Environmental Carbon-14 Monitoring in Marine Ecosystems Near Nuclear Power Plants

A Methodological Intercomparison of Accelerator Mass Spectrometry, Liquid Scintillation Counting, and SCAR Spectroscopy

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Abstract

Environmental monitoring of Carbon-14 (^{14}C) around nuclear power plants (NPPs) has traditionally prioritized gaseous emissions, leaving waterborne liquid discharges unmonitored due to low nominal volumes ($<0.5\%$) and analytical challenges using traditional techniques. However, liquid releases do not experience as effective dilution as gas releases, instead discharging directly into coastal ecosystems where ^{14}C accumulates in marine organisms, raising localized specific activities above regional background levels. This thesis presents a methodological intercomparison evaluating three analytical technologies for marine monitoring: Accelerator Mass Spectrometry (AMS), Liquid Scintillation Counting (LSC), and laser-based Saturated-Absorption Cavity Ring-down Spectroscopy (SCAR).

Biological and environmental samples collected near the Ringhals NPP cooling water outlets were analyzed across five international facilities in Lund (Sweden), Debrecen (Hungary), Mangalore (India), Borås (Sweden), and Florence (Italy). The technologies were evaluated based on, for example, precision, sample throughput, investment costs, labor requirements, and automation.

Findings confirm that while AMS offers superior precision, its high capital costs make it impractical for routine industrial use. Traditional LSC has low investment barriers but suffers from throughput limitations due to intensive manual chemistry and long decay-counting times. The more recent addition to the market, SCAR spectroscopy, effectively bridges this gap; by analyzing CO_2 gas directly and eliminating graphitization, it delivers near-AMS precision, high automation, and rapid turnaround times for a fraction of the capital investment. The study concludes that routine liquid emission monitoring using automated systems like SCAR is fully justified under the ALARA principle to ensure transparency, public safety, and institutional trust.

Ny laserteknik gör det enkelt att övervaka radioaktivt kol i havsmiljöer

Idag mäts inte flytande utsläpp av kol-14 från kärnkraftverk, trots att isotopen anrikas i marina organismer. Ett nytt examensarbete visar att modern laserteknik kan göra övervakningen både billig och effektiv.

Kärnkraftverk släpper regelmässigt ut små mängder radioaktivt kol, så kallat kol-14. Eftersom kol är livets byggsten tas det snabbt upp av allt levande. I Sverige mäts sedan 2002 de gasformiga utsläppen i luften, men de flytande utsläppen som spolas ut med kylvattnet i havet mäts inte. Detta beror på att de utgör en mycket liten andel, mindre än 0,5 %, av de totala nominella utsläppen och historiskt har varit svåra att analysera med traditionella metoder.

Men till skillnad från gaser i luften, som sprids och späds ut effektivt, hamnar de flytande utsläppen direkt i det lokala marina ekosystemet. Studier nära Ringhals kärnkraftverk visar att marina organismer som lever nära kylvattenutsläppet kan få halter av kol-14 som är flera gånger högre än den naturliga bakgrundsnivån. Att övervaka dessa utsläpp handlar om ren strålsäkerhet, men också om öppenhet och att allmänheten ska kunna känna förtroende för både kärnkraftsindustrin och tillsynsmyndigheter.

I detta examensarbete utvärderades tre olika mättekniker för att hitta en lämplig väg framåt för miljöövervakning av de flytande utsläppen av kol-14. Biologiska prover i form av blåmusslor, tång och rödspätta insamlade nära Ringhals analyserades vid fem olika laboratorier runtom i världen: i Lund, Borås, Ungern, Italien och Indien.

De tekniker som jämfördes var traditionell acceleratorbaserad masspektrometri (AMS), radioaktiv sönderfallsmätning (LSC) och en nyare laserteknik kallad SCAR-spektroskopi.

Resultaten bekräftar att den tekniskt avancerade partikelacceleratortekniken (AMS) erbjuder bäst precision, men dess enorma investeringskostnader gör den opraktisk för rutinmässig industrianvändning. Den äldre metoden (LSC) har lägre inköpskostnad men lider av begränsningar, särskilt om man vill uppnå hög precision, som stora provmägnder och hög tidsåtgång, såväl under mätningarna som under provförberedelsen. Den nya lasertekniken (SCAR) visar sig däremot överbrygga detta gap. Genom att analysera koldioxidgas direkt, och därmed korta ner provberedningstiden, levererar den de snabbaste svaren till högre precision än standard-LSC och till en bråkdel av kostnaden av AMS.

Arbetet drar slutsatsen att en rutinmässig mätning av flytande utsläpp med hjälp av automatiserade system som SCAR är möjlig och motiverad. Genom att införa detta kan industrin bättre leva upp till den grundläggande ALARA-principen, det vill säga att hålla stråldoser så låga som det bara är rimligt möjligt, och därmed säkerställa transparens och samhälleligt förtroende.

Preface

This master's thesis marks the final milestone of my five years of studies at the Faculty of Engineering (LTH), Lund University.

First and foremost, I would like to express my deepest gratitude to my supervisors, Kristina Eriksson Stenström from Lund University and Karin Aquilonius from the Swedish Radiation Safety Authority (Strålsäkerhetsmyndigheten). Thank you for your guidance and patience. Your encouragement and positive energy kept my spirits high throughout the entire process. Further, I want to extend a warm thank you to all the researchers and staff I met during my travels who welcomed me so generously into your laboratories and took the time to teach me so much.

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Finally, I want to thank my family and friends. Your support and understanding have been my foundation over the past five years in general, and during the course of this thesis in specific. I could not have completed this journey without you.

Lund, May 2026

Abbreviations and units

AGE Automated Graphitization Equipment

ALARA As Low As Reasonably Achievable

AMS Accelerator Mass Spectrometry

Bq Becquerel

CARER Centre for Advanced Research in Environmental Radioactivity

CHF Swiss franc

CO_2 Carbon dioxide

CRDS Cavity Ring-Down Spectroscopy

DIC Dissolved Inorganic Carbon

DOC Dissolved Organic Carbon

EA Elementar Analyzer

$F^{14}C$ Fraction Modern

LEA Low Energy Accelerator

LSC Liquid Scintillation Counting

MICADAS Mini Radiocarbon Dating System

NEC National Electrostatics Corporation

NPP Nuclear Power Plant

pMC Percent Modern Carbon

PMT PhotoMultiplier Tube

RISE Research Institutes of Sweden

SCAR Saturated-Absorption Cavity Ring-Down Spectroscopy

SEK Swedish Krona (Swedish Crowns)

SSAMS Single-Stage Accelerator Mass Spectrometer

USD United States Dollar

Contents

1 Introduction

Carbon-14 (^{14}C or radiocarbon) has a unique position in radiological protection and environmental monitoring due to its dual nature as a long-lived radionuclide, with a half-life of 5730 years, and a fundamental building block of all living matter [1]. While it is naturally produced in the upper atmosphere by cosmic ray interactions with nitrogen [2], anthropogenic activities have significantly increased the global ^{14}C inventory. After the atmospheric peaks caused by testing of nuclear weapons in the mid 20th century, the usage of Nuclear Power Plants (NPPs) now is the main ongoing source of anthropogenic ^{14}C releases [2]. Because of its long half-life and high mobility within the global carbon cycle, gaseous or liquid release of ^{14}C cannot be locally contained, increasing the global inventory and delivering low, multi-generational collective doses to human populations [3].

In Sweden, airborne emissions of ^{14}C via NPP ventilation systems have been measured since 2002 [4]. However, similar measurements for liquid releases into cooling water outlets does not currently exist. This is partly due to liquid releases constituting less than 0.5% of the total nominal discharge, along with the analytical challenges involved in monitoring the liquid releases [4]. Despite their low volume, liquid releases do not dilute in the same way as atmospheric releases, instead they are discharged directly into coastal waters, where they are assimilated into the marine food web [4]. Recent studies near facilities like the Ringhals NPP reveal that organisms that live close to the cooling water outlet, especially those higher up the trophic chain can show ^{14}C specific activities that are several times above background levels. This effective accumulation creates substantial localized variance, which highlights why there may be a need for environmental monitoring strategies for marine environments.

Quantifying ^{14}C has traditionally relied on two very different methodologies: Accelerator Mass Spectrometry (AMS) and decay counting methods, such as Liquid Scintillation Counting (LSC). AMS instruments count individual ions directly, giving them high sensitivity [5]. However, AMS requires massive capital investments, specialized personnel, and often intricate sample preparation. On the other hand, LSC, measuring radioactive decays rather than counting ions, provides ^{14}C-analysis for much smaller capital investment costs but in turn require a lot of time and sample material, that needs to go through extensive chemical pretreatment [6].

A much more recent addition to the market is Saturated-absorption Cavity Ringdown spectroscopy (SCAR). It provides an alternative to traditional accelerator-based or decay counting methods by targeting specific molecular transitions of $^{14}CO_2$ within an optical cavity with a laser [7]. SCAR has the potential of bridging the gap between AMS and LSC by approaching AMS precision and sample size requirements, for a lower capital investment cost.

This thesis presents a methodological intercomparison evaluating these three techniques for the purpose of environmental monitoring. Real biological and environmental samples collected from the vicinity of Ringhals NPP were processed and analyzed across multiple European and international facilities—including the SSAMS laboratory at Lund University in Sweden, the MICADAS and LEA laboratory at Atomki

in Debrecen, Hungary, the low-background LSC laboratory at CARER in Mangalore, India, and SCAR laboratories operated by RISE in Borås, Sweden and ppqSense in Florence, Italy. The aim of this study is to critically evaluate these technologies based on different operational dimensions—including precision, sample throughput, automation, and investment cost—in the end defining a pathway for modern nuclear environmental monitoring.

This thesis is structured as follows: Section 1 introduces the environmental relevance of Carbon-14 monitoring and outlines the core objective of the study. Section 2 provides a theoretical background on carbon isotopes, anthropogenic ^{14}C production mechanisms, and the underlying physical principles behind AMS, LSC, and SCAR. Section 3 details the materials and methods used, documenting the biological samples harvested near Ringhals NPP and describing the operational profiles of the participating laboratories in Lund, Debrecen, Mangalore, Borås, and Florence. Section 4 delivers the experimental findings and precision checks across the methodologies. Section 5 presents a critical discussion comparing the economic, practical, and throughput dimensions of the systems, concluding with recommendations for future environmental surveillance strategies.

2 Background

2.1 Carbon and its isotopes

Carbon is known as the building block of life. It has four valence electrons, enabling it to create strong covalent bonds with an array of different elements. Of all known chemical compounds, 94% contain carbon [8], and its presence in proteins, carbohydrates, lipids, and nucleotides makes it fundamental to the concept of life as we know it [8].

There are three naturally occurring carbon isotopes, of which ^{12}C is by far the most abundant, with a share of 98.9% [8] of all carbon. The second isotope of carbon that is found most frequently is ^{13}C with a share of 1.1% [8]. The third isotope, radioactive ^{14}C, is only found in trace amounts [8], but due to the large amount of carbon atoms present, this tiny share corresponds to a large number of ^{14}C-atoms. The global atmospheric inventory of ^{14}C is estimated to be $140 \cdot 10^{15}$ Bq [9], with the unit corresponding to one radioactive decay per second. Considering that the atmosphere is not the only host of ^{14}C – the isotope is also found in the geosphere, hydrosphere, and biosphere alike – and that the halftime of the isotope is 5730 years [10], meaning that the decay rate is slow, it becomes evident that ^{14}C exists in abundance all around us.

^{14}C undergoes β^--decay with the following procedure:

$$^{14}_{6}\mathrm{C} \rightarrow ^{14}_{7}\mathrm{N} + \beta^- + \bar{\nu}_e$$

This is considered a low energy decay, with maximum energy of 156 keV [11]. The three products of the decay share the released energy, which results in the β-particle, in this case an electron, being able to carry any energy lower than 156 keV according to the distribution seen in figure 1. The average energy of the β-particle is 49 keV. This reduces the penetrating power of the decay, with the β-particle having a maximum reach of 0.28 mm in human tissue [12], and 22 cm in air [13].

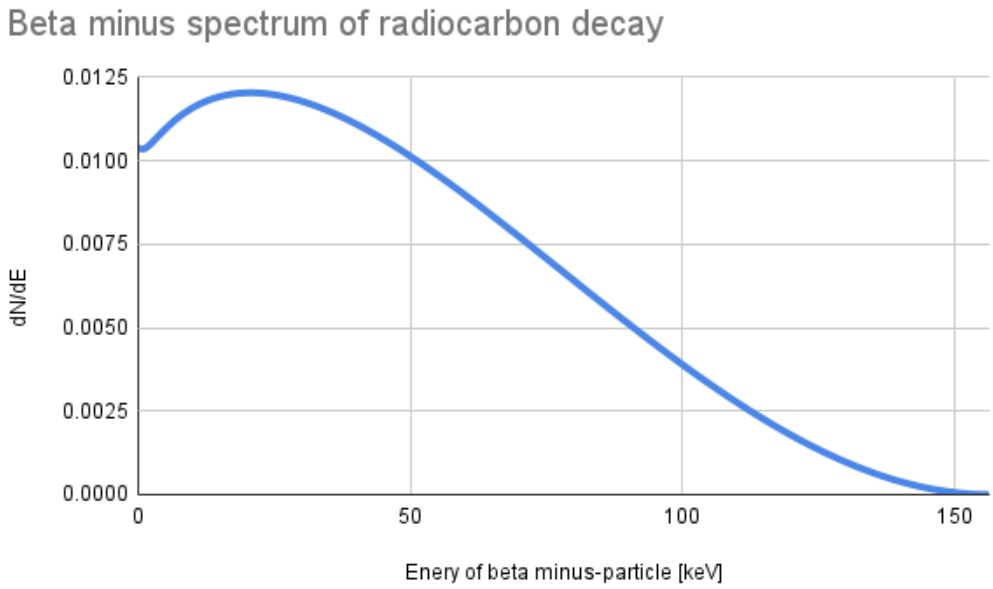


Figure 1: Differential energy spectrum (dN/dE) of β^--particles produced during the radioactive decay of ^{14}C, plotted as a function of particle kinetic energy. Data obtained from IAEA [11].

2.1.1 ^{14}C and its natural occurrence

^{14}C is produced in a continuous process in the upper troposphere. Cosmic rays, consisting of high-energy charged particles such as protons or atomic nuclei, interact with atmospheric nitrogen and provide enough energy to release neutrons from the nuclei, in a process known as spallation. These neutrons are, in turn, thermalized and absorbed by nitrogen atoms in a nuclear reaction, shown below, that yields ^{14}C.

$$^{14}\mathrm{N} + n \rightarrow ^{14}\mathrm{C} + p$$

The produced ^{14}C is rapidly oxidized to CO, and then to CO_2 [14]. This carbon dioxide then partakes in the carbon cycles, spreading both on land and in the ocean. The rate of which ^{14}C is produced in the atmosphere fluctuates with both the strength of the magnetic field of the Earth and of the Sun, both of which control the flux of cosmic rays reaching the atmosphere [9]. Although the production of ^{14}C varies spatially, the distribution of $^{14}CO_2$ easily spreads across the atmosphere.

2.1.2 ^{14}C and its production in the nuclear era

Apart from the natural and inevitable production of ^{14}C in the atmosphere, it can also have anthropogenic origins. During the detonation of nuclear weapons, neutrons are produced in a chain reaction. Some of these neutrons will be captured by the atmospheric nitrogen, inducing the same process as in the case with cosmic rays. During the decades of the 20th century in which nuclear weapons were either tested or used in warfare, more than twice the amount of ^{14}C that was previously present in the atmosphere is expected to have been released, $350 \cdot 10^{15}$ Bq [9]. Most of the ^{14}C released

from the testing of nuclear warfare has been dissolved in the oceans, but there is still a higher concentration of ^{14}C in the atmospheric carbon content, more than half a century later [4]. The concentration of atmospheric ^{14}C is decreasing consistently also due to the usage of fossil fuels, releasing large amounts of CO_2 into the atmosphere. Fossil fuels, because they are fossil, do not contain ^{14}C, so the massive use of, for example, oil, coal and natural gas reduces the atmospheric concentration of ^{14}C.

In contrast to the bomb-^{14}C, which has spread globally despite testings mainly having taken place in certain parts of the world, there may be higher concentrations of ^{14}C close to research-heavy, medical and nuclear facilities [15].

Nuclear power is the largest contributor to anthropogenic ^{14}C since testings of nuclear weapons have seized. The process is partly the same as with the naturally produced, and the bomb-produced, ^{14}C, with neutron activation of nitrogen. There are also two other processes present [2]:

$$^{17}O + n \rightarrow ^{14}C + \alpha$$

$$^{13}C + n \rightarrow ^{14}C + \gamma$$

These three processes together produced, in the 1990s, amounts of ^{14}C in the same order of magnitude as the production induced by cosmic rays in the atmosphere [2]. About 10% of the produced ^{14}C is estimated to be released from nuclear power plants to the environment [2]. Both releases into air, via ventilation, and into water, via the cooling water outlet of the NPP, occur [16]. The airborne releases are required to be measured by Swedish NPPs since 2002, but the liquid releases are not measured. This decision is partly due to analytical challenges but also since it is known that only a small fraction of the total releases are waterborne, around 0.5% [4].

Although Sweden does not currently reprocess any fuel from the three NPPs currently in operation, the reprocessing performed in other countries affect the levels of ^{14}C in the environment beyond the borders of the country in which the reprocessing takes place. France, as well as the UK, both reprocess nuclear fuel, from which formerly captured ^{14}C is released both to the air and water. These releases amount to several TBq annually. The waterborne releases into the North Sea then reach the Swedish coastal areas via Skagerrak and Kattegat. A consequence of this is that the northern part of the Swedish west coast, which is closest to the North Sea, may show up to 10% higher level of activity from ^{14}C than the southern Swedish west coast [17].

