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Transfer of Strontium in the Potato Plant, *Solanum tuberosum L.*, Following Single Foliar Wet Deposition: Field Experiments Performed in Eastern Norway and at the West Coast of Norway

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Abstract

^{90}Sr is one of the most important and frequent radioactive products from nuclear fission. If ingested, it is considered a major hazard to humans due to high retention in skeleton, leading to irradiation of cells and cancer development. The ecosystem behavior of ^{90}Sr is of great interest for countermeasure implementation after potential fallout scenarios where the aim is to understand and predict transfer in agricultural plants grown for consumption. This study used a known Sr-isotopic composition ($^{84}Sr/^{87}Sr$) to follow uptake and transfer of Sr in the potato plant, *Solanum tuberosum L.*, after single foliar wet deposition. Agricultural fields at two different NIBIO Research stations; Apelsvoll, situated in the east and Fureneset, situated at the western coast were used for potato cultivation to study how variation in climatic conditions and soil characteristics affect transfer of Sr. The main objectives were to (i) investigate if Sr concentration on foliage would be reduced over time, (ii) quantify and differentiate transfer of Sr at Apelsvoll and Fureneset, (iii) identify where in the potato tubers Sr would accumulate and (iv) determine if translocation from foliage to potato tuber occurs.

Following the wet deposition, leaves and stem were significantly ($p<0.05$) contaminated by ^{84}Sr at both sites. The ^{84}Sr contamination on foliage (leaf+stem) remained about constant during the following three weeks at Apelsvoll. At Fureneset, the foliage concentration of ^{84}Sr was slightly reduced ($p<0.052$), and the reduction was negatively correlated ($r=-0.95$) with the high precipitation rates. The uptake of ^{84}Sr in below soil plant tissue such as stolons, root hair and, to some extent, potato tubers was only significant ($p<0.05$) in plants grown at Fureneset, where ^{84}Sr was found to accumulate in the peel layer of the potato tuber. The higher uptake at Fureneset was found to be related to the low soil cation exchange capacity (CEC), plant available Ca (Ca-Al), plant available Sr (Sr-Al) and clay content in soil. Elevated levels of soil CEC, Ca-Al and clay content reduced uptake of Sr in below soil plant tissue at Apelsvoll, where increasing Ca-Al in soil was found to be the main factor for reduced Sr uptake. The contribution of temperature, precipitation and plant development for Sr uptake was also emphasized due to the differences in weather and plant biomass at Apelsvoll and Fureneset. Results indicated that ^{84}Sr uptake in below soil plant tissue mainly was attributed to supporting soil components, though a foliage to potato tuber translocation at Fureneset could not be disregarded. The much higher uptake of Sr in potato tubers at Fureneset, compared to Apelsvoll, indicated a greater vulnerability to radioactive fallout containing divalent Sr cations than usually assumed for Fureneset, situated in a coastal area with sea spray.

Norsk sammendrag

Norsk tittel: Overføring og opptak av strontium i potetplanten, *Solanum tuberosum L.*, etter våtavsetning på blader og stilk: feltforsøk utført på Østlandet og på vestkysten av Norge.

^{90}Sr er en av de viktigste og mest utbredte radionuklidene fra atomfisjon. Radionukliden anses som en stor fare for mennesker hvis den blir spist og tatt opp i kroppen fordi den lange oppholdstiden i skjelettet kan føre til utvikling av kreft. Derfor er det ønskelig å forstå oppførselen til ^{90}Sr i ulike økosystem, inkludert overføring og opptak i matplanter, for å kunne implementere tiltak etter et potensielt radioaktivt nedfall av ^{90}Sr. Et kjent isotopforhold mellom $^{84}Sr/^{87}Sr$ ble i dette forsøket brukt til å følge overføring og opptak av Sr i potetplanten, *Solanum tuberosom L.*, etter våtavsetning på blad og stilk. Forsøksfelt ved to NIBIO-forskningsstasjoner: Apelsvoll, lokalisert på Østlandet, og Fureneset, lokalisert på vestkysten, ble brukt til å dyrke potetplanter og studere hvordan variasjon i klima og jordegenskaper kunne påvirke overføring av Sr. Målet med forsøkene var å (i) se om Sr-konsentrasjonen på løvverk ble redusert over tid, (ii) finne forskjeller i opptak av Sr på Apelsvoll og Fureneset, (iii) identifisere hvor i poteten Sr akkumuleres og (iv) bestemme om translokasjon fra løvverk til potet finner sted.

Blad og stilk på både Apelsvoll og Fureneset var signifikant ($p<0.05$) forurenset med ^{84}Sr etter våtavsetning. ^{84}Sr-konsentrasjonen på løvverk (blad+stilk) var nokså konstant på Apelsvoll gjennom forsøksperioden. Dette var i motsetning til på Fureneset, hvor reduksjonen ($p=0.052$) i løvverk viste seg å være negativt korrelert ($r=-0.95$) med store mengder nedbør. Opptaket av ^{84}Sr i stoloner, rothår og potet var kun signifikant på Fureneset. Her ble ^{84}Sr akkumulert i potetskallet. Det høye opptaket på Fureneset sammenlignet med Apelsvoll ble forklart med lav kationbyttekapasitet (CEC), plantetilgjengelig Ca (Ca-Al), plantetilgjengelig Sr (Sr-Al) og leirinnhold i jorda. De høyere verdiene for CEC, Ca-Al og leirinnhold på Apelsvoll reduserte opptaket av ^{84}Sr og den høye fraksjonen av Ca-Al ble antatt å være hovedårsaken til redusert opptak av ^{84}Sr. Effekten av temperatur, nedbør og vekststadium har også blitt tydeliggjort grunnet forskjeller i klima og plantebiomasse på de to stedene. Resultatene antydet at hovedopptaket av ^{84}Sr i stoloner, rothår og potet kom fra jorda. Dette til tross for at antagelsen om translokasjon fra blad til potet på Fureneset ikke kunne forkastes. Det høyere opptaket av ^{84}Sr i potet på Fureneset, sammenlignet med Apelsvoll, indikerte en større sårbarhet for radioaktivt nedfall av toverdige Sr-kationer enn først antatt ettersom Fureneset er lokalisert i et kystområde og utsatt for sjøsprøyt.

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1. Introduction

Food safety is a luxury most people in the developed countries of the world take for granted. However, the journey from seed to field and plate is long and vulnerable. Crops (and meat) are exposed to several possible stressors that could represent risks to humans from dietary intake. The Norwegian Food Safety Authority is a national governmental body ensuring that food and drinks consumed in Norway are as safe as possible (Mattilsynet, 2020). To assess, evaluate and regulate the food commercially available in stores and markets, research and testing are important. Information about possible toxicants and their sources, ecosystem transfer and behavior (e.g. in crops), as well as effect concentrations in humans (if consumed) is essential for advisors in important decision-making processes if contamination occurs (IAEA, 1994). The fallout of radionuclides in Norway after the Chernobyl accident (1986) was a reminder that Norway was not prepared for the given scenario (Miljødirektoratet, 2020). Since then, a nuclear preparedness plan has been established to reduce the impact of future potential nuclear fallout in Norway.

Nuclear accidents like Chernobyl, Ukraine (1986), and Fukushima, Japan (2011), are examples of why research and countermeasure implementation on radionuclides associated with nuclear fallout in agriculture are important. Norway is a country with large differences in meteorological, geographical and geological conditions, and radionuclide fallout may have varying effects depending on fallout location (CERAD, 2016). For instance, fallout from potential accidents at Sellafield, a nuclear reprocessing facility in England, is modelled to have a much greater impact in the Western Norway compared to the Eastern Norway, being most contaminated by the Chernobyl fallout (Ytre-Eide et al., 2009). Consequently, assessing impacts and risks for environmental protection and emergency preparedness is necessary. This can be challenging if the information on potential radioactive sources, ecosystem transfer and site-specific biological effects of relevant radionuclides is scarce, thus the uncertainties associated with prognostic impact and risk modelling could be unacceptably large (Salbu, 2016). To overcome these knowledge gaps and uncertainties, experiments investigating contaminant distribution, transfer in different plant species, effect of plant growth stage and contaminant solubility are essential (Pröhl et al., 2012). Müller et al. (2003) state that model parameters must be adjusted to radioecological regions, depending on agricultural properties and climate. Even when adjusted with respect to relevant regional generic parameters, the modelling output may largely differ from what is observed in field.

Site specific information is therefore crucial for reducing the overall uncertainties in ecosystem transfer models in a nuclear fallout context.

One of the most important products from nuclear fission is a long-lived (29 years) beta-emitting radioisotope of strontium, ^{90}Sr. The radioactive element is present in the environment due to fallout after nuclear weapons testing (1945-1980), nuclear accidents and nuclear installation releases (Dorsey et al., 2004; James et al., 2011). The focus on ^{90}Sr and daughter yttrium (^{90}Y) increased when research showed that ^{90}Sr deposited on vegetation could be transferred to humans, either directly through ingestion of agricultural produce or indirectly through ingestion of animal products (e.g. milk) (Larson & Ebner, 1958).

^{90}Sr is considered to be the major hazard for consumers through ingestion of agricultural produce because of its long half-life and because the chemical and physical characteristics are similar to that of calcium (Ca) (Middleton, 1958). This leads to a high retention in the skeleton of humans once ingested, and as ^{90}Sr in the skeleton irradiates surrounding cells, it increases the chance of diseases and cancer development (EPA, 2020; Libby, 1956). As Sr is absorbed as Ca in the human body, ^{90}Sr is often expressed in a strontium unit, S.U (previously called sunshine unit), stating the number of microcuries of ^{90}Sr present in one gram of Ca (Larson & Ebner, 1958; NOU, 1987:1). 1 S.U = 37 Bq/kg in the human skeleton, where 100 strontium units (3700 Bq/kg) previously was set as the maximum safe level after exposure to ^{90}Sr (Gyllenbok, 2018; Larson & Ebner, 1958). Today (2020), the Norwegian food intervention level for ^{90}Sr is 100 Bq/kg (Codex, 2011).

To reduce the risk of ^{90}Sr ingestion, it is of great interest to define and characterize the uptake and transfer of Sr in plants after fallout and direct wet deposition. Direct contamination is either 1) deposition on the crop surface (external), or 2) internal contamination where Sr is absorbed (via surfaces or the root system) and relocated within the plant (Baratta, 1994). The latter is of greatest importance for human intake and exposure as the Sr is absorbed inside the plant and cannot be removed by washing or peeling before ingestion (Fozzy, 1962).

The contaminant concentration in plants depends on radionuclide concentration in fallout and the amount of water in contact with the plant´s aerial organs (Colle et al., 2009). After radiostrontium fallout and deposition, plant surfaces will be exposed to Sr. Due to transport via pores, Sr may translocate to other parts of the plant (Ambler, 1964). Translocation is

especially important in plants with only one edible part, like potato plants, as the initial Sr deposition will not come in contact with potato tubers as they grow below the soil surface. Potato tubers can incorporate radioactive elements, and if ^{90}Sr enters the food chain, ^{90}Sr-contaminated potato tubers could represent a significant threat to human health. Consequently, it is important to understand uptake, transfer and retention of ^{90}Sr in plants to assess potential hazards related to human consumption: if the uptake in potato tubers is negligible it can be used as food, but if radiostrontium accumulate in potato tubers the intake should be reduced / avoided after a fallout scenario.

Due to the long half-life (29 years), ^{90}Sr is not permitted to be used as a tracer in outdoor field experiments. Instead, stable strontium can be used as an analog for experiments concerning radiostrontium, as radiostrontium and stable strontium have similar behavior (Burger & Lichtscheidl, 2019). The usage of isotopic labeling to study nutrient absorption, transport and mobility is a well-known approach (Bukovac & Wittwer, 1957). However, stable Sr cannot be used directly as a tracer due to the interference of natural Sr-isotopes. Alternatively, a low abundant isotope of Sr (e.g. ^{84}Sr) could be utilized. Creating a new Sr isotope ratio with a unique fingerprint differing from naturally occurring Sr will make it possible to separate the fingerprint from natural Sr background through an advanced analytical setup and mathematical corrections (Wiech et al., 2018).

In the present work, a known Sr-isotopic composition (^{84}Sr/^{87}Sr) has been used to follow transfer and uptake of Sr in potato plants in two different geographical and meteorological areas of Norway. As the aim of the experiment was to find trends of ^{90}Sr behavior in different agroecological regions, and did not simulate a site-specific fallout episode, the introduction of Sr to potato plants was identical. Similar experiments with Sr have previously been done, but mainly in controlled environments like laboratories or greenhouses. Experiments included strontium applied directly to the soil solution (Andersen, 1967; Menzel, 1954; Rediske & Selders, 1953; Roca & Vallejo, 1995), or direct deposition of Sr on foliage and possible translocation (Ambler, 1964; Bukovac & Wittwer, 1957; Middleton, 1958; Moorby & Squire, 1963). However, there is not much information reported from in field experiments, where Sr is deposited on foliage (mainly) and soil from a simulated precipitation event. This gives a more realistic picture of how fallout Sr could move in an agricultural ecosystem, depending on a series of site-specific factors (e.g. meteorology (rainfall), geography (deposition density), time of contamination, growth stage of plant, soil type and agricultural practices).

Data collected from the present experiments should be viewed as a trend indicator useful for modeling purposes.

2. Objectives and hypotheses

The overall goal of this thesis was to quantify transfer and uptake of ^{84}Sr from precipitation into potato plants, including potential translocation from foliage to potato tubers. By comparing results from two experimental sites situated in different geographical, meteorological and agricultural regions, an additional aim for the future is to use the results found in this thesis for more accurate modelling and risk estimation of radiostrontium transfer in different radioecological regions in Norway.

Transfer and uptake of strontium in potato plants was addressed by the following four hypotheses:

1. The ^{84}Sr concentration *on* foliage will decrease over time
    a. Absorbed ^{84}Sr *in* leaf will not be removed by wash off with water
2. Higher uptake of ^{84}Sr in the potato tubers and / or other below soil plant tissues (roots and stolons) grown at the inland site compared to coastal areas, due to less competition of stable Sr in the soil
    a. Areas in ocean proximity are exposed to sea spray incidents, thus the Sr concentrations in soil is high, competing with the uptake of ^{84}Sr
3. Accumulation of ^{84}Sr in potato peel is expected due to higher Ca-concentrations in the peel
4. Translocation from foliage to potato tubers is not to be expected
    a. Uptake of ^{84}Sr in potato tubers is most likely related to uptake from soil contaminated with ^{84}Sr

3. Background

3.1 Strontium

3.1.1 Physical, chemical and radiological properties

Stable strontium (Sr), atomic number 38, is a divalent alkaline earth element occurring naturally as four isotopes in the environment; three stable isotopes (^{84}Sr, ^{86}Sr, ^{88}Sr) and one radiogenic (^{87}Sr). The relative abundances for ^{84}Sr, ^{86}Sr, ^{87}Sr and ^{88}Sr are 0.56 %, 9.87 %, 7.04 % and 82.53 % (Figure 3.1), respectively (Burger & Lichtscheidl, 2019; Capo et al., 1998). Its oxidation states are 0 (metallic) and +2, where metallic Sr is very reactive in contact with air. Consequently, it is the divalent Sr that is present in the environment (Dorsey et al., 2004).

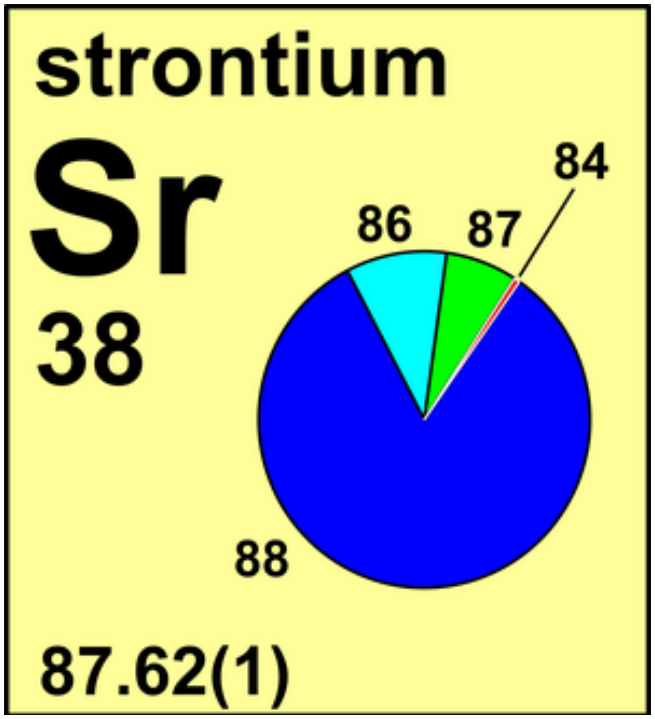

Figure 3.1: Natural abundance of the four stable Sr isotopes in the environment. Strontium is element 38 in the periodic table, with a standard atomic weight of 87.62 amu (CIAAW, 2020).

Several radioactive, not natural, isotopes of strontium are by-products from nuclear fission of ^{235}U, ^{238}U and ^{239}Pu. The isotopes range from ^{73}Sr to ^{107}Sr where the most significant isotopes are ^{85}Sr, ^{89}Sr and ^{90}Sr, with half-lives of 65 days, 51 days and 29 (28.79) years, respectively (Dorsey et al., 2004; Semenishchev & Voronina, 2019). This means it takes 65 days, 51 days and 29 years, respectively, before 50 % of the radioactive nuclei concentration has undergone

radioactive decay and changed into a new element. Most of the other radioactive isotopes have half-lives less than 10 hours (Semenishchev & Voronina, 2019). The radiogenic ^{87}Sr is stable in its ground state, while the exited state ^{87m}Sr is a gamma emitter produced from radioactive decay of ^{87}Rb ($^{87}Rb \rightarrow {}^{87}Sr$, β^-, $t_{1/2}$=48.4 Ga) (Knudson et al., 2010).

Through the release of beta particles, ^{90}Sr is formed from its radioactive precursor krypton-90 (^{90}Kr) (eq. 1) (Larson & Ebner, 1958). ^{90}Sr decays to radioactive yttrium-90 (^{90}Y), further decaying to stable zirconium-90 (^{90}Zr) (eq. 2). The long half-life of ^{90}Sr, as well as the beta radiation from its daughter ^{90}Y, makes ^{90}Sr the most hazardous of the radioactive Sr-elements.

^{90}Kr ($t_{1/2}$=33 sec) → ^{90}Rb ($t_{1/2}$=2.7 min) → ^{90}Sr eq. 1

^{90}Sr ($t_{½}$=29 yrs) → ^{90}Y ($t_{½}$=64 hrs) + β^- (0.546MeV) → ^{90}Zr (stable) + β^- (2.28 MeV) (99.98 %) eq. 2
^{90}Zr (stable) + β^- (0.523 MeV) + γ(1.75 MeV)

(Dorsey et al., 2004; Larson & Ebner, 1958).

The chemical properties of strontium are similar to the alkaline earth element calcium, e.g. close in radius (1.00 Å and 1.18 Å for Ca and Sr, respectively) and the same charge (+2), meaning they can substitute each other and attach to the same sites in the environment and in living organisms (Bowen & Dymond, 1955; Capo et al., 1998). Experiments with Ca is therefore a good indicator of Sr behavior, with Ca being a chemical homologue. However, transfer may not be identical as the concentration as well as movement mechanisms and pathways through the environment, e.g. in plants, may vary significantly (Bowen & Dymond, 1955; Busse & Palta, 2011; Handley et al., 1967; Smith, 1971).

3.1.2 Strontium isotope fractionation

Depending on mass, Sr-isotope fractionation may take place in nature due to thermodynamic properties and thereby influence the relative abundance of the isotopes (Urey, 1947 cited in Lewis et al., 2017). Swiss Alps plants incorporate approximately 0.3 ‰ more ^{86}Sr than ^{88}Sr, resulting in an $^{88/86}Sr$ ratio lower than the surrounding soil (de Souza et al., 2010). The accumulation of the lighter isotope in foliar organs was explained by Sr being fractionated during allocation. Furthermore, paleodietary studies use Sr-ratios for localization of ancient populations, and Sr-ratios is useful when dating rocks and sediments and determining trophic levels (Knudson et al., 2010). However, applying a stable Sr-ratio to follow absorption,

transport and mobility depends on one of two assumptions: (i) that stable strontium isotopes applied are not fractionated by biological processes or (ii) the isotope fractionation is predictable (Venkatraman, 2009). The insignificant fractionation in assumption (i) is based on the very small mass differences between Sr-isotopes compared to lighter isotopes like hydrogen ($^{1}H/^{2}H$), carbon ($^{12}C/^{13}C$) and nitrogen ($^{14}N/^{15}N$) (Lewis et al., 2017; Menzel, 1954). As a result, the isotope fractionation in applied isotope-ratio is considered negligible when studying the short-term mobility of Sr in biota (Capo et al., 1998; Dambrine et al., 1997; Knudson et al., 2010). This is in accordance with Flockhart et al. (2015), finding no fractionation of Sr between soil, plants and herbivores when testing monarch butterflies in a controlled greenhouse experiment. Furthermore, transfer factors and transfer rates of stable Sr and ^{90}Sr have been reported to be almost identical (Uchida et al., 2007). These findings could be due to the very small differences in uptake between the isotopes (permille (‰) level) and the slow equilibrium reactions (Lewis et al., 2017).

Using stable isotope ratios is a good method to trace uptake and transfer in living organisms. The benefits of adding a known isotopic ratio are several, e.g. no need for a control group, no issue with natural background concentrations or previously contaminated samples, no permittance needed to add radioactivity to the environment and no radioactive waste, and economically it is often cheaper to buy pure stable isotopes compared to radioactive tracers (Wiech et al., 2018; Wilschefski & Baxter, 2019). One problematic issue is that ascending background concentrations increase the measurement detection and quantification limits (Wiech et al., 2018). The challenge when working with stable isotope ratios is also to separate the added concentrations from the natural background concentrations. To do this, isotopic composition and site-specific variation (e.g. ratio in biota, soil and bedrock) must be determined (Capo et al., 1998).

3.1.3 Dry and wet deposition

Both stable and radioactive Sr are deposited as dry and wet deposition. Dry deposition is mainly particles affected by gravity (sedimentation) or mesoscale winds, with no water involved (Koranda & Robison, 1978). Wet deposition, with rainfall, is most important for ^{90}Sr deposition and was proven to be a significant source of ^{90}Sr transported by air following the Chernobyl accident (Hirose et al., 1993). This is because raindrops sweep up around 90 %

of the air volume, i.e. the rain-out processes will capture radioactive particles below the rain layer (Libby, 1956). Menzel et al. (1963) reported that rainfall is very important for fallout contamination as dry periods did not show accumulation of ^{90}Sr on plants. This despite elevated concentrations of ^{90}Sr measured in surrounding air. Frere et al. (1963) reported similar findings, stating that Sr in dry deposition was easily washed off plants compared to Sr from wet deposition. It has been assumed that 95 % of ^{90}Sr deposited far from the exploded Chernobyl reactor was available for uptake by biota (Koranda & Robison, 1978).

^{90}Sr is emitted as strontium oxide (SrO) and condensed onto particles after being released from nuclear sources (Koranda & Robison, 1978). Through reactions with moisture or CO_2 in the atmosphere it forms strontium hydroxide ($Sr(OH)_2$ or strontium carbonate ($SrCO_3$), respectively (Dorsey et al., 2004). $Sr(OH)_2$ will dissolve into Sr^{2+} and $SrOH^+$ when in contact with water, indicating that the main speciation of strontium after wet deposition will be free ions or $SrOH^+$, if not integrated in fuel particles. Operation Castle, in the U.S. in 1954, showed widespread fallout of particles containing ^{90}Sr more than 1.7 years after the initial nuclear bombing tests in March 1954 (Libby, 1956). If the fallout nuclides reached the stratosphere, the residence time was estimated to be 10 ± 5 years. Air concentration, particle solubility, drop size and precipitation rate influence the environmental distribution of strontium by wet deposition.

3.1.4 Soil characteristics and the effect on Sr behavior in soil

The cation exchange capacity (CEC), soil organic matter content (SOM) and the size distribution of the soil particles are important factors controlling if an element will sorb to available surfaces, bind as complexes and / or flow with water through pores. The particle size is important for physical properties like sorption and there are primarily three different size distributions in soil particles: sand (20 μm-2mm), silt (2 μm -20 μm) and clay (<2 μm) (Hu et al., 2011). Clay and organic matter have large surfaces that take part in sorption and / or exchange processes (vanLoon & Duffy, 2011). The CEC, a measure of cations sorbed to the soil surface, in the soil is affected by surrounding competing cationic elements in the soil water, their charge and concentration.

Strontium transformation in soils and sediment is affected by abiotic processes like sorption / desorption, redox conditions, ion exchange and complexation, as well as factors like organic matter, pH, temperature, ionic strength, solution speciation and biological organisms (Dorsey et al., 2004). The soil CEC is the dominating factor for strontium transformation and binding to surfaces in the short term. In the long run, the ions can become less exchangeable and can be sorbed onto sterically hindered, not exchangeable, sites. This was shown by Gastberger et al. (2000), where ^{90}Sr was so strongly bound in the soil matrix that it decayed before released by weathering.

The mobility of strontium in soils and sediments is relatively low, close to a source, as Sr could be associated with inert particles or be present as cations that could sorb to clay and metal oxides (AMAP, 2004; Dorsey et al., 2004). The inert particles with ^{90}Sr are a result of radioactive release from accidents and nuclear tests where ^{90}Sr have been integrated in particles made from uranium materials as carrier for fission, activation products and transuranics (Salbu, 2009). These fallout particles range from submicrons to fragments, where ^{90}Sr often has been present in inert fuel particles, reducing its mobility.

The migration rate in soil has been reported to be very low (4.2 mm/year with soil water percolation at 2500 mm/year in the Nagasaki area; 1.30 and 0.65 cm/year for wet and dry soil, respectively, in podzolic-gley soil in Belarus), and downward diffusion and migration of Sr between soil layers is often considered negligible (Arapis et al., 1997; Dambrine et al., 1997; Dorsey ey al., 2004; McHenry et al., 1956). This negligible vertical migration was seen in Chernobyl, where the top 10-20 centimeters of contaminated soil contained over 95 % of total ^{90}Sr (Ivanov et al., 1996 cited in Kashparov et al., 2001). The minor vertical migration was attributed to most of the ^{90}Sr being bound in fuel particles. Soils in the United States showed that all of ^{90}Sr after testing thermonuclear weapon in the 1950s was found in the top 12 centimeters of soil, with more than 50 % concentrated in the first 5 centimeters (Libby, 1956). As the inert fuel particles have started to weather over time, the deposited and previously immobilized ^{90}Sr, is becoming remobilized as divalent ^{90}Sr, being much more mobile in soil and biota (Salbu et al., 1994).

In contrast to weapon or accidental release of particles containing ^{90}Sr, the release from the reprocessing facility Sellafield, England, contained presumably ^{90}Sr cations (AMAP, 2004). Thus, ^{90}Sr was weakly bound to particle surfaces or complexes for long periods of time, and

was easily remobilized if conditions changed due to ion exchange processes (Wallace et al., 2012). High concentrations of iron, sulphates, carbonates and phosphates could transform Sr cations to insoluble compounds in soil and sediment through secondary sorption layers and precipitation (Rediske & Selders, 1957).

3.1.5 Environmental concentrations and human exposure to radioactive strontium

Stable strontium can be found everywhere in water, rocks, coal, plants, air, soil and oil, released from geological materials through weathering (Eisenbud, 1957). It has, on average, a concentration of 340-370 mg/kg in the Earth´s crust, and a soil concentration of 240 mg/kg (Capo et al., 1998; Dorsey et al., 2004). ^{90}Sr is also present almost everywhere due to global fallout from nuclear weapons tests (1945-1980), from nuclear accidents, and from authorized and unauthorized releases from nuclear installations (James et al., 2011).

Grains, dairy products and fresh vegetables are the three main dietary sources for ^{90}Sr, where the latter contribute to more than one third of a person´s yearly ^{90}Sr-intake (Dorsey et al., 2004). The concentration of ^{90}Sr in one kilogram fresh vegetables in the US is reported to be <0.3 Bq/kg, while the Norwegian Radiation and Nuclear Safety Authority report activity concentrations <LOD in Norwegian vegetables (Dorsey et al., 2004; Komperød et al., 2015). For Norwegian dairy products (milk, cheese, butter) and grains, the ^{90}Sr-concentration is reported to be 0.205 Bq/kg and <LOD, respectively (Komperød et al., 2015).

Ingestion is the main source of human exposure to radioactive strontium. The daily exposure is estimated to be 0.16 Bq/day, divided between food (96 %) and water (4 %) intake (Dorsey et al., 2004). Once ingested, water-soluble strontium dissolves and enters the bloodstream. It follows Ca behavior due to similar chemical and physical characteristics, accumulating on bone surface. 20-30 % of ingested ^{90}Sr is absorbed in the digestive tract, where 99 % of this absorbed fraction will deposit in the human skeleton (EPA, 2020). The elimination rate from the skeleton is low (Libby, 1956). Due to beta radiation from ^{90}Sr/^{90}Y situated in the bone marrow, negative effects such as reduced immune system and radiation sickness can occur, as well as increased probability of cancer (Wallace et al., 2012). This was seen in Russia, where releases of ^{90}Sr in the Techa River have been linked to increases in leukemia in the downstream population 5-20 years after the contamination occurred (Standring et al., 2009).

3.2 The potato plant

The potato plant, *Solanum tuberosum L.*, is the third most important crop globally (Patil et al., 2016). The potato tuber is a dominant ingredient in the diet of more than one billion people, and during the 1800s it was one of the main drivers for population growth in Norway (Sandvik, 2015). Since then, the Norwegian potato tuber consumption has decreased. Regular "food potatoes" have been reduced with over 75 % since the 1970s, while the processed fraction (fries, chips) has increased multiple times (Helsedirektoratet, 2019). In the 1950s, every Norwegian ate 92.5 kg "food potatoes" annually, compared to the 20.7 kg in 2018. Together with the processed potato tuber intake at 28.2 kg/year, every Norwegian eat a total of 49.1 kg potato tubers each year (Helsedirektoratet, 2019). As the potato tuber is a large contributor to the average Norwegian´s diet, information on the transfer of radiostrontium in the potato plant is important.

3.2.1 Potato plant development, growth and uptake of nutrients

The potato plant (Figure 3.2) normally reproduces vegetatively and is used as an annual herbaceous plant in agriculture, though naturally perennial (Struick, 2007).

