03 %
2	2060	34	0.362	0.07 %
3	652	11	0.117	0.02 %
4	670	11	0.117	0.02 %

It is possible to use eluate fraction activities [Bq] from Table 8 as standards for other ^{85}Sr measurements performed with the same extraction procedure and measured with the same gamma counter. The calibration points are presented in Figure , and the slope for both curves are presented below: equation 3.4.1. being the slope for non-extracted standards curve and equation 3.4.2. for the extracted standards curve.

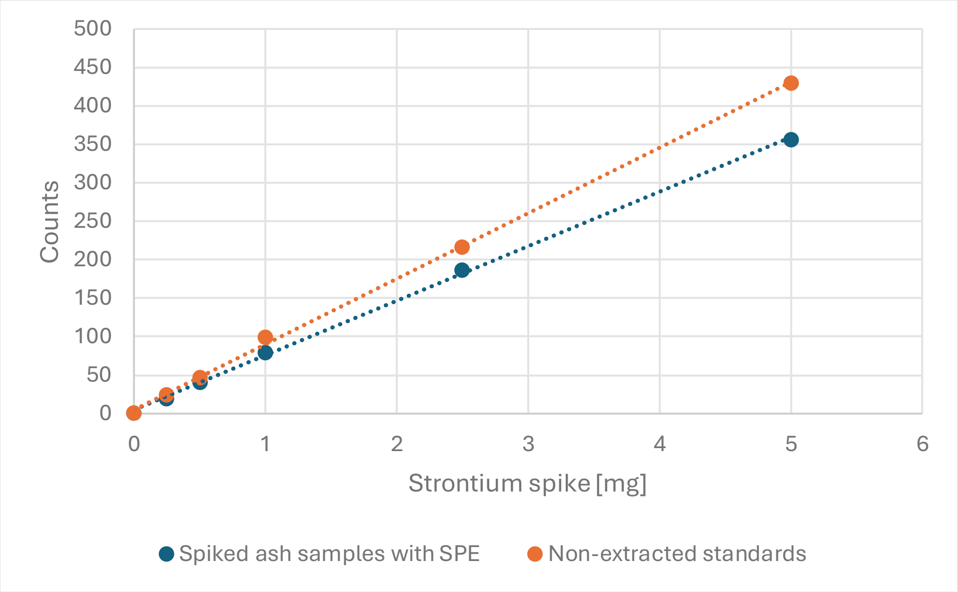


Figure 22: Gamma counted spiked and Sr-extracted ash samples together with non-extracted standards

$$y = 85.179x + 4.8486 \qquad (3.4.1.)$$

$$y = 70.864x + 4.918 \qquad (3.4.2.)$$

Due to the real bone samples having lower strontium content than most points in the calibration curve (Sr mass approximately 0.20 mg per sample), the corrected strontium mass calculation in Table 10 was performed with the 0.25 mg (20 Bq in the eluate fraction) spiked sample.

Table 10: Counted extracted strontium from bovine bones and Sr mass corrected and calculated with the 0.25 mg Sr standard from Table 8

Sample	^{85}Sr CPM [counts/minute]	^{85}Sr activity [Bq]	Sr mass **corrected** [mg]	% of bone ash mass **corrected**
1	692	12	0.250	0.05 %
2	2060	34	0.303	0.13 %
3	652	11	0.230	0.05 %
4	670	11	0.230	0.05 %

3.5. ^{90}Strontium measurement – solubility test and LSC

The goal of this experiment was to test the solubility of aqueous ^{90}Sr eluate solution with the Ultima Gold scintillation liquid after different number of evaporations till dryness of the weak nitric acid solution containing ^{90}Sr. The evaporation step is used to minimize the concentration of the nitric acid to reach better miscibility with the scintillation liquid minimizing the quenching effect. Considering these results, there seems to be more correlation with the number of samples mixed than with the number of evaporations.

The solution used for Sr elution was 0.01 mol/L of HNO_3, differing from the used solution (0.05 mol/L HNO_3) in extraction yield determination experiment, which shouldn't have much different Sr or Pb elution efficiencies according to existing literature (18).

Countings have been performed by the QuantaSmart™ software on the beta counter, with the efficiency presumed to be 100 % (18).