2.1.2.1 ^{14}C and its levels at Ringhals The released activity of ^{14}C to water from the Swedish nuclear power plant Ringhals was, in the years of 2022 and 2023, estimated to be nine and eight GBq respectively [18] [19]. Although the airborne releases of ^{14}C amount to almost all total releases of ^{14}C (around 99.5%), tests on the terrestrial biota of the area around Ringhals show little to no higher specific activity of carbon compared to tests performed at other places in southern Sweden [17]. This is partly due to the emissions being released high above the ground, and having time to dilute and spread before reaching the ground, but also because the releases are to a large extent organic compounds, which are not available to photosynthesize upon

[4]. This is especially the case for Ringhals NPP, as it operates pressurized water reactors (PWRs), unlike the Swedish boiling water reactors (BWRs) at Forsmark and Oskarshamn. PWRs release a larger share of their airborne ^{14}C in organic forms, primarily hydrocarbons such as methane, while BWRs release most airborne ^{14}C as carbon dioxide [20]. Since carbon dioxide is directly incorporated into plants through photosynthesis, releases from BWRs are generally more biologically available to terrestrial ecosystems than the releases from PWRs which feature more hydrocarbons.

Despite only a small fraction of the total ^{14}C releases being to water, the accumulation in biota appears to be much more effective in marine environments than in terrestrial environments around NPPs operating PWRs [4]. The releases contain both dissolved inorganic carbon (DIC) and dissolved organic carbon (DOC) [4].

DIC is more common in marine environments, as carbon dioxide is continuously dissolved in seawater. This makes the released DIC from the NPPs more diluted, contributing to a lower specific activity. DOC, on the other hand, is less diluted leading to a higher specific activity [4].

During a study conducted by scientists at Lund University for the Swedish Radiation Safety Authority, it was found that marine organisms near Ringhals show elevated levels of ^{14}C, but the enrichment depends on their position in the food web. Primary producers, such as seaweeds, which fix DIC such as dissolved carbon dioxide during photosynthesis, showed elevated specific activities of carbon compared to background levels, although the enrichment was smaller than for organisms higher up in the food chain [4]. Organisms that have higher trophic levels and consume or store organic carbon, showed significantly greater enrichments in ^{14}C [4]. For example, the seaweed *Fucus vesiculosus* collected near the cooling water outlet showed specific activities with an elevation of 50-100%, while the wrasse fish *Symphodus melops*, sampled at the same location, contained specific activities approximately 400% above background levels [4]. The fish species is non-migratory [21], which likely explains the prolonged exposure and resulting accumulation of ^{14}C in the organism.

2.1.3 ^{14}C and its effects on humans

The radiological significance of ^{14}C comes from its dual nature as a very long-lived radionuclide, with a half-life of 5730 years and carbon being a building block of all organic matter. Because of the longevity, any anthropogenic release of ^{14}C increases the global inventory for millennia to come. This contributes to a global collective dose that is multi-generational and exceeds the human timescales.

Once released, ^{14}C quickly enters the global carbon cycle, behaving just like the stable carbon isotopes (^{12}C and ^{13}C). It spreads through the atmosphere, for example as $^{14}CO_2$, ^{14}CO or $^{14}CH_4$. ^{14}C also dissolves in water as inorganic or organic carbon, and it is incorporated into biota via photosynthesis and bioaccumulation through the food chain. Because of this high environmental mobility, local releases of ^{14}C cannot be contained; instead they contribute to a global baseline [1].

When it is external, ^{14}C poses almost no radiological threat because the low-energy

β^- particles it emits, with an average energy of 49 keV, cannot penetrate the outer layer of human skin [22]. However, it can become hazardous when internalized. The main paths for ^{14}C to enter the human body are the ingestion of food and water [22].

Once inside the body, ^{14}C is metabolically incorporated into molecules, including proteins, lipids, and nucleic acids [22]. When an internal ^{14}C atom decays, it deposits all of its ionizing energy directly into the surrounding tissue. In addition to this, the decay of ^{14}C into ^{14}N within DNA can induce local bond rupture and damage, and may contribute to mutations or an increased long-term risk of cancer [23]. The hazardous potential of internal ^{14}C, together with the large amount of ^{14}C around us compensate for the low energy of a single decay.

Under the regulations enforced by the Swedish Radiation Safety Authority (SSM), radiation doses to the public from nuclear power plants are kept very low. Nevertheless, ^{14}C consistently is the radionuclide that delivers the largest effective dose to the public from the operation of NPPs [19]. The public doses are evaluated using models that account for parameters such as the discharged activity concentration and the rate of internalization in humans. However, while airborne ^{14}C releases are directly measured, liquid releases are only estimated [19]. Even though the absolute liquid releases are minimal, relying on estimations rather than physical measurements could introduce uncertainties into the regulatory dose calculations. As demonstrated in previous sections, even these low-level liquid releases are accumulated within local marine organisms. As a consequence of this, for certain individuals who experience exposure pathways such as through regular consumption of local seafood, this accumulation could give a disproportionately high contribution to the individual internal dose.

2.1.4 Units of measurement of ^{14}C and its activity

The concentration of ^{14}C in a sample can be expressed in several different ways, depending on which information is of necessity. If the activity of a sample is desired, the units could be Bq per sample, or specific activity such as Bq per unit of mass or volume. However, when the results of interest of a sample analysis lies in the excess of, or lack of, ^{14}C compared to the reference level, it is more important to compare the measured activity to a known standard. Two units, that both belong to the latter sort, are of special interest to this paper; The most widely used unit, percent Modern Carbon (pMC), and the unit to which all forecoming units in this paper will be converted, Fraction Modern, (F^{14}C).

The quantity percent Modern Carbon, pMC, is given by [24] [25] as:

$$\text{pMC} = \frac{A_S}{A_{abs} \cdot e^{(1950-y)/8267}} \cdot \left(\frac{1 + \frac{\delta^{13}\text{C}_{ref}}{1000}}{1 + \frac{\delta^{13}\text{C}_S}{1000}}\right)^2 \cdot 100\% \quad (1)$$

This is the ratio between the activity of the sample (A_S) and that of a predefined modern reference value. The modern reference is oxalic acid I which, during the year of reference, had a specific activity of $A_{abs} = 226$ Bq/kg of carbon [24]. The exponential part of the denominator represents the decay from the year of reference, 1950, to the year of measurement, y, with 8267 being the mean life of ^{14}C [24]. A_S in the equation is normalized for isotope fractionation, represented by the second term of

the equation. This correction needs to be performed because different isotopes engage differently in chemical reactions or physical processes due to their differences in mass. This leads to enrichment or depletion of certain carbon isotopes in relation to the others in the analyzed sample [24]. In the equation, δ^{13}C is the variable representing the isotope fractionation, expressing the deviation of the ratio between ^{13}C and ^{12}C from that of a limestone standard material (VPDB NBS19) [24]. As the values of the isotope fractionation factors vary between organisms and materials, the values can be experimentally determined for the specimen in question or tabulated values from literature can be used in calculations. The factors can also be measured specifically for the samples in question. The entire term is squared because the isotope fractionation for the ratio between ^{14}C and ^{12}C can be approximated with the square of the ^{13}C-counterpart [26].

When comparing ^{14}C concentrations, they are normalized to an isotope fractionation of a reference material of wood. Wood has a well-documented factor of $\delta^{13}C_{ref} = -25‰$ [24]. Equation 1 becomes:

$$\text{pMC} = \frac{A_S}{A_{abs} \cdot e^{(1950-y)/8267}} \cdot (\frac{0.975}{1 + \frac{\delta^{13}C_S}{1000}})^2 \cdot 100\% \tag{2}$$

When receiving results from analysis, in which the results have not been corrected for isotope fractionation, equation 3 can be used to normalize the activity for correct comparison between results.

$$A_{SN} = A_S(\frac{0.975}{1 + \frac{\delta^{13}C_S}{1000}})^2 \tag{3}$$

The other unit of measurement, F^{14}C, has a similar definition to that of pMC [24]:

$$\text{F}^{14}\text{C} = \frac{A_S}{A_{abs} \cdot e^{(1950-y)/8267}} \cdot (\frac{0.975}{1 + \frac{\delta^{13}C_S}{1000}})^2 \tag{4}$$

The similarities between the definitions make translation between the two units very easy

$$\text{F}^{14}\text{C} = \frac{\text{pMC}}{100\%} \tag{5}$$

Both of these units represent a deviation from a standard of modern carbon levels. Since they are corrected for the year of measurement within their definitions, the same sample can be measured many times, during different years, and still yield the same results.

2.2 Methods of measuring ^{14}C

This paper focuses on three fundamentally different analytical techniques that can be used in measuring and quantifying ^{14}C for environmental samples. The different methods are accelerator mass spectrometry (AMS); a decay counting method, in this case liquid scintillation counting (LSC); and a laser-based spectroscopic method, known as saturated-absorption cavity ring-down spectroscopy (SCAR). The different methods feature individual approaches to ^{14}C analysis. AMS individually counts the atoms of ^{14}C in relation to other isotopes of carbon, providing extremely high precision and low detection limit, but is also very complex, resource-intense and expensive. LSC, on the other hand, measures the radioactive decay of ^{14}C, and is more widely used. Standard LSC is both simpler and cheaper than AMS, although compromising on sensitivity. SCAR is a much more recent addition to the industry that could become a middle ground of the two other methods that are the currently most used techniques.

This chapter provides a technical description of each technique and variations in the systems used in certain facilities of special interest for this report.

2.2.1 Sample preparation

Although AMS, LSC, and SCAR use fundamentally different measurement principles, their sample preparation procedures share several steps. In all three techniques, the purpose of the preparation is to extract the carbon contained in the sample, minimize contamination, and convert the carbon into a form that is suitable for the respective analytical method. A flowchart illustrating the process before analysis is shown in figure 2.

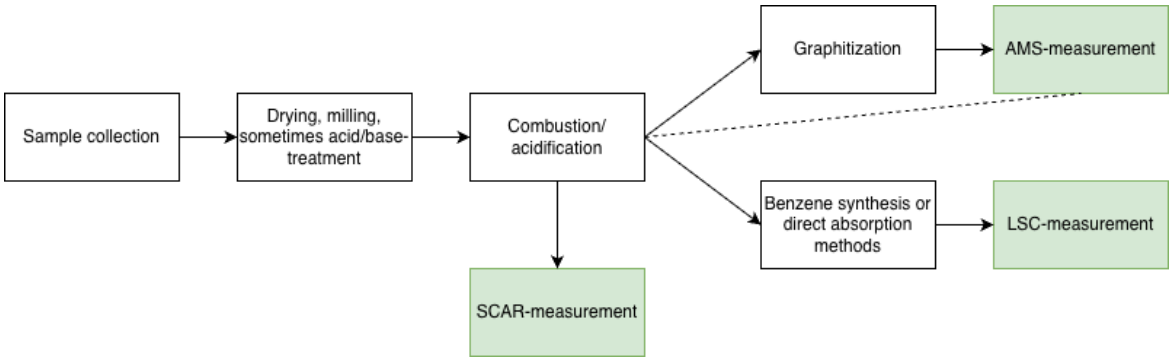


Figure 2: A flowchart showing the process for ^{14}C analysis from sample collection to measurements. The dotted line represents a possible pathway, in this case the possibility for some AMS-systems analyzing carbon dioxide directly.

The first step, mostly when handling archeological samples, is typically cleaning and pretreatment of the sample. This can include manually removing contaminants such as dirt, rocks, or plant material, as well as chemical treatment with acids and bases [5]. Hydrochloric acid is commonly used to dissolve unwanted carbonates, while sodium hydroxide can remove humic acids and other organic contaminants [27]. While this is an important step when handling archeological samples, the pretreatment with acids and bases for environmental samples can generally be skipped. The environmental

samples might still need to be cleaned.

For environmental samples in the proximity of NPPs, it is important for the sample to be homogenized, especially when using AMS and SCAR since very small amounts of material are used in analysis. This process is done by drying, grinding and thoroughly mixing the sample before the further preparation. The drying can be performed in ovens or in some cases by freeze drying. The oven used for drying and the mill used for grinding the samples at Medical Radiation Physics in Malmö are shown in figures 3a and 3b respectively.

(a) (b)

Figure 3: Two steps of sample preparation: drying and grinding of the samples. Pictures are taken at Medical Radiation Physics in Malmö. (a): An example of an oven that can be used to dry samples. (b): An example of a grinding mill that can be used to grind the dried samples.

After pretreatment, the carbon in the sample is converted into carbon dioxide, CO_2. For organic materials, this is typically done by combustion in an elemental analyzer (EA), shown in figure 4b. Small masses of samples, normally only one or two mg, are portioned into a foil, shown in figure 4a, before being inserted into the EA. The samples are then combusted at temperatures exceeding 1000°C in a stream of excess oxygen [28, 27]. The oxygen ensures complete combustion and formation of CO_2, while carrier gases such as helium transport the produced gas through the system where by-products such as H_2O and N_2 are removed [27]. For inorganic carbonates, CO_2 is instead released by acidification, commonly using orthophosphoric acid [27]. Up to this point, the preparation procedure is similar for AMS and SCAR. For LSC, combustion in an EA is not possible due to large sample mass requirements (several grams in the case of environmental samples). Instead, samples are combusted in, for example, pyrolysers.

The methods begin to differ after the production of CO_2, since each analytical technique requires carbon in a different form. Many AMS systems analyze graphite targets,

so the produced carbon dioxide is converted into graphite through graphitization. Automated graphitization systems are commonly used, such as the Ionplus AGE3, shown in figure 4c, where one preparation cycle producing seven graphite samples takes approximately five hours [27]. In this process, the CO_2 reacts with hydrogen gas in the presence of an iron catalyst at temperatures slightly below 600°C, forming graphite [5]. There are other automated graphitization devices, such as the CORgiS that can graphitize 30 samples per day [29], or the μGRAPHILINE that can both combust and graphitize 24 samples per day [30]. Many laboratories use own methods for graphitization, that are not automated. An example of what a manual graphitization method can look like is shown in figure 4d. These methods can offer higher throughput and flexibility, but require manual handling.

There are also AMS-systems that can perform analysis directly on CO_2. For these systems, graphitization can be skipped altogether.

For LSC, the carbon instead needs to be converted into a liquid form that is compatible with a scintillation cocktail, consisting of a solvent and a scintillator. A scintillator is a molecule that can absorb and be excited by radiation, after which deexcitation releases photons [32].

An important aspect of LSC sample preparation is minimizing quenching, where the light generated during scintillation is prevented from reaching the detector [33]. Quenching is generally categorized into two types: chemical quenching and color quenching. The former type occurs when the energy from the excited solvent is transferred to a non-scintillating molecule instead of the scintillator [33]. Instead of light, the energy is released as heat. A common quenching molecule is dissolved oxygen [34]. Color quenching occurs when the sample itself is not perfectly clear. The sample could, for example, contain plant pigments. These pigments reabsorb the light emitted by the scintillator before it can escape the vial, which distorts the measurement results [33].

One of the widely used preparation techniques for LSC is benzene synthesis, which is known for high precision and sensitivity [6]. After producing carbon dioxide, it is reduced with molten lithium metal to lithium carbide (Li_2C_2), which is then hydrolyzed with water to form acetylene gas (C_2H_2). The acetylene is then trimerized into benzene, C_6H_6, using catalysts such as vanadium or chromium [35]. Benzene provides high carbon content and low quenching effects, improving measurement precision [35]. However, benzene synthesis is labor-intensive, time-consuming, and involves hazardous chemicals [3, 6]. For example, the SUERC laboratory reported that only three to six benzene samples could be produced in one day [6]. An alternative preparation method is direct absorption of the released CO_2 into an alkaline solution, which can then be mixed with the scintillation cocktail [35]. This method is simpler and faster than benzene synthesis, although it generally captures less carbon and therefore results in poorer detection limits [35].

SCAR does not require any additional steps after CO_2 production, since the method measures carbon dioxide directly [36]. The produced CO_2 is purified and introduced directly into the spectroscopic system within the device [36]. The elemental analyzer used for combustion is often directly coupled to the SCAR instrument, making manual

(a)

(b)

(c)

(d)

Figure 4: Different stages of the graphitization process. From portioning the sample to combusting it and two different graphitization units: one automated and one manual. (a): Tin capsules used during sample preparation. Small amounts of sample are placed in tin foil and enclosed, after which they are combusted in an elemental analyzer. (b): An elemental analyzer (EA). Image by MaRufaru82, licensed under CC BY-SA 4.0 via Wikimedia Commons [31]. (c): The automated graphitization unit AGE3 by Ionplus used by the SSAMS laboratory in Lund. (d): An example of a manual graphitization line, this one used at Atomki in Debrecen.

handling for SCAR-sample preparation only required up until the point of combustion.

2.2.2 AMS

AMS is a technique that directly measures the ratio between ^{14}C and the other two isotopes of carbon, ^{12}C and ^{13}C, by counting the individual atoms within a sample. This enables detection of extremely low concentrations of ^{14}C with high precision (often better than 0.2%) [5], making AMS a well used method, especially for radiocarbon dating purposes. However, the technique includes particle acceleration, mass separation systems and extensive sample preparation. This leads to drawbacks such as high costs and infrastructure requirements.

2.2.2.1 The principles of AMS The purpose of the AMS is to count the individual carbon isotopes that the graphite samples contain. In a mass spectrometer the particles are separated by mass, which makes it of importance to suppress any isobars of ^{14}C before reaching the detector.

The graphite samples that have been produced, that usually contain 50 μg – 1 mg of graphite, are loaded into holders that are placed in a sample wheel inside the sputter ion source of the AMS-system. The wheel can contain many samples simultaneously, allowing for automated measurements in sequence. Some AMS-systems instead use gas ion sources, where CO_2 is measured directly without graphitization. The ion source operates under high vacuum conditions. Inside the sputter ion source, caesium is heated until it vaporizes, after which the caesium atoms are ionized by a very hot tantalum surface [5]. The resulting caesium ions are accelerated in a beam by an electric potential toward the graphite samples. When the caesium ions bombard the graphite, atoms and molecules are sputtered from the sample surface while also receiving one or multiple extra electrons, forming negative ions [5]. If any ^{14}N is present in the sample, it is suppressed already at this stage since ^{14}N cannot form stable negative ions [5].