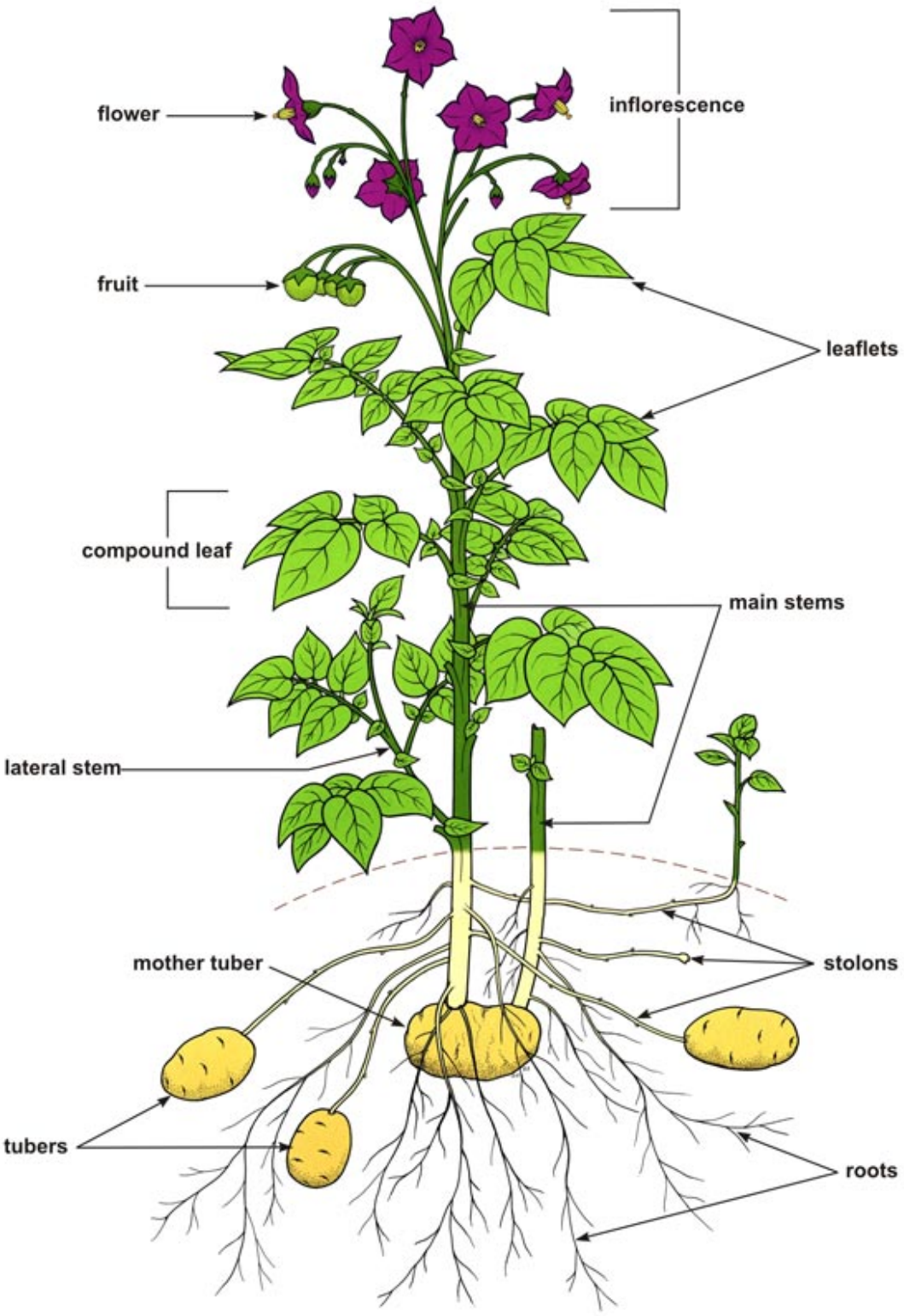

Figure 3.2: Illustration of the potato plant (Solanum tuberosum L.) (CIP, 2020).

The plant growth and development can roughly be divided into five stages, illustrated in Figure 3.3 (Patil et al., 2016). A more detailed overview of the phenological growth stages, based on BBCH-identification keys, is described by Hack et al. (1993) cited in Meier (2018). In **growth stage I** sprouts start to develop from old seed potato tubers, then roots develop and the plant grows upwards and out of the soil (Patil et al., 2016). The vegetative growth, such as leaves, stolons and roots begin to emerge in **growth stage II**. These first two stages last between 30-70 days depending on environmental factors and genotype. In **growth stage III** the potato tubers begin to initiate at stolon tips. This stage lasts for about two weeks, and the potato tubers do not enlarge during this time period. It is first at **growth stage IV** the potato tubers begin to bulk. The potato tuber cells accumulate water and nutrients. The uptake of nutrients is almost complete after this stage and very little is taken up during the final maturation **growth stage (V)** (Jackson & Haddock, 1959; Westermann, 2005). For nitrogen, almost two-thirds of the total nutrient requirement for the plant is taken up by the time potato tuber bulking begins, highlighting the importance of nutrient application timing for adequate growth (Horneck & Rosen, 2008). When the potato tubers are full grown, they detach from the stolons while developing new shoots, so called buds or eyes, that result in the next season´s yield (Patil et al., 2016).

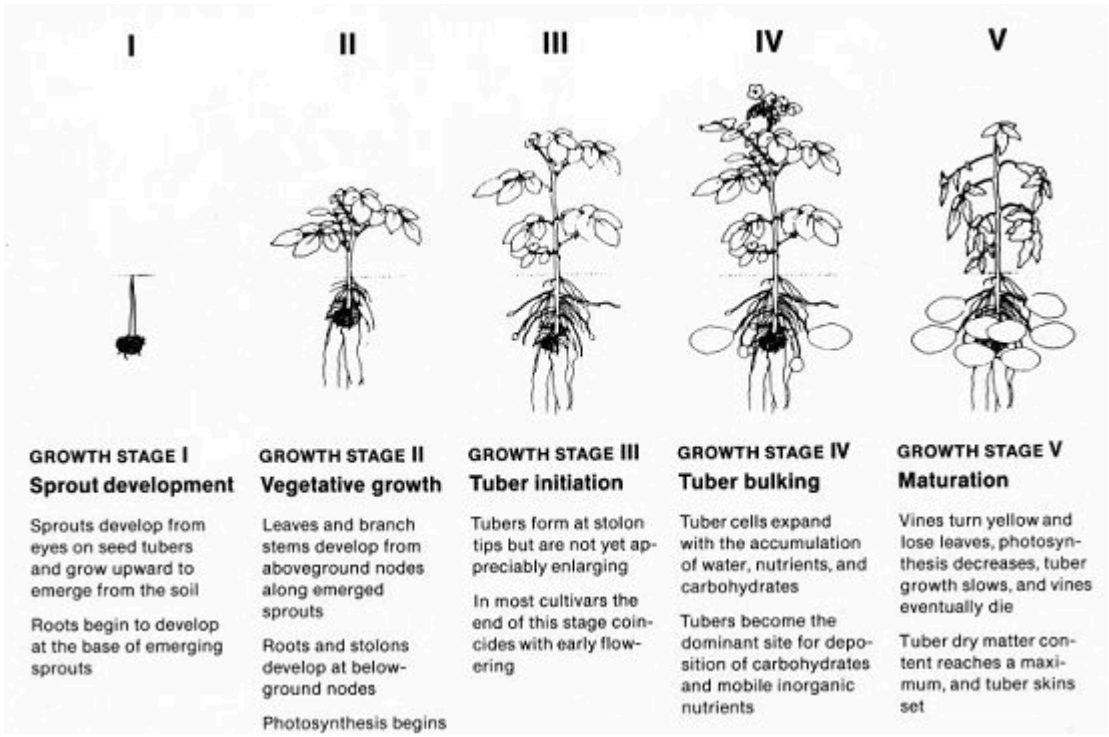

Figure 3.3: Illustration and description of the five growth stages for the potato plant (Patil et al., 2016).

Several stems are usually derived from one seed tuber, leading to a cluster of stems sharing and competing for resources like nutrients and light (Strucik, 2007). The stem is divided into main and lower stem, being above and below soil surface, respectively. The below-ground stems are often massive, compared to the hollow, triangle-shaped above-ground parts. As the main stem grows upwards, it holds the leaves, fruits and flowers. The leaves are irregularly odd pinnate and the plant has a rosette/semi-rosette habit (Patil et al., 2016). Three or four leaflets dominate each stem, with small ones in between (Struick, 2007). At the top of each plant there is one main leaf, being larger and deviating in form from the other leaves on the stem. The below ground basal stem nodes start spreading outwards as the plant grows and it is the source of stolons and roots. The stolons are the potato tubers´ rhizomes, starting to grow at the basal roots progressing upward on the lower stem (Plaisted, 1957; Struick, 2007). Potato tubers form from modified stems developing on the stolon, starting as a thickening on the stolon´s active apical bud (Figure 3.4) (Plaisted, 1957).

Figure 3.4: Development of a potato tuber from a stolon tip (left) to a potato tuber reaching maturity (right). Scale set to 1 cm. Photo taken at Fureneset, 26.05.2019.

The potato tubers form from cell division and radial cell enlargement, and tuberization is revealed by a reduction in the sugar metabolism as starch content and dry matter increase (Struick, 2007). The increase in size is primarily attributed to an increase in cell volume in the perimedullary region (Figure 3.5) (Peterson et al., 1985). The perimedullary region is cells that have developed differently from its originating stem tissue (pith) in the stolon. It is the cells in pith that transport and store nutrients throughout the plant. The potato tubers mainly get nutrients, like sugar, through internal phloem, stolons and tuber roots during potato tuber initiation (Peterson et al., 1985; Steckel & Gray, 1974 cited in Struick, 2007). The potato tuber development is influenced by environmental factors like precipitation, day length, soil fertility and temperature (Jackson & Haddock, 1956). A soil temperature of 16-19

°C is ideal for potato tuber initiation, and development is reduced with temperatures above 20 °C (Patil et al., 2016). Low temperatures generally result in a higher number of potato tubers per plant, while high temperatures generate larger potato tubers (Patil et al., 2016). Soil moisture over 65 % favors the greatest yields, often because a lower moisture percentage do not generate as many potato tuber sets (Patil et al., 2016).

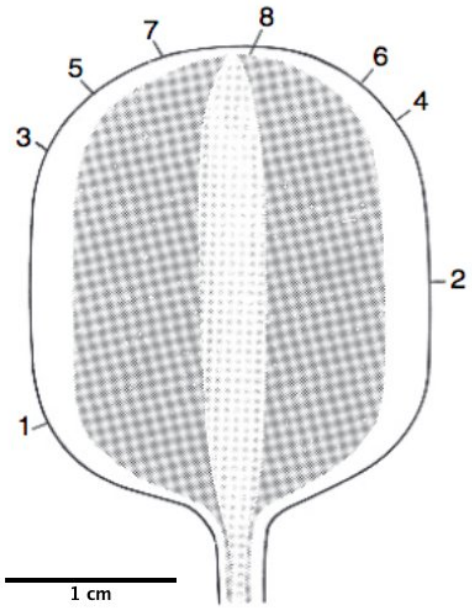

Figure 3.5: Illustration of radial cell enlargement and tuberization. The light shaded area in the middle is the pith, transporting and storing nutrients. The dark grey area surrounding it is the perimedullary region, containing the cells responsible for the increasing size of the potato tuber. The stolon attachment is at the bottom (Xu et al., 1998 cited in Struick, 2007, p. 240).

3.2.2 Transfer and uptake of strontium in plants

Uptake of nutrients

Potato tubers start to grow as tubers at the end of stolon roots. Through active and passive mechanisms, these tubers get nutrition from the aerial plant, roots and surrounding soil. The plant has two main transportation systems: xylem and phloem vessels (Westermann, 2005). The xylem vessel is a one-directional transportation system (upwards) mainly transporting water (with dissolved compounds) from roots to foliage. All nutrients are considered mobile in the xylem, and the transportation is an automatic, physical process. This in contrast to phloem transport which requires energy and is bidirectional. The phloem transports products from photosynthetic parts of the plant (leaves) to the non-photosynthesizing parts (roots) (Mohr & Schopfer, 1995). The products, like sugars (sucrose) and amino acids dissolved in

water, are transported for immediate use and storage in the below-soil plant tissue. The transportation is also referred to as translocation, internal transport from one part of the plant to another, and the element mobility is dependent on chemical characteristics of the respective element (Westermann, 2005).

Plant nutrients like nitrogen (N), phosphorus (P), sulphur (S) and potassium (K) are mobile, meaning they redistribute within the plant (Handley et al., 1967; Isermann, 1981; Westermann, 2005). Baker and Moorby (1969) demonstrated this by adding ^{32}P to a potato plant, showing how it relocated from the aerial plant to the stolon within 90 minutes. The same substantial movement of nutrients was seen for N, P and K in Russet Burbank potato tubers, where their concentrations decreased in the foliage while increasing in the potato tubers during growth stage IV (Jackson & Haddock, 1959). The alkaline earth elements (Ca, Ba, Sr) do not show the same mobility due to physiological reasons (Ambler, 1964; Bukovac & Wittwer, 1957; Isermann, 1981; Smith, 1971). Ca is an essential nutrient to plants, while Sr is not needed. However, as Ca is absorbed through the xylem vessel, Sr follows the same uptake pathway, explaining why Sr can be found in plant tissue (NCRP, 1984 cited in Watts & Howe, 2010).

Ca and Sr are transported by water (Palta, 2010). The main driver for Ca transportation in potato plants is transpiration (Colle et al., 2009). It is transported in the xylem vessels, and due to differences in water potentials in the plant, a concentration gradient is created where Ca is concentrated in the foliage (Win et al., 1999). The xylem connection is important for Ca transportation from roots to foliage and this transportation can be from all types of potato roots (i.e. root hair, stolons, tuber roots) (Busse & Palta, 2006). As both Ca and Sr are considered phloem immobile, a re-translocation through phloem transport would seldom occur (Bukovac & Wittwer, 1957; Kratzke & Palta, 1985).

Plant availability of Ca and Sr

The total concentration of ions in soil water can influence transfer of elements in plants. The concentration of other cations in soil solution strongly affects uptake of Sr, and the absorption of Ca^{2+} and Sr^{2+} occur relative to the plant available concentrations in soil solution (Isermann, 1981; McHenry et al., 1956; Menzel, 1954). As only the plant available fraction

of an element can be absorbed from soil solution to a plant, it is the fraction of mobile ^{90}Sr that is of interest for uptake (Korobova et al., 1998).

Plant available and labile Ca has been reported to be the main factor affecting root uptake and accumulation of radiostrontium in plants, where increasing plant available Ca (Ca-Al) concentrations result in small uptake of Sr due to competitive or antagonistic effects (Eisenbud, 1957; Fredriksson et al., 1970 cited in Lönsjö & Haak, 1975; Helal et al., 1997; Libby, 1956; Roca & Vallejo, 1995). Andersen (1967) indicated that plants are able to discriminate against Sr and favor Ca. The same Sr discrimination was found in humans (Eisenbud, 1957), possibly explained by Sr being 2.5 times heavier than Ca (Larson & Ebner, 1958). In equation 3, Menzel (1954) states how the ratio between Sr and Ca in plants is proportional to the available concentrations in soil, only affected by a distribution factor *k*. This distribution factor is an indicator of different release rates of elements in soil, mainly due to plant availability, root absorption and xylem and phloem transport. Watts & Howe (2010) emphasized this by stating that it was the relative concentration of Sr to Ca in soil water that determined uptake of Sr, and not the total Sr concentration in soil.

$$\frac{Sr\ in\ plant}{Ca\ in\ plant} = k\frac{available\ Sr\ in\ soil}{available\ Ca\ in\ soil} \qquad \text{eq. 3 (Menzel, 1954)}$$

Sr absorption takes place via the root system, but Sr is not retained *in* the root. Rediske and Selders (1957) showed that both dead and live roots accumulated 120 mikrogm/gm and 130 mikrogm/gm Sr, respectively, after being placed in a nutrient solution. This means that the Sr in roots is not a result of living tissue absorption, but rather a flocculation of Sr on the root surface. As radiostrontium accumulates in the surface soil, the root system of the potato plant is important for possible uptake. Yatazawa & Yamazaki (1957), cited in Frere et al., (1963), reported the following order for uptake of fission products from soil to be Leguminosae>Gramineae>Compositae>Solanaceae, where the potato plant belongs to the Solanaceae family. This indicates that uptake of Sr may be low, though reliability can be questioned as other studies did not find consistent differences between species (Nishita et al., 1960). The low uptake of Sr can be explained by the Sr speciation (particles or cations) or by a deeper root system for potato plants, showing increased growth and uptake of macronutrients like Ca after tillage (Nunes et al., 2006; Yaroson et al., 2019). This in contrast to shallow root systems showing a 70 % Sr-reduction after tillage (Milbourn et al., 1959).

Simultaneously, ploughing could lead to redistribution and availability of Sr bound to clay particles.

Translocation of Sr

Translocation describes how an element is internally redistributed to parts of the plant not in direct contact with the chemical substance. It happens after deposition on surfaces and absorption, and the two main factors affecting translocation are physiological behavior of an element and the growth stage of a plant (Colle et al., 2009). This includes, but is not limited by, humidity, age of leaves, temperature, physiological status of plant, plant growth stage and mobility of nutrient (Ambler, 1964; Kirchmann et al, 1966; Müller et al., 2003). Uptake and translocation pathways for Sr in a plant is illustrated in Figure 3.6. There is, at the moment (2020), no standardized experimental method for studying translocation.

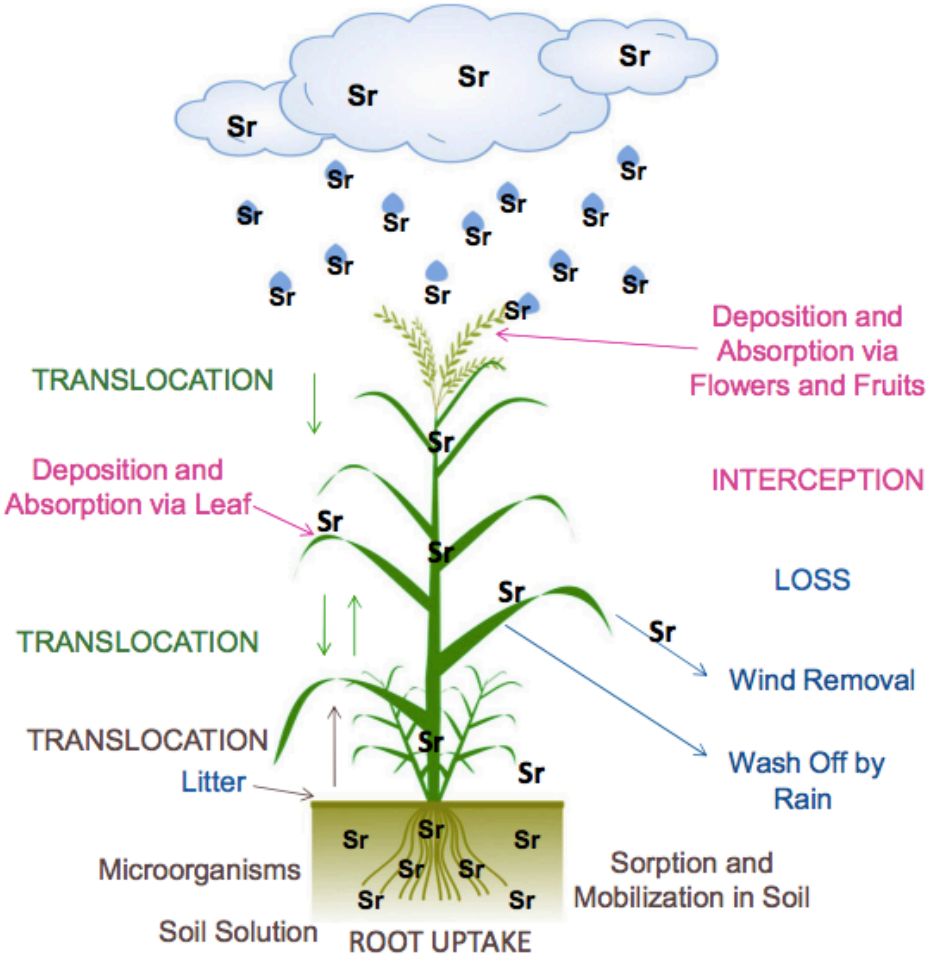

Figure 3.6: Illustration of the different uptake pathways for Sr in a plant after foliar wet deposition (Burger & Lichtscheidl, 2019).

After water-soluble fallout, Sr is expected to quickly absorb to leaf surfaces where the expected half-life is around 14 days due to weathering (Dorsey et al., 2004; Libby, 1956). The presence, and importance, of water for a limited ^{90}Sr translocation has previously been reported (Ambler, 1964; Handley et al., 1967; Kirchmann et al., 1966; Middleton, 1958). This is in accordance with Kirchmann et al. (1966) and Libby (1956) reporting that morning fog can enhance deposited ^{90}Sr concentrations in foliage as air humidity can facilitate ion penetration into the leaf as the cuticle changes, absorbing more ^{90}Sr than normal.

It is mainly the stolons and tuber roots that transport nutrients to the potato tuber. This was demonstrated by Busse & Palta (2006), who separated stolons from the soil and dipped them in Safranin O (red dye) and ^{45}Ca-containing water. Within a short period of time, the potato tubers showed increased levels of both (Figure 3.7). Additionally, there was no evidence of increased Ca-concentrations in the potato tubers when Ca was added to the main root system. Bamberg et al. (1993) reported that potato tubers have the highest Ca concentration in the peel. This concentration quickly decreased to nothing closer to the core, in accordance with Kratzke & Palta (1986). The high Ca concentrations in the periderm (peel) may be an explanation for the accumulation of red dye and ^{45}Ca in vascular tissue reported by Busse & Palta (2006). No evidence implied transportation of ^{45}Ca or red dye *across* the periderm, indicating that the periderm itself is not very penetrable (Busse & Palta, 2006). Barthakur et al. (2002) described similar findings, where the absorption of ^{90}Sr and ^{45}Ca stopped *in* the periderm. Hence, the cells within the periderm gladly adsorb and accumulate Ca and dye, leaving a sharp boundary marking the periderm (Figure 3.7) (Busse & Palta, 2006). These findings are in contrast to the significant ^{45}Ca diffusion through the periderm into the pith, reported by Habib & Donnelly (2002). Furthermore, Habib & Donnelly (2002) reported that basal roots contribute significantly to the potato tuber concentration of ^{45}Ca.

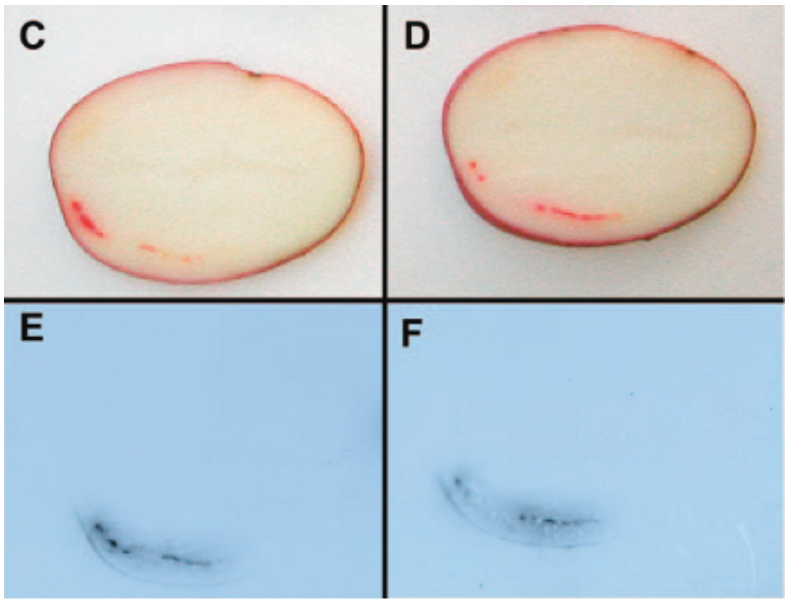

Figure 3.7: Distribution of the red dye Safranin O (C and D) and radioactive ^{45}Ca (E and F) in potato tubers after stolons were dipped in red dye and radioactive calcium (Busse & Palta, 2006).

4. Materials and methods

The experiment is part of the tracer field experiment project through CERAD, Center for Environmental Radioactivity, to study transfer of I-131 in grass, barely and potato plants. The field experiment with Sr was performed on potato plants only, looking to obtain information about transfer (sorption, uptake and translocation) in the plant after foliar wet deposition. The wet deposition simulated radioactive fallout, e.g. a nuclear accident or atomic bomb. Applying Sr to potato plants in natural field conditions gives a realistic introduction to the processes affecting Sr behavior in the given ecosystem. Plant tissues and soil were sampled and analyzed to assess how Sr is taken up and transferred in the plant, and how soil parameters and climate may affect this.

4.1 Experimental sites

The experiments took place in two climatically different Norwegian locations: Apelsvoll, Innlandet (60.7002º N 10.8695º E), and Fureneset, Vestlandet (61.2928º N 5.0443º E) (Figure 4.1). Both fields are run and maintained by NIBIO, the Norwegian Institute for Bioeconomy. One of the goals of the experiment was to compare transfer of Sr in different meteorological and agroecological areas of Norway to identify differences. Therefore, the site preparations and field management, e.g. fertilization, ploughing and potato cultivar (Table 1), were done as identically as possible to make differences in the climate and soil composition the main factors for different Sr-behavior.

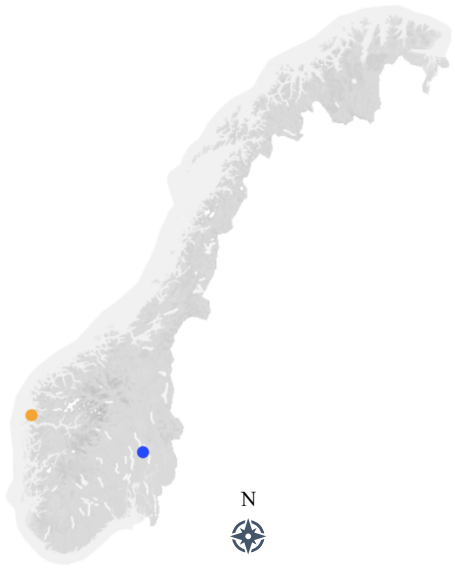

Figure 4.1: Geographical location of Apelsvoll (blue dot) and Fureneset (orange dot). ©kartverket/norgeskart.no

Apelsvoll represents the inland climate with large seasonal temperature differences, low precipitation rates and reduced wind speed. The low precipitation rates can lead to drought in the early summer months. Fureneset, located by the west coast, represents the Western part of Norway with its warm winters, heavy rain and much wind. This climate leads to strongly weathered soils often depleted in alkali and earth-alkali cations. The close proximity to the ocean is an important source of nutrient supply and it gives a constant $^{87}Sr/^{86}Sr$ ratio (0.7092) from the atmospheric precipitation (Dambrine et al., 1997; Knudson et al., 2010). General weather, precipitation and soil parameters for the two sites can be found in Table 1.

Table 1: Environmental parameters and field management for the potato field plots at Apelsvoll and Fureneset. Temperature (ºC) and weather are from the day Sr was sprayed on the plants: June 25th and June 26th 2019 for Apelsvoll and Fureneset, respectively. Data on annual precipitation, soil pH, soil classification and field management are given by the respective NIBIO research stations. ND = no data.

Parameter	Apelsvoll	Fureneset
Temperature*	16 ºC	14 ºC
Weather*	Cloudy, rain in the air, some wind	Cloudy, rain in the air, some wind
Meters above sea level	264 m.a.s.l.	7 m.a.s.l.
Precipitation, growth season (May-September)	319 mm	ND
Precipitation, yearly	600 mm	2010 mm
Soil	Silty loam	Loamy sand
Soil pH	6.0	5.8
Drift		
Potato cultivar	Arielle	
Date of sowing	10.05.2019	
Spraying	30.04.2019 Soil not sown earlier this growing season (2019)	
Potato hilling	14.06.2019	
Fertilizing Type Amount	10.05.2019 12-4-18 80 kg/da	

*the day of spraying

The plots were prepared and the potato tubers, (Arielle: early maturity, good initial development), were put in the soil May 10th, 2019. The potato plants were grown in a plot of 4 rows x 20 meters. At the time of Sr-deposition, the most mature potato tubers had reached growth stage IV.

4.1.1 Spraying

Simulating wet deposition, as fallout contaminants generally are deposited by rain, approximately nine liters of artificial rainwater was sprayed on the fields with estimated deposition of 0.776 mg $^{84}Sr/m^2$ (this equals 1 mg Sr/m^2). Sr was added as the mobile cation Sr^{2+}, though analysis of previous fallout of ^{90}Sr indicate that it could also be associated with inert particles after a nuclear fallout (AMAP, 2004; Dorsey et al., 2004). The ionic composition of the artificial rainwater is found in Table 2. It was made specifically for the two sites, mimicking actual precipitation data from the two sites in 2016 (Table C.2 in Appendix).

Table 2: Ionic composition of the artificial rainwater deposited on potato plants at Apelsvoll and Fureneset. The composition of main ions (cations) is based on a rainwater analysis from a pilot experiment in 2016 (Table C.2 in Appendix).

Site	Na (mg/L)	Mg (mg/L)	S (mg/L)	Ca (mg/L)	pH	Conductivity (µS/cm)
Apelsvoll	0.15	0.1	0.1	0.4	5.7	5
Fureneset	4.2	0.5	0.4	0.3	5.6	33

A Hardi BP 20 back pack sprayer (Figure E.1 in Appendix) with a boom of four nozzles was used to ensure homogeneous distribution while walking across the plot nine times, continually spraying nine liters of artificial rain water. Figure 4.2 visualizes how the nozzles distributed the spray evenly on the two middle rows, with reduced concentration on the outer two. The total area sprayed with artificial rainwater was 2 x 11 meters. Apelsvoll and Fureneset were sprayed in the morning of June 25th and 26th, respectively. The plants were still damp from the night before, and the cloudy weather ensured minimal evaporation.