Table 11: Samples preparation (evaporations till dryness) for LSC

Sample	Number of evaporations till dryness	End volume of the solution before mixing with ultima gold [mL]	Miscibility with 9 mL Ultima Gold scintillation liquid
1	0	5	Bad
2	1	2	Good
3	2	2	Good
4	3	2	Good
5	5	5	Bad

Table 12: LSC measurement of the spiked ash samples

Sample	Counts/ minute	Time from separation	Activity$_{total}$ [Bq]	Activity$_{90Sr}$ [Bq]	Amount of ^{90}Sr corrected with 0.85 extraction yield
1	209	1 h	3.48	3.45	0.794 pg
2	200		3.33	3.30	0.769 pg
3	235	4 h	3.92	3.76	0.876 pg
4	247		4.12	3.95	0.920 pg
5	238		3.97	3.81	0.876 pg

To calculate the ^{90}Y ingrown we use the equation:

$$A_Y\,(t) = A_0 * (1 - e^{-\lambda t}) \tag{3.5.1.}$$

With λ (^{90}Y decay constant) being:

$$\lambda = \frac{\ln{(2)}}{64.1} \sim 0.01\ per\ hour \tag{3.5.2.}$$

The ^{90}Y activity is calculated to be 4.23 % of initial activity of ^{90}Sr after 4 hours.

$$A_Y\,(4h) = A_0 * 0.0423$$

From the corrected ^{90}Sr using the following equation, mass has been calculated to be:

$$m[g] = \frac{\frac{A}{\lambda} * M}{N_A} = \frac{A[Bq] * 89.907}{7.63 * 10^{-10} * 6.022 * 10^{23}} = A[Bq] * 1.957 * 10^{-13} \tag{3.5.3.}$$

Using the equations 3.5.1., 3.5.2., and 3.5.3. (1) the strontium amount in the ^{90}Sr solution is determined to be 8.47±0.63 pg ^{90}Sr/mL stock solution. Even though samples 1 and 5 demonstrated poor miscibility with the scintillation cocktail, their activity did not deviate from the samples with good miscibility, hence their measured activities were used in the calculations.

4. Conclusion

The prospect of receiving bovine bones stemming from the area ~2 km from Fukushima NPP requires a unique method for the ^{90}Sr analysis in bovine bones. The method developed herein consists of the initial material conditioning – ashing, which is required to turn bones brittle for easier pulverization. The second part was acid digestion of the bones from which strontium can be extracted using a commercially available Sr-resin. The extraction yield was subsequently determined. Since the end-goal is quantifying ^{90}Sr from the bones, test samples were spiked with ^{90}Sr, followed by its extraction and counting by LSC – testing the miscibility of the sample solution and the scintillation cocktail Ultima Gold in the process.

Bones pyrolysis was effective at the tested temperature of 550°C for 3 h, which is suitable for the future pyrolysis of bovine bones stemming from Fukushima. The dry-bones mass loss of approximately 20 % will have to be exactly determined and accounted for during the strontium amount calculation. Pyrolyzed bone pieces were brittle enough to be pulverized with a pestle and mortar.

The ash dissolution performed in a microwave under the set program for bovine bones at 200°C for 30 min was demonstrated to be successful in dissolving bovine bones ash in 10 mL 8 M HNO_3.

For the strontium extraction a specific resin from Triskem® was used as the solid phase which specifically binds to strontium, allowing for selective separation. ^{85}Sr was used as a radiotracer to determine the extraction yield, which would be later used for calculating the total ^{90}Sr in bovine bones. This extraction yield was demonstrated to be 84 ± 2.5 %.

From the LSC tests, it was concluded that the miscibility of the samples with 9 mL of the scintillation cocktail Ultima Gold was best when the sample (aqueous) solution volume does not exceed 2 mL. Therefore, after the samples' elution following the strontium separation procedure, an evaporation till dryness step was required. This process also evaporates the nitric acid, lowering its concentration from the 0.01 mol/L elution solution.

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6. Appendix

Appendix A: List of Figures

Appendix B: List of tables

Appendix C: List of abbreviations

Bq	Becquerel
SI	International system of units
A	Activity
N(t)	Number of particles at the time t
t	Time
λ	Decay constant
$t_{1/2}$	Half-life
V	Volt
W	Watt
σ	Cross section
b	Barn
eV	Electron volt
NPP	Nuclear power plant
FDNPP	Fukushima Daiichi nuclear power plant
LSC	Liquid scintillation counting
Φ	Thermal neutron flux
N_A	Avogadro constant
t_a	Activation time
t_w	Time between the end of activation and the start of the count
BNFL	British Nuclear Fuels Ltd
HPGe	High purity germanium
Art.#	Article number
m	Mass
M	Molar mass
aq	Aqueous
NAA	Neutron activation analysis