The sputtered ions are accelerated in an electric field into a focused beam that enters the first of two magnets used for mass selection of the ions. The first of the two mass spectrometers is a low-energy magnet whose purpose is to separate unwanted ions from the ones of interest, which are ^{12}C, ^{13}C and ^{14}C. This separation is done by applying a magnetic field over the trajectory of the ions. When exposed to the magnetic field, the Lorentz force causes the ions to travel along curved paths, where the radius of curvature depends on the mass, m, energy, E, and charge of the particle, q, as well as the magnetic field strength, B, according to equation 6 [5]. Since the ions have the same charge (in this case, $q = -1$), ions with different masses will follow different trajectories if they are accelerated to the same energy. This makes it possible to separate the carbon isotopes from most unwanted ions. The magnetic field is kept constant while the ion energies are adjusted, allowing ions of different masses to sequentially follow trajectories that match the exit of the mass spectrometer. The switching between energies can occur many times per second [5]. However, some ions may still have slightly different energies than intended, allowing particles with incorrect masses to pass through the mass spectrometer [5]. In addition, molecular

ions with the same mass as the carbon isotopes, such as $^{12}CH_2$, can also pass through the low-energy magnet [5]. These remaining interferences are removed in later stages of the AMS-system.

$$r = \frac{\sqrt{2mE}}{qeB} \tag{6}$$

The mass-selected ions then reach the accelerator and the subsequent stripper. This is where variations of AMS usually differ. The common principle is acceleration of the particles to very high energies, from keV to MeV, and directing them through a narrow tube containing, most often, a gas, known as the stripper gas, usually argon or helium [5]. The negative ions that contain high kinetic energy will be subject to violent collisions with the gas particles, which leads to the "stripping" of electrons of the ion beam particles [5]. The ions are therefore from now on positive. During these collisions, molecules that have survived the first mass-selection, due to their mass coinciding with the mass of the carbon isotopes, will break apart into their constituents [5]. The result is a cleaner beam of positive ions. Developments have been made with the technique since its beginning, and more recent devices can still effectively strip the ion beam while lowering accelerator energies.

There are mainly two types of accelerators: the tandem accelerator and the single-stage accelerator [5]. In a tandem accelerator, shown in figure 5, the ions experience two accelerations. The stripping occurs at the high-voltage terminal, to which they have been attracted. After the stripping at the terminal the charge of the ions has switched, making the terminal voltage repel the ions instead [5]. The particles are therefore accelerated twice, giving them very high energies, sometimes up to the MeV-range, suppressing interferences effectively. The single-stage accelerator, however, accelerates the ions only once [5]. Since these systems operate with lower voltages, they do not require the large insulating pressure tanks used in tandem accelerators. The removal of the pressure tanks, and the lower energies allow for the systems to be smaller than for tandem accelerators.

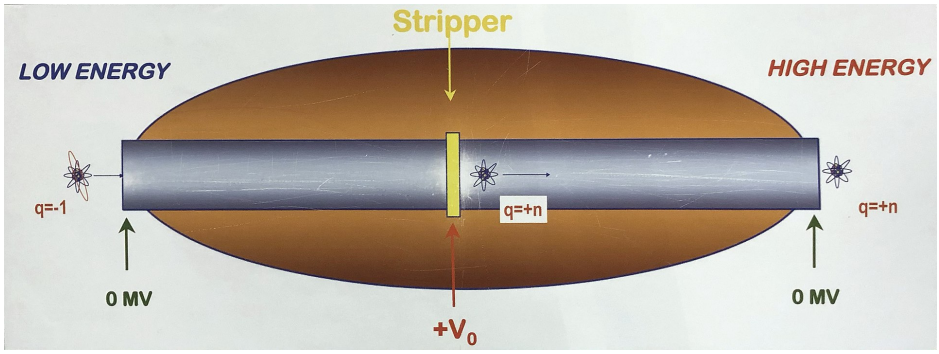


Figure 5: An illustration of a tandem accelerator. An ion comes in from the left. Because of its negative charge it is attracted by the positive terminal voltage in the middle, where it is stripped of electrons. The ion, now positive, is repelled by the voltage, being accelerated a second time. Image by Davide Mauro, licensed under CC BY-SA 4.0 via Wikimedia Commons [37]

The ion beam is then, once again, subject to a magnetic analyzer, this time a high-

energy one. The idea is in analogy with the previous magnetic analyzer. The aim is to select ^{14}C with a specific charge state and a well-defined energy, and only allowing it to reach the final detector. The magnetic field selects a certain mass-to-energy ratio, ensuring a bending pathway of the beam that steers it into an electrostatic analyzer [5]. The radii of curvature for ^{12}C and ^{13}C with the chosen charge state is known, and in their pathways Faraday cups are placed to collect the particles [5]. The measured amounts will be used in establishing the ratio between these isotopes and ^{14}C, which is the desired measurement result.

In the electrostatic analyzer, the beam passes an orthogonal electric field, that, equivalently to the magnetic analyzers, bend the path of the beam depending on the energy of the particles [5]. This stage eliminates ions that have the correct mass-to-charge ratio but not the correct energy [5]. The combination of these steps successfully suppresses any particles that could mimic ^{14}C from reaching the detector.

Lastly, the ions reach the detector. Since the ^{14}C usually just exists in trace amounts in a sample, it is not possible to measure the beam current of the ions [5]. They are rather measured individually when they appear. The detector is usually a gas-ionization detector or a silicon detector [5]. In a gas-ionization detector the beam will ionize gas particles in its path. The released electrons will induce a measurable signal when reaching an electrode [5]. This type of detector can, through the energy loss along the path of the ion, identify the ion in question. The silicon detector does not have the ability to identify the ion reaching it. The energy that is deposited in the semiconductor, creating electron-hole pair, gives rise to a signal proportional to the energy [5]. Finally the ratio between the different carbon isotopes can be established.

2.2.2.2 SSAMS The single-stage accelerator mass spectrometer, SSAMS, is a variation on the AMS. In contrast to most AMS applications, the SSAMS does not use a tandem accelerator. Instead, the ions are only accelerated once. The terminal voltage is 250 kV, accelerating the ions to a maximum of 275 keV [38]. The selected charge of the ions that want to be detected is +1, which lowers the need for acceleration compared to other AMS-systems. An illustration of an SSAMS is shown in figure 6.

The precision is reported to reach 0.3% by the producer of the first SSAMS, the National Electrostatics Corp., NEC [38]. The SSAMS produced by NEC uses either an argon or helium stripper and a silicon detector [38]. The single-stage accelerator makes the SSAMS smaller in comparison to other AMS-systems that were in use at the time of introduction; the SSAMS requires an area of 35 m^2 while there are other AMS-systems that require up to 165 m^2 [5].

The investment cost of the SSAMS if bought today by the same manufacturer is reported, by personal communication with the sales team, to be approximately 2.1 million USD, which, with the rate of 1 USD = 9.49 SEK (April 2026) is equivalent to just under 20 million SEK. The prize includes shipping and installation.

SSAMS devices can come with sample wheels that are able to contain varying amounts of samples. A laboratory in Lithuania reports that each sample typically is measured 18 times, for three minutes each [27]. If the sample wheel contains 40 samples, this

corresponds to 36 hours of measurements. The standard graphite target at the same SSAMS laboratory is reported to be 1 mg. It was mentioned in [27] that, after optimization, the SSAMS of the Lithuanian lab achieved a precision of 0.35%.

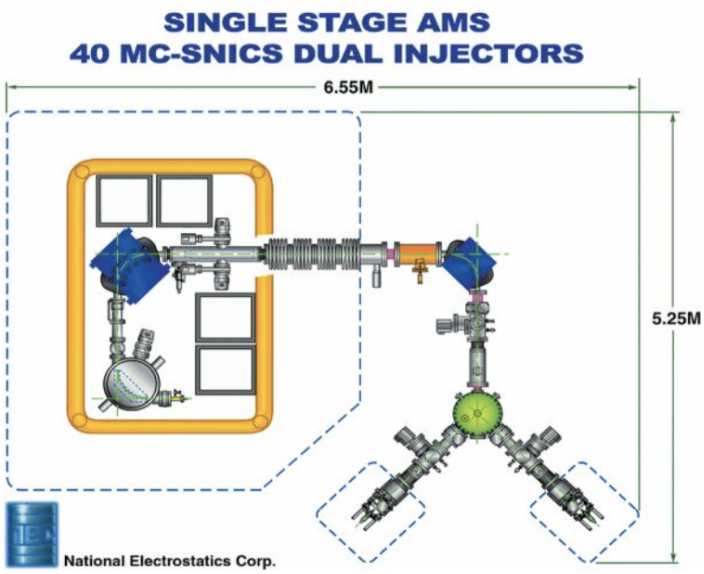


Figure 6: Illustration from above of an SSAMS device with two ion sources, just like the one at the facility in Lund. Picture courtesy of NEC.

2.2.2.3 MICADAS The mini radiocarbon dating system, MICADAS, is a more compact AMS-system commissioned in 2004, constructed especially for analysis of ^{14}C [39]. The aim was to reduce the acceleration energy and therefore the size of the system. MICADAS uses a tandem accelerator with a terminal voltage of around 200 kV. This reduction enables the entire system to be scaled down, since lower energies result in smaller radii of curvature according to equation 6 [39]. The MICADAS occupies 8 m^2 and weighs 4,500 kg [40], which is a significant reduction of footprint and weight compared to other AMS-systems. The stripping gas used is helium [41] and the selected charge of the ions is +1 [42]. When deliberately focusing on only the +1 ions, together with a denser but lighter stripping gas, the energy of the acceleration can be reduced. A gas-ionization detector corrects for unwanted ions reaching the detector. The precision of the device is "0.2% or better" according to the producer of the first MICADAS-system, Ionplus [40]. One report shows precisions below 0.1% [43].

With the rate of 1 CHF = 11.89 SEK that was the case in April of 2026, the prize of the MICADAS is roughly 22 million SEK, including shipment, installation and training. This was provided by personal communication with the company Ionplus.

A feature of the MICADAS is that it can take both graphite samples and measure carbon dioxide directly [40]. Standard sample sizes for the graphite targets are 0.9 mg C while using gas targets only requires 1-50 μg [44].

The MICADAS can be used for radiocarbon dating of samples that are more than 50,000 years old [40].

One MICADAS is operated at Atomki in Debrecen. Their device is shown in figure 7.

Figure 7: The MICADAS-device used at Atomki in Debrecen.

2.2.2.4 LEA The low energy accelerator, LEA, is a system based on the MICADAS, that was introduced to the markets in 2022 [45] [43]. Just like the MICADAS, a tandem accelerator is used, but the terminal voltage is reduced to as low as 50-60 kV [43]. This allows for the AMS to shrink even more, and LEA is to date the smallest AMS commercially available, occupying a space of around 4.5 m^2 and weighs 3,000 kg [46]. The stripping gas is helium with a higher local density, still effectively stripping the beam from unwanted ions [43]. The gas-ionization detector provides a precision on par with the MICADAS: 0.2% or better [46]. The power usage of the LEA is the lowest of all AMS systems, less than 2.5 kW [46], which is comparable to common household items. A quotation from Ionplus in April 2026 was at 1.7 million CHF, which, with the exchange rate of 1 CHF = 11.74 SEK (April 2026) gives a price of almost 20 million SEK. The price includes training, shipping, packing and installation.

LEA, just like its predecessor, can analyze both graphite and gas samples [46]. LEA also has roughly the same detection limit as MICADAS, with both devices being able to date archeological samples older than 50,000 years [46].

A Lithuanian lab operating a LEA reports that standard procedure is ten measurements for five minutes of each sample [27]. This results in 40 samples being analyzed in approximately 33 hours, slightly quicker than for the SSAMS at the same laboratory.

Atomki in Debrecen operates a LEA. Their device is shown in figure 8.

Figure 8: The LEA-device used at Atomki in Debrecen.

2.2.3 LSC

Liquid Scintillation Counting, LSC, is a classic and widely used method for quantifying low-energy β-emitting isotopes, such as ^{14}C. Unlike AMS, which determines the isotope ratio by counting individual carbon atoms based on their mass, LSC is a decay-counting method. It functions by measuring the actual radioactive decays occurring within a sample over a specific period of time [47].

A big challenge of measuring ^{14}C via decay counting is the low energy of its β-particles, which have a maximum energy of only 156 keV. These particles have a very short range and are easily absorbed by the sample material or the walls of a container before they can reach a detector. To overcome this, the LSC method requires the sample to be converted into a chemical form that can be distributed homogeneously within a liquid scintillation cocktail. By mixing the sample with the scintillation cocktail, ensuring that the sample is in direct contact with the scintillator, the released photons can be detected by the device instead of the emitted β-particles that would not reach the detector themselves.

2.2.3.1 The principles of LSC For the radioactive decay to be detectable using LSC, the sample needs to be integrated into a scintillation cocktail. The cocktail consists of a solvent, such as toluene or xylene, and a scintillator solution [6].

The mixing process involves combining the prepared sample with the cocktail in a vial at a specific ratio [48]. Following this, the vial is usually shaken thoroughly to ensure a uniform solution; uneven distribution can lead to quenching and therefore a reduction in counting efficiency [48]. In addition to this, for ^{14}C measurements, samples are often kept inside the LSC instrument for a period, often 24 hours, before counting to allow

any photoluminescence to decay [48]. The released photons during photoluminescence would otherwise disturb the measurements and could be mistaken for a radioactive decay from the sample.

The purpose of the sample preparation and the mixing is to bring the radioactive isotopes in the sample into direct contact with the scintillation medium [6]. When a ^{14}C atom undergoes β-decay, in accordance to the process described in section 2.1, it emits a β-particle with some kinetic energy. This particle travels through the cocktail and loses its energy by colliding with the molecules of the solvent, exciting them [34]. These excited solvent molecules then transfer the energy to the scintillator molecules [34].

As the excited scintillator molecules return to their ground state, they emit photons in random directions [34]. The number of emitted photons, i.e the intensity of the light pulse, is directly proportional to the energy the β-particle deposited in the cocktail [34]. These bursts of light are captured by photomultiplier tubes (PMTs), which convert the photons into electrical pulses [34]. In the PMT, the incident photon will trigger an electron release, that is multiplied at several dynodes along the way [6]. The scintillation cocktail and the PMTs therefore enable the energy from a radioactive decay to be transformed into photons, and then to electric pulses that can be collected and analyzed by computers. Ultimately, the amplitude of each electrical pulse is proportional to the original decay energy, allowing the LSC to accurately quantify the radioactivity present in the sample [34].

LSC devices usually include more than one PMT, either two or three. By having multiple PMTs it is possible to implement coincidence counting [49]. This is a noise-reduction method, that is used to reduce the risk of detection errors. PMTs can give rise to "dark current" which are random electrical pulses that are not generated by incident photons [6]. The chance of more than one PMT experiencing a dark pulse within the same narrow time period is very unlikely. However, the radioactive decay events give rise to many photons at once, that are released in many different directions, making the chance of more than one PMT recording a decay at the same time big. If only one PMT registers a "decay", it can therefore be discarded as noise [6].

Another measure that is taken to minimize noise and ensure the detection of only radioactive decay is that the device is shielded by metals such as lead, cadmium and copper to block background radiation [35]. The shielding makes even small devices very heavy.

2.2.3.2 1220 Quantulus The 1220 Quantulus LSC system produced by PerkinElmer is specialized on ultra-low background counting, making it particularly suitable for measuring low concentrations of radionuclides and achieving low backgrounds. The producer reports that the device is able to detect radiocarbon ages of up to 65,000 years, when using long counting times and large sample sizes [49].

The shielding includes a 650 kg lead block enclosing the PMTs, constructed from low residual activity lead and lined with copper and cadmium [49]. The system also contains extra PMTs to implement coincidence counting with a short coincidence resolu-

tion time [49]. In total, the device weighs 1000 kg and has a footprint of approximately 1 m^2 with a height of around 1.5 m [49].

While the required measurement time is highly specific for the sample and purpose, the device can measure a sample for up to 277 hours [49]. Archeological samples would require very long counting times and large sample sizes, while enriched samples, with higher decay rates, can be measured for shorter times and using smaller sample sizes. One article reports the Quantulus 1220 having precision of 1.7-3.5%, accuracy of 1.7-14.1% and relative uncertainties of 3.7-9.5% for samples that were measured for three hours [48]. It is important to mention that the precision, accuracy and uncertainty increases with counting time.

2.2.4 ^{14}C-SCAR

Saturated-absorption cavity ring-down spectroscopy, SCAR, is a laser-based spectroscopic technique used to determine the ^{14}C content of carbon dioxide that is released during the combustion of a sample [36]. The method is based on sensitive optical absorption measurements of the molecule $^{14}CO_2$ [50]. Sample preparation follows the procedure described in chapter 2.2.1. Combustion occurs in an EA that is directly coupled to the SCAR-device [36].

The produced gas is introduced into an optical cavity formed by highly reflective mirrors, known as a Fabry-Pérot etalon [36]. An infrared laser, whose frequency can be tuned, is used to excite a molecular transition that is specific to $^{14}CO_2$ [51]. Because different carbon isotopes exhibit slightly different rovibrational transition frequencies, it is possible to measure the absorption associated with only $^{14}CO_2$ while minimizing interference from the much more abundant $^{12}CO_2$ and $^{13}CO_2$ [7].