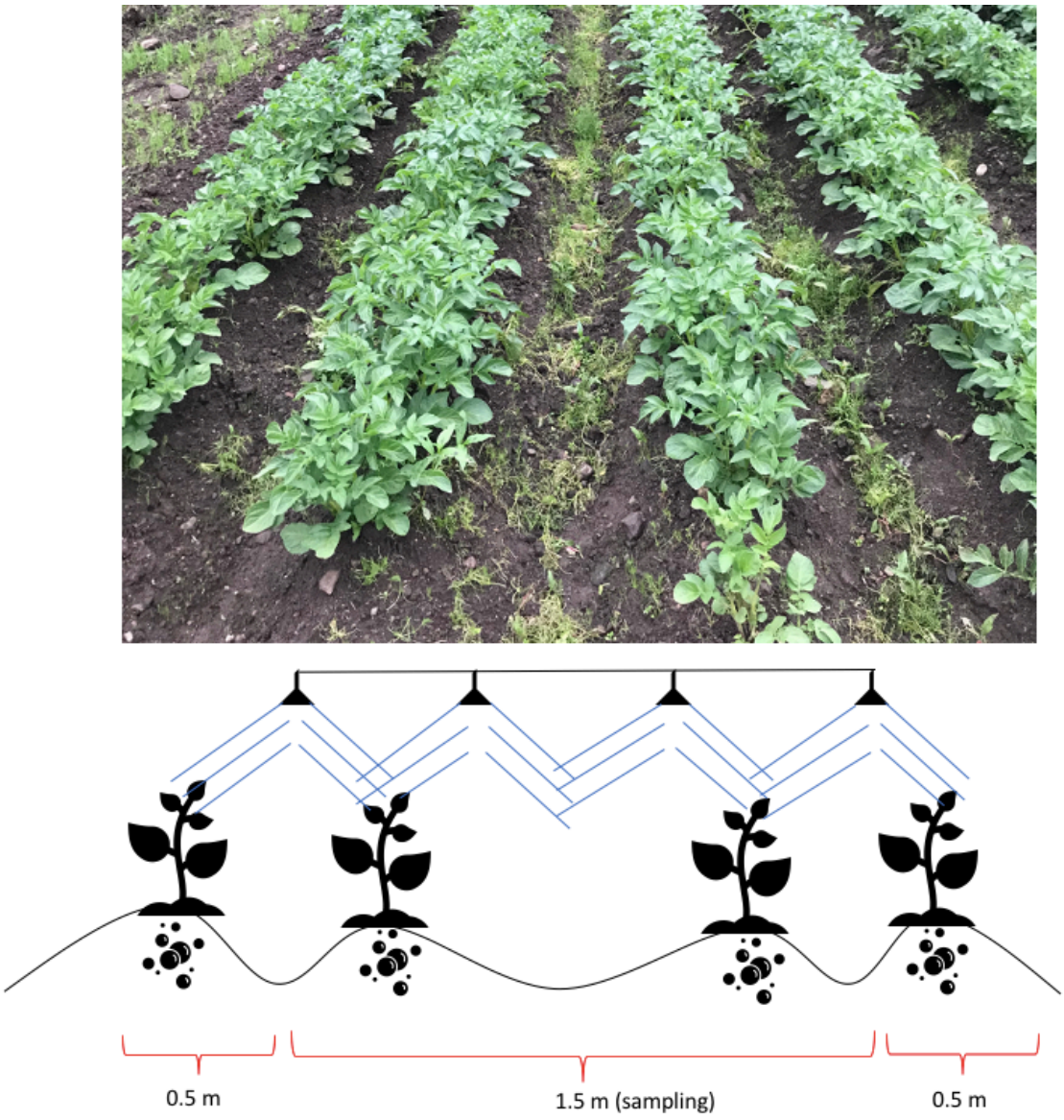

Figure 4.2: Photo (top) and illustration (bottom) of how the potato plants were planted and the distribution of the artificial rainwater by the four nozzles on the Hardi BP 20 back pack sprayer boom. The person spraying the plants walked up and down the middle row. Only the two rows in the middle (1.5m) were sampled for analysis. The photo is taken at Fureneset (26.06.2019), but the planting and deposition of rainwater were done identically at Apelsvoll.

4.1.2 Sampling

Control samples of soil, leaf, stem, stolons, root hair and potato tubers were taken the day before spraying (Table 3). Of soil, three different soil profiles were collected representing soil layers 0-5 cm, 5-10 cm and 10-15 cm. Leaves and stem were put directly into 20 mL plastic vials. Soil, stolons, root hair and potato tubers were put in plastic bags. After spraying, only samples from the two middle rows were taken to ensure homogeneous Sr distribution (Figure 4.2). Samples were collected after 3 hours, 48 hours, 1 week, 2 weeks, 3 weeks and 9 weeks

(in August). Three parallels of each plant tissue (Figure 4.3) and associated soil profile (Figure 4.4a) were sampled from three plants at each sampling: They all contained soil, leaf, stem, stolons, root hair and potato tubers except for the August sampling (Table 3). In August, the growth period was over, resulting in a sampling of potato tubers and soil only. Control samples in August were taken from the remaining 9 meters that were not sprayed. All potato tubers (⌀>1.5 cm) from one potato plant were collected in a plastic bag, while 20 mL containers / plastic bags of sample material from each tissue (leaf, stem, stolon, root hair, potato tuber (⌀<1.5 cm)) represent one replicate of plant tissue that was sampled in triplicate from each plant.

Table 3: Number of replicates collected for soil layers and plant tissues at each sampling time after spraying (3 hours, 48 hours, 1 week, 2 weeks, 3 weeks, 9 weeks). The three replicates represent three plants with associated soil. The August (nine week) sampling only contained soil and full-grown potato tubers (⌀>1.5 cm). Letters indicate sampling container and other details.

Plant tissue	Control	3 hours	48 hours	1 week	2 weeks	3 weeks	9 weeks
Soil 0-5 cm[a]	5	3	3	3	3	3	3[b]
Soil 5-10 cm[a]	5	3	3	3	3	3	3[b]
Soil 10-15cm[a]	5	3	3	3	3	3	3[b]
Leaf[c]	5	3	3	3	3	3	
Stem[c]	5	3	3	3	3	3	
Stolon[d]	5	3	3	3	3	3	
Root hair[d]	5	3	3	3	3	3	
Potato tuber[d, e]	5	3	3	3	3	3	3

[a] plastic bag as container, manual removal of larger fragments before placed in plastic vials (20 mL), [b] Apelsvoll did not separate the soil layers resulting in a total of three soil replicates (0-15 cm) at nine weeks, [c] 20 mL plastic vial as container, [d] plastic bag as container. Transferred to plastic vials after soil removal (washing) in the lab, [e] small potato tubers (⌀<1.5 cm) for all samplings except August (full-grown potato tubers (⌀>1.5 cm)).

Figure 4.3: Different plant tissues circled in separate colors as illustration. Leaf in red, stem in yellow, stolon in blue, root hair in orange and potato tuber (⌀<1.5 cm) in green. Photos taken at Fureneset, 26.06.2019.

The chance of cross-contamination when sampling was large. Trying to avoid this there was a focus on not touching the different parts of the plant while sampling. Soil samples were collected first. Leaves were cut with a scissor before uprooting the plant. Uprooting was done by only holding on to one part of the stem, while a second person used scissors to cut pieces of stem, stolons, root hair and potato tubers directly into plastic vials or bags. Soil profile samples were taken with a metal Auger soil sampler (⌀=5 cm), which was dried off between each sampling (Figure 4.4b). The soil samples were taken close to the plant being uprooted, as visualized in Figure 4.4a. A metal knife was used to separate the soil layers (0-5 cm, 5-10 cm, 10-15 cm).

Figure 4.4: a) Showing where the soil sample was taken in relation to the plant. b) An example of a soil column taken at Fureneset, before being split up into three different soil layers (0-5 cm, 5-10 cm, 10-15 cm). Photos taken at Fureneset, 26.06.2019.

The team from NMBU was present at the two first sampling times (control and three hours) at both sites. The remaining samples were collected by employees at NIBIO Apelsvoll and NIBIO Fureneset and shipped to NMBU throughout the summer.

4.1.3 Weather data

Weather data was collected from eklima.no, the database of the Norwegian Meteorological Institute. Temperature and precipitation were measured at the respective stations at Apelsvoll (station number 11500) and Fureneset (station number 56420), logged and transferred to a data base. Precipitation during the summer months was measured at ground level with a tipping bucket rain gauge (ARG 100 from EML, resolution ± 0.2mm) (Agrometeorology Norway, 2020). Temperature was measured at 2 m above ground level, with a HMP45A Vaisala combination sensor (accuracy ± 0.2 °C) or a PT500/PT100- sensor (accuracy ± 0.1 °C). The daily values used for temperature (TAM) is the arithmetic mean of the 24 hours. Wind was measured 2 meters above ground level with a Vector or Friedrics anemometer. The values represent the arithmetic mean for the first ten minutes after 00.00, 06.00, 12.00 and 18.00.

4.2 Laboratory sample preparation

The soil and plant tissues (after washing) were immediately dried after arrival at NMBU: soil and plants in the warm room (40 ºC) and freeze dryer (Christ Epsilon 2-4 LSC), respectively. Fresh and dry weight was obtained where necessary.

4.2.1 Preparation of soil samples

Analysis of soil properties like grain size, organic matter, plant available nutrients and pH were either performed in the lab at NMBU or by Eurofins. This to identify how these soil properties could influence the transfer and uptake of Sr in soil and plants.

Soil fractionation

The soil was fractionated (sand, silt, clay) in the three vertical layers from sampling: 0-5 cm, 5-10 cm and 10-15 cm. One subsample for each sampling time (3 hours, 48 hours, 1 week, 2 weeks, 3 weeks, n=5) were mixed and homogenized to represent each layer. This gave a total of nine replicates for each site (three replicates of sand, three of silt and three of clay). The fractionation procedure was done as described in 1-5:

1. The dry soils were sieved at 200 mm.
2. 10 g of sieved soil was weighed in 500 mL beakers and heated to 75-80 ºC on a sand bath with 20 mL distilled water and 15 mL H_2O_2 (5 mL at the time for three rounds) to remove organic matter. Solution was cooled down (20 ºC) before filtering the sand fraction through a 63 µm sieve. Sand fraction set to dry.
3. The clay and silt fraction left in solution was set to sediment for 6 hours. After 6 hours the two fractions were separated because silt sedimented while clay remained suspended in the water. The clay containing water was sucked out (into a 500 mL beaker) with a peristaltic pump. This procedure was repeated 4 times until water cleared, stirring the water and soil thoroughly before each sedimentation round. Silt fraction set to dry.
4. 2 mL 1 M NaCl was added to flocculate clay in the clay containing water (500 mL beaker). The clay fraction was set to sediment for 5 days before most of the overlaying water was sucked out. Clay fraction set to dry.

5. All samples (sand, silt, clay) were dried at 105 °C in the laboratory drying oven (Thermaks T1119). Dry weight was obtained and percentage distribution of all fractions in their respective layers were calculated.

Total element extraction in soil

Dried soil samples were manually sorted by removing stones and other fragments > 2 µm (Figure 4.5). 0.25 g of sieved soil (<2 µm) was transferred to acid-treated Teflon tubes, weighed, added 5 mL ultrapure nitric acid (HNO_3) and 2 mL deionized water before digestion.

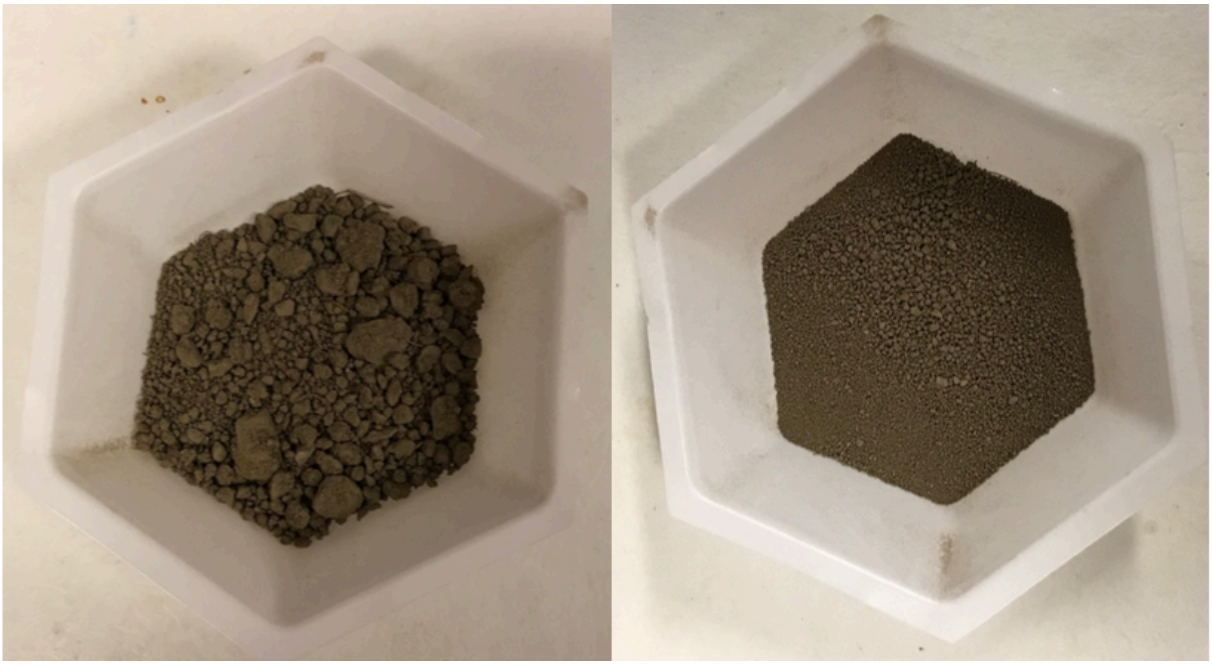

Figure 4.5: Dried soil directly from the 20 mL plastic vial (left) and after manual separation of larger fragments (right). Only the smaller fragments on the right were put in Teflon tubes and digested.

Plant available element extraction in soil

The AL-method described in Krogstad (2009) was used to determine the assumed plant available fraction of elements in the soil.

Dry and sieved soil (dried in dry room and sieved at 2 mm) was extracted with an AL-solution of 0.1 M ammonium lactate and 0.4 M acetic acid (pH=3.75). Soil:solution was 1:20. Sample preparation (1-4) was done by Senior Engineer Marit Nandrup Pettersen:

1. 2.0 g of air-dried soil (<2mm) was transferred to 50 mL plastic vials (PP, Sarstedt) and added 40 mL AL-solution.
2. The plastic vials were immediately put in a shaker. The vials were placed laying, in the oscillating direction, on the oscillating board with a speed of 100 shakings / minute for 90 minutes at 20 ºC.
3. Suspension was filtered through a 2 µm paper filter (Whatman 602 H, 125 mm). The filter was cleaned with 40 mL clean AL-extraction solution beforehand.
4. Extract was stored in 50 mL plastic vials (PP, Sarstedt) at 4 ºC until analysis.

The samples used to quantify the added ^{84}Sr in soil (^{84}Sr-Al) were diluted ten times (1+9) with distilled water before analysis.

The samples of plant available elements analyzed at Eurofins were prepared similar according to Krogstad (2009) and analyzed by the following standards:
Ca-Al; Mg-Al; K-Al; P-Al - DIN EN ISO 11885:2009:09
Na-Al - SS 028310:1995-12
Loss on ignition - EN 15935 (S33): 2012-11
pH - ISO 10390: 2005-12.

4.3 Preparation of plant tissue

All samples were digested before analysis on the ICP-MS (Agilent 8900 QQQ ICP-MS (Hachiōji, Japan)), following these sample preparations:

- Drying
- Homogenization (crushing, weighing, mixing)
- UltraCLAVE preparations
- Dilution
- Analysis

A test screening was done to get an overview and indication on where Sr accumulates. This to categorize and prioritize the correct samples, minimizing unnecessary laboratory work and economic costs. Soil, leaf, stem and potato tuber (⌀ <1.5 cm) samples from Apelsvoll after

three hours and three weeks, and August potato tubers (⌀ >1.5 cm) from Apelsvoll and Fureneset, were used for the screening. The samples used for the screening were prepared as described below.

Stolons, root hair and potato tubers were collected in plastic bags and thoroughly washed with water to remove soil before they were put in 20 mL laboratory plastic vials for drying and storage. Leaf and stem arrived in 20 mL plastic vials from the research stations and were dried in these. Three of the potato tubers (⌀<1.5 cm) from each outtake (control, 3 hours, 48 hours, 1 week, 2 weeks, 3 weeks), 18 in total, were separated into tuber core and tuber peel/rest (n=6 per outtake)(Figure 4.6). The same was done on 12 of the potato tubers (⌀>1.5 cm) (6 control and 6 treated, n=12) from the August sampling from Fureneset, based on results from the test screening. Figure 4.6 shows where the division between core and peel/rest is. The same line was present in the small potato tubers (⌀<1.5 cm).

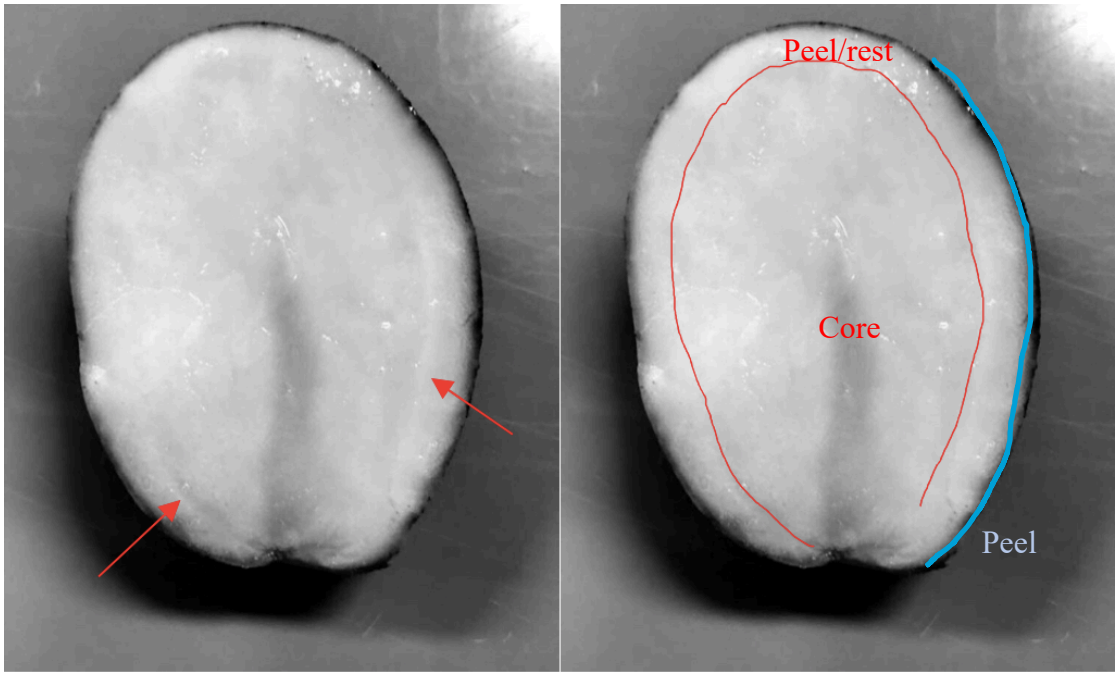

Figure 4.6: Red arrows and red line indicate the separation between core and peel/rest in a potato tuber. Blue line shows the thin outer layer representing the peel in the separate peel samples.

Separate peel samples (blue line in Figure 4.6) were taken of the potato tubers (⌀>1.5 cm) from August. 24 potato tubers from both Apelsvoll and Fureneset were peeled using a metal cheese slicer. Two potato tubers were made to represent one replicate, giving 12 replicates of peel in total for each site. The 12 replicates represent three control plants and three sprayed plants, as illustrated in Figure 4.7. The peeled August samples were visually divided into

small, medium and large (Figure E.2 in Appendix). After preparation, August samples of core, peel/rest and peel were put in plastic bags and dried in the freeze dryer. Gloves were used at all times to avoid contamination.

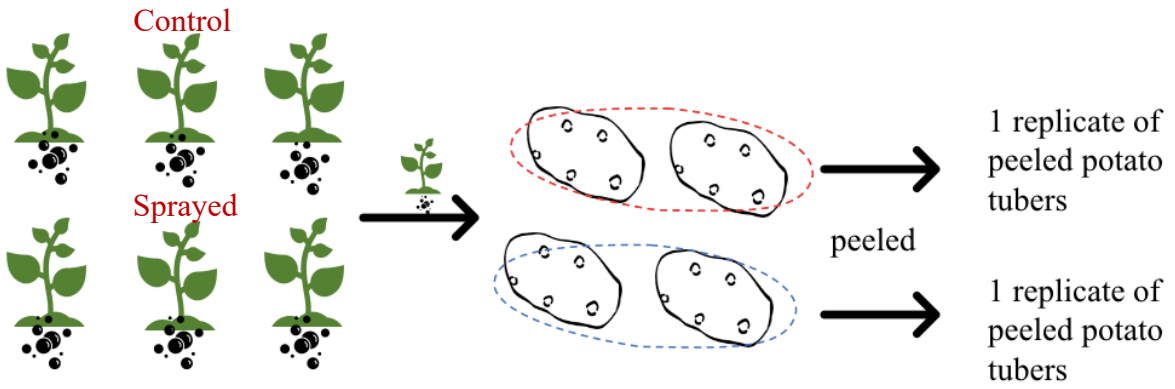

Figure 4.7: Illustration of how the 12 replicates of peeled August potato tubers represent 6 plants (3 control and 3 sprayed). Four potato tubers from each plant were divided in two groups, with two potato tubers in each group. Two potato tubers were peeled and dried together, making up one replicate of peeled potato tubers. Consequently, one potato plant had two replicates of peeled potato tubers. A total of 12 replicates represent all six potato plants (3 control and 3 sprayed) in one site (potato tuber outline: ©downloadclipart.net)

4.3.1 Homogenization

To ensure homogeneous samples, the freeze dried plant tissues (leaf, stem, stolon, root hair, potato tuber) were transferred from plastic vials to plastic bags, where the tissue was crushed with a mortar (Figure 4.8). Stem, stolons, root hair and peel were cut into small pieces with a scalpel when crushing was challenging. The crushed samples were mixed, by hand, to ensure homogenization inside the plastic bags.

Figure 4.8: Sample preparation and homogenization: a) freeze dried leaves from a plastic vial transferred to a plastic bag and crushed with a mortar, b) freeze dried potato core before and after crushing.

4.4 Digestion of samples

Samples were digested using ultraCLAVE (Milestone UltraCLAVE IV (Sorisole, Italy)). To end up with about the same concentration of ^{84}Sr in digested samples, the amount of sample material used varied between type of sample: leaf, stem, stolon, root hair (0.15-0.25 g dw), soil (0.25 g dw) and potato tuber (0.35-0.50 g dw). A metal spatula was used to move the sample powder from plastic bags to acid-treated Teflon tubes. To avoid cross-contamination, the metal spatula was cleaned with a paper towel between samples and plastic foil was used to cover the Teflon tubes throughout preparation to avoid contamination from lids.

After transfer to Teflon tubes, the respective samples, certified reference material (CRM) and blanks were added 5 mL ultrapure nitric acid (HNO_3) and 2 mL deionized water before digestion. The Teflon tubes (maximum 40 tubes per round) were placed in a load containing 15-20 mL H_2O_2, 5 mL HNO_3 and 350-400 mL deionized water, being heated and microwaved at a constant high temperature (260 ºC) and pressure (start pressure of 50 bar, increases to approximately 120 bar). A more detailed instrument description is found in Appendix A.1. After digestion, the samples were diluted to 50 mL with deionized water. All soil samples were further diluted ten times (1+9) with 10 % (V/V) nitric acid due to very high natural Sr concentrations.

4.5 Instrumental analysis

4.5.1 Quantification using ICP-OES

Elemental quantification of plant available elements in soil was performed using ICP-OES (Agilent 5110 VDV ICP-OES (Mulgrave, Australia)) in radial view by Senior Engineer Karl Andreas Jensen. The prepared soil solutions were not diluted before analysis. General principles of the instrument can be found in Appendix A.2.

4.5.2 Quantification using ICP-MS

Elemental quantification of Sr-isotopes in samples was performed using a triple quadrupole ICP-MS. The ICP-MS analyses were done by Senior Engineer Karl Andreas Jensen. Concentration of ^{84}Sr is a result of both natural concentration and addition by spraying. The added concentration by spraying is determined as ^{84}Sr-spike throughout the thesis.

It was only possible to quantify ^{84}Sr (and ^{87}Sr, used to calculate natural background concentrations of Sr) in plant tissue and soil with a triple quadrupole ICP-MS due to the two mass filtrations, making it possible to remove isobars. Figure 4.9 illustrates how the constituent elements in the sample were ionized when passing through the plasma source. Mass filter one (Q1) removed all masses different from 87 amu, leaving the isobars ^{87}Sr and ^{87}Rb as analyte. A mix of nitrous oxide, N_2O, and hydrogen gas, H_2, was then added to the reaction cell to generate polyatomic strontium hydroxide, $SrOH^+$ ($^{87}Sr^{16}O^1H^+$ = 104 amu). Through an RF-field the ions of interest ($^{87}Sr^{16}O^1H^+$) were directed towards the second mass filter (Q2), removing remaining interferences (amu ≠ 104) through a second mass filtration. Only the mass of interest, ^{87}Sr, was then, indirectly counted on the detector. The same procedure was done with ^{84}Sr ($^{84}Sr^{16}O^1H^+$ = 101 amu), where the isobar was ^{84}Kr (originating from the argon gas). A more detailed description of the instrument is described in Appendix A.3.

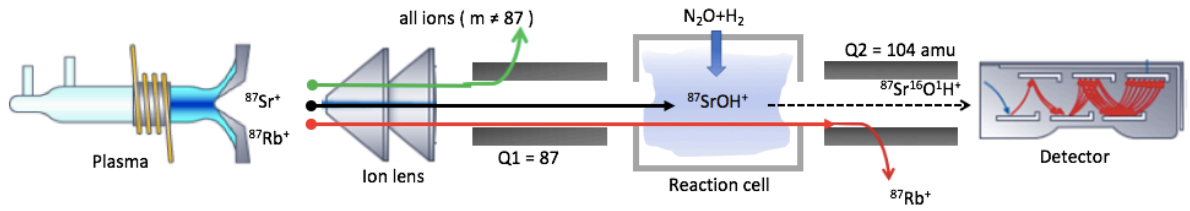

Figure 4.9: Illustration of how ^{87}Sr was quantified by mass filtration in a triple quadrupole ICP-MS. The same principles applied when quantifying ^{84}Sr (Karl Andreas Jensen, 2020. NMBU).

To ensure precise determination of Sr concentration, the pulse mode detector was used (pulse < 1.2 Mcps/s < analog), and all analog counts were diluted due to nonlinear calibration between the two modes. Based on screening of samples, samples were diluted by hand until pulse detector was reached (diluted 1, 2 or 3 times) or with argon gas (eight times online) for more precise measurements.

A total of 312 samples of plant tissues and 39 soil subsamples (+26 soil samples for plant available ^{84}Sr (^{84}Sr-Al)) were dried, homogenized, digested, diluted and quantified using ICP-MS for ^{84}Sr. Results are presented as concentration of ^{84}Sr per kilogram dry weight sample.

4.6 Mathematical calculations and isotope corrections

Karl Andreas Jensen, Senior Engineer at NMBU, created an equation for finding the added ^{84}Sr-spike, as shown in equations 7-14. The ^{84}Sr-spike could be estimated by calculating the natural Sr-background concentrations, and natural and spiked $^{84/87}$Sr-ratios. The two isotopes ^{84}Sr and ^{87}Sr were used because they have the lowest abundances naturally. Low abundance means lower concentrations and reduced counts, which is beneficial when using the pulse detector. The relative mass abundances for ^{84}Sr were 0.005 and 0.069 for natural and spiked, respectively. For ^{87}Sr, the natural and spiked mass abundances were 0.775 and 0.019 respectively. This gave a constant mass abundance of 0.006 for natural $^{84/87}$Sr-ratio (IRn_{84}). The $^{84/87}$Sr-ratio added in the artificial rainwater (IRs_{84}) had a mass abundance ratio of 3.63. The equations 7-14 were based on the natural ^{84}Sr/^{87}Sr ratio being identical in both sites due to negligible Sr-fractionation in plants (Capo et al., 1998; Dambrine et al., 1997; Knudson et al., 2010). A 1:1 uptake of Sr-isotopes was therefore assumed.

The calculated ratios use mass fractions instead of moles, as this also includes the mass bias in the mass spectrometer system (Wiech et al., 2018). Sample volumes (V) were identical and could be ignored (eq. 4), meaning concentration (C) equaled mass (m) (eq. 5). This resulted in a mass fraction (IR) that was determined from concentrations (eq. 6):

$$C_1 = \frac{m_1}{V_1} \quad , \; C_2 = \frac{m_2}{V_2} \quad \rightarrow \quad V_1 = V_2 \qquad \text{eq. 4}$$

$$C_1 = m_1 \quad , C_2 = m_2 \qquad \text{eq. 5}$$

$$IR = \frac{C_1}{C_2} = \frac{m_1}{m_2} \qquad \text{eq. 6}$$

The total concentration of all four natural Sr isotopes were defined as the sum of natural background (Cn) and enriched artificial rainwater (Cs):

$$C_{84} = Cn_{84} + Cs_{84} \qquad \text{eq. 7}$$

$$C_{86} = Cn_{86} + Cs_{86} \qquad \text{eq. 8}$$

$$C_{87} = Cn_{87} + Cs_{87} \qquad \text{eq. 9}$$

$$C_{88} = Cn_{88} + Cs_{88} \qquad \text{eq. 10}$$

Only C_{84} and C_{87} were needed to calculate the ^{84}Sr-spike concentration. The mass fraction isotopic ratios for natural (IR*n*) and spiked (IR*s*) strontium were:

$$IRn = \frac{Cn_{84}}{Cn_{87}} \quad \text{eq. 11}$$

$$IRs = \frac{Cs_{84}}{Cs_{87}} \quad \text{eq. 12}$$

Four unknowns (C_{84}, C_{87}, Cs_{84}, Cn_{87}) gave four equations. The equations were solved with software wxMaxima 16.12.0: http://andrejv.github.io/wxmaxima, where equations 13 and 14 calculated total background Sr and spiked ^{84}Sr, respectively. In Equation 13, the spiked ^{84}Sr-concentration is the only unknown, calculating the amount of added ^{84}Sr in plant tissue or soil, depending on total measured concentration (C_{84}, C_{87}) and isotopic ratios (IRn_{84}, IRs_{84}):

$$Cn_{87} = \frac{C_{84} - C_{87} IRs_{84}}{IRn_{84} - IRs_{84}} \quad \text{eq. 13}$$

$$Cs_{84} = -\frac{C_{84} IRs_{84} - C_{87} IRn_{84} IRs_{84}}{IRn_{84} - IRs_{84}} \quad \text{eq. 14}$$

Where:
- Cn_{84}/Cn_{87} is $^{84/87}$Sr-ratio of natural concentration of the isotopes
- C_{84}/C_{87} is $^{84/87}$Sr-ratio of total concentration of the isotopes (natural + spike)
- IRn_{84} is natural $^{84/87}$Sr-isotope ratio
- IRs_{84} is spiked $^{84/87}$Sr-isotope ratio
- Cs_{84} is spiked ^{84}Sr-concentration.

Isotope abundances for the four stable Sr-isotopes, both natural and spiked, can be found in Table 4. The different isotopic ratios of natural and spiked Sr resulted in an atomic weight of 87.6 and 84.7 amu, respectively. The mass abundances were used to calculate IR*s* and IR*n*.

Table 4: Isotope abundance of the four stable Sr-isotopes (^{84}Sr, ^{86}Sr, ^{87}Sr and ^{88}Sr) in natural environments and in the artificial rainwater deposited on potato plants at Apelsvoll and Fureneset.

	^{84}Sr	^{86}Sr	^{87}Sr	^{88}Sr
Isotope abundance, natural	0.56 %	9.86 %	7.0 %	82.58 %
Isotope abundance, spike*	78.3 %	4.34 %	1.91 %	15.45 %

(Burger & Lichtscheidl, 2019; *Neonest AB – Certificate of Analysis No. 190610-0081 (Figure B.1 in Appendix)).