The SCAR technique is derived from conventional cavity ring-down spectroscopy, CRDS, a well-established method for highly sensitive gas absorption measurements [51] [52]. In CRDS, a short pulse of laser light with narrow bandwidth is sent into a Fabry-Pérot etalon [52]. The mirrors of the etalon can have transmission coefficients as low as $T = 10^{-6}$ in the visible and infrared spectral regions [53]. The transmission represents the loss per reflection. The remaining share, which in this case is almost all light, is reflected back into the etalon. Once the short laser pulse enters the cavity it is reflected repeatedly between the mirrors, producing an effective optical path length of several kilometers [52]. At each reflection a small fraction of the light is transmitted through the mirror [52]. The light that escapes the etalon is measured by the detector, a photodiode [52]. After the initial laser pulse has entered the cavity, no additional light is supplied and the intensity of the trapped light decreases exponentially as energy is gradually lost with every reflection [52]. This decay of intensity is referred to as the ring-down event.

Instead of measuring the absolute absorption of the sample, CRDS determines the rate at which the light intensity decays inside the cavity [52]. When no absorbent is present, the decay time, τ, depends only on the reflectivity of the mirrors R and the cavity length d [54]. When an absorbing sample is introduced into the cavity, additional losses occur due to absorption of the light by the sample molecules, resulting in a

shorter ring-down time. The decay time can be expressed as

$$\tau = \frac{d}{c((1-R)+\alpha)} \tag{7}$$

where c is the speed of light and α is the absorption coefficient of the sample [54]. By comparing the ring-down transients in the presence of the sample, τ, with that of an empty cavity, τ_0, the absorption coefficient can, according to [54], be determined by:

$$\alpha = \frac{1}{c\tau} - \frac{1}{c\tau_0}. \tag{8}$$

The minimum detectable absorption is determined by the ability to measure small changes in the decay time and is expressed by [54] as:

$$\alpha_{min} = \frac{\Delta\tau_{min}}{c\tau_0^2} \tag{9}$$

Because the mirrors are highly reflective, the effective optical path length becomes extremely long, allowing trace concentrations of absorbing gases to be detected [52]. CRDS is therefore particularly well suited for measuring very small concentrations of particles.

To enable selective detection of ^{14}C, CRDS is combined with the principle of saturated absorption, forming the SCAR technique shown in figure 9 [55]. Different isotopes of carbon dioxide have slightly different rovibrational transition frequencies in the infrared region [7]. By tuning the laser to a transition that is unique to $^{14}CO_2$, the system can excite this isotope while avoiding transitions of $^{12}CO_2$ and $^{13}CO_2$ [56]. At sufficiently high laser intensities the targeted transition becomes saturated, meaning that the rate of excitation equals the rate of relaxation [56]. In other words, the population of excited molecules reaches its maximum and steady state. Under these conditions the absorption signal becomes highly specific to ^{14}C, improving selectivity despite the overwhelming abundance of other carbon isotopes [57].

In practice, the presence of $^{14}CO_2$ in the cavity leads to more of the laser light being absorbed, making the ring-down event occur more rapidly than in a cavity with no $^{14}CO_2$ [52]. The measured ring-down time therefore depends on the concentration of $^{14}CO_2$ in the introduced carbon dioxide sample [51]. By comparing the measured signal with calibration samples with known ^{14}C content, the ^{14}C concentration of the sample can be determined.

The time to complete one measurement is usually 15 minutes of averaged measurements, but depends on the precision wanted [36]. For longer measurement times, the precision can reach levels of 0.03 $F^{14}C$ according to the producers of the first available SCAR-device, ppqSense [36]. The detection limit is 0.01 $F^{14}C$, and samples with activities up to 1,000 $F^{14}C$ can be measured [58].

The SCAR-device is comparatively small and light, with a footprint of only 1-2 m^2

[58]. The first generation of the device weighed 600 kg and has since been reduced [36]. The standard sample size that is combusted is 1 mg of carbon [58], which usually is 2-3 mg of dried organic sample.

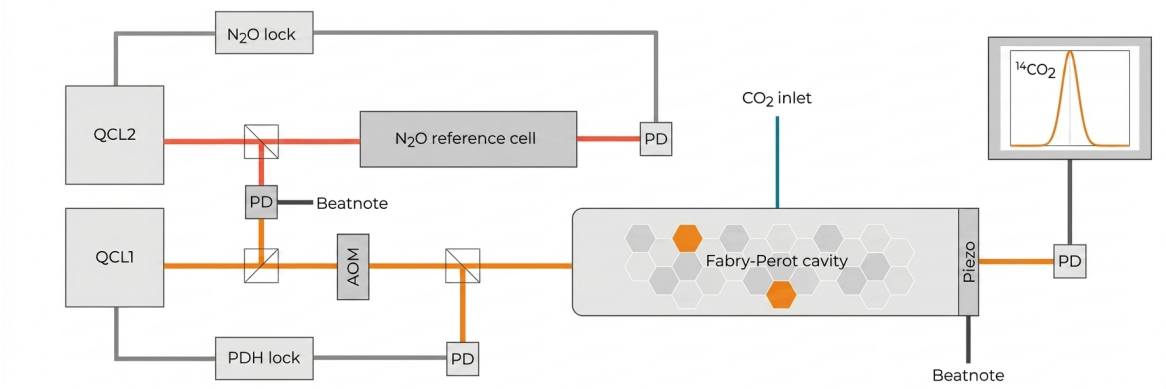


Figure 9: A graphic overview of the SCAR-technique. Adapted from [59] and used with permission from ppqSense.

3 Materials and methods

3.1 Samples

Four different types of organism were selected to be analyzed. These were the meat of *Mytilus edulis* (blue mussel), *Fucus serratus* (toothed wrack), *Fucus vesiculosus* (bladderwrack) and the meat of *Pleuronectes platessa* (European plaice). Illustrations of the organisms are found in figure 10. The samples had been collected by the Division of Particle and Nuclear Physics at Lund University and SLU, the Swedish University of Agricultural Sciences, prior to the start of this project, and have initial measurement results performed by the SSAMS in Lund. The samples, with more information, are presented in table 1. The order of the samples is decided by the distance from the cooling water outlet of Ringhals NPP found by using Google Maps. A map of the collection locations is found in figure 11. All samples are given an identifier that is used for this project.

The last sample in table 1 is a fish caught somewhere in the Kattegat sea. Since the fish is not bound to one location, a distance to the cooling water outlet of Ringhals cannot be defined. Due to a possible mix up of samples in the SSAMS laboratory in Lund, no initial measurement results are given for the last sample.

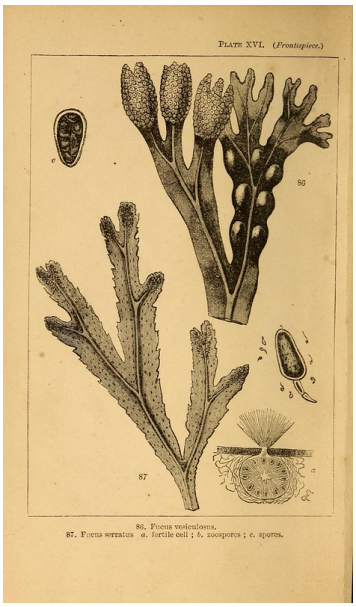
All seaweed samples were pretreated, being cleaned and acid/base/acid-treated, before initial measurements. The pretreatment removes any carbonate pieces or other impurities to ensure that the measured activity is from the organic material of the sample alone. This was done with samples of plant material, but not with animal material as impurities could have stuck to the outer side of the seaweed but not the meat of the fish and mussels.

The sample FV_Sär contains only the bladders and the upper part of the seaweed.

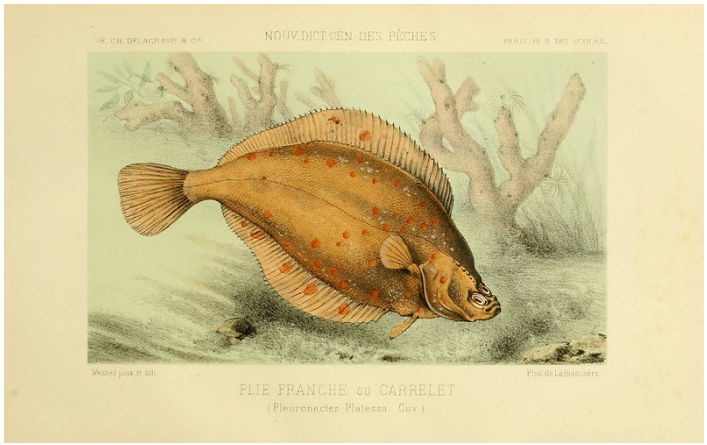
Table 1: Samples analyzed in this report. The table contains the short Latin name for the species; the collection date and location (Loc.); the distance (Dist.) from the collection site to the cooling water outlet of Ringhals; the ID given by the SSAMS laboratory in Lund (Lab ID); the given ID:s for this project (ID); the activities (Act.) and standard deviations (σ) found during initial measurements with the SSAMS in Lund.

Species	**Date**	**Loc.**	**Dist.** [km]	**Lab ID** (LuS)	**ID**	**Act.** [$F^{14}C$]	σ
Mytilus edulis	230908	Outlet	0	19468	ME_Out_1	2.812	0.010
Mytilus edulis	230908	Outlet	0	19469	ME_Out_2	2.795	0.010
Fucus serratus	250505	Bua	1	19056	FS_Bua_a	1.235	0.005
Fucus vesiculosus	200303	Särdal	66	15827	FV_Sär_a	1.048	0.006
Fucus vesiculosus	230830	Lysekil	120	19214	FV_Lys_a	1.068	0.006
Mytilus edulis	221012	Båteviken	197	18885	ME_Båt_1	1.056	0.005
Mytilus edulis	221012	Båteviken	197	18886	ME_Båt_2	1.059	0.005
Pleuronectes platessa	231001	Kattegat	-	21174	PP_Kat	-	-

(a)

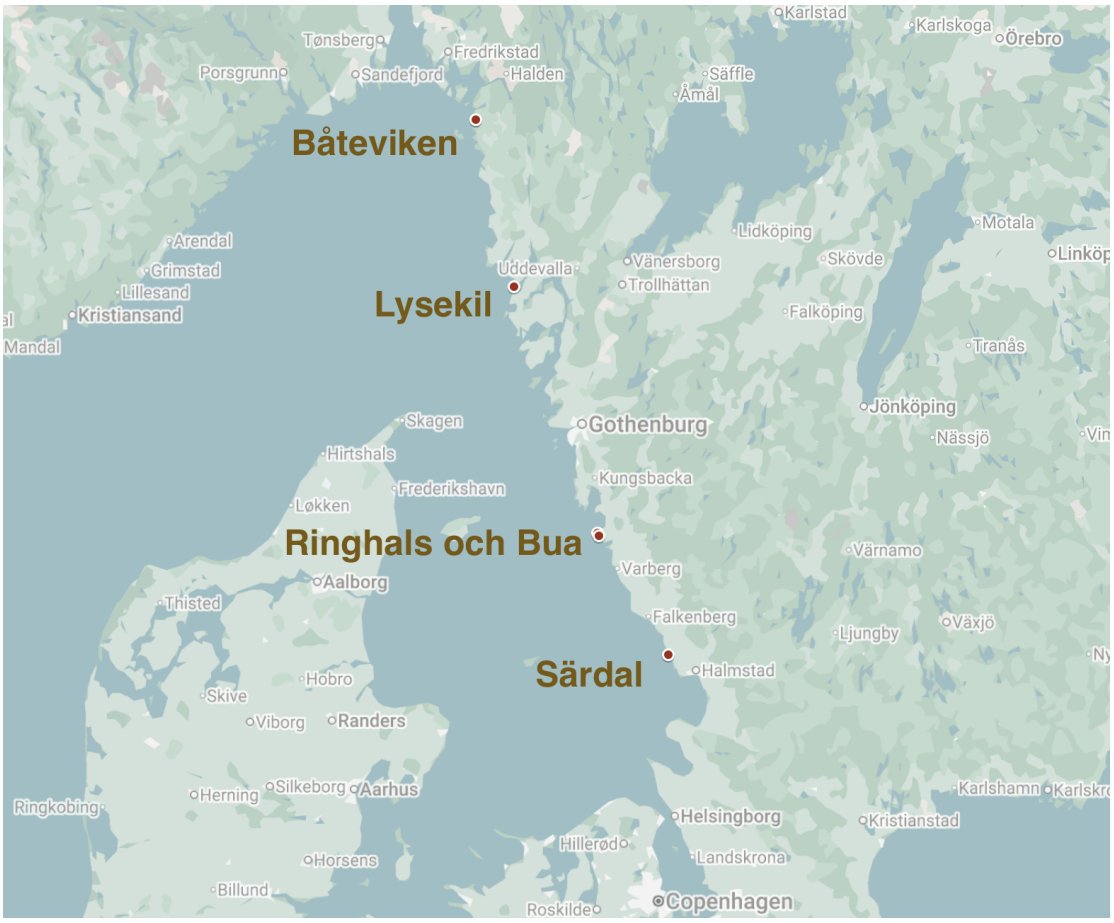


(b)

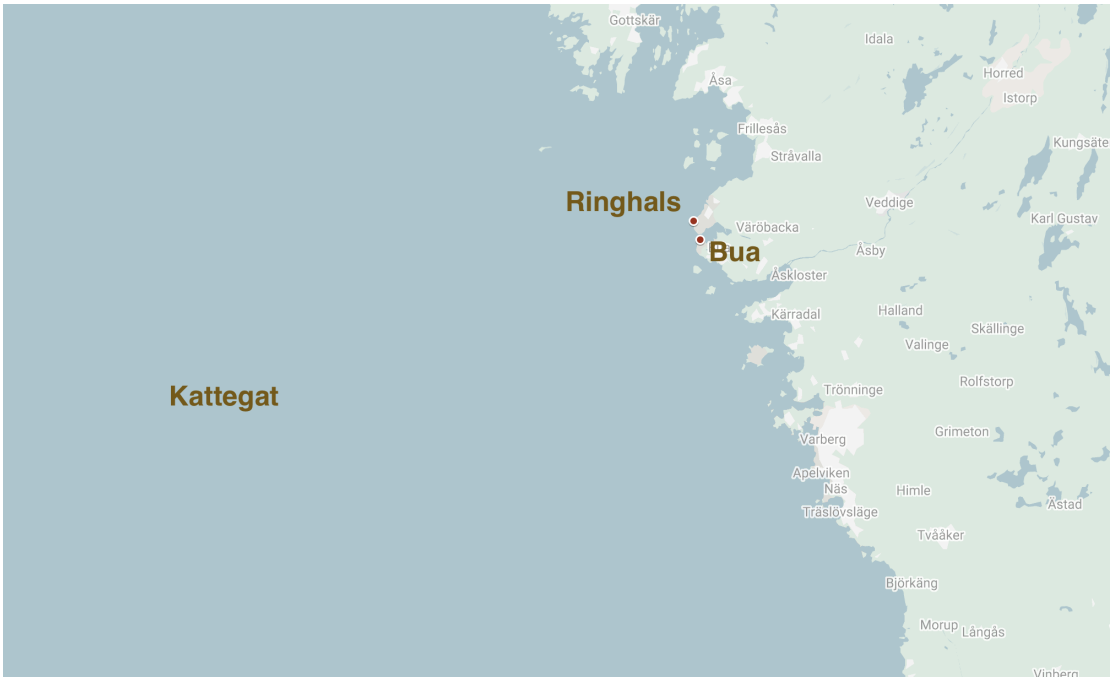


(c)

Figure 10: Illustrations of the four different organisms studied in this report. The illustrations have been downloaded from the Biodiversity Heritage Library [60], licensed under CC0 1.0 Universal (CC0 1.0) Public Domain Dedication license. (a): Illustration of *Mytilus edulis*, blue mussel, the top illustration. (b): Illustration of both *Fucus vesiculosus*, bladderwrack, in the top right, and of *Fucus serratus*, toothed wrack, in the bottom left. (c): Illustration of *Pleuronectes platessa*, European plaice.



(a)



(b)

Figure 11: Two maps showing the sampling locations used. Maps were created using Google MyMaps. (a): A map of the Swedish west coast with the different sample locations marked out. Map data ©2026 GeoBasis-DE/BKG (©2009), Google. (b): A zoomed in map of the area around Ringhals NPP. Bua is approximately 1 km SE of Ringhals. Map data ©2026, Google.

3.2 AMS

To test the homogeneity of the samples, an additional four measurements were conducted by the SSAMS in Lund: two subparts of sample **PP_Kat** and two subparts of sample **ME_Out_2**. The samples for the homogeneity test were handed in to the lab on the 13th of March, with results being reported on the 8th of April.

The samples were analyzed during different runs with the SSAMS. From all measurements at the facility in Lund, the results for the different standards that are used were acquired. These were used to evaluate the precision of the measurements.

For two of the seaweed samples, FS_Bua and FV_Lys, measurements were carried out also without the acid and base pretreatment that was done for the initial measurements. This was done to better be able to compare the results with the other analytical methods, since acid and bases are not used in their pretreatment protocols.

In addition to the measurements carried out with the Lund University SSAMS system, samples **PP_Kat**, **FS_Bua** and **FV_Lys** were analyzed at Atomki in Debrecen. The measurements were performed using the MICADAS and LEA devices using the two different graphitization methods: by using the sealed tube method or by using the AGE. All three samples were graphitized using the sealed tube method twice and analyzed using both the MICADAS and the LEA. All three samples were also graphitized using the AGE and analyzed using both the MICADAS and the LEA. The samples were delivered to Atomki on the 7th of April, with measurement results being reported on the 13th of May.

Two study visits were carried out for this thesis: one at the SSAMS-facility in Lund, and one at Atomki in Debrecen. The gathered information is presented in the sections below.

3.2.1 The SSAMS facility in Lund

The following information has been gathered through personal communication with the facility.