4.7 Quality assurance

4.7.1 CRM, house standard and blanks

Certified reference materials (CRM) were used to control the accuracy of the method. The CRMs for plant tissue were apple leaf (NIST 1515), spinach leaves (NCS ZC73013; NIST 1570a) and tea (NCS ZC73014). The CRMs were adjusted for 97 % dry weight. For the plant available fraction in soil, house standards from round robin (A – sandy silt, B – organic rich soil) were used as reference material. The house standard, 1643H, was used to control systematical and/or random errors in the calibration of the instruments.
Blank samples (n=10) and CRMs (n=8) were prepared and analyzed like plant tissue and soil samples.

4.7.2 Calibration and internal standard

A three-point external calibration was used in this experiment, using a range of known analyte concentrations (5 µg/l, 50 µg/l and 500 µg/l) to generate a linear regression calibration curve. The standards used differed based on ^{84}Sr-concentration in given plant tissue, i.e. 500 µg/l for leaf, stem and soil, and 50 µg/l for potato tubers. Indium (In) was used as an internal standard (IS) because the ionization potentials for In and Sr are 5.79 eV and 5.69 eV, respectively. IS was added online to the instrument, adjusting for matrix and drift effects.

4.7.3 Limit of detection and limit of quantification

The limit of detection (LOD) and limit of quantification (LOQ) for spiked ^{84}Sr were calculated from control group concentrations, i.e. controls of leaf, stem, stolons, root hair and potato tubers after the ^{84}Sr-spike was quantified through isotope corrections (eq. 14). By applying eq. 14 on control samples of plant tissue, the ^{84}Sr-spike concentration would be close to zero. Method blanks were not subtracted from the control samples as all blanks were <LOQ for ^{84}Sr. The LOD and LOQ were calculated from average weight of all samples within each tissue separately.

LOD_{84Sr} = 3 x standard deviation of equation corrected control (eq.14) for given plant part
LOQ_{84Sr} = 10 x standard deviation of equation corrected control (eq.14) for given plant part

LOD and LOQ for natural background concentrations (e.g. Ca) and plant available concentrations in plant tissue and soil were calculated from method blanks.

LOD = 3 x standard deviation of method blanks
LOQ = 10 x standard deviation of method blanks

4.8 Statistical analysis

Statistical analysis were performed in Microsoft Office Excel 2019 (version 16.33 (20011301)) and Minitab 18. Diagrams were produced in Excel to visualize the data. A two-tailed, paired t-test was run in Excel to find significant differences between control groups and treated groups, and within treated groups in one site. Independent t-tests were used to find differences between Apelsvoll and Fureneset at specific sampling times. The hypotheses in the paired t-test were H_0: control = treated, with the alternative hypothesis H_1: control ≠ treated. Within treated groups (e.g. peel and core of treated potato tubers), the hypotheses were H_0: $treated_1 = treated_2$, with the alternative hypothesis H_1: $treated_1 \neq treated_2$. The hypotheses in the independent t-test were H_0: Apelsvoll = Fureneset and H_1: Apelsvoll ≠ Fureneset. The groups were defined as statistically different if $p<0.05$ (95 % confidence interval) unless otherwise is stated.

A student´s t-test can only be run on normally distributed data. To meet this criteria, quantified data was log transformed (log_{10}) before analysis. The log transformation normalized the data with skewness and kurtosis values between -2 and 2 (Table D.1.1 in Appendix) (West et al., 1995).

Pearson´s correlation test in Minitab was used to evaluate the strength of the linear relationship between different variables, e.g. concentrations of Ca and natural Sr in plant tissue. A correlation coefficient (r) with its p-value was obtained, and the correlation was defined as significant when $p<0.05$. Multiple regression was used to see which variables that could affect the dependent variable ^{84}Sr. The independent variables used in the test were concentrations of Ca, Ba, background Sr and time. Data was converted to log_{10} before correlation analysis.

To handle values below LOD and LOQ statistically, the numbers have been modified. Simple replacement has been used on values <LOD, using ½ of the detection limit (½LOD) (Croghan & Egeghy, 2003). For values <LOQ, the median between LOD and LOQ for the respective plant tissue and soil was used. The values used are presented in Table 5. Overall uncertainties with <LOD and <LOQ have been tested by running statistics (t-test) with LOD, LOD/2 and LOD/10 to see if the value replacing <LOD had a significant effect on the results. It did not. Results can be found in Table D.2.1 in Appendix.

Table 5: LOD and LOQ values for ^{84}Sr used in statistical analysis.

Plant tissue	Replacing <LOD for ^{84}Sr (µg/kg)	Replacing <LOQ for ^{84}Sr (µg/kg)
Soil	0.25	1.05
Leaf	2	8.5
Stem	1.5	7
Stolon	1	4.0
Root hair	5	23.5
Potato tuber	0.15	0.7
Core/peel (tubers)	0.2	0.8

Some replicates were considered to be excluded in final data or re-analyzed as some results show large standard deviations. Due to reduced lab access since March, the latter was challenging. After thorough consideration the removal of outliers was not done to show the big deviaitons and emphazise the possibly low reliability of the few replicates analyzed. The deviations are most likely not due to errors with sampling, lab preparations or measurements, but a result of random variation.

5. Results and discussion

The aim of these experiments was to investigate transfer and uptake of Sr in the potato plant (*Solanum tuberosum L.*) after single foliar wet deposition. Here, a short discussion on quality of sampling and analysis is reviewed first. Then, the general characteristics (climate and soil) are presented separately for Apelsvoll and Fureneset, followed by a comparison of the two sites. Soil concentrations of ^{84}Sr are presented before the focus is shifted to plants. The plant biomass differences in the two sites is emphasized before the background concentrations in plant tissue are presented separately, followed by a comparison of background concentrations at Apeslvoll and Fureneset. Then the ^{84}Sr concentrations in plant tissue are presented and discussed; separately at first, then together in a comparison. Lastly, uptake in potato tubers is discussed, followed by a discussion on whether or not the findings could be attributed to translocation.

The deposited Sr simulating ^{90}Sr is referred to as ^{84}Sr throughout the chapter. Background concentrations of Sr is referred to as tot-Sr (or tot-Sr-Al), calculated from ^{87}Sr (eq. 14, p. 39). In soil, only the plant available fraction of ^{84}Sr (^{84}Sr-Al) is presented.

5.1 Quality of sampling and analysis

The experimental fields at Apelsvoll and Fureneset have been utilized for similar experiments, using iodine tracer, the last four years. Thus, the field has been well characterized. Sampling took place from 24/25.06-30.08.2019 and the harvesting of soil and plant tissue was performed by trained scientists at the research farms according to a sampling protocol. Three separated plants were collected and was assumed to be representative of the experimental plot. The screening test and theory indicated that the peel was where Ca and Sr would concentrate in the potato tubers. Consequently, the peel has been a plant tissue of focus in the large potato tubers from the August sampling.

5.1.1 Limit of detection and limit of quantification

The detection and quantification limits for ^{84}Sr are presented in Table 6, where increasing background concentrations of tot-Sr increased LOQ (e. g. root hair). The LOD and LOQ values were the same for small (⌀<1.5 cm) and big (⌀>1.5 cm) potato tubers.

Table 6: Limits of detection (LOD), limits of quantification (LOQ), mean ± SD and range (min-max) for tot-Sr in control groups in soil and different plant tissues.

Plant tissue	LOD ^{84}Sr (µg/kg)	LOQ ^{84}Sr (µg/kg)	Tot-Sr in control group (mg/kg) (mean ± SD)	Range (min-max), tot-Sr control (mg/kg)	n
Soil	0.5	1.6	14.3 ± 3.19	9.12-19.8	30
Leaf	4	13	100.5 ± 39.86	42.1-145	6
Stem	3	11	112.8 ± 39.64	64.6-160	6
Stolon	2	6.1	50.1 ± 17.31	29.3-77.9	8
Root hair	10	37	206.4 ± 15.14	188-229	5
Potato tuber	0.3	1.1	25.2 ± 13.38	13.8-47.7	8
Core/peel (tuber)	0.4	1.2	8.8 ± 5.28	3.8-15.5	6

5.1.2 CRM and house standards

The relative error for the plant available elements in the soil house standard A is big (Table 7). House standard B shows low or no relative error compared to control soil, except for P. The CRMs and house standards used showed that Ca and Sr were within their respective reference values in plant tissue (Table 8). The relative accuracy for Ca and Sr was within 3 % variation of true value (except Sr in NCS ZC73014 with 13 % relative error), indicating high accuracy.

Table 7: Plant available fraction (indicated with -Al)(g/kg) for elements in control soil A (sandy silt) and control soil B (organic rich soil) quantified on ICP-OES. House standard A is the control soil A analyzed with plant available soil samples. House standard B is the control soil B analyzed with the plant available soil samples. Values are a mean ± SD.

Element	Control soil (g/kg)	n	House standard (g/kg)	n	Relative error (%)
Soil A – sandy silt					
Ca-Al	1.86 ± 0.09	20	1.71 ± 0.03	3	8.1
K-Al	0.08 ± 0.005	37	0.06 ± 0.001	3	25
Mg-Al	0.06 ± 0.003	27	0.07 ± 0.003	3	17
Na-Al	0.007 ± 0.003	10	- [a]	-	-
P-Al	0.10 ± 0.008	34	0.07 ± 0.002	3	30
Soil B – organic rich soil					
Ca-Al	3.7 ± 0.10	6	3.77 ± 0.01	3	1.6
K-Al	0.11 ± 0.007	9	0.11 ± 0	3	0
Mg-Al	0.31 ± 0.02	10	0.31 ± 0.001	3	0
Na-Al	0.02 ± 0.004	12	- [a]	-	-
P-Al	0.11 ± 0.005	11	0.0944 ± 0	3	14.2

[a] Na was not possible to quantify due to sample contamination in method.

Table 8: Measured concentrations (mg/kg) presented as mean ± SD (n=3) and certified concentrations (mg/kg) in certified reference material (CRM) used to control accuracy of method using ICP-MS. The reference value is the value given in the CRM.

Certified reference material	Sr in sample (mg/kg) Mean ± SD	Reference value Sr (mg/kg)	Ca in sample (mg/kg) Mean ± SD	Reference value Ca (mg/kg)
Apple leaves (NIST 1515)	25.7 ± 0.6	25 ± 2.0	15.7 ± 0.3	15.2 ± 0.2
Spinach leaves (NIST 1570a)	55 ± 1.2	55.6 ± 0.8	15.4 ± 0.5	15.2 ± 0.4
Tea (NCS ZC73014)	7.9 ± 0.2	9.1 ± 1.2	3.15 ± 0.066	3.2 ± 0.08
Spinach (NCS ZC73013)	84.3 ± 0.3	87 ± 5.0	6.8 ± 0.2	6.6 ± 0.3
House standard (1643 H)	0.332	0.32 ± 0.004	32.04	32.3 ± 1.1

5.1.3 Homogeneity of samples

Blank samples indicated no cross contamination as ^{84}Sr was not detected, and values for tot-Sr, Ca and Ba were <LOD. All control samples gave concentrations <LOD for ^{84}Sr in all plant tissues except stem and root hair at Fureneset (1 and 2 values <LOQ (n=5), respectively). Contamination was, however, not an issue of importance when analyzing ^{84}Sr and working with isotopic ratios as contamination has a natural background ratio different from the tracer, and will be subtracted (Wiech et al., 2018).

The homogeneity of the crushed samples was tested twice. Three parallels of the same leaf sample gave an ^{84}Sr concentration of 940, 930 and 910 µg/kg, giving a relative standard error (RSE) of 1.6 %. For tot-Sr, Ca and Ba, the relative standard errors were 1.4 %, 1.1 % and 2.7 % in the same three leaf samples, respectively. Two parallels of the same potato tuber sample gave 3.7 µg ^{84}Sr/kg and 4.2 µg ^{84}Sr/kg (RSE=8.8 %). Though the RSE for the potato tubers was >5 %, the leaf sample results indicated a good homogenization.

5.1.4 Sample matrix effects

The sample matrix effects were not found to be an issue when quantifying ^{84}Sr in plant tissue. The mass ratio of ^{84}Sr to ^{87}Sr may have been slightly adjusted due to the lower mass of ^{84}Sr, increasing the chance of ^{84}Sr to be preferentially removed due to the isotope effect. However, this effect was small enough to be adjusted for with calibration curves, internal standards and mathematical isotope corrections.

High natural background concentrations of tot-Sr, matrix interferences and space-charge effects in the instrument made quantification of ^{84}Sr in soil (samples containing all elements) challenging. Dilution was needed to get pulse mode on the detector, but when diluting 10 and 100 times, the expected $^{84}Sr/^{87}Sr$ ratio changed. This can be explained by dissolution using strong acids, dissolving soil minerals where fractionation of Sr-isotopes may be evident (Capo et al., 1998). Hence, as the background $^{84}Sr/^{87}Sr$ changed the isotope correction equations (eq. 7-14, p. 38) did not work as they are based on a constant ratio. The space charge effect may also be an explanation for the shift in $^{84}Sr/^{87}Sr$ ratio, as ^{84}Sr was repelled in the instrument due to lower mass than ^{87}Sr. Consequently, only the ^{84}Sr-Al fraction in soil is presented as no data on tot-^{84}Sr was obtained. The total concentration of Sr-Al quantified on

ICP-MS and ICP-OES showed a correlation of 98.3 %, emphasizing quantification accuracy on both instruments.

5.1.5 Field variability

The field variability is likely to have an effect on transfer and uptake of ^{84}Sr in the plant (Müller et al., 2003; Salbu, 2016). Soil parameters may vary within short distances and the fractionation of sand, silt and clay can be different within short distances and depths. For Apelsvoll and Fureneset, it is assumed that the top 20-30 centimeters are homogeneous due to regular ploughing. Hence, the sampling of different layers (0-5, 5-10, 10-15 cm) was done to follow vertical movement of ^{84}Sr in soil.

Strontium concentrations sprayed on the leaves were 1966 ± 306 µg/kg dw and 2233 ± 153 µg/kg dw for Apelsvoll and Fureneset, respectively, after three hours. The similar concentrations indicated that wet deposition of ^{84}Sr was homogeneously and equally distributed by spraying at both sites. Weather data presents a daily average of temperature and total precipitation throughout the day. The exact time (hour) of precipitation during the day throughout the experiments is not known, which would be an uncertainty factor as contact time between leaf and element is essential for sorption (Kirchmann et al., 1966).

5.1.6 Comments on laboratory work

To reduce sample contamination, de-ionized water, clean agate mortars, acid-washed Teflon tubes and plastic vials and bags were used to avoid material contamination, as suggested by Capo et al. (1998). Blanks were not prepared with each batch. ^{84}Sr was, however, not detected in the blanks prepared, indicating that this was not an issue.

The plant available fraction of elements in soil was determined by the method described in Krogstad (2009). This method is not specifically designed for Sr, but is based on the assumption that Sr and Ca are chemically similar, and that the method and results also should reflect the plant available fractions of Sr (Sr-Al) in soils.

For the ICP-MS, N_2O and H_2 was found to be the best gas mixture for removing isobar interferences, other polyatomic interferences and doubly charged interferences (e.g. ^{174}Y for ^{87}Sr and ^{168}Er for ^{84}Sr) (Karl Andreas Jensen, personal communication, March 2020). To separate the isobars (^{87}Sr and ^{87}Rb, ^{84}Sr and ^{84}Kr) in the ICP-MS, Sr-hydroxides was formed. Hydroxides was preferred to oxides as the response was improved and the chance of interferences reduced. The use of N_2O gas in the reaction cell ensured complete reaction with Sr due to its very low oxygen affinity, whereas Rb did not react. Pulse mode was desirable for this experiment due to its high precision when quantifying very small isotope ratios.

5.2 General characteristics in the experimental fields

5.2.1 Climate

Mean temperatures for the growth months May and June were 8 ± 4 ºC and 14 ± 3 ºC, respectively, at Apelsvoll. The temperature for the whole sampling period after wet deposition (24.06-30.08) was 16 ± 3 ºC (Figure 5.1). Precipitation from June 24th to August 30th added up to 101.9 mm (Figure 5.1), where average wind the first three weeks was 1.3 m/s (Appendix, Table C.1).

Fureneset showed mean temperatures of 8 ± 3 ºC and 13 ± 2 ºC for May and June, respectively. For the nine-week sampling period (25.06-30.08) an average temperature of 15 ± 3 ºC was found (Figure 5.1). The sampling period gave a precipitation total of 364 mm with a wind average of 1.7 m/s during the first three weeks after wet deposition/spraying.

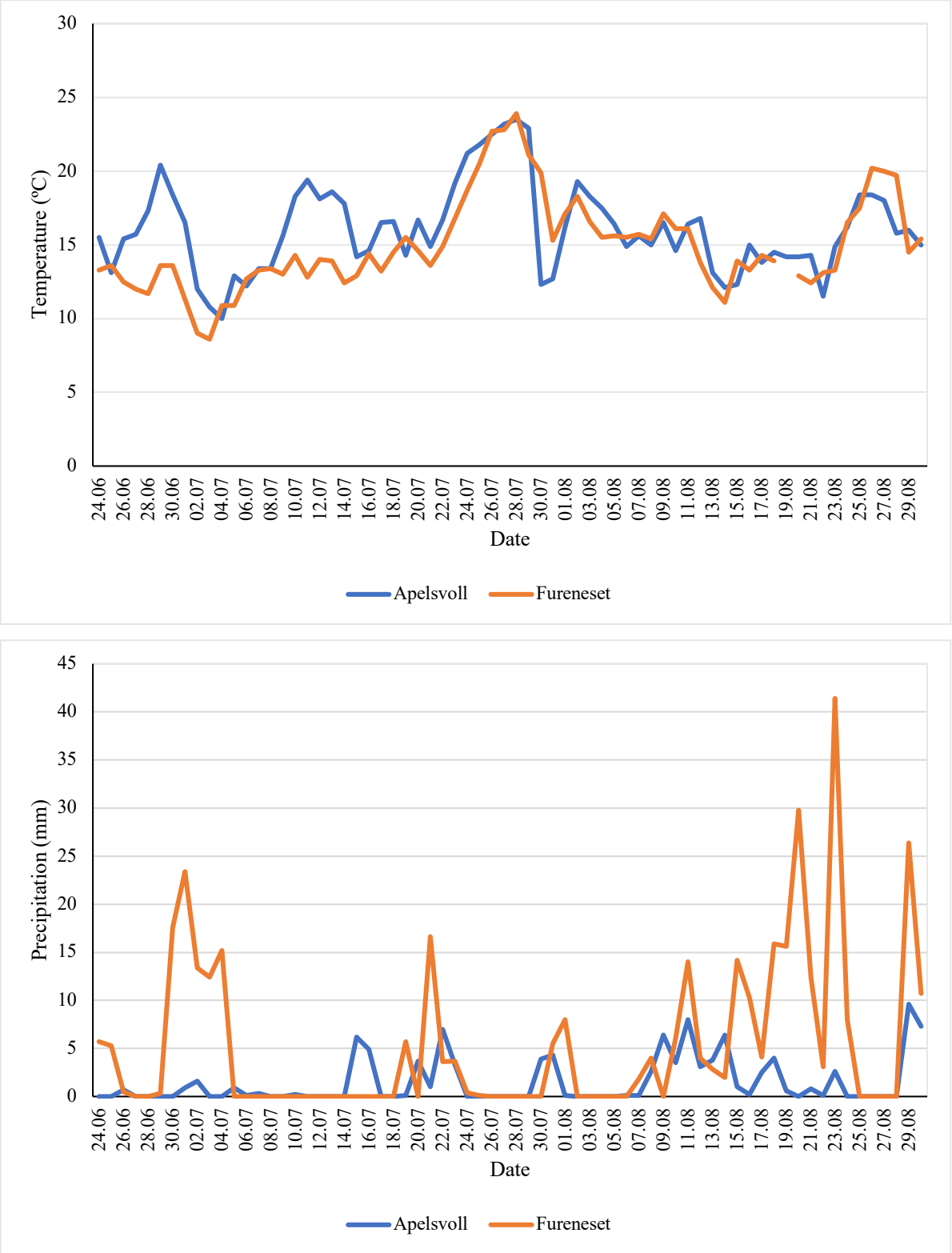

Figure 5.1: Temperature in celcius, °C, (top) and precipitation in millimeters, mm, (bottom) at Apelsvoll and Fureneset. The two parameters are presented measured from June 24th to August 30th 2019. The gap in temperature for Fureneset on August 19th is due to missing data.

The daily mean temperatures for the experimental period 24.06-30.08.2019 are significantly different (p=0.0003) at the two experimental sites, where Apelsvoll showed an overall higher temperature. Precipitation at Fureneset was 3.5 times higher than at Apelsvoll. High temperatures and increased air humidity could increase nutrient absorption in plants. Initial potato tuber development and growth is ideal at a given soil temperature (16-19 ºC), and temperatures above or below this may reduce potato tuber growth and number (Patil et al., 2016). Figure 5.1 shows only air temperatures, but as soil holds a more constant, and often higher, temperature than air due to the heat capacity (Zheng et al., 1993) it is likely that the growth conditions for the potato plants have been good in both sites.

5.2.2 Soil characteristics

The soil at Apelsvoll is classified as a silty loam by NIBIO, in accordance with the obtained results for sand, silt and clay in Table 9 (raw data on fractionation is found in

Table G.*1* in Appendix). The clay content of 12 % may indicate a high CEC, in accordance with high concentrations of the four most important plant available cations Ca, Mg, K and Na (vanLoon & Duffy, 2011). The CEC was not analyzed for the soil at Apelsvoll, but previous measurements reported a CEC of 11.2 ± 2.7 cmol+/kg for the same site (Korsaeth, 2005). This is a modest level, which could partly be explained by the low soil organic matter content (Table 9). Using the reported CEC of 11.2 ± 2.7 cmol+/kg gave a base saturation of 16 %. The total concentration of Sr, Ca and Ba in soil at Apelsvoll was 0.05 ± 0.02 g/kg, 3.6 ± 0.9 g/kg and 0.50 ± 0.07 g/kg, respectively (Table 9). The plant available fraction of Sr (Sr-Al) and Ca (Ca-Al) made up 32 % and 42 %, respectively.

The soil at Fureneset is a sandy loam with high soil organic matter, 10 %, indicating surface soil from a high productivity field (vanLoon & Duffy, 2011). The CEC of 6.2 cmol+/kg in the soil at Fureneset is, however, low. This is in accordance with the low content of clay and low concentration of the exchangeable cations Ca-Al and Mg-Al giving a base saturation of 11 %. The total concentrations of Sr, Ca and Ba were 0.35 ± 0.02 g/kg, 19 ± 1 g/kg and 0.09 ± 0.01 g/kg, respectively, where the plant available fraction was 3 % for both Sr-Al and Ca-Al (Table 9).

Table 9: Information on soil characteristics at Apelsvoll and Fureneset. Soil type, pH, CEC (cation exchange capacity), SOM (soil organic matter) and nitrate (NO_3, NH_4) are analyzed by Eurofins. Percentage distribution of sand, silt and clay is based on the average of three replicates for each layer (0-5, 5-10, 10-15 cm) (n=3). Plant available elements (indicated with -Al) are analyzed by NMBU and Eurofins. Eurofins´analysis at Apelsvoll represents one sample from 2018, from a field 10 meters north of the experimental site. NA - no analysis.

Parameter	Apelsvoll				Fureneset			
Soil type	Silty loam				Loamy sand			
pH	5.5				5.7			
CEC	NA				6.2 cmol+/kg			
SOM	6 %				10.10 % ± 0.009			
Sand (%)	57 %				66 %			
Silt (%)	31 %				31 %			
Clay (%)	12 %				3 %			
Element	NMBU	n	Eurofins	n	NMBU	n	Eurofins	n
Tot-Sr (g/kg)	0.05 ± 0.02	12	NA		0.35 ± 0.02	28	NA	
Sr-Al (g/kg)	0.016 ± 0.001	27	NA		0.010 ± 0.002	27	NA	
Tot-Ca (g/kg)	3.6 ± 0.9	12	NA		19 ± 1	28	NA	
Ca-Al (g/kg)	1.5 ± 0.3	27	1.3	1	0.5 ± 0.2	27	0.62 ± 0.03	3
Tot-Ba (g/kg)	0.50 ± 0.07	12	NA		0.09 ± 0.01	28	NA	
K-Al (g/kg)	0.11 ± 0.02	27	0.16	1	0.11 ± 0.07	27	0.14 ± 0.04	3
Mg-Al (g/kg)	0.102 ± 0.008	27	0.12	1	0.04 ± 0.01	27	0.048 ± 0.002	3
Al-Al (g/kg)	0.29 ± 0.02	27	NA		0.94 ± 0.07	27	NA	
Na-Al (g/kg)	NA[a]		0.027	1	NA[a]		0.061 ± 0.002	3
NH_4-N (g/kg)	NA		1.73[b]	1	NA		NA	
NO_3-N (g/kg)	NA		2.85[b]	1	NA		NA	
P-Al (g/kg)	0.05 ± 0.01	27	0.069	1	0.07 ± 0.02	27	0.09 ± 0.02	3

[a] Not possible to quantify due to Na-contamination in method, [b] the nitrogen samples were taken in the area in 2019.

The CEC was low in both sites. The difference between 11.2 cmol+/kg and 6.2 cmol+/kg is sufficiently large to give distinct soil characteristics (Krogstad, T, personal communication, April 2020), mainly attributed to high clay content and exchangeable cations at Apelsvoll. Tot-Sr and tot-Ca were higher at Fureneset compared to Apelsvoll, though plant available Sr-Al and Ca-Al were significantly higher at Apelsvoll ($p=5.3*10^{-10}$ and $p=1.8*10^{-18}$ for Sr-Al and Ca-Al, respectively). The difference between tot-Sr and Sr-Al in soil at the two sites was large, where the plant available Sr-Al at Apelsvoll contributed 32 % of tot-Sr in soil, compared to 3 % at Fureneset. Ca-Al made up 42 % and 3 % of tot-Ca at Apelsvoll and Fureneset, respectively.

The very low fraction of tot-Sr as Sr-Al at Fureneset was unexpected due to the ocean proximity. Sea spray elements are believed to be plant available, and Andersson et al. (1990) concluded that 90 % of Sr on snow near the coast in Scandinavia came from the Atlantic

Ocean. This led to the assumption that Sr-Al would be very high at Fureneset, as tot-Sr was more than 6 times higher than at Apelsvoll. Furthermore, the high concentrations of tot-Sr and tot-Ca, together with the low $^{87}Sr/^{86}Sr$ ratio (0.717 ± 0.002, n=27) in soil indicated a marine Sr source ($^{87}Sr/^{86}Sr \sim 0.7092$) (Frei & Frei, 2013). However, the precipitation analysis for Fureneset showed no elevated concentrations of Sr compared to Apelsvoll (Table C.2 in Appendix). Weather during the precipitation sampling is unknown, though low winds and good weather may be an explanation for the low Sr concentrations as sea spray would especially occur during storm events in the autumn and winter months.

The reason for low Sr-Al and Ca-Al at Fureneset can be explained by its soil minerals / bedrock as Fureneset is dominated by Precambrian gneiss, containing minerals like quartz, biotite and plagioclase, where the latter is rich in Ca. Quartz and plagioclase (feldspar) have a low CEC range of 1-2 cmol+/kg, further explaining reduced CEC at Fureneset (vanLoon & Duffy, 2011). Three thoroughly examined sites ~60 km east of Fureneset showed a mineralogy containing 30 % plagioclase (rich in Ca), strengthening the theory that Sr and Ca are incorporated in the mineral structure (Lund, 1999). The bedrock also contains biotite, with rubidium-87 (the mother of ^{87}Sr), incorporated in the structure and being unavailable for plants. The proximity to the sea, with possible marine sediments precipitated with inorganic Ca, can also be of importance (Capo et al., 1998). High Ba-concentrations at Apelsvoll could be explained by the calcareous Cambro-Silurian sediments often rich in Ba (Bjørlykke & Griffin, 1973 cited in Nordrum et al., 2003).

The higher clay content and CEC at Apelsvoll indicated a high potential for adsorption / absorption to clay and competition from other divalent cations in the soil. The presence of relatively mobile divalent cation metals would increase the potential for ion exchange, and interfere with uptake. The opposite was true for Fureneset, with low clay content and low CEC. The potential for sorption of mobile cations, and the competition of other base cations, is consequently reduced, while uptake should be promoted.

5.3 ^{84}Sr in soil at Apelsvoll and Fureneset

There were traces of plant available ^{84}Sr (^{84}Sr-Al) in the topsoil (0-5 cm) in both sites (Table 10). For Apelsvoll, a significant difference (p=0.002) was found between control group and treated group in the top layer. No significant concentrations were detected in the two bottom layers (5-10 cm, 10-15 cm), indicating soil deposition of ^{84}Sr after spraying but no vertical movement of ^{84}Sr-Al. Fureneset also showed a significant difference between control group and treated group (p=0.0006), with no vertical movement. The results indicated that ^{84}Sr-Al found in soil was deposited on topsoil directly after wet deposition, or later as wash off by rain, as suggested by Isermann (1981) and Moorby & Squire (1963).

Table 10: Plant available ^{84}Sr-Al (µg/kg) and tot-Sr-Al (mg/kg) in soil at Apelsvoll and Fureneset three weeks after wet deposition of ^{84}Sr. The values are a mean ± SD, n=4 [1].

Soil layer	^{84}Sr-Al (µg/kg)		Tot-Sr-Al (mg/kg)	
	Apelsvoll	Fureneset	Apelsvoll	Fureneset
Soil, 0-5 cm	4 ± 1	1.5 ± 0.5	17.2 ± 0.4	7.3 ± 0.2
Soil, 5-10 cm	<LOQ	<LOD	16.7 ± 0.8	7.9 ± 0.5
Soil, 10-15 cm	<LOQ	<LOD	17 ± 1	9 ± 1

Table 10 shows the concentrations of plant available ^{84}Sr-Al and tot-Sr-Al in soils three weeks after deposition. If ^{84}Sr was removed from foliage to soil with water, one would assume the highest concentration of ^{84}Sr in soil at Fureneset due to more precipitation over the three weeks (Apelsvoll and Fureneset with 15.8 mm and 82.8 mm, respectively, in the three weeks after spraying). Values in Table 10 shows that this was not the case, and the ^{84}Sr-Al concentration in surface soil at Apelsvoll was significantly higher than at Fureneset (p=0.01). The ratios of ^{84}Sr-Al:tot-Sr-Al and ^{84}Sr-Al:Ca-Al were not significantly different between the two sites (Table 11). The concentration of ^{84}Sr-Al in topsoil made up 0.02 % of tot-Sr in topsoil at both sites, and 2.3 % and 2.1 % of tot-Sr-Al for Apelsvoll and Fureneset, respectively.

[1] The values in Table 10 represent 12 of the 27 soil samples analyzed (at each site) for plant available elements in Table 9. The concentrations of tot-Sr-Al in the three layers (mean ± SD) in Table 10 are within the concentrations reported for tot-Sr-Al in Table 9.