The single-stage accelerator mass spectrometry (SSAMS) facility is used for high-precision measurements of ^{14}C. The system was purchased in 2003 and installed in 2004. The laboratory infrastructure required for chemical sample preparation already existed prior to installation, meaning that the needed investment was the accelerator system itself together with a graphitization unit (Ionplus AGE 3) used to convert carbon dioxide into graphite targets for AMS measurements. The automated graphitization unit was installed after the AMS, and replaced previous own methods. The SSAMS instrument cost 1169315.74 USD which, with the mean USD-to-SEK exchange rate of 2003, corresponded to approximately 9.46 million Swedish crowns [61]. Adjusted for inflation, that is just over 14.3 million SEK in present value [62]. The device contains two ion sources and has a sample wheel with the ability to host 40 samples.

The price to have one sample analyzed at the facility is roughly 4,000 SEK, and it

increases with additions such as fast handling. No profit margin is included in the price.

The accelerator system occupies a floor area of 6.5 m × 4.5 m and is housed in a controlled laboratory environment. The facility requires temperature regulation to prevent the electronics from overheating, with room temperatures maintained below approximately 25 °C. Humidity control is also necessary, as both too high and too low humidity can cause electrical discharges in the high-voltage power supply. Therefore both a humidifier and a dehumidifier is installed. The system does not require special radiation shielding despite generating small amounts of X-radiation internally, and no specialized radiation protection equipment is necessary during operation.

The facility currently analyzes approximately 800–900 samples annually. Individual AMS measurement runs typically require approximately 2.5–3 days to complete, and measurements are usually performed roughly once per week. The main bottleneck in the analytical workflow is not the accelerator measurement itself but the chemical preparation of samples, particularly the conversion of carbon dioxide into graphite targets used for analysis. Turnaround time for samples is typically a few months for standard analysis, while customers paying for faster analyses can expect results delivered within a few weeks.

Maintenance requirements are moderate but regular. The ion source must be cleaned around four times annually, which results in a few days of downtime on each occasion. In total, planned maintenance may lead to several weeks of instrument downtime each year. Additional unplanned downtime may also occur, although many issues can be fixed internally by laboratory employees. Service support from the manufacturer is available, but has only occurred a few times since installation. Spare parts may become more difficult to obtain as the instrument ages.

The primary application of the SSAMS facility is radiocarbon dating, which makes up the majority of the analyzed samples. The system is capable of measuring extremely low ^{14}C concentrations corresponding to ages of up to approximately 43,000 years. Highly active samples, typically with $F^{14}C>10$ are generally avoided because very high ^{14}C activities could contaminate the ion source and affect later measurements. The facility has not been routinely used for environmental monitoring programs such as NPP environmental monitoring, apart from university research. However, in principle the laboratory could analyze such samples if required, depending on sample throughput and delivery time requirements.

3.2.2 The MICADAS and LEA facilities at Atomki

The following information regarding the AMS facility at Atomki in Debrecen, Hungary, has been gathered through personal communication and technical observations of their setup during a visit carried out in April of 2026. Atomki is the Institute of Nuclear Research of the Hungarian Academy of Sciences in Debrecen. The facility is unique in that it operates both a MICADAS- and a LEA-type AMS.

The facility is operating partly as a research institute, with a closely connected com-

pany, Isotoptech Zrt. The MICADAS device that is operated here is the third available MICADAS device. While MICADAS nowadays is produced by the company Ionplus, the device was bought from ETH Zürich before Ionplus was formed. Due to the device being bought from the university, rather than the succeeding company, the price for the device, that was installed in 2011, does not properly reflect the price of the device today. Because of the early design of the device, it still has features that the current models of the MICADAS have altered. One such feature is the use of electromagnets in the mass selectors, instead of permanent magnets that Ionplus currently equips MICADAS with. This makes the device rely on a cooling water system, that is reported to cause some problems.

The LEA that is operated at the facility is the first commercially available LEA installed in 2022. It was bought in a cooperation deal between Atomki and Ionplus, not necessarily reflecting the price of the device on the market. Both the devices, LEA and MICADAS, are used interchangeably, but approximately two thirds of the samples are measured in the MICADAS, which is fully booked, and the rest is analyzed using LEA, which is only booked for approximately 50% of the time. The MICADAS and LEA differ on several points at Atomki. The LEA analyzes gas instead of graphite targets, with an EA directly coupled to the device. Due to the LEA carrying out gas measurements, the facility uses the LEA to analyze smaller samples than with the MICADAS or if very quick measurements should be carried out; analyzing with the LEA does not require the graphitization process which saves a lot of time. Other differences between the operation of LEA and MICADAS are due to the latter being many years older than the former; the LEA is described to be more user-friendly with a newer software, the ion current being more stable and the system requiring less manual tuning than the MICADAS version at Atomki. The LEA is also reported to be in need of less maintenance than the MICADAS.

The price to analyze a standard organic sample is 360 EUR which, with the conversion rate of 1 EUR = 10.83 SEK (April 2026) corresponds to roughly 3,900 SEK. Carbonates and water samples are cheaper, while certain materials or additions are more expensive. Since the facility is both research-based and offers commercial services through the company, the prizes are adjusted to conditions on the market with interest of profit and do not only reflect the cost of actually analyzing a sample. While the cost of labor and consumables produced in Hungary are cheaper than alternatives in Sweden, the instrument, having been produced in Switzerland, is equally as expensive in Sweden as in Hungary.

Maintenance costs can be high if service from Ionplus is needed. However, this is rarely needed, and occurs approximately once every three years. Due to the facility having skilled engineers, scientists and technicians, most problems that occur can be solved internally. While running the instrument routinely is carried out by people with various scientific backgrounds at the facility, being able to troubleshoot and perform maintenance requires more advanced technical skills such as by trained engineers or scientists with PhDs toward instrumentation.

Sample preparation for the MICADAS is partly carried out as at the SSAMS facility in Lund, using an elementar analyzer and AGE graphitization device to make the graphite targets. In addition to using the AGE graphitization, the facility at Atomki also

manages a home-built sample preparation line that converts the sample to graphite. The following section is a brief description of the procedure.

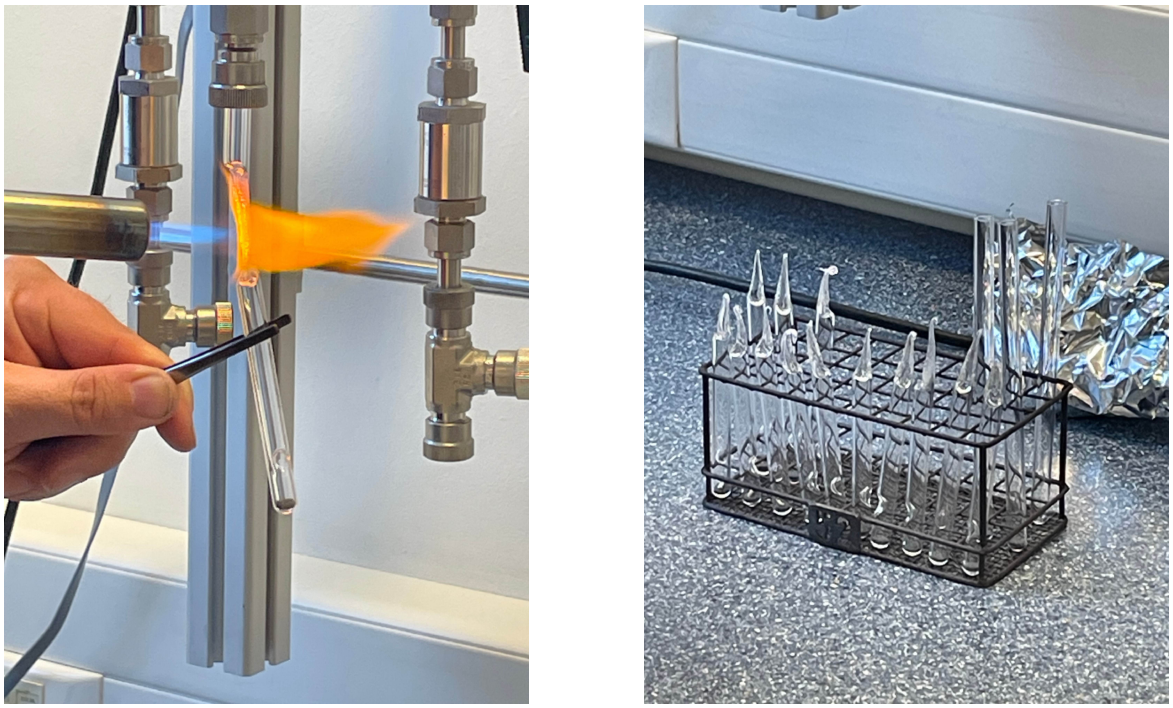
The process starts by measuring the needed amount of sample and adding it to a glass vial. Instead of combusting the sample in a stream of oxygen gas, manganese dioxide, MnO_2, is added to the vial as well, with oxygen contents that well exceed the stoichiometric ratio, ensuring complete combustion [63]. The vials are then inserted into a vacuum line, in which the air that resides in the vial is pumped out. The glass vial is then sealed using a torch flame. What remains in the vial is then simply the sample and MnO_2. The next step is combusting the samples in a furnace at maximum 550°for 12 hours, during which the oxygen releases and is used to combust the sample, producing CO_2. When the vials have cooled, they can be inserted into a gas-handling line, where a hole is poked through the glass allowing the gasses within the vial to enter the line. A sequence of traps, using first isopropyl-aclohol and dry ice then liquid nitrogen, is used to trap water and then freeze only the CO_2. Vacuum is maintained in the line using scroll pumps.

The next step of the process is to graphitize the produced CO_2 that has been isolated. A reactor tube is used, consisting of a combination of two glass tubes; one with a bigger diameter and length, to which a smaller tube, with shorter diameter and length is inserted. The outer tube is loaded with zinc and titanium hydrate, TiH_2, while the inner tube contains iron [64]. The isolated CO_2 from the previous step is inserted to the reactor tube, and the outer tube, that totally encompasses the inner tube is sealed in the same manner as previously performed, with a torch flame. The reactor tubes containing the CO_2 from all samples then enter the furnace once more. First, they are heated to 500°for three hours, allowing the H_2 to be released from the TiH_2. The temperature is then raised to 550°during which the zinc acts as a reducer, and the iron catalyses the process turning CO_2 into elemental carbon in the form of graphite.

The sample combustion and graphitization that is carried out with the sealed tube method is described to be both cheaper and more effective and flexible than graphitization with the EA and AGE. While they with the AGE only can produce a maximum of 14 graphite targets per day, one person could, with the sealed tubed method, produce up to 30 graphite targets per day. On the other hand, graphitization with the AGE requires less manual labour and produces targets of more consistent quality.

By having and using two methods of graphitization, of which one is comparatively quick and can be scaled up or down based on demand, the facility has a much higher throughput than, for example, the one in Lund. At Atomki the annually measured samples amount to about 4,500 to 5,000.

Atomki has experience analyzing samples of various different sorts and for different causes. Together with the company, Isotoptech Zrt., samples are tested both commercially and as a part of research. Many of the analyzed samples are archeological, but they also carry out bio-based testing, as well as environmental monitoring both nationally and internationally. Because of this the facility has experience measuring samples with activities that vary greatly, from archeological samples to highly enriched samples. Handling of high activity samples could lead to contamination within the laboratories. At the facility there are two sample preparation laboratories, one of



(a) (b)

Figure 12: Pictures showing the beginning of the sealed tube method for graphitization as used at Atomki in Debrecen. (a): Sealing a tube containing a sample and MnO_2 using a torch. (b): A number of sample vials that have been sealed and are ready for combustion.

which handles any sample that is suspected to be above 10 $F^{14}C$. Between the two laboratories there cannot be any crossovers. If contamination should occur, it could affect all measurements negatively, forcing a total clean-out of the laboratory, which the facility has had to do in the past.

Hungary currently has one NPP, which is situated along the Danube. The environmental monitoring that is carried out by Atomki related to ^{14}C focuses on the releases in gas-form. Releases to the Danube are not measured, but the ground water is measured regularly.

3.3 LSC

Three samples were sent to CARER, Centre for Advanced Research in Environmental Radioactivity, in Mangalore, India. The samples that were sent were **PP_Kat**, **FS_Bua**, and **FV_Lys**. The LSC used for the measurements was the Quantulus 1220. The samples arrived to CARER on the 24th of March, with results being reported back on the 21st of May.

3.3.1 The Quantulus 1220 facility at CARER in Mangalore

The following information has been gathered through personal communication with the facility.

The liquid scintillation counting (LSC) facility at CARER in Mangalore uses a Quantulus 1220 ultra-low-background liquid scintillation counter for low-level environmental ^{14}C measurements. The instrument was installed in 2014 and has since been used primarily for environmental sample analyses, although the laboratory also has experience with radiocarbon dating of archaeological samples. The facility mainly focuses on low-level environmental monitoring. The approximate investment cost of the Quantulus system was estimated by the laboratory to be around 200,000 USD, which , with the rate of 1 USD = 9.49 SEK (April 2026) is equivalent to 2.5 million SEK when adjusted for inflation.

The sample preparation procedure is based on thermal combustion followed by direct absorption of the generated CO_2 into an absorber-scintillator mixture. The combustion process takes approximately five hours, followed by centrifugation and oven drying requiring an additional six hours. Carbon dioxide regeneration takes roughly one hour. After preparation, the samples are stored in darkness for approximately twelve hours before measurement. In total, the complete analytical procedure for one sample, including reporting of results, takes approximately three days.

The facility currently analyzes approximately 600–800 samples annually. According to the laboratory staff, the instrument is operated almost continuously and the present workload is close to the maximum capacity. The main bottlenecks are the long counting times required for low-activity environmental samples together with the manual work involved in sample preparation. Environmental samples are typically measured for 8 hours and 20 minutes, divided into ten counting cycles of fifty minutes each. Increasing the counting time improves the counting statistics and reduces the measurement uncertainty.

A significant amount of manual labour is involved in the analytical workflow, particularly during sample preparation and processing. The complete procedure requires approximately two employees. According to the laboratory, personnel with a scientific background, especially in chemistry or physics, can learn the procedure after suitable training.

The cost of one analysis is mainly driven by consumables rather than by the instrument itself. The laboratory charges industrial customers approximately 250 USD per sample, corresponding to around 2,400 SEK.

The Quantulus system requires regular maintenance but is generally regarded as stable and reliable. The laboratory reports that the instrument has functioned reliably for more than ten years. Measurement uncertainties are mainly influenced by sample preparation errors, although instrument-related factors can also contribute.

3.4 ^{14}C-SCAR

Three samples were analyzed at RISE, the Research Institute of Sweden, at the ^{14}C-SCAR facility located in Borås. The samples were **PP_Kat**, **FS_Bua**, and **FV_Lys**. The samples arrived to RISE on the 11th of March, and the results were reported on the 17th of March.

In addition to this, all samples except for **ME_Out_2** were tested by the company that produces the C14-SCAR-device, ppqSense, at their facility in Florence using more recent versions of the device compared to the one in Borås. Four of the samples, **PP_Kat**, **FS_Bua**, **FV_Lys**, and **ME_Out_1**, were analyzed in March, while the rest, **FV_Sär**, **ME_Båt_1**, and **ME_Båt_1**, were analyzed during a study visit in April. During the study visit, a feature of the device that is not yet available to customers was tried to determine the isotope fractionation. While the four samples that were analyzed in March were corrected using δ^{13}C-factor found in literature or by analysis of similar samples, the samples that were analyzed during the study visit were corrected using the factor determined by the device itself. δ^{13}C for seaweed samples were estimated to be -15 ‰ through personal communication with Kristina Stenström found by averaging measurements using an IRMS. The δ^{13}C used for blue mussels was estimated to be -22 ‰ from [65] and for the fish, -19 ‰ from [4].

The samples were sent to ppqSense from Sweden on the 24th of February with the results being reported on the 9th of March

Two study visits were carried out, both at RISE in Borås and at ppqSense in Florence. The gathered information is presented below.

3.4.1 The ^{14}C-SCAR facility at RISE in Borås, Sweden

The following information has been gathered through personal communication with the facility.

The SCAR facility at the Research Institutes of Sweden (RISE) operates the earliest commercially available SCAR instrument, produced by the Italian company ppqSense. The system was installed in 2022 with a total investment cost of approximately 7 million SEK, not adjusted for inflation. This included the required sample preparation equipment. The investment is expected to amortize over approximately 7–8 years. Annual maintenance costs are relatively low compared to the initial investment, estimated at roughly 40,000 SEK per year, excluding routine laboratory consumables.

The cost of having one sample analyzed at the facility is roughly 5,500 SEK. While RISE is fully owned by the Swedish state it is still run as a company with interest for profit. The laboratory is also accredited by Swedac, which raises the price for customers.

The instrument is installed in a standard laboratory environment without special radiation shielding requirements. The facility maintains a temperature-controlled environment of 22 °C. No other infrastructure is necessary that was not provided with the

machine. The SCAR instrument itself has design features that reduce external disturbances such as vibrations. Consequently, the system can be operated in a typical analytical laboratory setting rather than requiring a highly specialized facility.

Routine operation of the instrument can be carried out by laboratory personnel after short training, although troubleshooting and maintenance require greater experience. Daily calibration is performed using control and calibration samples to ensure measurement quality.

The current throughput of the facility is 100–200 samples annually, with tests typically being performed in batches when the instrument is operating, which is not daily. It is estimated that seven samples could be analyzed each day with the current standard amount of scans (two), which would result in a much higher throughput. This does, however, require more allocated working hours; the current team of two people do not only work with the SCAR-laboratory. Sample preparation and combustion can be performed while measurements are ongoing, which means that minimal downtime between measurements can be acquired.

The time from the submission of the sample to the reporting of the results in communicated as 10 working days. The downtime of the instrument is relatively limited; unscheduled maintenance is roughly estimated to be required once every three months, and most of the problems that occur can be solved within one day. Service from the manufacturer has been required twice within the four-year time span of operation. Maintenance of the vacuum pumps and the elemental analyzer are reported to require more attention. More challenges were encountered in the beginning of the implementation of the instrument, but it has grown to be more reliable with the experience of the employees.

The facility is capable of measuring ^{14}C levels much smaller than modern background levels, with routine reporting down to 3 pMC with a relative expanded uncertainty of $\pm 3\%$, representing two standard deviations. The technique is not used by RISE for radiocarbon dating applications, instead the focus is on testing a vast range of samples, such as fuels or materials, for their fossil and biogenic content; by measuring the concentration of ^{14}C the share of contents stemming from fossil or modern sources can be determined. Since the ^{14}C in fossil sources have decayed completely by now, a plastic bag or a fuel made completely by fossil oil will show no ^{14}C concentration. Customers of the facility at RISE are mainly companies or authorities demonstrating the climate friendliness of products, often to receive a certificate or prove the upholding of commitments. While it is technically possible for the instrument to analyze samples with much higher ^{14}C concentration than modern background levels, the facility at RISE has chosen not to accept those types of samples, for work environment reasons. A facility using the same instrument and more experience with samples with higher levels of radioactivity would be able to carry out measurements of samples with more ^{14}C concentration.

3.4.2 The ^{14}C-SCAR facility at ppqSense in Florence, Italy

ppqSense is an Italian company that specializes in developing instruments used in laser spectroscopy. One of their developments is their C14-SCAR, which is an implementation of the SCAR-technology for ^{14}C. At the moment of writing (April 2026), they are building their ninth and tenth devices, belonging to the second generation of the device. The development from the first to the second generation is mostly due to the incorporation of mirrors whose reflectance is better. This allows for the cavity to shrink to less than half of the original size, and the rest of the system to be scaled down. The time to perform one scan has also decreased from about 25 minutes to 15 minutes, while simultaneously reducing the uncertainty.

While they primarily work as a manufacturer of the devices they also perform sample analysis with their devices as a sideline, both for research and commercially. In a year, they analyze somewhere around 100 to 150 samples. The cost of analyzing a sample by ppqSense is 350 euros, approximately 3,800 SEK (1 euro = 10.87 SEK in May 2026). Most of the samples tested are different materials or fuels for which the fossil to bio-based ration is determined. Customers can, for example, be oil companies looking to prove their incorporation of biofuels or the fashion industry testing textiles. A future application can be companies proving their emissions of CO_2 being of non-fossil nature, within the EU emissions trading system. A growing interest has been noticed in using the technique for environmental monitoring. For example, a Swiss customer has implemented the SCAR for continuous measurement of the ^{14}C content of air. The applications of the SCAR within the pharmaceutical industry is also under research. While the detection limit of the devices are low, the company does not seem to focus on competing with the AMS for archeological radiocarbon dating purposes.

The SCAR devices do not measure the isotope fractionation directly. The ability to include a feature that measures it is in progress at the company. However, if needed, the frequency of the laser can already be shifted by the staff to a molecular transition of $^{13}CO_2$, and its concentration being measured instead, giving a δ^{13}C-coefficient that can be used to correct the measurement results for better intercomparison with AMS-results.

3.5 Statistical analysis

To determine whether the measurement results from different analytical techniques were in statistical agreement with each other, pairwise comparisons were conducted for some of the measurement results using a modified z-score, sometimes referred to as a Zeta (ζ) score [66]. When comparing two independent measurement results, x_1 and x_2, each with their own reported standard deviation, σ_1 and σ_2, the z-score for the difference is calculated as follows:

$$z = \frac{x_1 - x_2}{\sqrt{\sigma_1^2 + \sigma_2^2}} \tag{10}$$

This statistic normalizes the absolute difference between the two methods by their

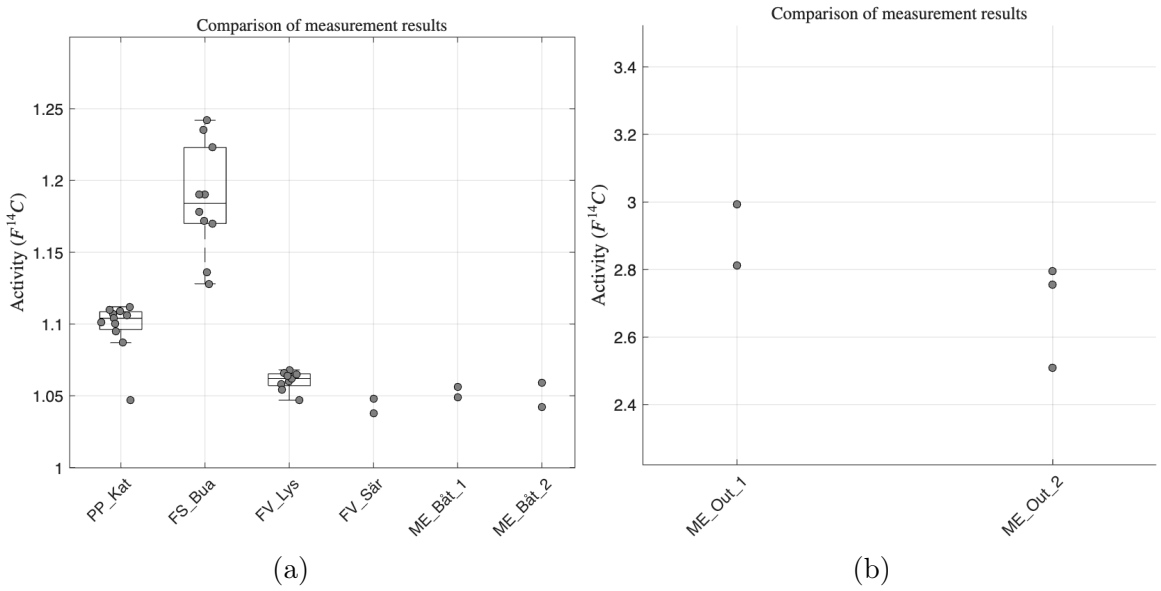
combined standard uncertainty. The resulting z-score is interpreted using normal distribution criteria to evaluate the compatibility of the methods:

- $|z| \leq 2$: Indicates agreement. The difference between the two measurements is statistically insignificant and can be entirely explained by the reported measurement uncertainties.
- $2 < |z| < 3$: Indicates a questionable difference, implying a potential minor systematic discrepancy or an underestimation of uncertainty by one of the laboratories.
- $|z| \geq 3$: Indicates unsatisfactory agreement. The two methods significantly disagree, implying a strong systematic error or sample inhomogeneity.

4 Results

4.1 Overview

Tables containing all measured results is found in Appendix A. A plot illustrating all of the results is seen in figure 13. For samples that were measured four times or more, a box plot was used to show the spread of the results, while, for the samples with three or less measurement results, the results are shown without a box plot. The figure is divided in two subfigures, since two of the samples show much higher activities than the rest.



(a) (b)

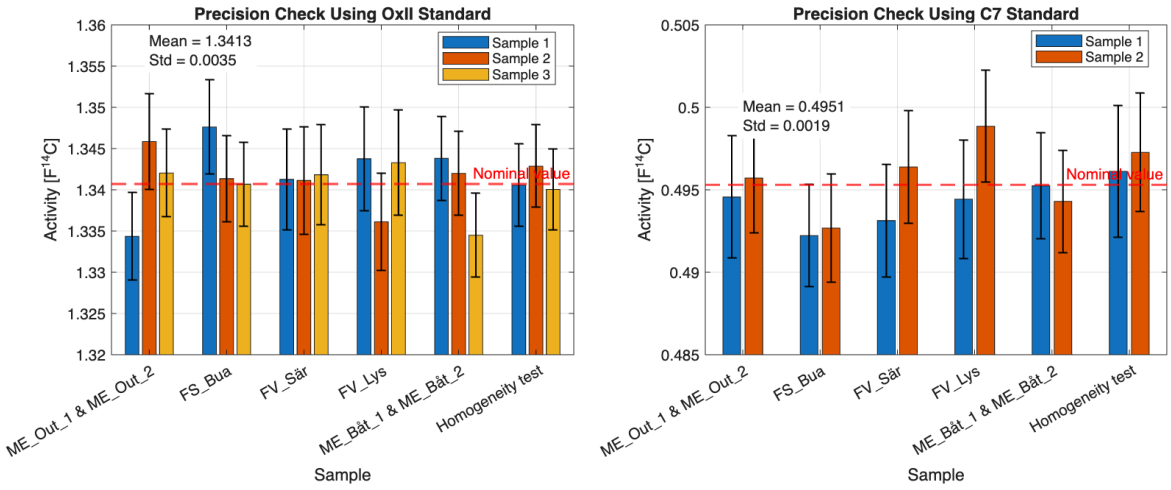
Figure 13: Measured activities of all samples using the different analytical methods. Box plots are used to illustrate the spread of the results, for samples with more than three measurement results. The numerical values that are plotted can be found in tables A1 and A2. (a): Six of the samples and their measurement results plotted either by themselves or together with a box plot. (b): Two of the samples and their measurement results plotted either by themselves or together with a box plot.

From the box plots, there is a strong indication that sample FS_Bua is inhomogeneous. The large spread of measurement results could also be due to lacking precision from the analytical methods, but the precision appears to be decent for other samples. Despite FV_Lys having many measurement results, all results are concentrated in close proximity. PP_Kat also shows high precision results and high homogeneity, except for one measurement result, which, for now, can be considered an outlier. The rest of the samples show similar results, but the deviation increases for the two blue mussel samples that were collected right by the cooling water outlet. With the constantly fluctuating levels of ^{14}C, they are expected to be inhomogeneous.

A breakdown of all results using the different analytical methods for each sample will be provided in the following sections.

4.2 Precision check for SSAMS

The activities for the standards that were used during analysis of the samples in this thesis were retrieved and plotted in figure 14. Some of the samples were measured on the same wheel in the AMS. Three samples of the OxII-standard and two samples of the C7-standard were measured on each sample wheel. The nominal values for the OxII-standard and the C7-standard are 1.3407 $F^{14}C$ and 0.4953 $F^{14}C$ respectively.



(a) (b)

Figure 14: Precision checks carried out for two of the standards used during measurements at the SSAMS laboratory in Lund. Some samples were graphite targets on the same sample wheel. (a): Precision check for the OxII standard that has a nominal value of 1.3407 $F^{14}C$. Three samples of the standard were measured on each sample wheel. (b): Precision check for the C7 standard that has a nominal value of 0.4953 $F^{14}C$. Two samples of the standard were measured on each sample wheel.

The precision check for the analyses using the SSAMS in figure 14 show good results: for the OxII standard the mean is 1.3413 $F^{14}C$ compared to the nominal value of 1.3407 $F^{14}C$ and the standard deviation is 0.0035 $F^{14}C$ which, converted to a relative measure, only is 0.26%. For the C7 standard the mean is 0.4951 $F^{14}C$ compared to the nominal value of 0.4953 $F^{14}C$ and the standard deviation is 0.0019 $F^{14}C$ which, converted to a relative measure, only is 0.38%. The uncertainties are in both cases minor.

4.3 Precision check for SCAR at ppqSense

All results for the standard OxII that were measured on the day of analysis of the samples FV_Sär, ME_Båt_1 and ME_Båt_2 at ppqSense in Florence are plotted in figure 15 together with the nominal value of 1.3407 $F^{14}C$.

The precision check for the SCAR at ppqSense before the measurements of samples FV_Sär, ME_Båt_1 and ME_Båt_1 shown in figure 15 shows decent results. Especially the accuracy of the measurements stands out: the mean of the 12 scans is only

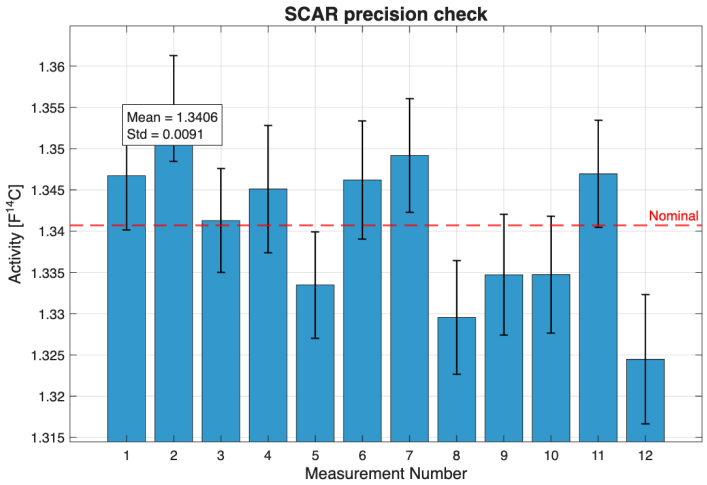


Figure 15: Precision check for the OxII standard that has a nominal value of 1.3407 $F^{14}C$. Twelve measurements were carried out at ppqSense in Florence of the standard on the day of measuring the samples FV_Sär, ME_Båt_1 and ME_Båt_2.

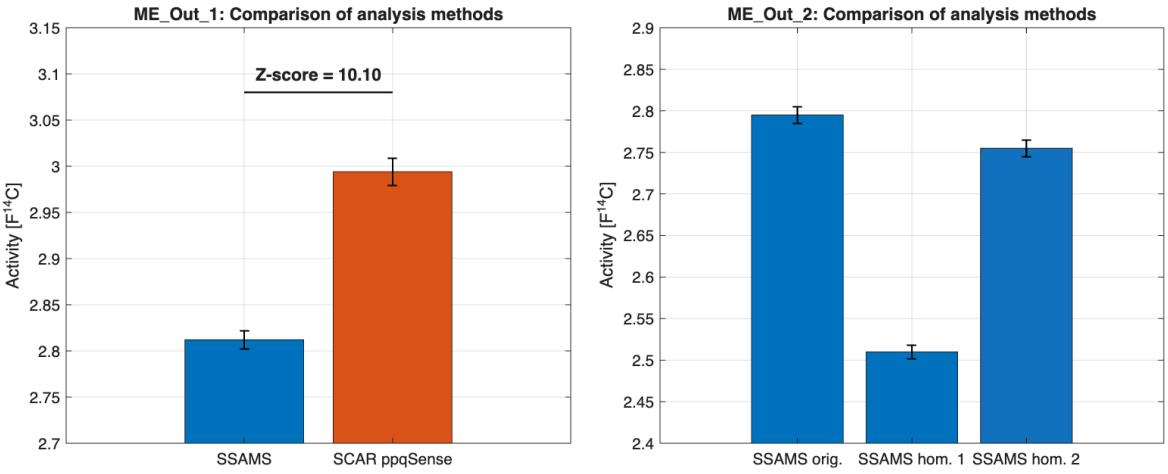
0.0001 $F^{14}C$ off from the nominal value of the standard. The standard deviation of the 12 scans is 0.0091 $F^{14}C$, which is a relative deviation of 0.068%. This result is slightly higher than the reported precision. When looking at the figure, it appears that most scans yield results close to the nominal value, but a few scans have larger deviations increasing the standard deviation. These can occur for slight shifts in laser frequency, pressure or temperature. However the accuracy and precision are still good enough for most applications.

4.4 Intercomparison

4.4.1 ME_Out_1, ME_Out_2, ME_Båt_1 and ME_Båt_2

Results for all samples of blue mussel using different methods of analysis are shown in figures 16 and 17. The results for the blue mussel samples that were collected by the cooling water outlet of Ringhals NPP are shown in figure 16a and 16b. For figure 16a, the SCAR measurement result from ppqSense is the corrected value from table A1, using an estimation of the $\delta^{13}C$ from literature. In the same figure, a Z-score is presented that compares the SSAMS-result to the SCAR-result. Figure 16b shows the original SSAMS-results together with the results for the homogeneity tests.

Figure 16b shows multiple measurement results using the SSAMS. A homogeneity test was conducted and the results are compared to the original measurement results. The activities clearly differ more than they should given the small uncertainties. A statistically significant difference between the results is determined. Considering that all measurements were conducted on the same sample, with the same procedures, it is highly unlikely that the differences come from a lack of accuracy or precision of the SSAMS. Instead, the results should be regarded as a strong indicator of sample inhomogeneity. This is especially likely considering that the blue mussel sample was collected by the cooling water outlet of Ringhals NPP, where the ^{14}C-levels fluctuate



(a) (b)

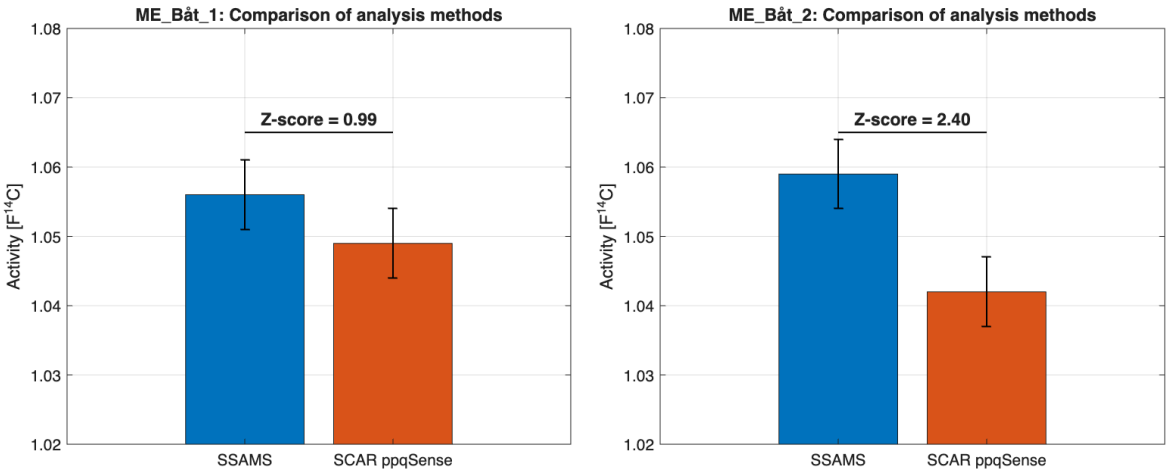
Figure 16: Results for the two blue mussel samples collected by the cooling water outlet of Ringhals NPP, measured using the SSAMS in Lund and the SCAR at ppqSense in Florence. (a): Measurement results for ME_Out_1 using the SSAMS in Lund and the SCAR in Florence. The SCAR-results are corrected using a δ^{13}C estimation from literature. A Z-score is also displayed, evaluating the difference between the analytical methods. (b): Measurement results for ME_Out_2 using the SSAMS in Lund. The original measurement result is presented together with the results from the homogeneity test.

greatly, which can explain the inhomogeneity of the sample. This is further strengthened by the fact that the other homogeneity test that was performed, on PP_Kat in figure 20, shows no inhomogeneity. The European plaice fish, in contrast to the blue mussel, did not live by the cooling water outlet of Ringhals NPP, and was probably not exposed to as large fluctuations in ^{14}C activity. The meat of the fish should also show more homogeneous spread of ^{14}C.