Table 11: ^{84}Sr-Al:Ca-Al and ^{84}Sr-Al:tot-Sr-Al ratios in top soil (0-5 cm) at Apelsvoll and Fureneset, n=4.

Ratio	Apelsvoll	Fureneset
^{84}Sr-Al:Ca-Al	$2.5 * 10^{-6}$	$4.1 * 10^{-6}$
^{84}Sr-Al:tot-Sr-Al	$2.3 * 10^{-4}$	$2.1 * 10^{-4}$

The concentrations of ^{84}Sr-Al deeper in the soil were insignificant at both sites. However, the mobility was expected to be higher at Fureneset than at Apelsvoll as the soil at Fureneset had a lower clay content, reducing the fixation capacity of Sr. Together with the sandy soil, this could indicate an increased vertical migration through the soil profile, transporting ^{84}Sr away with pore water (Baratta, 1994). This vertical movement was not identified experimentally, but could be explained by soil pore water transporting ^{84}Sr downwards and out of the soil. In contrast, Roca & Vallejo (1995) reported sandy soils to sorb almost four times as much Sr as loamy soil. Wind could also play a role in distributing and diluting ^{84}Sr over a larger area (Jourdain, 2009). High wind speed (1.7 m/s) together with the heavy rainfall (17.6 mm) on June 30th could be attributed to the lower ^{84}Sr-Al concentrations in the supporting soil at Fureneset.

The values presented so far represent the plant available fraction, indicating the presence of an unavailable plant fraction of ^{84}Sr in soil[2]. Data on total ^{84}Sr in soil should therefore reflect variations in the concentrations of ^{84}Sr as Reitemeier (1958) reported that increased contact time between ^{90}Sr and soil increased fixation and immobilization of ^{90}Sr. However, fixation of deposited Sr in soil is generally of minor importance, though this may vary between soil types (Baratta, 1994). Only small fractions of ^{90}Sr were immobilized over a three-year period (Russel, 1963 cited in Baratta, 1994), indicating that immobilization of ^{84}Sr over three weeks should be negligible.

[2] Tot-^{84}Sr in soil was challenging to determine on ICP-MS, being especially problematic at Apelsvoll. One assumption for this was that the Sr-isotope ratios in soil changed as the soil minerals were dissolved in acid. The low Sr and Na, together with a high ^{87}Sr/^{86}Sr ratio (0.735 ± 0.004, n=12) in soil indicated mineral weathering (minimum ^{87}Sr/^{86}Sr ~ 0.720) (Frei & Frei, 2013) at Apelsvoll, strengthening the theory of dissolved minerals changing the Sr isotope ratios in soil.

5.4 Plant biomass at Apelsvoll and Fureneset

The foliage (leaf+stem) biomass at Apeslvoll was significantly higher ($p=1.07*10^{-8}$) than at Fureneset throughout the three-week experimental period (Figure 5.2). Average foliage biomass at Apelsvoll and Fureneset was 42.7 ± 9.8 g dw and 18.7 ± 8.6 g dw, respectively. Though significantly higher temperatures were found at Apelsvoll, it seems unlikely that temperature is the only explanation for the larger plants. Factors like precipitation, soil humidity, hours of direct sunlight and longer development time should also be of importance (Struick, 2007).

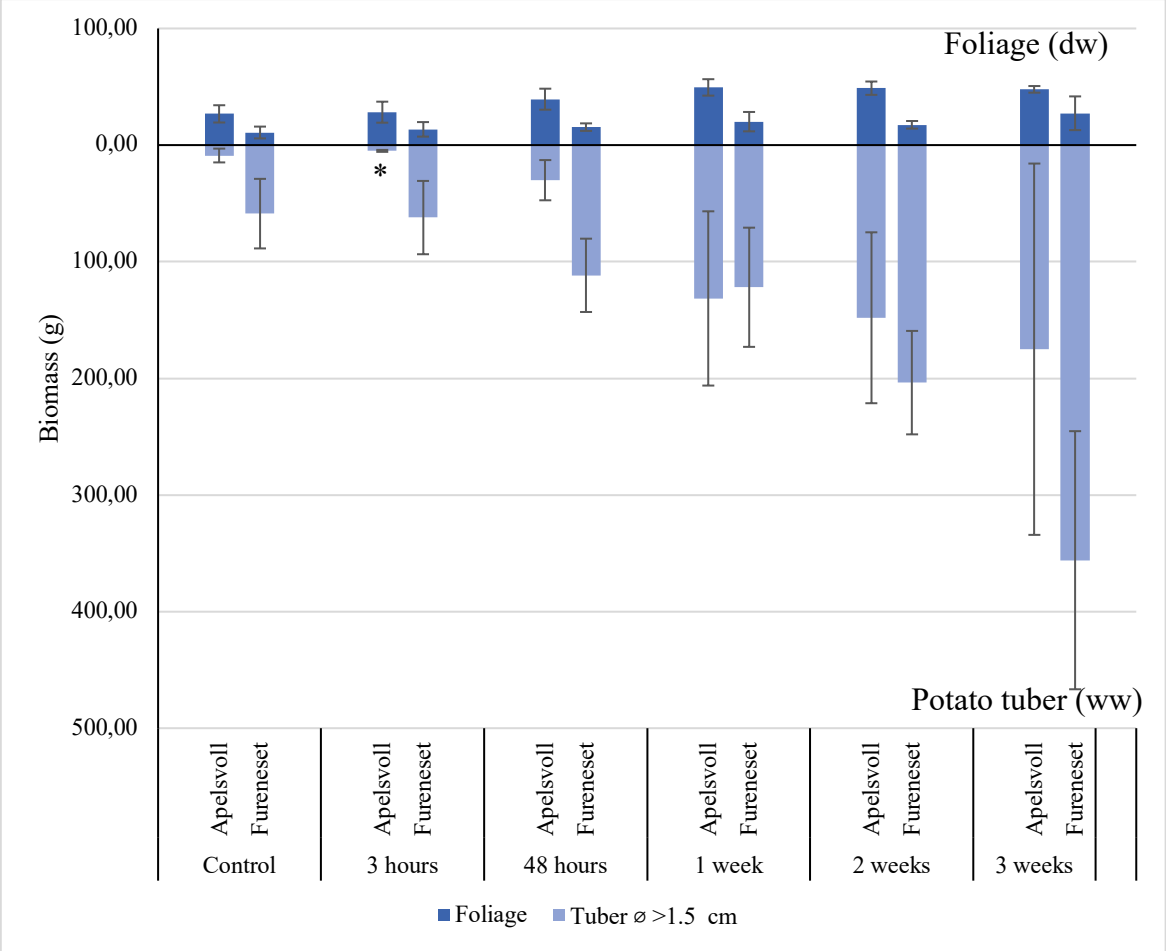

*Figure 5.2: Foliage (leaf+stem)(g dw) and potato tuber (⌀ >1.5 cm) (g ww) biomass at Apelsvoll and Fureneset over the three weeks (mean ± SD, n=3. For control: n=5). * n=2.*

The average biomass for potato tubers (⌀ >1.5 cm) was significantly higher (p=0.0005) at Fureneset compared to Apelsvoll, showing an opposite trend than above ground biomass (Figure 5.2). Potato tuber (⌀ >1.5 cm) biomass at Apelsvoll increased from 9 ± 6 g ww to 175 ± 159 g ww over the three weeks. This compared to 59 ± 30 g ww (n=5) and 356 ± 111 g ww

(n=3) for the control and three weeks, respectively, at Fureneset. The increased root biomass and reduced foliage biomass at Fureneset may be an indicator of a nutrient-deficient soil (McConnaughay & Coleman, 1999; Müller et al., 2000). As the potato tubers from August were visually smaller at Fureneset compared to Apelsvoll, the extra weight may be due to a higher number of potato tubers per plant. This is in accordance with Patil et al. (2016) reporting that low temperatures will generate a higher number of potato tubers per plant. The lower temperatures over the growing period at Fureneset compared to Apelsvoll may therefore partly explain this finding.

5.5 Background concentrations of Sr and Ca in plant tissue

At Apelsvoll, the Sr and Ca background concentrations in foliage (leaf+stem) were 0.17 ± 0.04 g/kg and 25 ± 5 g/kg, respectively. Both elements are concentrated in the foliage, giving a foliage:tuber (⌀<1.5 cm) ratio of 11.04 and 18.4 for Sr and Ca, respectively (Figure 5.3 and Figure 5.4). The ratio between tuber core and peel/rest (⌀<1.5 cm) was 3.9 and 3.4 for Sr and Ca, respectively. The Sr:Ca ratio in leaf and stem gave a stem:leaf ratio of 1.7, indicating that Sr concentrations accumulate in the stem whereas Ca is transported to the leaves.

Fureneset had foliage (leaf+stem) concentrations of 0.28 ± 0.03 g/kg and 18 ± 2 g/kg for total Sr and Ca, respectively. The foliage:tuber (⌀<1.5 cm) ratio for Sr and Ca was 15.9 and 21.6, respectively. For tuber core:peel/rest (⌀<1.5 cm), a ratio of 3.63 and 2.87 for Sr and Ca was obtained. The Sr:Ca ratio in stem:leaf was 1.8. Significant differences in Sr and Ca concentrations between Apelsvoll and Fureneset throughout the three-week sampling period was found.

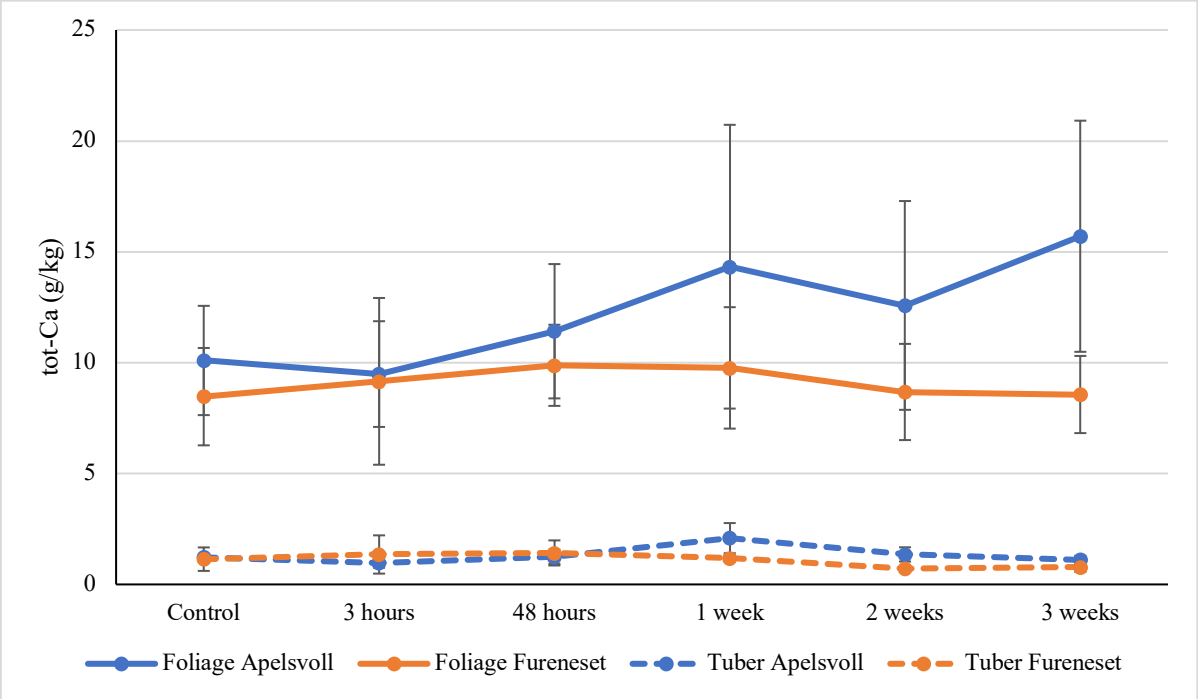

Figure 5.3: Tot-Ca concentrations (g/kg) in foliage (leaf+stem) and potato tubers (⌀<1.5 cm) at Apelsvoll and Fureneset. Values are mean ± SD, n=3.

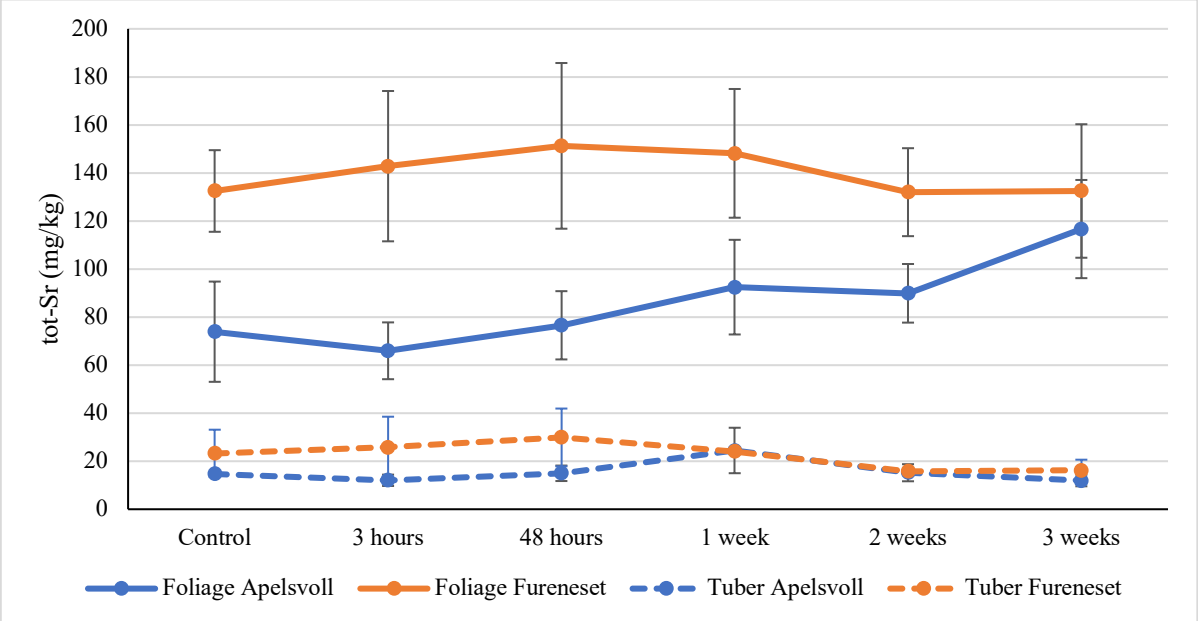

Figure 5.4: Tot-Sr (mg/kg) in foliage (leaf+stem) and potato tubers (⌀<1.5 cm) at Apelsvoll and Fureneset. Values are mean ± SD, n=3.

The accumulation in the above-soil plant tissue at both sites, as seen in Figure 5.3 and Figure 5.4, is due to Sr and Ca being phloem immobile, concentrating in foliage as a result of water potential differences (Westermann, 2005; Win et al., 1999). The higher concentrations of Ca and Sr in the tuber periderm was in accordance with Bamberg et al. (1993) and Busse & Palta (2006) reporting Ca concentrations to be much higher in the periderm/peel layer than tuber core. A Pearson correlation of r=0.96 was found between tot-Sr and Ca in plant tissue for both Apelsvoll and Fureneset. This is an important correlation emphasizing the similarity in chemistry and biochemical behavior. It strengthens the theory that Sr follows the chemistry of Ca, and that Ca can be used as a chemical analogue in the experimental fields.

The concentration of Sr in soil and plant tissue was much lower than the concentration of Ca at both sites (Table 12). This was expected as the concentration of Sr-Al also was much lower than Ca-Al in soil, in addition to Sr not being an essential nutrient and consequently not actively taken up by plants. The background Sr:Ca ratio in different plant tissues varied between, but was rather similar within, the two sites. The Sr:Ca ratio in plant tissue was equivalent to the plant available fraction of Sr:Ca in the respective soil (Table 12). This is expected if soil water is the source for uptake in plant (Ben-Bolie et al., 2014; Nye, 1966). This could be due to a linear adsorption-diffusion of nutrients between soil water and plant, resulting in a nearly proportional uptake of nutrients from soil solution (Menzel, 1954). The results showed that this could fit for the vegetative parts at Apelsvoll and Fureneset.

The only plant tissue deviating from the plant available Sr:Ca ratio in the soil was leaves, deviating at both sites. The ratio in leaf was found to be around half the ratios observed in the respective soil, stem, stolon and potato tubers (⌀ <1.5 cm), in accordance with Baratta (1994). Higher Sr:Ca ratios closer to the soil (e.g. in stem) emphasized the reduced in-plant mobility of Sr compared to Ca in the xylem vessel (Ben-Bolie et al., 2014; Choi et al., 2009). This is in agreement with Capo et al. (1998) reporting that Sr generally has a relatively high retention in soil and biota and is more strongly bound than Ca.

Table 12: Sr:Ca ratio in soil (plant available fraction of Sr and Ca) and plant tissues at Apelsvoll and Fureneset. Values are a mean ± SD of Sr:Ca ratios for n replicates.

Sr:Ca ratio	Apelsvoll	n	Fureneset	n
Soil	0.011 ± 0.001	27	0.02 ± 0.001[a]	27
Leaf	0.006 ± 0.0007	18	0.0116 ± 0.0007[a]	22
Stem	0.01 ± 0.001	18	0.021 ± 0.002[a]	20
Stolon	0.0114 ± 0.0007	20	0.022 ± 0.001[a]	20
Root hair	NA		0.021 ± 0.002	20
Small tuber (⌀<1.5 cm)	0.0117 ± 0.0009	18	0.021 ± 0.002[a]	18
Big tuber (⌀>1.5 cm)	0.0098 ± 0.0019	18	0.023 ± 0.002[a]	18

[a] significantly higher

The uptake of Sr in plant tissue was twice as high at Fureneset compared to Apelsvoll, as seen in Table 12, initially attributed to the external Sr source (sea spray). However, as the ratio of Sr:Ca in soil and plant tissue was similar, and lower in leaf, the assumption that Sr is deposited on leaves and transferred to other plant parts in the plant may not be correct. Hence, the source of Sr and Ca in plant tissue appeared to be soil rich in plant available Sr and Ca at both sites. As Sr-Al in soil at Fureneset was lower than at Apelsvoll, the much higher uptake of Sr in plant tissue at Fureneset could be attributed to Fureneset´s low concentrations of Ca-Al and other base cations (Table 9) (Menzel, 1954), and thereby low competition from other base cations that would increase the chance of Sr uptake in plants.

5.6 Transfer and uptake of ^{84}Sr in plant tissue

Discussion on the uptake of ^{84}Sr in plant tissue is presented separately for Apelsvoll and Fureneset in the next two subchapters, followed by a comparison and possible explanation for the different findings of ^{84}Sr in foliage and below soil plant tissue. It is important to mention that radionuclide fallout most often contains radionuclides as well as a series of metals, while in this experiment the focus is put only on depositing traces of ^{84}Sr. Hence, a multi stressor scenario would be expected to affect and change the behavior of the element analyzed here (Salbu, 2016).

Unless otherwise stated, all means ± SD are based on three replicates. The standard deviations at different sampling times were often large. The large standard deviations were found in above and below soil plant tissue, making discussions somewhat uncertain. Results indicated that quantification of ^{84}Sr, especially in below soil plant tissue, was rather random and emphasized the uncertainty of the experiment when having few replicates.

5.6.1 ^{84}Sr in plant tissue at Apelsvoll

The ^{84}Sr concentration was found to be highest in leaves at Apelsvoll, with concentrations of 1967 ± 306 µg/kg dw and 983 ± 102 µg/kg dw three hours and three weeks after wet deposition, respectively (Figure 5.5). This gave an ^{84}Sr reduction (p=0.002) of 50 % three weeks after wet deposition. Leaf concentrations were significantly higher (p=0.0005) than stem concentrations with 850 ± 566 µg/kg dw and 1033 ± 635 µg/kg dw after three hours and three weeks, respectively. Both leaf and stem were significantly different from their respective control groups after three weeks, indicating absorption of ^{84}Sr after wet deposition (Table D.2.1 in Appendix). The ^{84}Sr concentrations in foliage (leaf+stem) remained rather stable over the sampling period (p>0.05), as shown in Figure 5.5.

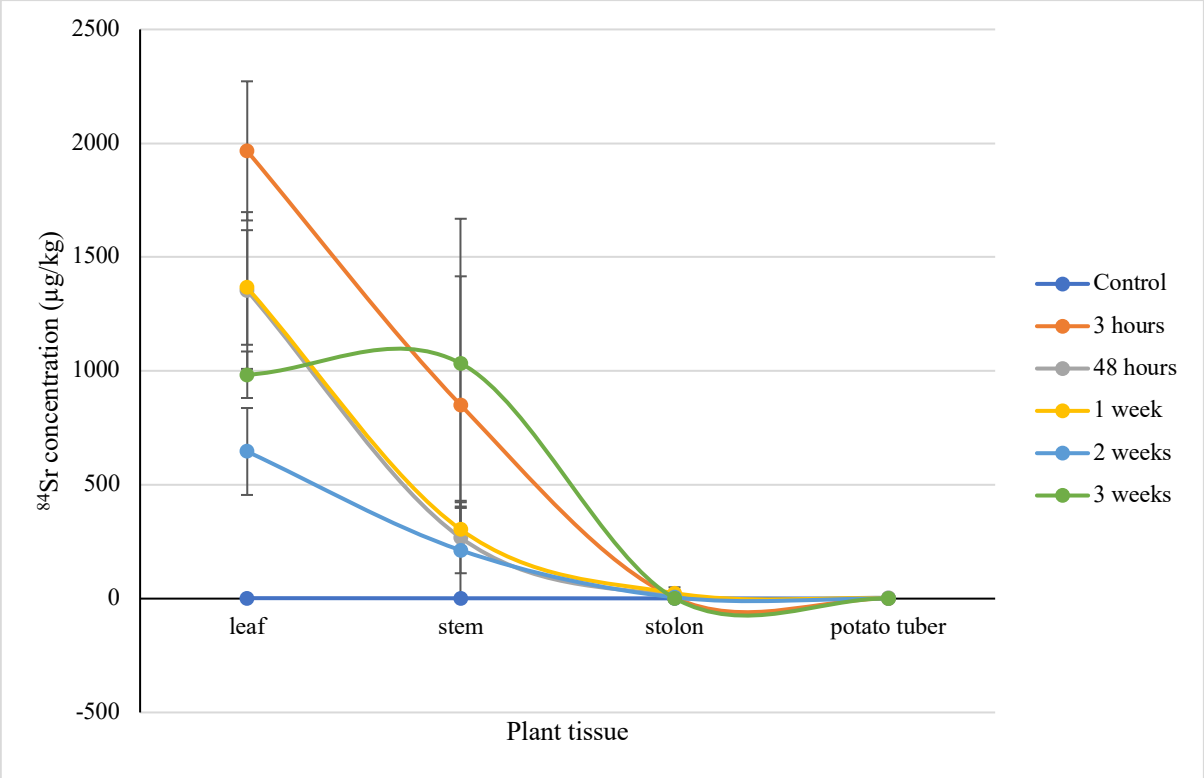

Figure 5.5: ^{84}Sr concentrations (µg/kg dw) in leaf, stem, stolon and potato tuber(⌀<1.5 cm) at the different sampling times at Apelsvoll. Mean ± SD, n=3.

Stolons and potato tubers did not show a significant difference compared to their respective control after three weeks, as visualized in Figure 5.5. After three hours all concentrations were <LOQ, which was expected after a foliar deposition of ^{84}Sr. In the next three sampling points (48 hours, 1 week, 2 weeks), ^{84}Sr was quantified in one or more replicates for stolons. After three weeks all three replicates gave concentrations <LOQ. The values fluctuated greatly and showed no clear trend. The same was shown for potato tuber (⌀ <1.5 cm) concentrations of ^{84}Sr, with 0.3 ± 0.4 µg/kg dw after three hours compared to 1 ± 2 µg/kg dw after three weeks. Big potato tubers (⌀ >1.5 cm) from August did not contain any significant amount of ^{84}Sr in peel or in the whole potato tubers.

5.6.2 ^{84}Sr in plant tissue at Fureneset

Leaves, stem, stolons and root hair were all significantly different ($p<0.05$) than their respective controls three weeks after spraying at Fureneset (Table D.2.1 in Appendix). The concentration of ^{84}Sr was highest in leaves, showing a steady reduction ($p=0.03$) from 2233 ± 153 µg/kg dw three hours after wet deposition to 980 ± 302 µg/kg dw after three weeks (Figure 5.6). The stem concentration was more similar with 1063 ± 212 µg/kg dw after three hours and 353 ± 258 µg/kg dw after three weeks.

Stolons and root hair showed a rather stable ^{84}Sr-spike concentration throughout the three weeks (Figure 5.6). After wet deposition, the concentration increased faster in root hair with 94 ± 49 µg/kg dw after three hours, compared to 12 ± 12 µg/kg dw for stolons. The high uptake in root hair after three hours was significantly reduced ($p=0.03$) between one and two weeks. The stolons showed an opposite trend where the ^{84}Sr concentration increased ($p=0.04$) after two-three weeks and the concentration was found to be correlated with time ($p=0.027$). The concentrations in stolons and root hair were not significantly different after three weeks with 50 ± 30 µg/kg dw and 48 ± 29 µg/kg dw for stolons and root hair, respectively.

The potato tuber (⌀ <1.5 cm) concentration of ^{84}Sr was the only plant tissue concentration not significantly different from its control after three weeks at Fureneset. Concentrations of 3 ± 3 µg/kg dw and 2 ± 2 µg/kg dw were quantified three hours[3] and three weeks after spraying, respectively. The values fluctuated greatly, and no trend was visible in the small tubers.

[3] n=2

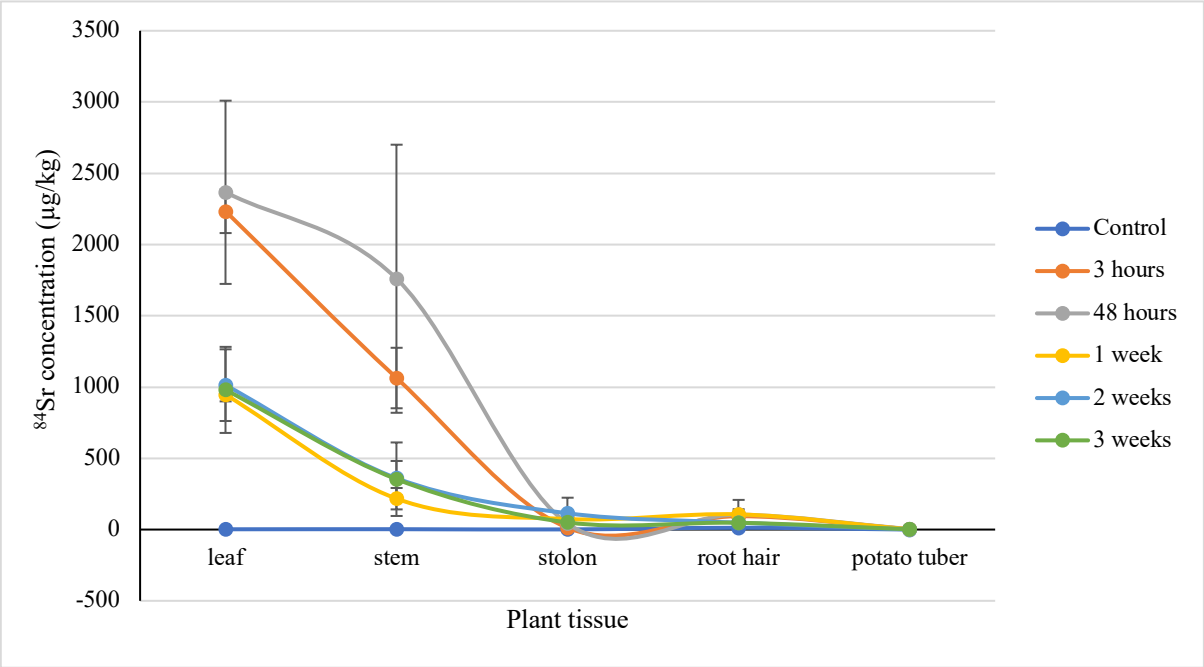

Figure 5.6: ^{84}Sr concentrations (µg/kg dw) in leaf, stem, stolon, root hair and potato tuber (⌀ <1.5 cm) at the different sampling times at Fureneset. Mean ± SD, n=3 (for control, n=5; for tuber after 3 hours, n=2).

With leaf fall in August, it was assumed that nutrients from the leaf was transferred to the soil, and then to roots and potato tubers, possibly increasing contamination of below-ground plant tissue (Watts & Howe, 2010). The August sampling showed an increase of ^{84}Sr in the sprayed potato tubers compared to the control at a 96 % confidence interval (p=0.051). Considering the fact that only a small fraction of the freeze-dried potato tuber was analyzed, and homogeneous distribution of ^{84}Sr was not ensured, a 96 % confidence level was justified. One sample may have contained more peel than another, which would most likely influence the results significantly.

When analyzing the potato peel separately, which according to literature is where Sr will accumulate, a significant difference (p=0.006) was evident between control group and treated group; ^{84}Sr clustered in the peel. The division of tubers into small, medium and large (Figure E.2 in Appendix) gave no clear dilution trend for ^{84}Sr (n=6). Few potato tubers resulted in a mix with few replicates of different sizes (small+medium), and a more consequent focus on size as well as more potato tubers could give different results. The differences between core and peel/rest in the treated group (see Figure 4.6, Materials and Methods p. 33) and between the August potato tubers (⌀ >1.5 cm) and small potato tubers (⌀ <1.5 cm) after three weeks were not significant.

5.6.3 Comparison of ^{84}Sr uptake in plant tissue at Apelsvoll and Fureneset

The concentration of ^{84}Sr in leaves and stem at Apelsvoll and Fureneset was similar, Figure 5.7, and significantly different from their respective control group after three weeks. The big drop in the ^{84}Sr concentration in stem after one week at Fureneset was not seen in leaves (Figure 5.7). This could be due to weaker absorption / retention on stem surfaces compared to leaf, as the stem surface is smoother. Apelsvoll showed no significant uptake of ^{84}Sr in stolons or potato tubers throughout the three-week sampling period, compared to the increased concentrations in stolons, root hair and potato tubers at Fureneset (Figure 5.7). Ca was the only independent variable explaining the uptake of ^{84}Sr in samples at Apelsvoll ($p=0.001$, $R^2=54.48\%$) whereas the increased concentrations of ^{84}Sr at Fureneset may be related to background concentrations of Sr ($p=0.001$), Ca ($p=0.001$) and Ba ($p=0.001$) ($R^2=48.16\%$).