For the sample ME_Out_1, whose results are presented in figure 16a, there is a statistically significant difference between measurements performed using the SSAMS and the SCAR. The SCAR result is corrected using an estimated isotope fractionation value. From the discussion regarding sample ME_Out_2 in figure 16b it is unlikely that the difference between the SSAMS and SCAR is due to lacking accuracy or precision for any of the measure. The difference is instead highly linked to the inhomogeneity of the blue mussel samples from the cooling water outlet of Ringhals NPP.

The results for the blue mussel samples that were collected in Båteviken are shown in figure 17a and 17b. The SCAR measurement results from ppqSense are the corrected values from table A1, using the δ^{13}C measured by the SCAR-device. In the figures, a Z-score is presented that compares the SSAMS-result to the SCAR-result.

For the samples of blue mussel that were collected roughly 200 km away from Ringhals NPP, ME_Båt_1 and ME_Båt_2 (figure 17) the SSAMS and SCAR measurement results agree more than for ME_Out_1. For ME_Båt_1 the results show a Z-score of less than one, showing good agreement, while for ME_Båt_2 there is an indication of



(a) (b)

Figure 17: Results for the two blue mussel samples collected in Båteviken, measured using the SSAMS in Lund and the SCAR at ppqSense in Florence. (a): Measurement results for ME_Båt_1 using the SSAMS in Lund and the SCAR in Florence. The SCAR-results are corrected using the δ^{13}C measured by the SCAR-device. A Z-score is also displayed, evaluating the difference between the analytical methods. (b): Measurement results for ME_Båt_2 using the SSAMS in Lund and the SCAR in Florence. The SCAR-results are corrected using the δ^{13}C measured by the SCAR-device. A Z-score is also displayed, evaluating the difference between the analytical methods.

a questionable difference between the measurement results. Although the blue mussel has shown a tendency to have inhomogeneous spread of ^{14}C, the large distance from Båteviken to Ringhals should mean fairly stable levels of ^{14}C in the waters, leading to less inhomogeneity. One measurement result deviation is not enough to draw any conclusions about differences between AMS and SCAR and their accuracies; unless this type of difference proves to be a trend it is more likely that the deviating results are due to something like sample preparation or memory effects within the systems.

4.4.2 FS_Bua

The measurement results for FS_Bua using different analysis methods are shown in figure 18. The sample was pretreated with acid-and-base treatment before the original SSAMS-measurement, in contrast to all of the other measurements. A measurement was also carried out with the SSAMS without the sample pretreatment with acids and bases. The samples analyzed with MICADAS and LEA were graphitized using either their own sealed tube method or by using the automated AGE. Both the SCAR-measurements from RISE and from ppqSense were corrected using an average δ^{13}C from IRMS-measurements performed by Kristina Stenström that was provided through personal communication.

For FS_Bua the results are deviating greatly. This is a strong indication that the toothed wrack sample is inhomogeneous. The sampling location, Bua, is only roughly 1 km from Ringhals which causes variations in the ^{14}C exposure for the seaweed. The variations cause uneven distribution of ^{14}C within the plant, and since AMS and SCAR

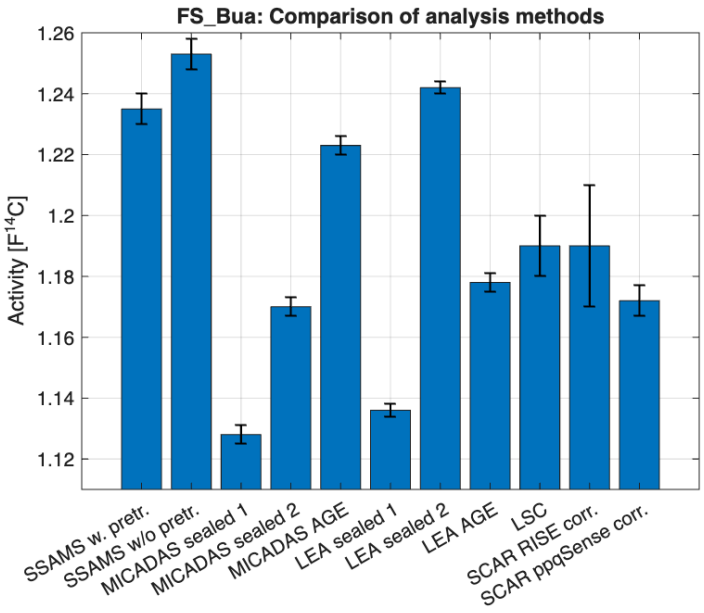


Figure 18: Measurement results for FS_Bua using the SSAMS in Lund, the MICADAS and LEA at Atomki in Debrecen, the LSC at CARER in Mangalore and the SCARs at RISE and ppqSense in Borås and Florence. The SSAMS-measurements were both with (w.) and without (w/o) acid-and-base pretreatment (pretr.). "Sealed" refers to the sealed tube graphitization method used for some of the samples analyzed at Atomki. Both the SCAR-measurements from RISE and from ppqSense were corrected using an average δ^{13}C from IRMS-measurements performed by Kristina Stenström that was provided through personal communication.

only require tiny samples to be able to perform a measurement, it is possible that the tiny parts that are measured do not fully represent the sample as a whole.

There are variations even between measurements using the same analytical methods. For example, the sealed tube measurements for both the MICADAS and the LEA show statistically significant differences. This is not the case for other samples, such as FV_Lys and PP_Kat, which further strengthens the theory that the measurement result variations are due to inhomogeneity.

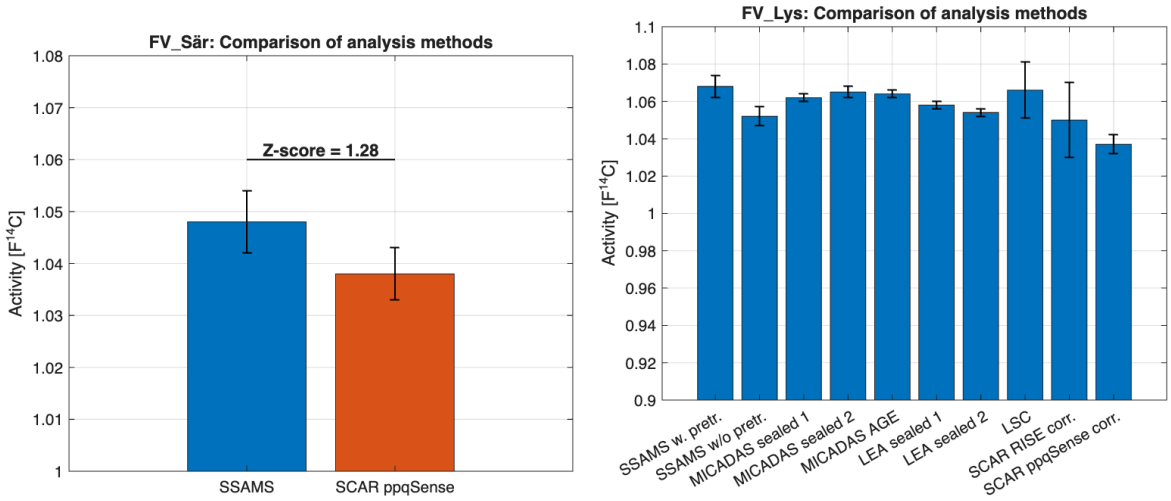
The measurement results with the SSAMS in Lund also show differences with a z-score of $z = \frac{1.253-1.235}{\sqrt{0.005^2+0.005^2}} \approx 2.55$ which implies a potential systematic discrepancy between the results using acid-and-base pretreatment. A conclusion cannot be drawn based on this sample alone, due to its inhomogeneity.

Evaluation of the different analytical methods cannot be made by studying the sample FS_Bua alone, since it likely is very inhomogeneous. One thing worth noting, however, is that the accuracy of LSC should benefit from an inhomogeneous sample compared to the other techniques, since LSC requires much larger sample sizes. The sample that was tested by the LSC should therefore have a greater chance of including parts of both higher and lower activities to the analysis. If this is the case, the LSC measurement could be more representative for the sample as a whole, and its results be considered more of an average for the sample than for the other analytical methods. For this sample, the LSC result is indeed in the middle of the range.

4.4.3 FV_Sär and FV_Lys

Results for the two samples of bladderwrack using different methods of analysis are shown in figures 19a and 19b. The results for the bladderwrack sample that were collected in Särdal are shown in figure 19a. For figure 19a, the SCAR measurement result from ppqSense is the corrected value from table A1, using the δ^{13}C measured by the SCAR-device. In the same figure, a Z-score is presented that compares the SSAMS-result to the SCAR-result. Figure 19b shows the measurement results for the bladderwrack collected in Lysekil. Both the SCAR-measurements from RISE and from ppqSense were corrected using an average δ^{13}C from IRMS-measurements performed by Kristina Stenström that was provided through personal communication.

For both samples, the SSAMS-measurements were carried out on samples that had been acid-and-base-treated. For sample FV_Lys measurements were also performed without the pretreatment.



(a) (b)

Figure 19: Results for the two samples of bladderwrack collected in Särdal and Lysekil, measured with different methods. (a): Measurement results for FV_Sär using the SSAMS in Lund and the SCAR in Florence. The SCAR-result is corrected using the δ^{13}C provided by measurements at ppqSense. A Z-score is also displayed, evaluating the difference between the analytical methods. (b): Measurement results for FV_Lys using the SSAMS in Lund, the MICADAS and LEA at Atomki in Debrecen, the LSC at CARER in Mangalore and the SCARs at RISE and ppqSense in Borås and Florence. The SSAMS-measurement was performed both with (w.) and without (w/o) acid-and-base pretreatment (pretr.). "Sealed" refers to the sealed tube graphitization method used for some of the samples analyzed at Atomki. The AGE-graphtized sample measured with LEA failed. Both the SCAR-measurements from RISE and from ppqSense were corrected using an average δ^{13}C from IRMS-measurements performed by Kristina Stenström that was provided through personal communication.

Based on the data for the bladderwrack samples FV_Sär and FV_Lys, figure 19, the sample appears to be homogeneous. All AMS measurements show agreement with each other, regardless of the AMS type or the preparation used (seales tube method and AGE). It is worth noting, however, that the SSAMS results yield a z-

score of $z = \frac{1.068-1.052}{\sqrt{0.006^2+0.005^2}} \approx 2.05$ when comparing results with and without acid-base pretreatment, indicating a small but noticeable shift in activity. While the LSC and SCAR RISE results align closely with the values of the AMS, they show lower precision due to their larger measurement uncertainties. The SCAR ppqSense measurements show a slight systematic deviation, resulting in unsatisfactory agreement with z-scores exceeding 3 against the majority of the AMS data. A slight error could have been introduced due to the isotopic fractionation correction, as the ppqSense data relied on an average δ^{13}C value from previous IRMS runs rather than a sample-specific measurement.

4.4.4 PP_Kat

The measurement results for PP_Kat using different analysis methods are shown in figure 20. The sample was tested for homogeneity with the SSAMS in Lund. The samples analyzed with MICADAS and LEA were graphitized using either their own sealed tube method or by using the automated AGE. Both the SCAR-measurements from RISE and from ppqSense were corrected using an average δ^{13}C from literature.

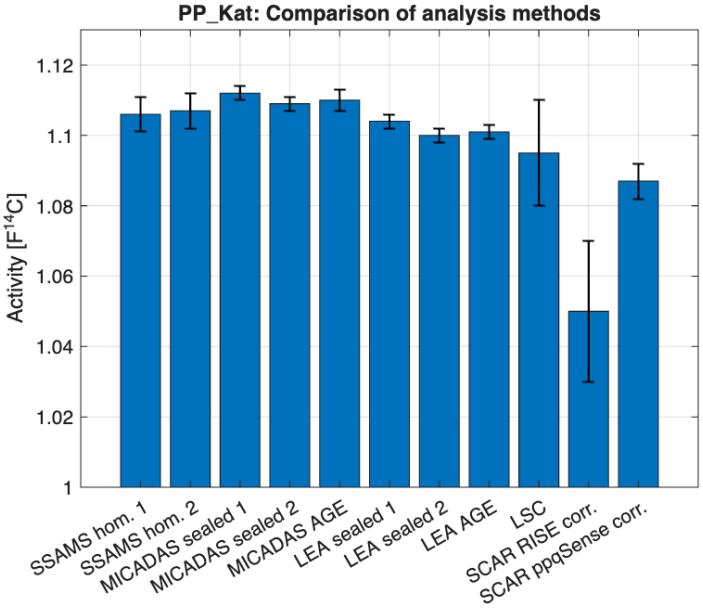


Figure 20: Measurement results for PP_Kat using the SSAMS in Lund, the MICADAS and LEA at Atomki in Debrecen, the LSC at CARER in Mangalore and the SCARs at RISE and ppqSense in Borås and Florence. "Sealed" refers to the sealed tube graphitization method used for some of the samples analyzed at Atomki. Both the SCAR-measurements from RISE and from ppqSense were corrected using an average δ^{13}C from literature.

For the last sample, PP_Kat, the AMS-measurements show very similar results. The SCAR measurements deviate, especially the RISE measurements. It should be noted that the SCAR results have been corrected using an estimated isotope fractionation value. Both SCAR results still show statistically significant differences to at least some of the AMS measurements. The LSC result sits between the AMS and SCAR results.

5 Discussion

5.1 Accuracy and precision

Precision in ^{14}C analysis refers to the ability to reproduce measurement results of a sample when the conditions for it has not changed. In contrast to the accuracy, which refers to how close the result is from the actual value, precision is expressed as a standard deviation, σ, and evaluates how consistent and stable the measurement system is. When evaluating methods of analysis for ^{14}C, precision is used to determine which method can separate an actual signal from noise the best; high precision is needed to distinguish subtle enrichments, and fluctuations over time, in samples.

5.1.1 AMS

AMS is generally considered to be the most precise ^{14}C-measurement technique available. The high precision comes from the technical approach of direct atom-counting. Modern AMS systems can achieve uncertainties on the order of a few permille for modern samples. The precision is influenced by, for example, ion source stability, beam transmission and sample preparation. The high precision of AMS is one of the main reasons the technique has become the standard method for radiocarbon dating.

The SSAMS is capable of performing comparatively high-precision measurements. NEC states that the achievable precision is 0.3%. It was mentioned in [27] that, after optimization, the SSAMS of the Lithuanian lab achieved a precision of 0.35%.

From the measurement results presented in tables A1 and A2 in Appendix A the SSAMS measurements carried out at the laboratory in Lund have uncertainties of 0.005-0.010 $F^{14}C$ for samples with activities ranging from 1.048-2.812 $F^{14}C$. These uncertainties correspond to a precision of roughly 0.4-0.5%.

The MICADAS is a more recent and optimized alternative in AMS technology compared to the SSAMS. It is known for being stable and having improved striping efficiency. While the product page states that the precision is better than 0.2% [40], one report shows precisions below 0.1% [43].

In total, nine measurements were performed with the MICADAS at Atomki in Debrecen for this report. The standard deviations varied between 0.002-0.003 $F^{14}C$ for activities ranging between 1.062-1.223 giving a relative uncertainty of roughly 0.15-0.3%. The precision seems, by the results, to be indifferent of graphitization method.

Samples FS_Bua, FV_Lys and PP_Kat were analyzed using the MICADAS and the results can be seen in tables A1 and A2 in Appendix A as well as figures 18, 19b and 20. While the MICADAS results varu greatly for FS_Bua, the MICADAS results only show small differences, within the uncertainties for samples FV_Lys and PP_Kat. The variations in FS_Bua are likely due to inhomogeneity. This is a strong implication that the MICADAS is precise for more homogeneous samples such as PP_Kat and FV_Lys. No conclusion that there is a difference between the sealed

tube method of graphitization and the automated AGE can be drawn.

The LEA focuses on maximizing efficiency at reduced voltages. The precision is reported to be on par with the predecessor MICADAS, less than 0.2%, while at the same time being smaller and less energy consuming.

In total, eight measurements were performed with the LEA at Atomki in Debrecen for this report, since the planned ninth one failed. The standard deviations varied between 0.002-0.003 $F^{14}C$ for activities ranging between 1.054-1.242 with a relative uncertainty of roughly 0.15-0.25%. The precision seems, by the results, to be indifferent of graphitization method.

Samples FS_Bua, FV_Lys and PP_Kat were analyzed using the LEA and the results can be seen in tables A1 and A2 in Appendix A as well as figures 18, 19b and 20. In figure 18 the LEA results differ greatly, unlike in figures 19b and 20. This is in analogy with the MICADAS measurements, making it certain that the sample is inhomogeneous. For the other figures, 19b and 20, the LEA results only show small differences, within the uncertainties. To conclude, LEA appears to give accurate and precise measurement results, regardless of the graphitization method.

Based on both literature and the measurements carried out, the AMS systems appear to provide highly reproducible results with small uncertainties.

5.1.2 LSC

LSC generally results in lower precision than AMS for ^{14}C measurements. Since the method relies on detecting radioactive decay events, the precision is fundamentally limited by counting statistics. A way to improve precision is to use longer counting times. Factors such as quenching, background radiation and sample preparation can also affect the precision negatively. For some purposes, standard-LSC can still achieve sufficiently good precision. The method is especially well suited for samples with higher activities where counting statistics are less limiting. As previously mentioned, three hour measurements using the Quantulus 1220 showed precisions of 1.7-3.2%. Inhomogeneity in a sample can be less of a problem for LSC, due to the large sample sizes.