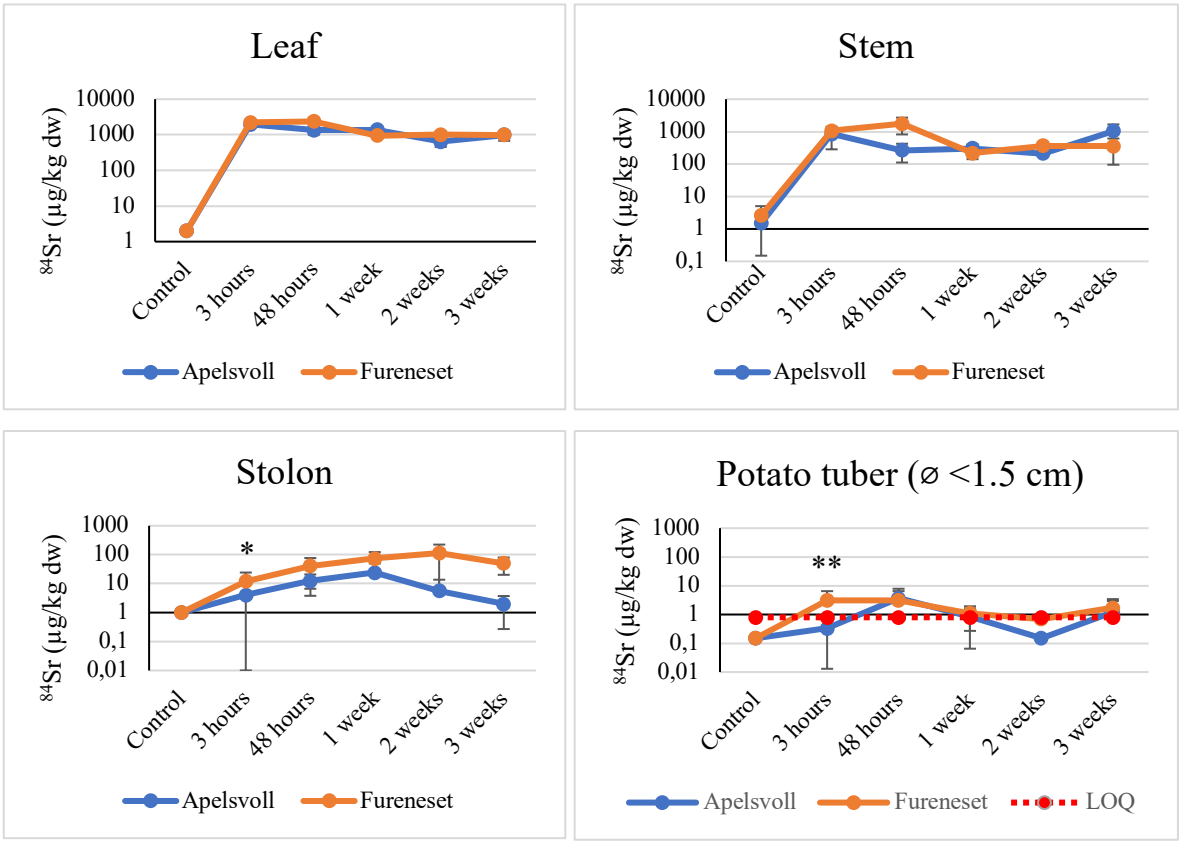

*Figure 5.7: Logarithmic concentrations of ^{84}Sr (µg/kg dw) in leaves, stem, stolons and potato tubers (⌀<1.5 cm) at Apelsvoll and Fureneset. Values are a mean ± SD, n=3 (for control at Fureneset, n=5). Y-axis is 10 times lower for stolons and potato tubers. The dotted red line in the potato tuber graph shows the LOQ value (0.8), emphasizing that the ^{84}Sr concentration in tubers at Fureneset generally were above LOQ while ^{84}Sr concentrations in tubers at Apelsvoll fluctuated more. * indicates one value <LOD for Fureneset, resulting in a big standard deviation. ** indicates two values <LOD for Apelsvoll resulting in a big standard deviation.*

An increase in ^{84}Sr in August potato tubers was only found at Fureneset. For Apelsvoll, the large potato tubers (⌀ >1.5 cm) from the August sampling contained no ^{84}Sr, possibly explained by biodilution as the tubers were bigger. Tot-Sr showed a decrease from 16 ± 6 mg/kg in small tubers (⌀ <1.5 cm) to 2.9 ± 0.9 mg/kg in full grown potato tubers at Apelsvoll, giving a tot-Sr reduction of 82 %. The Ca concentration was reduced by 78 % from small to big potato tubers. This compared to 60 % and 69 % reduction for tot-Sr and Ca, respectively, at Fureneset, explained by less dilution (see Results and Discussion 5.4) or the generally higher uptake of Sr.

5.6.4 Comparison of ^{84}Sr:Ca ratio in plant tissue and soil solution

The ^{84}Sr:Ca ratio can be helpful in understanding where uptake of ^{84}Sr in plant tissue comes from, and why the uptake is higher at one site compared to another. The ^{84}Sr and Ca concentrations were correlated at Apelsvoll (r=0.69), and weakly correlated at Fureneset (r=0.46). Figure 5.8 shows that the ^{84}Sr:Ca ratio in plant tissue was significantly higher at Fureneset throughout the three weeks and the trend at both sites was that the ratio decreased from leaf > stem > stolon > root hair (only at Fureneset, not in figure) > potato tuber. This in comparison to the tot-Sr:Ca ratio being lowest in leaves and twice as high in stem, stolons, root hair, potato tubers and soil (Table 12). As the two ratios were opposite, and the ^{84}Sr:Ca was highest in leaf, it became evident that the high ^{84}Sr concentration found in foliage was a direct result of the foliar wet deposition.

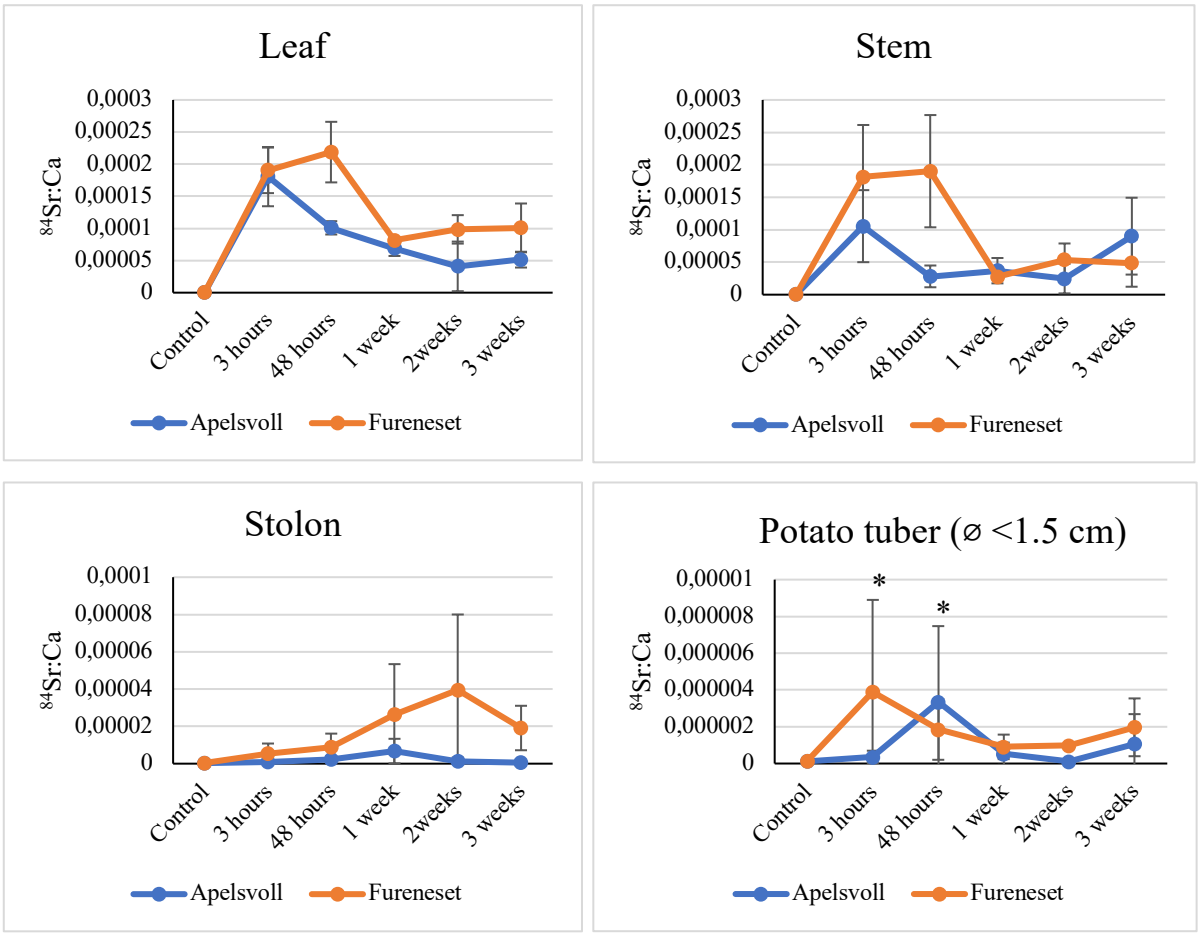

*Figure 5.8: ^{84}Sr:Ca ratio in leaf, stem, stolon and potato tuber (⌀ <1.5 cm) at Apelsvoll and Fureneset (mean ± SD, n=3). The y-axis for stolons and potato tubers is different by a factor of 10. Potato tuber replicates = 2 at 3 and 48 hours at Fureneset. * indicates one value <LOQ giving a large standard deviation.*

Three weeks after spraying, the average ^{84}Sr:Ca ratio in foliage (leaf+stem) was $7.07*10^{-5}$ (total plant: $3.58*10^{-5}$) and $7.47*10^{-5}$ (total plant: $3.53*10^{-5}$) at Apelsvoll and Fureneset, respectively. The leaf ratios were higher than in the topsoil (0-5 cm) with ^{84}Sr:Ca ratios of $2.5*10^{-6}$ and $4.1*10^{-6}$ at Apelsvoll and Fureneset, respectively (Table 11). Small potato tubers (⌀ <1.5 cm) at Apelsvoll[4] and Fureneset[5] had ^{84}Sr:Ca ratios of $1.07*10^{-6}$ and $1.96*10^{-6}$, respectively (n=3), after three weeks. Big potato tubers (⌀ >1.5 cm) at Fureneset[6] had an ^{84}Sr:Ca ratio of $6.83*10^{-6}$ (n=9). As the soil ratios at three weeks were in accordance with the potato tuber (⌀ <1.5 cm) ^{84}Sr:Ca ratios found in the two sites, it could indicate a soil to plant transfer of ^{84}Sr. This was further emphasized by the ratio in whole potato tubers (⌀ >1.5 cm) from the August sampling at Fureneset being similar to the soil ratio.

[4] Two values <LOD for ^{84}Sr
[5] Two values <LOQ for ^{84}Sr
[6] Four values <LOD for ^{84}Sr, two values <LOQ for ^{84}Sr

Libby (1956) reported that the ^{90}Sr:Ca ratio was twice as high in plants compared to the supporting soil after continuous foliar deposition. This mainly because fallout was directly deposited on foliage. The ^{84}Sr:Ca ratio between plants and soil at the two experimental sites was much higher than this with an ^{84}Sr:Ca ratio of 14.3 and 8.6 for Apelsvoll and Fureneset, respectively. However, due to a one-time deposition of ^{84}Sr, a comparison with long term deposition may not be accurate.

5.6.5 Comparison of ^{84}Sr on foliage

Although the ^{84}Sr concentrations in foliage was similar at both sites, the total ^{84}Sr content in leaves was higher at Apelsvoll throughout the period, as shown in Figure 5.9. The higher content at Apelsvoll could be attributed to the higher foliage biomass (Figure 5.2), as Menzel et al. (1963) reported that ^{90}Sr uptake in foliage was greatest in the largest plants. Only leaf concentrations were used to determine ^{84}Sr content, as the stem surface area and weight are small in comparison to the leaves (Poorter et al., 2015). The choice of leaf sample is crucial as Struick (2007) reported that there were three to four large leaflets dominating each plant, with a major leaf at the top probably sorbing much of the contaminant. If these were sampled on one plant, compared to the small in-between leaflets on another, the concentrations may vary greatly between plants.

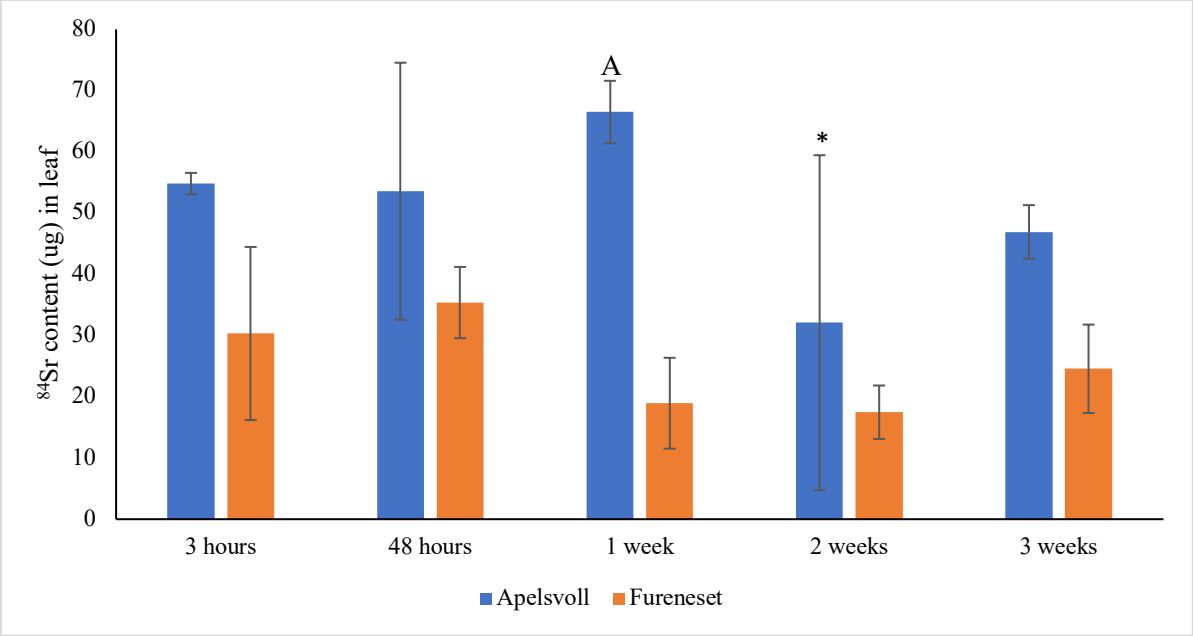

*Figure 5.9: ^{84}Sr content (µg) in leaf after wet deposition at Apelsvoll and Fureneset (mean + SD, n=3). "A" indicates significant differences ($p<0.05$) between sites. The visual increases in ^{84}Sr content over the three weeks were never significant. * indicates one value <LOQ.*

The reduction of ^{84}Sr in foliage (leaf+stem) during the first week was 41 % and 65 % for Apelsvoll (p=0.02) and Fureneset (p=0.002), respectively (Figure 5.10). Much rain directly after deposition would most likely lead to wash-off of ^{84}Sr from surfaces, transporting the tracer to the soil instead of the foliage (Moorby & Squire, 1963). Hence, the higher reduction rates at Fureneset may be attributed to precipitation. The precipitation downfall of 17.6 mm at Fureneset, happening on the fourth day after spraying, may therefore be the main explanation for reduced foliage ^{84}Sr concentrations between 48 hours and one week (Figure 5.10). A big drop in ^{84}Sr:Ca ratio between 48 hours and one week (Figure 5.8) for leaves and stem strengthened this assumption. The possible effect of precipitation has been reported by Middleton (1958), describing how rain reduced ^{89}Sr uptake on foliage by a factor of six. Additionally, plants grown in irrigated areas were reported to be less sensitive to Sr contamination due to dilution and wash off of Sr (Libby, 1956). Due to much precipitation at Fureneset, ^{84}Sr may not have had sufficient time to be absorbed. This is in accordance with Kirchmann et al. (1966) reporting that the relative fraction percentage of an element leaked from foliage decreased with increasing contact time between ion and foliage (Kirchmann et al., 1966).

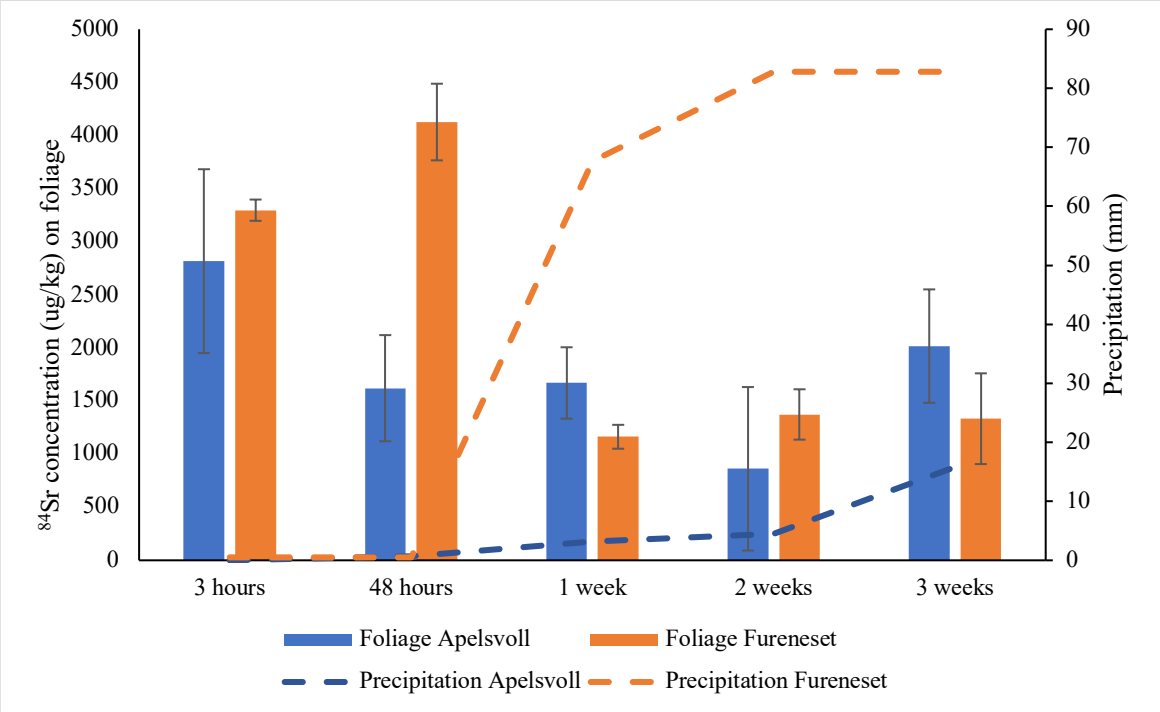

Figure 5.10: Concentration of ^{84}Sr (ug/kg) in foliage (leaf+stem)(mean ± SD, n=3) and total precipitation (mm) over the three weeks. Precipitation is the sum of precipitation at given time after wet deposition of ^{84}Sr. The precipitation line for Apelsvoll starts at 48 hours because there was no precipitation between 3 and 48 hours.

Fureneset showed a reduction (p=0.01) of more than 50 % from foliage (leaf+stem) two weeks after spraying (Figure 5.10), which is similar to the reported half-life of ^{90}Sr on foliage (Dorsey et al., 2004). Apelsvoll also showed more than 50 % reduction in ^{84}Sr concentration, although not significant. These values represent the average for three plants per site, which may not be comparable with half-lives measured by analyzing the change in Sr-concentration in *one* plant over time. At three weeks after spraying, foliage reduction of ^{84}Sr was only significant at Fureneset using a 94 % confidence interval. The total reduction (p=0.052) after three weeks was 60 %. The reduction at Apelsvoll was not significant after two-three weeks, reflecting that Sr deposited was to a certain degree retained. A higher number of replicates would most likely be efficient to reduce the variation in foliage concentration and further emphasize the differences in ^{84}Sr absorption at the two sites.

Translocation from the surfaces of leaves to other plant parts may occur, but is not likely to explain the large reduction in foliage concentrations as phloem transport from foliage to potato tuber has been reported to be very low (Bukovac & Wittwer, 1957; Kratzke & Palta, 1985). This left precipitation as the main explanation for the large difference in reduced ^{84}Sr concentrations in foliage at Apelsvoll (28 %, p=0.4) and at Fureneset (60 %, p=0.052) three weeks after spraying. The effect of precipitation on divalent ^{84}Sr cation absorption in foliage was further emphasized by the negative correlation of r=-0.95 between total precipitation and ^{84}Sr on foliage (leaf+stem) found at Fureneset (Table 13). No correlation was found between precipitation and ^{84}Sr concentration on foliage at Apelsvoll.

Table 13: Precipitation (mm), ^{84}Sr concentration (µg/kg) in foliage (average leaf+stem, n=3) and correlation at Apelsvoll and Fureneset. Precipitation is total amount (sum) of precipitation after wet deposition. Precipitation and ^{84}Sr concentrations are not converted to log_{10}.

Sampling time	Apelsvoll		Fureneset	
	Precipitation (mm)	^{84}Sr (µg/kg)	Precipitation (mm)	^{84}Sr (µg/kg)
3 hours	0	2816.6	0.5	3296.6
48 hours	0.7	1620.6	0.5	4126.6
1 week	3.2	1670	67.6	1163.3
2 weeks	4.5	861.7	82.8	1373.3
3 weeks	15.8	2016.6	82.8	1333.3
Correlation	-0.07		-0.95	

5.6.6 ^{84}Sr in stolons, root hair and potato tubers

^{84}Sr could be quantified in potato tubers (⌀ <1.5 cm) grown at both sites. Significant differences in ^{84}Sr concentration from the control group were found for Fureneset only, though not significantly different from Apelsvoll for small (⌀ <1.5 cm, p=0.2) or big (⌀ >1.5 cm, p=0.09) potato tubers. This was most likely due to large standard deviations. Figure 5.11 confirms that ^{84}Sr, like Ca, accumulated in the periderm/peel layer of newly developed potato tubers (⌀ <1.5 cm) at Fureneset, as reported by Kratzke & Palta (1986) and Busse & Palta (2006). The increased concentration in peel/rest compared to control group was only significant at two weeks (p=0.05). The high concentrations of ^{84}Sr in stolons, root hair and to some extent, potato tubers, at Fureneset may also be explained by increased soil moisture (IAEA, 2010 cited in Burger & Lichtscheidl, 2019; Wang et al., 2000). The assumption that soil moisture was higher at Fureneset than at Apelsvoll, as a result of higher precipitation rates, is logical although no data on soil moisture was obtained.

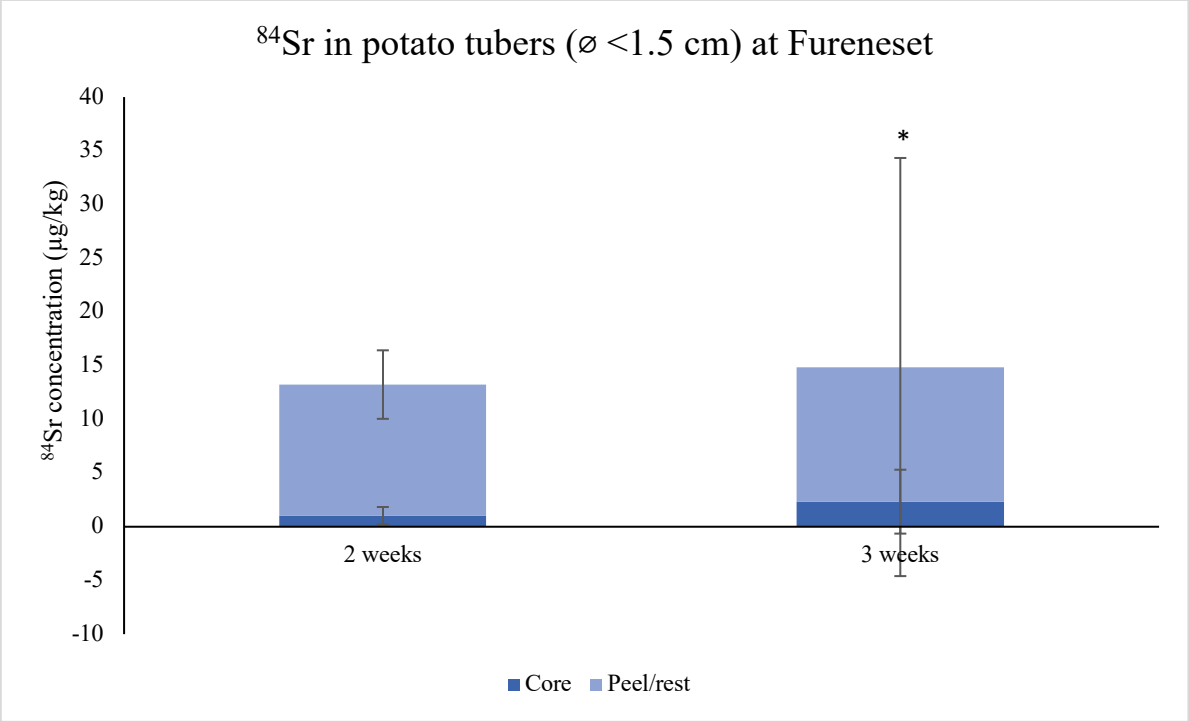

*Figure 5.11: ^{84}Sr concentrations (µg/kg) in core and peel/rest of potato tubers (⌀ <1.5 cm) at Fureneset two and three weeks after wet deposition (mean ± SD, n=3).The small potato tubers (⌀ <1.5 cm) were divided in two and separated into core and peel/rest (See Figure 4.6, Materials and Methods p. 33). * indicates one peel/rest value <LOQ for ^{84}Sr.*

A decrease in ^{84}Sr concentrations in below soil plant tissue after three weeks, compared with big potato tubers from August, was found in the following order for Fureneset[7]: stolons > root hair >> August potato tubers (⌀ >1.5 cm) > potato tubers (⌀ <1.5 cm). The difference in ^{84}Sr concentration between small (⌀ <1.5 cm) and big (⌀ >1.5 cm) potato tubers was not significant. The significantly higher uptake of ^{84}Sr in stolons ($p=0.01$) and root hair ($p=0.0002$) compared to potato tubers have been found for other contaminants like Cd, Pb and Cu (Dunbar et al., 2003; Zheng et al., 2018). This indicates that nutrients and contaminants accumulate in stolons and root hair, and that the transfer to potato tubers was low. This was further emphasized by the generally lower concentrations of Sr and Ca in potato tubers. However, as the potato tubers (⌀ <1.5 cm) should be in growth stage IV, the much lower uptake of ^{84}Sr was somewhat unexpected as nutrients should have been transferred from stolons and accumulated in potato tubers during this bulking period (Jackson & Haddock, 1959; Westermann, 2005).

As the ratio of ^{84}Sr:Ca in stolons, root hair, potato tubers (⌀ <1.5 cm) and soil at Fureneset was similar, it strengthened the assumption that soil-root transfer was the basic transport mechanism for ^{84}Sr. Elevated soil concentrations were attributed to high precipitation rates, in accordance with Isermann (1981) reporting that the increased uptake in below soil plant tissue after foliage application was explained by wash-off of Sr from leaves onto soil. Moorby & Squire (1963) reported similar findings. From the soil, ^{84}Sr could be transferred to potato tubers through stolons / root hair or direct diffusion. The latter was reported by Habib & Donnelly (2002) describing how Ca diffused directly from soil through the periderm and into the potato tuber over a 5-day period. Hence, direct contact with the deposited Sr, either through soil or direct deposition onto potato tubers, could be an explanation for contamination after fallout, as suggested by Müller et al. (2003). The low uptake found in potato tubers after soil contamination in this experiment was in accordance with Menzel et al. (1963) reporting that ^{90}Sr penetrating the crop cover and entering the soil was not an important pathway of potato tuber contamination. Additionally, concentrations of ^{90}Sr in potato tubers have been reported to be very low when ^{90}Sr was added directly to the soil (Andersen, 1967).

[7] Apelsvoll had few replicates with quantifiable ^{84}Sr concentrations in stolons and potato tubers. Root hair was not analyzed.

If increased concentrations of ^{84}Sr in below soil plant tissue was from contaminated soil, the difference in uptake in below soil plant tissue must be related to soil characteristics. The higher plant available Ca-Al, clay fraction and CEC at Apelsvoll were three soil characteristics that could strongly influence the uptake, where increasing levels of Ca-Al and CEC would reduce uptake of radiostrontium (Fredriksson et al., 1968 cited in Lönsjö & Haak, 1975). This could explain the reduced uptake of ^{84}Sr in below soil plant tissue at Apelsvoll, despite higher topsoil concentrations of ^{84}Sr-Al three weeks after spraying. A high CEC level is beneficial for ^{84}Sr removal as Sr was almost completely removed if the initial concentration of cations was <0.01 of total saturation capacity in soil (McHenry et al., 1956). The higher concentrations of the divalent cations Mg and Ba in soil may also increase ion competition and reduce the ^{84}Sr uptake.

For Fureneset, the low plant available Ca-Al, low clay content, low base cation concentrations and sandy soil appeared to be important factors for the increased ^{84}Sr uptake. The lower topsoil (0-5 cm) concentrations of ^{84}Sr-Al at Fureneset compared to Apelsvoll may partly be explained by the increased uptake in below soil plant tissue. Low plant available Ca-Al appeared to be the main reason for increased uptake, in accordance with Fredriksson et al. (1970) cited in Lönsjö & Haak (1975), Helal et al. (1997), Libby (1956), Menzel (1954) and Roca & Vallejo (1995).

5.6.7 Translocation of ^{84}Sr from foliage to potato tuber

If translocation occurred, then the concentration of ^{84}Sr in foliage would decrease as the uptake in other plant tissues such as stem, stolons, root hair and potato tubers would increase. An increase in below soil plant tissues such as stolons, root hair and potato tubers was only significant at Fureneset, where the ^{84}Sr reduction in foliage (leaf+stem) was 1963 µg/kg three weeks after wet deposition. Theoretically, this gave an estimated translocation from foliage to potato tuber of about 0.09 % (n=3), assuming total potato tuber concentration originated from foliar translocation. The theoretical translocation was within the foliage to potato tuber translocation of 0.01-0.5 % reported by Middleton (1958) and Moorby & Squire (1963) after single foliar application. The low translocation rate for Sr was also found in bean and corn plants, where only 0.006 % of ^{85}Sr translocated all the way from foliage to roots (Ambler, 1964). A higher translocation rate was found when Sr was applied during early fruit

development. For wheat, barley and rye, the foliar to root translocation gave a maximum of 2 % with a 95 % confidence interval (Colle et al., 2009). Translocation factors and absorption of Sr in foliage for different plant species may not always be comparable as leaf waxiness and maturity development vary greatly (Frere et al., 1963). However, a universal element behavior in plants is reported, justifying this comparison (Colle et al., 2009).

The higher content of ^{84}Sr in foliage (Figure 5.9) at Apelsvoll, together with lower potato tuber biomass, indicated that if translocation was happening, the highest concentrations of ^{84}Sr in potato tubers should have been at Apelsvoll. However, no significant ^{84}Sr concentrations were found in potato tubers at this site. Furthermore, the ^{84}Sr concentrations in stolons were negligible, and as transfer of ^{84}Sr from foliage to potato tuber would have to go through stolons (Busse & Palta, 2006), the translocation assumption was rejected at Apelsvoll. This was further emphasized by the stable foliage concentration of ^{84}Sr during the three weeks. The stable Sr concentrations in foliage was in accordance with Bukovac & Witter (1957) and Ambler (1964) reporting that once Sr was absorbed / deposited on a point of a leaf, it was rarely exported from the leaf. It could, however, be transported to adjacent leaf tissue or younger leaves.

The increase in stolons at Fureneset indicated that translocation from stolon to potato tuber was possible here, though no correlation (r=-0.09) between stolon and potato tuber concentrations was found. Furthermore, the low estimated translocation at Fureneset, despite higher reduction in ^{84}Sr foliage concentrations, indicated that the reduced concentrations in foliage was attributed to rainfall and wash off of ^{84}Sr. This assumption was supported by the ^{84}Sr concentrations detected in the topsoil (0-5 cm) after three weeks. Translocation factors from soil to leaf have been reported to be higher than leaf to soil, and the soil contamination and root uptake is assumed to be the dominating uptake pathway if contaminant is deposited at an early growth stage (James et al., 2011). This was shown by Choi et al. (2009) reporting that the transfer factor for Sr from soil to potato tuber was highest 30 days after planting (day 0-planting, day 90-harvesting)[8]. The potato tubers showed translocation factors from soil of $5.5*10^{-4}$ m^2/kg, $1.6*10^{-3}$ m^2/kg and $2.0*10^{-4}$ m^2/kg with soil contamination at 4, 31, and 63 days after planting, respectively.

[8] The article did not specify which growth stage the potato tubers were in when planted.

The presence of water is important for Sr-translocation. Dry bean and corn plants showed a translocation below 0.01 %, compared to 0.5-0.6 % after periodically rewetting (Ambler, 1964). Wheat plants doubled the Sr-translocation after rainfall, while dry conditions gave no translocation of ^{85}Sr in rye-grass and red clover (Kirchmann et al., 1966; Middleton, 1958). This indicates that if translocation was happening, it should be higher at Fureneset due to higher precipitation rates and more humidity. The lower temperatures at Fureneset compared to Apelsvoll may, however, reduce the translocation, as Kirchmann et al. (1966) found that temperatures <21 ºC stopped translocation in rye grass and red clover.

It is challenging to investigate if translocation of ^{84}Sr occurred in the outdoor experiments as both spraying and weather conditions can distribute ^{84}Sr differently on plant and soil surfaces in the experimental plot. The low / negligible translocation found for ^{84}Sr in this experiment was, however, in accordance with results reported by Busse & Palta (2006), who used ^{45}Ca (chemical homologue for Sr) to study transportation of Ca from foliage to potato tubers. After 57 days the potato tuber concentration of ^{45}Ca was at background levels. The same reduced movement towards the basipetal has also been found for Sr after foliage application (Ambler, 1964; Bukovac & Witter, 1957; Isermann, 1981; Smith, 1971). This despite observed concentration gradients within the plant (Rediske & Selders, 1953). Results from this experiment indicated that translocation of Sr from foliage to potato tuber after direct deposition on foliage was of minor importance, as previously reported by Isermann (1981), Moorby & Squire (1963), Müller et al. (2003), Oestling et al. (1989) and Rediske & Selders (1953).

5.7 Relevance of deposited ^{84}Sr concentration as ^{90}Sr activity

Converting the ^{84}Sr concentrations (µg/kg) deposited on soil and plant surfaces to ^{90}Sr, assuming the same concentrations, and then to activity concentrations of ^{90}Sr gave an indicator of how relevant, with respect to nuclear fallout, the deposited concentrations of ^{84}Sr in these experiments were. The calculation gave a ^{90}Sr activity of almost 4 GBq/m^2 (see Appendix F) which was extremely high and not relevant to potential nuclear fallout episodes in Norway. In comparison, a maximum fallout of ^{90}Sr on soil in Norway was about 3 kBq/m^2 after the nuclear weapons tests in the mid 1950s and 1960s (NOU, 1987:1, p. 54). The topsoil

(0-5 cm) concentrations of ^{84}Sr equaled 20 400 kBq/kg[9] and 7 650 kBq/kg[9] for ^{90}Sr at Apelsvoll and Fureneset, respectively (Table F.1 in Appendix). This is much greater than the reported ^{90}Sr soil activity in the Fukushima exclusion zone (3 ± 0.3 – 23.3 ± 1.5 Bq/kg) and areas in close proximity to the Chernobyl power plant after the accident in 1986 (Al-Rayyes & Mamish, 1999; Kashparov et al., 2001; Sahoo et al., 2016). Kashparov et al (2001) reported that the total content of ^{90}Sr in the Chernobyl exclusion zone (30 km radius) added up to 810 TBq, emphasizing how unrealistic a deposition of 4 GBq/m^2 in Norway would be.

As Norway is, geographically, far away from potential nuclear sources, lower activity concentrations of ^{90}Sr in precipitate can be expected after potential nuclear accidents. Though lower concentrations, the fraction of Sr in ionic form could potentially be higher than observed in Chernobyl, thus most of the concentrations deposited could be plant available (Blume et al., 2016). The expected fallout concentration is, however, strongly dependent on the source term (physico-chemical form of ^{90}Sr), season as well as the weather conditions (heavy precipitation) at the time of a potential accident.

5.8 Human exposure to radiostrontium after potato tuber ingestion

The very high deposited activity of ^{90}Sr gave extremely high potato tuber activities at Fureneset; the average concentrations of ^{84}Sr in potato tubers after three (⌀ <1.5 cm) and nine weeks (August) (⌀ >1.5 cm) would reflect ^{90}Sr activities of 8 670 kBq/kg[9] and 14 280 kBq/kg[9], respectively (Table F.1 in Appendix). Tough higher activity was found in the August potato tubers, the quantified values were not statistically different at three and nine weeks. The values are only theoretical and they were *much* greater than the range of measured ^{90}Sr concentrations[10] in Innlandet (Apeslvoll) and Vestlandet (Fureneset) after the Chernobyl accident in 1986 (NOU, 1987:1, p. 33). Grass and precipitation contained 31 Bq/kg[10] ^{90}Sr and 2 Bq/kg[10] ^{90}Sr, respectively, in the county where Fureneset is located. The county where Apelsvoll is located, further to the east and closer to the fallout source, showed higher concentrations, e.g. 68 Bq/kg[10] ^{90}Sr in grass.

[9] Assuming 1 µg ^{84}Sr/kg = 5.1 kBq/g ^{90}Sr (see Appendix F).
[10] Assuming ^{90}Sr is 1 % of the fallout concentration of ^{137}Cs (Salbu, B, personal communication, May 2020).

Ingestion of one kilogram mature, edible potato tubers from August would be much greater than the limit of 100 Bq/kg set for ^{90}Sr in internationally traded commercial food (Codex, 2011). The potato tuber concentration would also exceed the previous limit of 100 strontium units (SU), as 8 670 * 10^3 Bq/kg is higher than 1 SU[11]. Intake of one kilogram potato tubers with activity limit concentration of 100 Bq/kg ^{90}Sr would lead to a radiation dose of 0.5 mSv and 0.2 mSv in children and adults, respectively, the first year after an accident (Codex, 2011). This is lower than the dose limit of 1 mSv/year set for the general public (DSA, 2018). If contamination occurs, ^{90}Sr is estimated to have an ecological half-life of 10-29 years in potato tubers (Aarkrog et al., 1992; Pröhl et al., 2006). If ingested and absorbed, a biological half-life of 18 years could be expected for ^{90}Sr in the human body (Sahoo et al., 2016). However, as most of the ^{84}Sr taken up in potato tubers appeared to accumulate in the periderm layer, and the activity in this layer was low, peeling may be sufficient to remove Sr contamination in potato tubers before ingestion, as suggested by Barthakur et al. (2002).

5.9 Future implications

In order to conclude if ^{84}Sr found in stolons, root hair and potato tubers was a result of soil uptake or translocation, a controlled model experiment would be necessary. Pot experiments applying Sr directly to the soil, or covering the soil and applying Sr directly onto leaves, would give adequate data on this. Using different isotopic labeling for Sr would make it possible to distinguish between soil uptake and foliar translocation within the same plant (Wiech et al., 2018). To determine the importance of water, separating plants with water on foliage and no water on foliage (only to soil) can be done. The radioactive ^{85}Sr can be used to follow different phases of potential translocation or soil uptake through autoradiography.

Adding lime to increase the plant available Ca-Al at Fureneset could be done to investigate countermeasure and remediation methods if a nuclear fallout would occur. Usually, this would not reduce the Sr uptake by more than a factor of three and is only beneficial as long as the base saturation does not exceed 70 % of total base CEC in the soil (Baratta, 1994; Fredriksson et al, 1970 cited in Lönsjö & Haak, 1975). Following the same plot over multiple growing seasons could also be important for countermeasure as tillage of the soil could lead

[11] Calculation based on Chernousenko (1991, p. 252) stating that 1 SU = 0.001 μCi in the skeleton of adults. 1 Bq = $2.7*10^{-11}$ Ci.

to increased Sr uptake in the years following a nuclear fallout (Nunes et al., 2006; Yaroson et al., 2019).

6. Conclusion

Hypothesis one is supported as the results showed that the deposited ^{84}Sr concentration on leaves decreased ($p<0.05$) with time. Hypothesis two is rejected as the uptake of deposited ^{84}Sr in below soil plant tissue was found to be highest at Fureneset and negligible at Apelsvoll. At Fureneset, the ^{84}Sr taken up in potato tubers accumulated in the peel, supporting hypothesis three. The higher uptake in potato tubers and other below soil plant tissues was mainly attributed to elevated soil concentrations of ^{84}Sr. However, hypothesis four cannot be rejected as the elevated ^{84}Sr concentrations in stolons ($p=0.01$) and potato tuber periderm layer ($p=0.05$) at Fureneset could be from translocation.

The results obtained provided information about the safety associated with eating edible parts of the potato plant after fallout of ^{90}Sr following a nuclear event. A worst-case scenario was presented as all of the ^{84}Sr deposited was soluble. The hazard related to uptake in potato tubers after fallout was shown to be affected by factors like soil characteristics, precipitation rate and plant growth stage. The key parameter in determining uptake appeared to be the relationship between the plant available radiostrontium and its chemical homologue Ca, where high plant available Ca-Al reduced the ^{84}Sr uptake.

Significant differences in ^{84}Sr concentrations were found for control and treated groups in leaves and stem at Apelsvoll and Fureneset ($p<0.05$). A foliage (leaf+stem) reduction ($p=0.052$) of ^{84}Sr was only found at Fureneset. The reduction was correlated with precipitation ($r=-0.95$), emphasizing the effect of precipitation before complete absorption of ^{84}Sr in plant tissue. The uptake of ^{84}Sr in below-soil plant tissue, including potato tubers, was only significant at Fureneset ($p<0.05$). This was explained by low plant available Ca-Al, sandy soil and high precipitation rates transporting ^{84}Sr to the soil where it was available for uptake.

The findings indicated that Western Norway and the coastal areas are more vulnerable than Eastern Norway, such as Apelsvoll and the areas around Mjøsa, in terms of uptake of divalent Sr in plants after wet deposition, assuming Fureneset was a good representative for this region. This is an important finding for further research and also for emergency preparedness related to this agroecological region of Norway.

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Appendix

Appendix A Instruments

A.1 UltraCLAVE

Digestion of solid material into liquid form can be done through microwave digestion in an UltraCLAVE. Acid diluted samples are put in tubes (Teflon, glass or quartz) placed in a rack in a reaction chamber. The chamber holds a mixture of water and acids that absorb microwaves and heats up the samples to a constant high temperature (250 °C) and pressure (50 bar) (Milestone, 2008). The pressure is applied by an inert gas to avoid boiling of samples and cross-contamination. Temperature and pressure are gradually reduced, until atmospheric pressure and room temperature is reached, when digestion is done. Digestion in an UltraCLAVE is very beneficial for large sample masses as the only manual operation is loading the rack with tubes into the reaction chamber.

A.2 ICP-OES

The inductively coupled plasma-optical emission spectrometry (ICP-OES) determines element composition in samples by analyzing the excitation spectra (ThermoFisher, 2020). The sample is transformed into an aerosol and placed in plasma where the elements are ionized. When the excited elements return to their natural state, photons (light) are released. As every element has a characteristic emission spectrum, the spectrometer measures the different wavelengths and determines the element composition. Through a calibration the wavelength intensity can be converted into an element concentration.

A.3 Triple quadrupole ICP-MS

Quantification of elements and isotopes can be determined by means of inductively coupled plasma mass spectrometry (ICP-MS). The accuracy is high and the lower detection limits (LOD) are reduced, making it possible to work with challenging trace determination (Wiech et al., 2018). The instrument decomposes the aqueous samples and the constituent elements, in gaseous form, are ionized when passing through a plasma source at 10.000 K. These ions are sorted according to mass/charge (m/z) ratio in the mass spectrometer before being identified and quantified by an electron multiplier detector. The detector creates a signal large enough to be read as a pulse through secondary emission: the analyte strikes a dynode and emits electrons, and these electrons go on to release more electrons in the next dynode, gradually generating an amplified signal (Wilschefski & Baxter, 2019).

A triple quadrupole ICP-MS is more advanced than normal ICP-MS due to its´ greater mass interference removal capabilities, i.e. separating isobaric elements. It has two quadrupoles instead of one, and the additional quadrupole at the front of the instrument is set to selectively analyte mass in the first stage of mass filtration. The first quadrupole preselects the analyte by rejecting unwanted ions and low mass precursors (H, N, O, C) (ThermoFisher, 2017). This removal of the mass precursors eliminates the generation of new, unwanted interferences in the reaction cell. This removal is very important as many elements, e.g. strontium, are prone to polyatomic interferences and overlapping with ions like oxides and chlorides (Wilschefski & Baxter, 2019). The two quadrupoles consist of four cylindrical metal rods around an electrical field (ThermoFischer, 2017). It is the electrical field that filters out masses of interest by creating a specific mass stability voltage range for the given elements. The voltage range is created by applying DC (direct current) and RF/AC (alternating current) voltages. Only a given m/z ratio is stable at a given voltage, and unwanted analyte masses will collide with the metallic rods and be removed. Consequently, only the ion of interest will pass through the quadrupoles and be quantified on a pulse or analog detector.

Appendix B Characteristics of strontium enriched product

CERTIFICATE of Analysis № 190610-0081

Date of Issue: 10 June 2019

Description of product: Strontium-84; Sr-84; ^{84}Strontium; ^{84}Sr. (VP-1)

Chemical form: Sr-84 carbonate ($^{84}SrCO_3$).

Supplier: Neonest AB/www.BuyIsotope.com

Consignee: Norwegian University of Life Sciences (NMBU), Faculty of Environmental Sciences and Natural Resource Managemenet (MINA), Environmental Chemistry – Isotope laboratory, Fougnerbakken 1, NO-1430, Ås, Norway.

CHARACTERISTICS OF THE ISOTOPE-ENRICHED PRODUCT:

1. **Weight of enriched isotopic mixture.**
Amount: Element weight: 50 mg;

2. **Isotopic content.**

Isotope:	Sr-84	Sr-86	Sr-87	Sr-88
Percentage (at. %):	78.30±0.8	4.34	1.91	15.45

3. **Chemical impurities.**

Elements	Cr	Ca	Na	Mg	Al	Si	Ni	Cu	Mn	Pb
Content (weight %)	<0.001	0.024	0.002	0.004	< 0.004	0.006	<0.004	0.001	<0.001	0.01

Elements	Fe	Ba	Mo
Content (weight %)	<0.004	0.006	<0.002

4. **Remarks:**
It is hereby certified that the product has the above characteristics.

Figure B.1: Certificate and characteristics of the ^{84}Sr solution added to the artificial rainwater and sprayed on potato plants at Apelsvoll and Fureneset.

Appendix C Experimental sites – weather data

Table C.1: Average daily wind speed (m/s) at Apelsvoll and Fureneset for the three-week sampling period after spraying (eklima.no).

Date	Apelsvoll (m/s)	Fureneset (m/s)
25.06.2019	0.4	0.8
26.06.2019	1.6	0.9
27.06.2019	1.3	1.5
28.06.2019	1.2	1.0
29.06.2019	1.3	1.1
30.06.2019	1.9	1.7
0.1.07.2019	2.6	2.3
02.07.2019	3.3	1.7
03.07.2019	2.4	1.3
04.07.2019	1.8	2.4
05.07.2019	1.4	1.7
06.07.2019	1.3	1.3
07.07.2019	1.1	1.2
08.07.2019	1.2	2.2
09.07.2019	1.8	1.3
10.07.2019	1.5	1.1
11.07.2019	1.4	0.9
12.07.2019	1.5	1.0
13.07.2019	1.1	0.9
14.07.2019	1.4	1.0
15.07.2019	1.9	0.8
16.07.2019	1.4	1.7
17.07.2019	1.5	1.0

Table C.2: Concentrations (mg/L or µg/L, w/V) of elements, pH and conductivity (µS/cm) in actual rainwater at Apelsvoll and Fureneset, and the NMBU house standard 1643H. Precipitation was sampled in 2016 and the numbers shown are an average of two separate precipitation samples.

Element	Apelsvoll	Fureneset	1643H
Na (mg/L)	0.165	4.2	22
Mg (mg/L)	0.1225	0.495	8.3
Al (µg/L)	39.9	18	160
S (mg/L)	0.2965	0.375	2.5
K (µg/L)	0.735	0.245	1.9
Ca (mg/L)	0.75	0.295	33
V (µg/L)	0.117	0.051	39
Cr (µg/L)	0.175	<LOQ	21
Mn (µg/L)	5.385	0.595	40
Fe (µg/L)	56.95	21	99
Co (µg/L)	0.0739	0.011	28
Ni (µg/L)	0.625	0.19	64
Cu (µg/L)	18	4.1	23
Zn (µg/L)	7.1	2.85	79
As (µg/L)	0.154	<LOD	62
Se (µg/L)	0.084	0.0295	13
Sr (µg/L)	4	3.2	340
Mo (µg/L)	0.1585	<LOQ	130
Ag (µg/L)	<LOQ	<LOD	1.1
Cd (µg/L)	0.01545	<LOQ	6.9
Sb (µg/L)	0.315	0.00985	63
Cs (µg/L)	<LOQ	<LOD	1.1
Ba (µg/L)	8.3	0.405	20
Pb (µg/L)	0.08815	0.29	20
Bi (µg/L)	<LOD	<LOD	<LOD
Th (µg/L)	<LOQ	<LOD	1.1
U (µg/L)	0.00385	0.0135	1.1
pH	5.7	5.7	-
Conductivity (µS/cm)	5	33	-

Appendix D Statistical analysis

D.1 Lognormal distribution

Table D.1.1 shows how ^{84}Sr concentrations in plant tissue were normalized (kurtosis and skewness values between -2 and 2 (West et al., 1995)) when transforming to $\log_{10}$.

Table D.1.1: Normal distribution and lognormal distribution of ^{84}Sr in plant tissue at Apelsvoll and Fureneset.

		Normal distribution			Lognormal distribution		
	n	Kurtosis	Skewness	Mean	Kurtosis	Skewness	Mean
Apelsvoll							
Leaf	18	-0.595	-0.273	1053	0.245	-1.45	2.55
Stem	18	0.945	1.42	444	-0.116	-1.11	2.16
Stolon	20	10.4	3.02	7.5	0.124	0.988	0.676
Potato tuber	18	11.2	3.20	1.07	3.56	1.92	0.215
Fureneset							
Leaf	20	-0.563	0.440	1131	-0.156	-1.31	2.51
Stem	20	2.37	1.70	563	-0.670	-0.889	2.15
Stolon	20	6.65	2.33	44.1	-1.43	-0.248	1.52
Root hair	20	2.29	1.37	63.3	-0.441	-0.642	1.62
Potato tuber	16	1.71	1.71	1.46	0.242	1.12	0.311

D.2 Overall uncertainties

Table D.2.1 shows the p-value after paired two-tailed t-test for different replacement values of <LOD (LOD, LOD/2, LOD/10) at Apelsvoll and Fureneset. The t-test compared the ^{84}Sr-spike concentrations (n=3) in the control with the final concentrations after three weeks (n=3). All values are lognormal (ug/kg). No significant differences were found within a 95% confidence interval for the different values, except for root hair at Fureneset only showing significant differences with LOD/2. Potato tuber samples were the small potato tubers placed in vials (⌀ <1.5 cm). Significant difference between control group and treated plant tissue at three weeks ($p<0.05$) is marked with red numbers.

Table D.2.1: The p-value in different plant tissues after paired two-tailed t-test for different values of <LOD (LOD, LOD/2, LOD/10) at Apelsvoll and Fureneset. The control group is compared with the respective treated group after 3 weeks. Red numbers indicate a significant difference between control and treated group ($p<0.05$).

Plant tissue	LOD	LOD/2	LOD/10
Apelsvoll			
Leaf	0.000123	0.000102	$7.97 * 10^{-5}$
Stem	0.000113	0.00768	0.00622
Stolon	0.423	0.423	0.423
Potato tuber (⌀ <1.5 cm)	0.423	0.423	0.423
Fureneset			
Leaf	0.00116	0.000960	0.000749
Stem	0.0151	0.0182	0.0243
Stolon	0.0170	0.0129	0.00996
Root hair	0.0776	0.000726	0.0612
Potato tuber (⌀ <1.5 cm)	0.215	0.164	0.134

Appendix E Additional figures

Figure E.1: Illustration of a Hardi BP 20 back pack sprayer with a boom of 4 nozzles (last nozzle behind the legs on the woman in the photograph) (Agronom, 2015).

Figure E.2: Peeled potato tubers visually divided into small (left), medium (middle) and large (right).

Appendix F Conversion from ^{84}Sr (µg/kg) to ^{90}Sr (Bq/g)

To calculate activity (Bq) from mass:

$$Bq = [\frac{mass}{mass\ number}\ x\ Avogadro\ constant]x[\frac{\ln 2}{half\ life}]$$

Bq has units:	s^{-1}
Mass has units:	g
Mass number has units:	g mol^{-1}
Avogadro constant has units:	mol^{-1}
Half-life has units:	s
Mass ^{90}Sr:	89.907738 g mol^{-1}
Half-life ^{90}Sr:	28.80 ± 0.07 years (LNHB, 2005)
One year:	365.2422 days
One day:	86 400 seconds

This gave:

$$Bq = \left[\frac{0.776\ x\ 10^{-3}}{89.907\ 738}\ x\ 6.022\ 140\ 76\ x\ 10^{23}\right] x\left[\frac{\ln 2}{28.8\ x\ 365.2422\ x\ 86\ 400}\right]$$

$$Bq = 3.964\ x\ 10^{9}$$

When converting the added ^{84}Sr concentration to ^{90}Sr the activity was almost 4 GBq. This is extremely high and means that 1 µg ^{84}Sr/kg equals 5 100 Bq/g for ^{90}Sr (Karl Andreas Jensen, personal communication, April 2020). The average concentrations in Table F.1 were multiplied by 5 100 Bq/g to get the activity in soil and potato tubers.

Table F.1: Average concentration (µg/kg) of ^{84}Sr and theoretical activity of ^{90}Sr (Bq/g) in soil and potato tuber at Apelsvoll and Fureneset.

	Apelsvoll		Fureneset	
	^{84}Sr (µg/kg)	Sr (Bq/g)	^{84}Sr (µg/kg)	Sr (Bq/g)
Soil (0-5cm), three weeks	4	20 400	1.5	7 650
Potato tuber, three weeks (⌀ <1.5 cm)	-		1.7	8 670
Potato tuber, August (⌀ >1.5 cm)	-		2.8	14 280

Appendix G Raw data

G.1 Soil fractionation

Table G.1: Weight (g) and percentage distribution of the different soil fractions in the different layers. Three soil samples from each site, representing the three different soil layers (0-5, 5-10, 10-15 cm), adding up to six samples in total. The table shows that there is not much variation for the respective fractions in the different soil layers, which is expected due to continuous ploughing over several years.

Fraction	Detail	Weight (g)	Sum fraction (g)	Fraction % [a]
Apelsvoll				
Sand	0-5 cm	5.57	17.3	57
Sand	5-10 cm	5.86		
Sand	10-15 cm	5.87		
Silt	0-5 cm	3	9.22	31
Silt	5-10 cm	3		
Silt	10-15 cm	3.22		
Clay	0-5 cm	1.23	3.66	12
Clay	5-10 cm	1.26		
Clay	10-15 cm	1.17		
Total		30.2		
Fureneset				
Sand	0-5 cm	6.3	18.04	66
Sand	5-10 cm	5.91		
Sand	10-15 cm	5.83		
Silt	0-5 cm	2.88	8.64	31
Silt	5-10 cm	2.68		
Silt	10-15 cm	3.08		
Clay	0-5 cm	0.28	0.75	3
Clay	5-10 cm	0.24		
Clay	10-15 cm	0.23		
Total		27.4		

[a] calculated as (sum fraction weight/total soil weight) x 100

G.2 Plant tissue at Apelsvoll

Table G.2: Raw data for plant tissue at Apelsvoll, quantified using a triple quadrupole ICP-MS. ^{84}Sr is the Sr added with artificial rainwater, where the ^{84}Sr concentration in leaf tissue is calculated as described in Materials and Methods 4.6. Control samples in August are called "August C".

Time	Plant tissue	Weight (g dw)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
Control	leaf	0.251	<LOD	94.6	13.3	140
Control	leaf	0.252	<LOD	68.8	11.6	69.9
Control	leaf	0.254	<LOD	42.1	10.6	39.2
3 hours	leaf	0.253	1900	48.7	9.45	51.8
3 hours	leaf	0.254	1700	74.4	13.3	66.6
3 hours	leaf	0.253	2300	56.2	10.8	57.2
48 hours	leaf	0.147	1600	76.2	14.2	81.4
48 hours	leaf	0.171	1500	86.3	16.1	84.5
48 hours	leaf	0.162	960	49.7	9.91	46.8
1 week	leaf	0.161	1400	96.4	17.7	99.4
1 week	leaf	0.189	1100	105	19.6	119
1 week	leaf	0.216	1600	121	22.5	130
2 weeks	leaf	0.202	1100	68.2	14.2	61.2
2 weeks	leaf	0.214	830	107	18.6	93.3
2 weeks	leaf	0.199	<12.6	101	17	99.4
3 weeks	leaf	0.252	940	150	24.3	150
3 weeks	leaf	0.250	910	115	17.4	127
3 weeks	leaf	0.252	1100	104	17.4	112
Control	stem	0.147	<LOD	98.9	8.22	203
Control	stem	0.245	<LOD	64.6	6.37	85.4
Control	stem	0.211	<LOD	74.5	10.5	89.1
3 hours	stem	0.257	580	63.4	6.38	86.8
3 hours	stem	0.251	470	75.1	7.99	85.4
3 hours	stem	0.251	1500	78	8.99	102
48 hours	stem	0.152	390	81.6	8.9	112
48 hours	stem	0.165	320	89.5	10.6	114
48 hours	stem	0.155	92	76.2	8.79	98.4
1 week	stem	0.165	240	86.1	9.22	121
1 week	stem	0.167	250	82.3	9.88	105
1 week	stem	0.198	420	64	7.06	105
2 weeks	stem	0.158	440	99.2	9.56	129
2 weeks	stem	0.155	190	76.8	6.96	102
2 weeks	stem	0.198	<12.7	86.6	9.16	135
3 weeks	stem	0.251	1400	128	13.8	184
3 weeks	stem	0.251	1400	90.9	9.8	146
3 weeks	stem	0.251	300	112	11.5	178

Time	Plant tissue	Weight (g dw)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
Control	stolon	0.358	<LOD	36.7	2.88	94.9
Control	stolon	0.382	<LOD	42.8	3.48	97.1
Control	stolon	0.351	<LOD	35.1	3.49	81.8
Control	stolon	0.302	<LOD	51.2	5.37	89.6
Control	stolon	0.306	<LOD	29.3	2.33	78.9
3 hours	stolon	0.339	<7.4	40.2	3.35	80.7
3 hours	stolon	0.316	<8	45.9	3.86	86.4
3 hours	stolon	0.292	<8.6	50.2	4.45	77.2
48 hours	stolon	0.319	<7.9	48.6	4.25	85.8
48 hours	stolon	0.311	21	55.6	5.16	84.7
48 hours	stolon	0.359	12	57.5	5.5	81.2
1 week	stolon	0.312	<8.1	37.5	3.3	73
1 week	stolon	0.313	53	43.1	3.78	78.6
1 week	stolon	0.356	16	35.2	3.18	71.8
2 weeks	stolon	0.377	<LOD	81.1	7.22	149
2 weeks	stolon	0.409	<LOD	47.7	3.9	95.1
2 weeks	stolon	0.377	15	45.3	4.15	82.3
3 weeks	stolon	0.306	<LOD	41.6	3.6	72.7
3 weeks	stolon	0.348	<7.2	42.1	3.66	85.3
3 weeks	stolon	0.379	<LOD	26	2.29	61.4
Control	tuber (⌀ <1.5 cm)	0.350	<LOD	15.2	1.19	52.9
Control	tuber (⌀ <1.5 cm)	0.279	<LOD	15.1	1.17	47.6
Control	tuber (⌀ <1.5 cm)	0.355	<LOD	13.8	1.34	43.3
3 hours	tuber (⌀ <1.5 cm)	0.184	<LOD	9.95	0.77	28.3
3 hours	tuber (⌀ <1.5 cm)	0.440	<1.2	11.6	0.936	31.5
3 hours	tuber (⌀ <1.5 cm)	0.225	<LOD	14.6	1.19	44.5
48 hours	tuber (⌀ <1.5 cm)	0.369	<1.4	14.2	1.07	39.6
48 hours	tuber (⌀ <1.5 cm)	0.168	8.5	12.2	1.05	36.6
48 hours	tuber (⌀ <1.5 cm)	0.348	2.1	18.5	1.62	48.4
1 week	tuber (⌀ <1.5 cm)	0.347	1.7	15.7	1.4	42.1
1 week	tuber (⌀ <1.5 cm)	0.061	<LOD	34.5	2.76	69.3
1 week	tuber (⌀ <1.5 cm)	0.376	<1.4	23.2	2.1	55.1
2 weeks	tuber (⌀ <1.5 cm)	0.336	<LOD	11.2	1	35
2 weeks	tuber (⌀ <1.5 cm)	0.375	<LOD	18	1.63	50.9
2 weeks	tuber (⌀ <1.5 cm)	0.413	<LOD	16.4	1.43	45.7
3 weeks	tuber (⌀ <1.5 cm)	0.353	<LOD	14.5	1.3	38.3
3 weeks	tuber (⌀ <1.5 cm)	0.352	3.2	11.7	1.09	36.7
3 weeks	tuber (⌀ <1.5 cm)	0.352	<LOD	9.7	0.93	25.6
Control	core (⌀ <1.5 cm)	0.237	<LOD	3.8	0.335	19.2
Control	peel/rest (⌀ <1.5 cm)	0.350	<LOD	12	0.978	36.9

Time	Plant tissue	Weight (g dw)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
Control	core (⌀ <1.5 cm)	0.075	<LOD	4.38	0.422	16.8
Control	peel/rest (⌀ <1.5 cm)	0.277	<LOD	13	1.3	41.4
Control	core (⌀ <1.5 cm)	0.415	<LOD	4.09	0.418	18
Control	peel/rest (⌀ <1.5 cm)	0.424	<LOD	15.5	1.4	36.2
3 hours	core (⌀ <1.5 cm)	0.146	<LOD	4.89	0.424	16.8
3 hours	peel/rest (⌀ <1.5 cm)	0.369	1.7	14.5	1.19	39.1
3 hours	core (⌀ <1.5 cm)	0.131	<LOD	4.25	0.346	21.5
3 hours	peel/rest (⌀ <1.5 cm)	0.359	<1.4	13	0.947	40.9
48 hours	core (⌀ <1.5 cm)	0.419	<1.2	2.87	0.307	15.1
48 hours	peel/rest (⌀ <1.5 cm)	0.501	<1	8.36	0.734	25.6
48 hours	core (⌀ <1.5 cm)	0.378	<1.3	2.44	0.342	5.71
48 hours	peel/rest (⌀ <1.5 cm)	0.430	3.4	9.68	0.907	15.1
48 hours	core (⌀ <1.5 cm)	0.382	<1.3	3.24	0.354	17.4
48 hours	peel/rest (⌀ <1.5 cm)	0.532	1.4	16.8	1.58	38.4
1 week	core (⌀ <1.5 cm)	0.427	<LOD	0.839	0.101	5.4
1 week	peel/rest (⌀ <1.5 cm)	0.541	1.3	5.61	0.488	16.6
1 week	core (⌀ <1.5 cm)	0.446	<1.1	2.49	0.215	15.7
1 week	peel/rest (⌀ <1.5 cm)	0.489	2.4	11	0.807	29.4
1 week	core (⌀ <1.5 cm)	0.435	<LOD	1.69	0.192	7.9
1 week	peel/rest (⌀ <1.5 cm)	0.502	<1	6.75	0.566	19.6
2 weeks	core (⌀ <1.5 cm)	0.476	<LOD	0.755	0.0833	5.16
2 weeks	peel/rest (⌀ <1.5 cm)	0.533	<LOD	4.98	0.442	12.8
2 weeks	core (⌀ <1.5 cm)	0.515	<LOD	1.47	0.154	10.8
2 weeks	peel/rest (⌀ <1.5 cm)	0.452	<LOD	8.87	0.805	23.5
2 weeks	core (⌀ <1.5 cm)	0.522	<LOD	2.06	0.21	12.7
2 weeks	peel/rest (⌀ <1.5 cm)	0.576	<0.9	9.88	0.923	24.6
3 weeks	core (⌀ <1.5 cm)	0.449	<LOD	1.48	0.148	7.38
3 weeks	peel/rest (⌀ <1.5 cm)	0.668	<0.8	5.71	0.522	15.9
3 weeks	core (⌀ <1.5 cm)	0.538	<LOD	1.56	0.152	4.36
3 weeks	peel/rest (⌀ <1.5 cm)	0.532	<LOD	6.68	0.623	11.9
3 weeks	core (⌀ <1.5 cm)	0.504	<LOD	1.26	0.126	8.2
3 weeks	peel/rest (⌀ <1.5 cm)	0.414	<LOD	6.67	0.629	19.8
August C	tuber (⌀ >1.5 cm)	0.354	<LOD	1.16	0.178	1.81
August C	tuber (⌀ >1.5 cm)	0.358	<LOD	2.94	0.362	7.46
August C	tuber (⌀ >1.5 cm)	0.354	<LOD	2.42	0.253	5.08
August C	tuber (⌀ >1.5 cm)	0.354	<LOD	2.04	0.245	2.65
August C	tuber (⌀ >1.5 cm)	0.354	<LOD	2.48	0.272	7.11
August C	tuber (⌀ >1.5 cm)	0.353	<LOD	3.06	0.365	8.21
August C	tuber (⌀ >1.5 cm)	0.353	<LOD	1.49	0.208	1.53
August C	tuber (⌀ >1.5 cm)	0.352	<LOD	2.7	0.323	7.68

Time	Plant tissue	Weight (g dw)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
August C	tuber (⌀ >1.5 cm)	0.354	<LOD	4.08	0.507	11.7
August	tuber (⌀ >1.5 cm)	0.349	<LOD	3.21	0.28	2.45
August	tuber (⌀ >1.5 cm)	0.357	<1.13	3.11	0.282	4.6
August	tuber (⌀ >1.5 cm)	0.352	<LOD	3.34	0.314	9.62
August	tuber (⌀ >1.5 cm)	0.358	<LOD	3.44	0.313	11.4
August	tuber (⌀ >1.5 cm)	0.360	<LOD	4.74	0.43	14.5
August	tuber (⌀ >1.5 cm)	0.359	<LOD	3.21	0.24	6.17
August	tuber (⌀ >1.5 cm)	0.355	<LOD	3.37	0.329	10.8
August	tuber (⌀ >1.5 cm)	0.356	<LOD	2.38	0.207	7.42
August	tuber (⌀ >1.5 cm)	0.352	<LOD	2.59	0.202	4.92
August C	peel (⌀ >1.5 cm)	0.560	<LOD	10.8	0.976	18.8
August C	peel (⌀ >1.5 cm)	0.417	<LOD	10.7	1.19	22.2
August C	peel (⌀ >1.5 cm)	0.413	<LOD	9.46	0.965	26,00
August C	peel (⌀ >1.5 cm)	0.433	<LOD	8.7	0.872	16,00
August C	peel (⌀ >1.5 cm)	0.419	<LOD	8.94	0.9	17.4
August C	peel (⌀ >1.5 cm)	0.443	<LOD	10.2	0.972	25.4
August	peel (⌀ >1.5 cm)	0.493	<1	13.5	1.21	39.2
August	peel (⌀ >1.5 cm)	0.453	<1.1	10.6	0.972	23.5
August	peel (⌀ >1.5 cm)	0.475	<LOD	8.71	0.848	21.7
August	peel (⌀ >1.5 cm)	0.472	<LOD	7.81	0.6	24.3
August	peel (⌀ >1.5 cm)	0.488	<LOD	10.3	0.831	32.9
August	peel (⌀ >1.5 cm)	0.541	<LOD	7.06	0.578	22.5

G.3 Plant tissue at Fureneset

Table G.3: Raw data for plant tissue at Furenesetl, quantified using a triple quadrupole ICP-MS. ^{84}Sr is the Sr added with artificial rainwater, where the ^{84}Sr concentration in leaf tissue is calculated as described in Materials and Methods 4.6. Control samples in August are called "August C". Rows in yellow indicate that the same replicate is analyzed.

Time	Plant tissue	Weight (g dw)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
Control	leaf	0.163	<LOD	145	10.8	39.9
Control	leaf	0.173	<LOD	137	12	43.2
Control	leaf	0.198	<LOD	114	10	39.9
Control	leaf	0.173	<LOD	109	9.62	30.9
Control	leaf	0.199	<LOD	112	9.69	34.9
3 hours	leaf	0.146	2100	104	9.11	37.9
3 hours	leaf	0.158	2200	153	12.4	51.8
3 hours	leaf	0.147	2400	182	14.7	56
48 hours	leaf	0.161	1900	95.7	8.61	26.5
48 hours	leaf	0.154	2100	144	12.3	48.5
48 hours	leaf	0.178	3100	143	11.7	47.6
1 week	leaf	0.152	930	125	11.7	44.7
1 week	leaf	0.151	910	144	10.7	39.2
1 week	leaf	0.147	1000	142	12.6	44.4
2 weeks	leaf	0.200	910	130	11.5	34.5
2 weeks	leaf	0.199	830	107	8.85	32.6
2 weeks	leaf	0.193	1300	126	10.6	40.2
3 weeks	leaf	0.232	700	123	10.4	28.8
3 weeks	leaf	0.159	1300	102	9.15	29.7
3 weeks	leaf	0.159	940	109	10.1	29.7
3 weeks	leaf	0.158	930	107	9.95	29
3 weeks	leaf	0.219	910	106	9.87	28.1
Control	stem	0.156	<LOD	160	6.76	63.9
Control	stem	0.172	<11.3	129	6.12	61.3
Control	stem	0.149	<LOD	150	7.19	67.2
Control	stem	0.142	<LOD	138	6.63	62.9
Control	stem	0.153	<LOD	131	5.85	64.4
3 hours	stem	0.177	1300	107	4.75	62.1
3 hours	stem	0.143	1000	164	7.55	79.9
3 hours	stem	0.165	890	147	6.43	74.2
48 hours	stem	0.193	2200	170	9.26	82.4
48 hours	stem	0.144	2400	200	9.89	91
48 hours	stem	0.144	680	155	7.52	77.4
1 week	stem	0.144	260	164	8.38	81.9
1 week	stem	0.172	130	121	5	64.1
1 week	stem	0.152	260	193	10.2	92.3

Time	Plant tissue	Weight (g dw)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
2 weeks	stem	0.203	500	125	6.1	59.2
2 weeks	stem	0.154	290	143	6.4	78.2
2 weeks	stem	0.160	290	161	8.62	86.4
3 weeks	stem	0.159	140	134	5.96	76
3 weeks	stem	0.150	280	177	8.71	105
3 weeks	stem	0.161	640	150	7.06	96.2
Control	stolon	0.189	<LOD	47.7	2.08	8.75
Control	stolon	0.176	<LOD	39.7	1.92	7.34
Control	stolon	0.222	<LOD	77.9	3.81	16.3
Control	stolon	0.289	<LOD	68.9	3.37	10.1
Control	stolon	0.230	<LOD	59.1	2.61	14.1
3 hours	stolon	0.202	6.3	45.9	2.27	8.14
3 hours	stolon	0.228	<11.1	45.2	2.08	10.8
3 hours	stolon	0.300	26	48.3	2.24	10.2
48 hours	stolon	0.157	73	104	4.82	19.2
48 hours	stolon	0.249	48	93	4.57	21.6
48 hours	stolon	0.236	<10.7	94.3	4.4	27.5
1 week	stolon	0.243	40	71.5	3.32	17.4
1 week	stolon	0.158	130	57.2	2.26	14.2
1 week	stolon	0.217	52	106	5.5	25.1
2 weeks	stolon	0.258	61	57.1	2.7	11.9
2 weeks	stolon	0.145	240	64.9	2.8	13.8
2 weeks	stolon	0.242	42	86.4	4.09	15.6
3 weeks	stolon	0.311	45	52.7	2.33	10.3
3 weeks	stolon	0.413	82	55.4	2.65	10.3
3 weeks	stolon	0.301	23	69.6	3.24	16.6
Control	root hair	0.170	<14.8	188	8.53	44.3
Control	root hair	0.197	<12.8	229	13.6	116
Control	root hair	0.204	<LOD	199	9.79	40.8
Control	root hair	0.218	<LOD	206	9.88	41.6
Control	root hair	0.260	<LOD	210	9.89	49.1
3 hours	root hair	0.182	62	179	8.38	39.9
3 hours	root hair	0.086	150	186	9.17	40.1
3 hours	root hair	0.251	70	177	8.92	43.3
48 hours	root hair	0.192	74	213	10.8	43.9
48 hours	root hair	0.167	220	179	9.22	44.4
48 hours	root hair	0.223	20	140	7.05	34.9
1 week	root hair	0.225	100	208	9.9	47.4
1 week	root hair	0.211	100	182	7.54	42.4
1 week	root hair	0.168	120	178	8.95	40.5

Time	Plant tissue	Weight (g dw)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
2 weeks	root hair	0.282	54	167	7.57	36.3
2 weeks	root hair	0.247	32	162	6.81	36.1
2 weeks	root hair	0.271	59	201	9.37	40.3
3 weeks	root hair	0.210	63	190	8.45	28.2
3 weeks	root hair	0.228	66	153	7.03	36.4
3 weeks	root hair	0.210	14	195	9.56	35.3
Control	tuber (⌀ <1.5 cm)	0.340	<LOD	19.2	0.844	4.35
Control	tuber (⌀ <1.5 cm)	0.356	<LOD	16.1	0.823	2.98
Control	tuber (⌀ <1.5 cm)	0.181	<LOD	34.5	1.75	3.18
3 hours	tuber (⌀ <1.5 cm)	0.255	<1.59	34.8	1.96	4.5
3 hours	tuber (⌀ <1.5 cm)	0.381	5.5	16.8	0.754	3.17
48 hours	tuber (⌀ <1.5 cm)	0.315	5.5	38.4	1.82	7.85
48 hours	tuber (⌀ <1.5 cm)	0.366	<1.11	21.4	1.02	3.74
1 week	tuber (⌀ <1.5 cm)	0.349	1.6	23.4	1.16	4.34
1 week	tuber (⌀ <1.5 cm)	0.149	<LOD	22.6	1.05	4.52
1 week	tuber (⌀ <1.5 cm)	0.321	1.6	26.3	1.34	6.34
2 weeks	tuber (⌀ <1.5 cm)	0.356	<1.14	14.7	0.635	2.92
2 weeks	tuber (⌀ <1.5 cm)	0.215	<1.89	17.1	0.742	3.25
2 weeks	tuber (⌀ <1.5 cm)	0.338	<1.2	15.5	0.764	2.5
3 weeks	tuber (⌀ <1.5 cm)	0.371	<1.09	17.3	0.792	2.49
3 weeks	tuber (⌀ <1.5 cm)	0.343	<1.18	11.4	0.559	2.61
3 weeks	tuber (⌀ <1.5 cm)	0.33	3.7	20	0.981	4.66
3 weeks	tuber (⌀ <1.5 cm)	0.327	4.2	21.8	1.05	4.86
Control	core (⌀ <1.5 cm)	0.378	<LOD	5.27	0.322	1.24
Control	peel/rest (⌀ <1.5 cm)	0.369	<LOD	17.8	0.701	2.93
Control	core (⌀ <1.5 cm)	0.389	<LOD	5.55	0.29	1.46
Control	peel/rest (⌀ <1.5 cm)	0.397	<LOD	20.2	1.03	3.72
Control	core (⌀ <1.5 cm)	0.402	<LOD	5.35	0.309	1.25
Control	peel/rest (⌀ <1.5 cm)	0.375	<LOD	16.6	0.737	2.99
3 hours	core (⌀ <1.5 cm)	0.408	<LOD	6.58	0.366	1.52
3 hours	peel/rest (⌀ <1.5 cm)	0.386	<LOD	23.4	1.14	3.32
3 hours	core (⌀ <1.5 cm)	0.407	<LOD	5.83	0.313	1.77
3 hours	peel/rest (⌀ <1.5 cm)	0.376	<LOD	19.4	0.792	4.43
48 hours	core (⌀ <1.5 cm)	0.353	<1.15	5.35	0.287	1.64
48 hours	peel/rest (⌀ <1.5 cm)	0.377	2.4	19.5	0.923	3.58
48 hours	core (⌀ <1.5 cm)	0.397	<LOD	4.61	0.304	1.17
48 hours	peel/rest (⌀ <1.5 cm)	0.403	<1.01	21.6	0.949	3.2
48 hours	core (⌀ <1.5 cm)	0.362	<1.12	5.02	0.283	1.47
48 hours	peel/rest (⌀ <1.5 cm)	0.403	2	15.7	0.653	3.17
1 week	core (⌀ <1.5 cm)	0.358	<LOD	5.46	0.264	1.59

Time	Plant tissue	Weight (g dw)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
1 week	peel/rest (⌀ <1.5 cm)	0.387	<1.05	17.9	0.783	3.31
1 week	core (⌀ <1.5 cm)	0.381	<1.07	4.54	0.251	1.42
1 week	peel/rest (⌀ <1.5 cm)	0.409	2.4	17.6	0.768	3.25
1 week	core (⌀ <1.5 cm)	0.355	<LOD	4.24	0.21	1.49
1 week	peel/rest (⌀ <1.5 cm)	0.371	<LOD	14.3	0.546	3.19
2 weeks	core (⌀ <1.5 cm)	0.382	1.1	4.73	0.264	1.27
2 weeks	peel/rest (⌀ <1.5 cm)	0.376	2.4	15.4	0.674	2.67
2 weeks	core (⌀ <1.5 cm)	0.397	1.8	4.13	0.216	1.19
2 weeks	peel/rest (⌀ <1.5 cm)	0.369	7.5	12.5	0.495	2.38
2 weeks	core (⌀ <1.5 cm)	0.414	<0.98	6.41	0.378	1.49
2 weeks	peel/rest (⌀ <1.5 cm)	0.383	2.3	24.2	1.12	3.29
3 weeks	core (⌀ <1.5 cm)	0.411	6	2.9	0.149	0.967
3 weeks	peel/rest (⌀ <1.5 cm)	0.398	35	10.5	0.458	2.23
3 weeks	core (⌀ <1.5 cm)	0.414	<LOD	2.07	0.13	0.668
3 weeks	peel/rest (⌀ <1.5 cm)	0.366	<1.11	11.3	0.437	1.94
3 weeks	core (⌀ <1.5 cm)	0.398	<LOD	2.77	0.16	0.798
3 weeks	peel/rest (⌀ <1.5 cm)	0.395	1.8	15.7	0.711	1.94
August C	tuber (⌀ >1.5 cm)	0.357	<LOD	8.67	0.479	1.71
August C	tuber (⌀ >1.5 cm)	0.353	<LOD	7.56	0.423	1.44
August C	tuber (⌀ >1.5 cm)	0.351	<LOD	4.39	0.257	0.935
August C	tuber (⌀ >1.5 cm)	0.355	<LOD	4.32	0.21	1.15
August C	tuber (⌀ >1.5 cm)	0.351	<LOD	11.7	0.579	1.75
August C	tuber (⌀ >1.5 cm)	0.354	<LOD	3.62	0.168	0.816
August C	tuber (⌀ >1.5 cm)	0.351	<LOD	5.02	0.229	1.41
August C	tuber (⌀ >1.5 cm)	0.353	<LOD	4.09	0.208	0.979
August C	tuber (⌀ >1.5 cm)	0.354	<LOD	5.67	0.291	1.11
August	tuber (⌀ >1.5 cm)	0.355	<LOD	7.01	0.319	1.76
August	tuber (⌀ >1.5 cm)	0.355	<1.14	7.01	0.319	2.83
August	tuber (⌀ >1.5 cm)	0.353	8	9.28	0.405	1.69
August	tuber (⌀ >1.5 cm)	0.356	<LOD	6.37	0.319	1.99
August	tuber (⌀ >1.5 cm)	0.353	<LOD	7.09	0.34	1.39
August	tuber (⌀ >1.5 cm)	0.358	<LOD	7.88	0.355	1.88
August	tuber (⌀ >1.5 cm)	0.358	12	13.9	0.718	2.56
August	tuber (⌀ >1.5 cm)	0.355	3.3	3.78	0.195	0.774
August	tuber (⌀ >1.5 cm)	0.358	<1.13	3.28	0.168	0.755
August C	peel (⌀ >1.5 cm)	0.450	<1.1	14.9	0.799	3
August C	peel (⌀ >1.5 cm)	0.488	<LOD	18.9	1.04	4.37
August C	peel (⌀ >1.5 cm)	0.543	<LOD	9.4	0.451	1.95
August C	peel (⌀ >1.5 cm)	0.458	<LOD	13.7	0.613	2.2
August C	peel (⌀ >1.5 cm)	0.455	<1.1	22.5	1.03	3.93

Time	Plant tissue	Weight (g dw)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
August C	peel (⌀ >1.5 cm)	0.423	<LOD	17.6	0.773	4.13
August	peel (⌀ >1.5 cm)	0.500	1	14.6	0.619	3.97
August	peel (⌀ >1.5 cm)	0.484	1.3	15.3	0.669	4.54
August	peel (⌀ >1.5 cm)	0.459	1.6	23.1	1.01	4.01
August	peel (⌀ >1.5 cm)	0.519	1.5	18.5	0.951	6.51
August	peel (⌀ >1.5 cm)	0.495	6.4	11	0.5	3.07
August	peel (⌀ >1.5 cm)	0.487	2.4	10.3	0.45	3.94
August C	core (⌀ >1.5 cm)	0.387	<LOD	0.979	0.0702	0.23
August C	peel/rest (⌀ >1.5 cm)	0.499	<LOD	4.4	0.225	0.778
August C	core (⌀ >1.5 cm)	0.430	<LOD	0.987	0.0579	0.386
August C	peel/rest (⌀ >1.5 cm)	0.509	<LOD	2.95	0.129	0.862
August C	core (⌀ >1.5 cm)	0.408	<LOD	1.41	0.0928	0.379
August C	peel/rest (⌀ >1.5 cm)	0.461	<LOD	3.66	0.177	0.748
August	core (⌀ >1.5 cm)	0.425	<LOD	1.74	0.101	0.385
August	peel/rest (⌀ >1.5 cm)	0.428	<0.95	4.21	0.203	0.84
August	core (⌀ >1.5 cm)	0.432	<0.94	1.96	0.114	0.472
August	peel/rest (⌀ >1.5 cm)	0.487	<0.83	5.31	0.262	1.04
August	core (⌀ >1.5 cm)	0.419	<LOD	0.714	0.0642	0.219
August	peel/rest (⌀ >1.5 cm)	0.492	<0.82	3.65	0.164	0.971

G.4 Total element fraction in soil quantified using ICP-MS

Table G.4: Raw data for all soil samples analyzed using a triple quadrupole ICP-MS. For August samples: August C = control and August T = treated (sprayed with ^{84}Sr).

Time	Soil layer	Weight (g)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
Apelsvoll						
Control	0-5 cm	0.254	<LOD	49.3	3.09	537
Control	0-5 cm	0.253	<LOD	42	3.18	472
Control	0-5 cm	0.252	<LOD	58.9	5.01	536
3 weeks	0-5 cm	0.258	<LOD	50.9	3.34	558
3 weeks	0-5 cm	0.259	<LOD	47.9	3.42	515
3 weeks	0-5 cm	0.258	<LOD	49	3.89	481
August C	0-15 cm	0.267	<LOD	79.1	4.87	577
August C	0-15 cm	0.296	<LOD	62.8	4.14	533
August C	0-15 cm	0.275	<LOD	59.2	4.25	538
August T	0-15 cm	0.255	<LOD	34.4	2.19	290
August T	0-15 cm	0.267	<LOD	63.2	2.88	490
August T	0-15 cm	0.277	<LOD	49.9	2.48	517
Fureneset						
Control	0-5 cm	0.261	<LOD	407	19.6	91.8
Control	0-5 cm	0.252	<LOD	376	19.1	85.8
Control	0-5 cm	0.280	<LOD	364	20.2	82.2
1 week	0-5 cm	0.255	<LOD	333	18.8	92.1
1 week	0-5 cm	0.269	<LOD	370	20.5	89.2
1 week	0-5 cm	0.279	<37	384	19.9	87.4
3 weeks	0-5 cm	0.265	<38	383	21.5	97.9
3 weeks	0-5 cm	0.304	<LOD	336	19.8	102
3 weeks	0-5 cm	0.293	<35	371	19.9	85.6
August C	0-5 cm	0.249	<LOD	340	17.8	96.7
August C	5-10 cm	0.263	<LOD	322	16.9	95.2
August C	10-15 cm	0.257	<LOD	331	17.5	86.9
August C	0-5 cm	0.260	<LOD	329	19.7	88.1
August C	5-10 cm	0.249	<LOD	342	20.4	87.2
August C	10-15 cm	0.262	<LOD	361	20.4	96.9
August C	0-5 cm	0.271	<38	323	17.7	81.8
August C	5-10 cm	0.256	<LOD	350	18.5	87.3
August C	10-15 cm	0.273	<LOD	352	18.7	83.8
August T	0-5 cm	0.274	<LOD	364	18.7	91.8
August T	5-10 cm	0.275	<37	312	17.6	82.7
August T	10-15 cm	0.268	<LOD	343	19.6	136
August T	0-5 cm	0.259	<LOD	309	18.1	89.8
August T	5-10 cm	0.256	<LOD	337	18.1	90.8

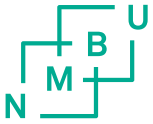
Time	Soil layer	Weight (g)	^{84}Sr (µg/kg)	Tot-Sr (mg/kg)	Ca (g/kg)	Ba (mg/kg)
Fureneset						
August T	10-15 cm	0.275	<LOD	355	19.4	97.8
August T	0-5 cm	0.299	<34	315	19.1	87.8
August T	5-10 cm	0.261	<LOD	328	18.9	83.6
August T	10-15 cm	0.283	<LOD	334	18.9	89.7

G.5 Plant available fraction in soil quantified using ICP-OES

Table G.5: Plant available fraction of elements in soil at Apelsvoll and Fureneset. All elements are quantified using ICP-OES, except for ^{84}Sr-Al quantified using ICP-MS. Each sampling time layer is represented by two replicates analyzed multiple times.

Time	Soil layer	Al-Al (g/kg)	Ca-Al (g/kg)	K-Al (g/kg)	Mg-Al g/kg	P-Al (g/kg)	Tot-Sr-Al (g/kg)	^{84}Sr-Al (µg/L)
Apelsvoll								
Control	0-5 cm	0.283	1.19	0.12	0.0878	0.0432	0.0147	<1.6
Control	0-5 cm	0.264	1.2	0.114	0.103	0.0396	0.0146	<LOD
Control	0-5 cm	0.307	1.24	0.121	0.0922	0.0469	0.0153	<LOD
Control	0-5 cm	0.262	2.02	0.0903	0.12	0.0489	0.0192	<LOD
Control	0-5 cm	0.262	1.97	0.0909	0.113	0.0491	0.0188	<LOD
Control	5-10 cm	0.291	1.22	0.102	0.0906	0.0402	0.0145	<LOD
Control	5-10 cm	0.295	1.24	0.104	0.0916	0.0426	0.0149	<LOD
Control	5-10 cm	0.26	1.83	0.126	0.116	0.061	0.0189	<LOD
Control	5-10 cm	0.258	1.83	0.126	0.116	0.06	0.0186	<1.6
Control	5-10 cm	0.291	1.85	0.0901	0.107	0.0527	0.0186	<LOD
Control	10-15 cm	0.303	1.79	0.0726	0.101	0.0426	0.0164	<LOD
Control	10-15 cm	0.297	1.79	0.0734	0.105	0.0408	0.0164	<LOD
Control	10-15 cm	0.293	1.78	0.0738	0.106	0.04	0.0164	<LOD
Control	10-15 cm	0.313	1.29	0.11	0.098	0.0523	0.0121	<LOD
Control	10-15 cm	0.315	1.29	0.111	0.0998	0.0535	0.0122	<LOD
3 weeks	0-5 cm	0.299	1.41	0.134	0.102	0.0549	0.0174	4.4
3 weeks	0-5 cm	0.27	1.35	0.126	0.101	0.0465	0.0161	5.2
3 weeks	0-5 cm	0.309	1.51	0.134	0.106	0.0535	0.0171	2.5
3 weeks	0-5 cm	0.307	1.5	0.134	0.106	0.0535	0.0168	2.5
3 weeks	5-10 cm	0.329	1.29	0.161	0.0978	0.0778	0.0173	<1.6
3 weeks	5-10 cm	0.303	1.25	0.151	0.0989	0.0681	0.0164	<1.6
3 weeks	5-10 cm	0.299	1.4	0.103	0.0973	0.0468	0.0155	<LOD
3 weeks	5-10 cm	0.313	1.42	0.104	0.0981	0.0492	0.0159	<LOD
3 weeks	10-15 cm	0.259	1.24	0.0886	0.0917	0.0386	0.0147	<LOD
3 weeks	10-15 cm	0.313	1.36	0.096	0.101	0.0492	0.0164	<1.6
3 weeks	10-15 cm	0.324	1.55	0.0784	0.109	0.0626	0.0181	<LOD
3 weeks	10-15 cm	0.336	1.51	0.0762	0.101	0.0695	0.0179	<LOD
Fureneset								
Control	0-5 cm	1.08	0.536	0.142	0.038	0.0794	0.0105	<LOD
Control	0-5 cm	0.95	0.47	0.128	0.0308	0.0734	0.0094	<LOD
Control	0-5 cm	0.948	0.498	0.135	0.0334	0.0695	0.00976	<LOD
Control	0-5 cm	1.02	0.582	0.219	0.0434	0.0874	0.0121	<LOD
Control	0-5 cm	0.973	0.534	0.207	0.0368	0.0898	0.0117	<LOD
Control	5-10 cm	0.979	0.647	0.161	0.042	0.104	0.0129	<LOD
Control	5-10 cm	0.983	0.655	0.162	0.0438	0.104	0.013	<LOD

Time	Soil layer	Al-Al (g/kg)	Ca-Al (g/kg)	K-Al (g/kg)	Mg-Al g/kg	P-Al (g/kg)	Tot-Sr-Al (g/kg)	^{84}Sr-Al (µg/L)
Fureneset								
Control	5-10 cm	0.925	0.57	0.225	0.0669	0.0699	0.0107	<LOD
Control	5-10 cm	0.988	0.598	0.225	0.0667	0.081	0.0119	<LOD
Control	5-10 cm	0.938	0.474	0.106	0.037	0.0527	0.00912	<LOD
Control	10-15 cm	0.969	0.791	0.0429	0.0462	0.0652	0.0145	<LOD
Control	10-15 cm	1.04	0.857	0.0473	0.0513	0.0697	0.0155	<LOD
Control	10-15 cm	1.07	0.855	0.0477	0.0501	0.0726	0.0156	<1.6
Control	10-15 cm	0.914	0.635	0.153	0.0724	0.0794	0.0118	<LOD
Control	10-15 cm	0.956	0.639	0.16	0.072	0.0794	0.0117	<LOD
3 weeks	0-5 cm	0.864	0.354	0.139	0.0306	0.0564	0.00727	1.8
3 weeks	0-5 cm	0.891	0.378	0.139	0.036	0.0585	0.00765	1.9
3 weeks	0-5 cm	0.809	0.328	0.0885	0.0328	0.0636	0.00711	<1.6
3 weeks	0-5 cm	0.828	0.332	0.0911	0.0316	0.0638	0.00725	<1.6
3 weeks	5-10 cm	0.96	0.434	0.0698	0.0334	0.047	0.00819	<LOD
3 weeks	5-10 cm	0.966	0.45	0.0726	0.0364	0.0478	0.00835	<LOD
3 weeks	5-10 cm	0.874	0.366	0.0509	0.0368	0.0605	0.00755	<LOD
3 weeks	5-10 cm	0.895	0.354	0.0495	0.0336	0.057	0.00737	<LOD
3 weeks	10-15 cm	0.925	0.426	0.029	0.0336	0.0513	0.00781	<LOD
3 weeks	10-15 cm	0.933	0.414	0.0284	0.0308	0.0466	0.00753	<LOD
3 weeks	10-15 cm	0.948	0.492	0.0324	0.0382	0.0585	0.0102	<LOD
3 weeks	10-15 cm	0.824	0.476	0.0308	0.0408	0.0425	0.0091	<LOD



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