Three measurements were carried out with LSC at CARER in Mangalore for this report. The results had uncertainties of 0.010-0.015 $F^{14}C$ for activities ranging between 1.066-1.190 $F^{14}C$. The relative uncertainties are roughly between 0.8-1.4%.

5.1.3 SCAR

SCAR is an emerging technique whose precision today can match the precision of some AMS systems. The first generation of C14-SCAR-devices produced by ppqSense, like the one available at RISE in Borås, provides worse precision, 0.015-0.02 $F^{14}C$ for the samples analyzed for this thesis. For the second generation of the device, the precision was improved, and all results that were performed with the second generation had a reported uncertainty of 0.005 $F^{14}C$. For the first generation measurements, the

relative precision was between 1.5-2% at RISE and 0.5% at ppqSense. For the second generation the relative precision was roughly 0.45%.

The reported results from ppqSense and RISE do not correct for isotope fractionation, as previously explained. Isotope fractionation occurs naturally in chemical or physical processes when the carbon isotopes engage differently due to their mass differences. This essentially means that the ratio between ^{14}C and ^{12}C in a sample is not the same ratio that is present in the surrounding atmosphere or water at the time. Since AMS is well used for radiocarbon dating purposes, where the sample activity is linked to the atmospheric ^{14}C content, all samples need to be corrected for isotope fractionation to be able to match the sample to its age correctly. Apart from this reason, AMS also introduces an isotope fractionation between the different carbon isotopes. Since AMS also measures ^{13}C and ^{12}C apart from ^{14}C, it becomes very evident exactly how large the introduced isotope fractionation is, and it can be effectively corrected. When comparing the results from a SCAR measurement to an AMS measurement, the former also needs to be corrected. In the SCAR-device there is no available method of measuring ^{13}C-content to customers, but the staff at the company are able to conduct the measurements by shifting the laser to a transition frequency specific to ^{13}C. This was done to the samples measured during the study visit: FV_Sär, ME_Båt_1 and ME_Båt_2, after which they were corrected accordingly, and their corrected values being found in table A2. For the previous samples measured at ppqSense and at RISE the correct isotope fractionation coefficient was not measured. Instead, the samples were corrected using either values found from literature, or from measurements conducted with IRMS and communicated by Kristina Stenström. This correction is of course not entirely correct. The samples affected by this correction are ME_Out_1, FS_Bua, FV_Lys and PP_Kat.

For three samples, SCAR-measurements were performed using both the device at RISE and the devices at ppqSense, the samples were FS_Bua, FV_Lys and PP_Kat. For the two former samples, the differences in the results are small, at least compared to the broad uncertainty of the RISE measurements, with the ppqSense results being within the uncertainty bars for the RISE results. For PP_Kat, a sample that should be quite homogeneous when looking at the AMS-results, the difference is larger. However, it is not a statistically significant difference.

5.1.4 Intercomparison

Overall, the results strongly indicate that AMS provides the highest precision and generally also the highest accuracy of the different techniques. From the AMS systems, MICADAS and LEA showed the best precision, with relative uncertainties typically below 0.3%, while SSAMS also showed consistently high precision around 0.4–0.5% for standard procedures. SCAR showed good performance as well, particularly for the newer ppqSense generation, where the reported uncertainties approached those of SSAMS and the agreement with AMS results was often good for homogeneous samples. LSC, in contrast, showed larger uncertainties for the standard procedure at CARER, but the results did not deviate much from other results. The method may benefit from averaging when analyzing inhomogeneous samples due to its larger sample sizes. For environmental monitoring close to nuclear power plants, where relatively small

enrichments and variations could be of interest, AMS is the most reliable technique, with SCAR not far behind. LSC may primarily be suitable when lower analytical precision is acceptable, or if the ability to detect large releases or enrichments above background levels is the main purpose, unless larger sample sizes or longer counting times are possible.

5.2 Detection limit and range

When comparing different analytical methods for ^{14}C detection, the detection limit is close at hand as a scale of comparison of their performance.

AMS is regarded to be the benchmark when it comes to very low-activity samples, which explains why it is the method of choice for archeological purposes. What makes the method unique in its ability to detect very low activities is its unrelatedness to the decay of the ^{14}C-isotopes; instead the individual atoms are counted. This has proved to be a successful approach, leading to activities as low as 0.001 $F^{14}C$ being detectable. The SSAMS system is able to determine the age of samples as old as up to 50.000 years, while the MICADAS and LEA can go even further than that. Things that could affect the sensitivity of the device is the sample preparation, contamination and memory effects within the system.

The LSC on the other hand relies on a technique where the decay of the isotope is measured, rather than the individual atoms. This is inherently a limitation of the sensitivity, as ^{14}C decays with a very low rate, and during measurement of low activity samples any background radiation or quenching effect can disturb measurements. While there are LSC devices that are especially well suited for low activity samples, such as the Quantulus 1220, that could be on par with — or even better than — AMS, these low activities require much larger sample sizes compared to AMS and SCAR for the decays to increase in quantity, and much longer measurement times, for the decays to have time to occur and be measured.

SCAR represents a more recent approach, detecting $^{14}CO_2$- molecules via isotope-specific optical transitions. Its detection limit is around 0.01 $F^{14}C$.

Despite these differences, the relevance of detection limit must be evaluated in relation to the desired application. In environmental monitoring near nuclear power plants, the aim is not to detect the absolute lowest possible concentrations of ^{14}C, but to identify deviations from natural background levels, which are on the order of 1 $F^{14}C$. In this context, it is more important with, for example, high precision and reproducibility rather than extremely low detection limits.

Consequently, while AMS is still the recommended technique for applications requiring ultra-low detection limits, such as dating archeological samples, its unique selling point is not necessary for routine environmental control. Instead, techniques such as SCAR and LSC, even LSC devices that are not specialized for ultra-low concentrations of ^{14}C, provide sufficiently low detection limits. Since the aim of the environmental monitoring of the vicinity of the NPP is to detect an excess, not a lack, of ^{14}C, the

ability of the technique to handle enriched samples, and the dynamic range, is more of interest for this purpose. The chosen method needs to be able to measure activities around background levels, but also potentially high activity samples.

AMS performs well for enriched or modern ^{14}C-samples, although the limiting factors differ from those at low activity. The constraints could be source stability, beam current, and accurate correction of isotopic ratios. The MICADAS, for example, can have total ^{14}C-counts reaching up to 10^6. Many AMS-laboratories handle samples of both low and high activity, but to accommodate for both types, separate handling to avoid cross-contamination should be implemented. Enriched samples may also require to be handled differently, for example to be diluted, or the time of analysis of the sample being shortened in AMS to avoid excessive count rates or detector limitations, making high-activity measurements somewhat less convenient than low-level ones.

LSC is inherently well suited for enriched samples because it directly measures β-decay events, meaning higher activity leads to improved counting statistics and it could also mean smaller sample sizes and shorter analysis times. For higher activities, the limiting factors could be count-rate effects such as dead time or pulse pile-up. Corrections for these effects, together with optimized sample preparation such as changing the ratio between sample and cocktail, are essential to maintain good results. In conclusion, LSC is robust and practical for medium to high activity samples as well.

SCAR performs particularly well for enriched samples. Because it measures $^{14}CO_2$-molecules optically instead of counting decay events or ions, increasing activity directly enhances the signal strength, making measurements results rapid without the need for dilution. SCAR-devices have been shown to be able to handle activities up to 30 $F^{14}C$. Just like for the other techniques, accurate results depend heavily on clean sample preparation without cross contamination.

In conclusion, the ability of AMS to detect very low activities is not necessary for this context. For the range of interest, SCAR or LSC, that perform even better for enriched samples than for low activity ones, are better suited.

5.3 Economics

5.3.1 Investment cost

The investment cost is a key factor when comparing the different techniques. The investment must correspond to the benefit, and too high investment costs are difficult to justify or defend for the investors.

AMS represents the, by far, highest investment cost out of the three techniques. The cost reflects the advanced technology requiring a powerful accelerator, ion source, vacuum system and detectors. Apart from the instrument itself, AMS requires supporting infrastructure: power supply, cooling systems, gas handling, and laboratory space in which sample preparation can be performed. The sample preparation requires additional investment, especially for graphitization; either using an own graphitization line or by investing in an automated device, such as an AGE, which costs around 2.4

million SEK. Investment costs for the AMS-instruments are typically tens of millions of SEK. The SSAMS in Lund was bought for a cost of 14.3 million SEK, when adjusted for inflation. The same device, if bought today, is on the market for just under 20 million SEK. The newer and more compact alternatives, MICADAS and LEA, are sold for approximately 22 million SEK and 20 million SEK respectively. These costs include training, packing, shipment and installation. Overall, AMS is a technique that requires a very large investment cost, which is justified mainly for specialized, and often shared, laboratories.

LSC systems have significantly lower investment costs compared to AMS. The Quantulus 1220 is roughly worth 2.5 million SEK. In addition to the device, a suitable lab also needs to be established and a pyrolyser to combust the samples is necessary which increases the investment cost. Compared to AMS, LSC is still a low-cost investment, making it accessible to a wider range of laboratories.

SCAR has a middle position between AMS and LSC in terms of investment cost. While RISE invested roughly 7 million SEK for the first commercially C14-SCAR-instrument in 2022, not adjusted for inflation, the price has reduced and a device can currently be bought for 6 million SEK, which includes the EA. Overall, SCAR represents a middle ground investment cost, positioned between LSC and AMS.

In the context of environmental monitoring near nuclear power plants, the choice of technique should balance cost with the analytical need. The most expensive alternatives, the AMS-systems, may be difficult to justify, especially when there already are laboratories that provide AMS services in Sweden that could be used.

5.3.2 Cost of analysis

The cost per analysis is another important parameter when evaluating the economics of the different analytical techniques for ^{14}C monitoring. While the investment cost determines if a technique is reasonable to invest in, the cost of each individual measurement determines the long-term economic stability of the method. The total analysis cost includes factors such as consumables and electricity, but also labor, maintenance, amortization, and depreciation of the instrument over its lifetime. For routine environmental monitoring programs, repeated measurements over long periods are carried out, making low operational costs desirable.

The analysis cost for AMS is driven by, for example, the sample preparation that requires working hours; consumables; infrastructure, such as vaccuum pumps, temperature regulators and more; margins for maintenance; and amortization and depreciation of the accelerator system itself. If graphitization is performed, it requires chemicals, gasses, catalysts and reaction vessels. If graphitization is carried out using own methods such as the sealed tube method, the investment cost is lower but the cost of labor increases since it uses more personnel hours. Automated graphitization systems reduce manual labor but contribute more to instrument amortization and depreciation costs which are reflected on the cost for analysis. During operation, AMS systems consume electricity, cooling water, vacuum pump oil, and stripper gases. Maintenance of ion sources, detectors, and vacuum systems contributes to the operational costs and are

also reflected in the cost of analysis for the customer. The price for a regular customer to analyze a standard sample without any extras is just under 4,000 SEK at both the SSAMS-laboratory in Lund as well as at Atomki in Debrecen. The laboratory in Lund is not for-profit, which should be reflected in the price. On the other hand, the age of the SSAMS should lead to higher maintenance costs. Atomki, together with the company Isotoptech Zrt., are for-profit, meaning that a margin for profit is included in the price. Despite the interest for profit, and that the machines are newer and probably still being amortized, the cost of analysis is the same. The prices being equal could be contributed partly to the fact that wages are lower in Hungary than in Sweden. Another explanation could be that the much higher throughput at Atomki - more than five times the throughput of the laboratory in Lund - distributes the fixed costs, such as the infrastructure, making the cost per sample decrease.

LSC usually has lower analysis costs compared to AMS, but the exact cost depends on sample preparation and counting times. Compared to the other techniques, LSC relies on more consumables. These include the scintillation cocktail, vials and chemicals for benzene synthesis or CO_2 absorption. Scintillation cocktails can cost a few thousand SEK per liter, but one liter can last for 50-100 samples. Electricity consumption and instrument maintenance are relatively low compared to AMS. However, the long counting times limit the throughput and reduce the instrument availability, indirectly increasing the cost per sample. The labor costs for the sample preparation can become significant. The prize for sample analysis at CARER in India, where cost of labour is significantly lower than in Sweden, is 2,400 SEK. Labour is roughly 10 times more expensive in Sweden compared to India, according to self-reported Indian engineering salaries on Indeed [67] [68].

SCAR operates directly on CO_2 gas, avoiding graphitization and reducing not only the need for a graphitization line or an AGE, but also the need for labor, consumables and infrastructure associated with AMS graphitization. Operational costs are mainly carrier gasses, consumables for combustion, electricity, maintenance and instrument amortization and depreciation. Labor requirements are generally lower than for AMS and LSC due to a more automated workflow and shorter measurement times. Compared to LSC, SCAR avoids scintillation cocktails and the chemicals used during sample preparation, reducing consumable costs. Because measurements are rapid, instrument utilization can be high, which can reduce the cost per sample. The price for having one sample analyzed by SCAR at RISE in Borås is just under 6,000 SEK while a sample being analyzed at ppqSense costs just under 4,000 SEK. Both RISE and ppqSense have interest for profit. ppqSense is not primarily a laboratory that analyzes samples for customers; this is just a sideline. RISE do not operate even close to their full capacity which likely is the reason for the price of analysis being higher. They are also an accredited laboratory, which likely increases the prices of their services.

By comparing the costs for having analysis done at the different facilities, LSC appears to be the cheapest while SCAR and AMS are fairly equal. It is, however, difficult to draw any conclusions since the cost of labour, which is a significant part of the total analysis costs, varies immensely between the different countries. Another important factor is that some facilities have interest for profit while others do not.

5.4 Throughput

Another way of comparing the different techniques is by comparing the possible throughput of a laboratory using each of the different methods.

The throughput of AMS is primarily limited by sample preparation rather than measurement time, with graphitization being the main bottleneck. Even in a well-optimized AMS laboratory, graphitization using, for example, one AGE, will constrain the number of samples that can be graphitized to 14 per day. The number of graphitized samples per day using one AGE cannot be matched with the number of samples that can be analyzed by an AMS, creating a bottleneck. Solutions that could increase the throughput would be multiple graphitization devices, or alternatives to the AGE that could handle more than seven samples at a time. Many laboratories use own methods of graphitization, such as the described sealed tube method used at Atomki in Debrecen. Such methods, although more labor-intensive, could allow for up to 30 samples being graphitized per person per day. At Atomki, where the MICADAS and the LEA almost are fully booked, the annual throughput can be up to 5,000 samples. This is largely due to them having both an AGE and using their own graphitization method. In addition to this, their LEA does not use graphite targets but is directly coupled with an EA and analyzes CO_2, meaning that the time consuming graphitization process can be skipped for analysis using the LEA. The laboratory in Lund uses one AGE to graphitize. The graphitization bottleneck leads to the SSAMS not being able to be used every day. Instead, it is running roughly once per week, making the total throughput about 800-900 samples annually. Overall, AMS throughput is best described as moderate, with the bottleneck clearly located in preparation rather than in the accelerator itself. Solutions to this bottleneck, such as investing in graphitization devices with higher capacity, is expensive, while own graphitization methods are labor-intensive. Another alternative is to invest in an AMS that analyses CO_2 instead of graphite, which could be done with, for example, LEA.

Throughput in LSC is decided by counting time and instrument capacity. Many instruments, including the Quantulus 1220, are equipped with sample changers that can handle many vials that are measured sequently and switched automatically. However, samples require counting times that could be several hours, depending on the desired precision and activity level. For environmental ^{14}C-measurements at low activity, long counting times limit the throughput significantly, while enriched samples can be measured more quickly. Sample preparation and leaving the samples in the dark for their flourescence to decay also limit the throughput. In practice, LSC throughput is low to moderate, with the main limitation being the time required to accumulate enough decay events for reliable counting. The laboratory at CARER in Mangalore operates at nearly full capacity, yet still only manage to analyze about as many samples as the SSAMS laboratory in Lund, in which the SSAMS only runs roughly once a week.

SCAR has the potential to offer a higher throughput due to both the sample preparation time and measurement time being comparatively quicker than the other techniques. Since the method operates directly on CO_2 gas and does not require graphitization, preparation is relatively fast and automated after entering the EA. While one sample is analyzed, another combusted sample can be stored within the SCAR-device in the cryogenic trap, ready to enter the cavity after the previous sample analysis

is finished. At the same time, other samples can be combusted in the EA, which is directly coupled to the SCAR. Measurement times themselves are short; the second generation of the device performs one scan in 15 minutes, which means that results with low uncertainty can be given in less than one hour. Since the steps in the process are roughly equally time consuming, there is no real bottleneck in the process. Neither the SCAR laboratory at RISE in Borås, nor at the company ppqSense in Florence, operate at full capacity, and they both manage to analyze roughly 100-200 samples per year. A staffed laboratory with a fully booked SCAR operating working days with an average of three scans per sample should be able to analyze 2,000 samples per year, by a simple calculation. In other words, SCAR could achieve a high throughput.

The three methods differ in what limits their throughput. AMS is constrained mainly by graphitization, if used, in the sample preparation, even though the actual measurement is relatively fast. LSC is limited by counting statistics, requiring long measuring times per sample, particularly at low activity levels. SCAR, in contrast, benefits from both faster measurements and simpler preparation, making it the most efficient in terms of samples processed per unit time. To conclude, SCAR has the potential to achieve the highest throughput. AMS can achieve equal or even higher throughput if sample preparation and graphitization can keep up with the rate of analysis with the AMS, which could be expensive in terms of both money and time. AMS can therefore be considered to fall in the intermediate position. LSC generally has the lowest throughput potential.

For environmental monitoring near NPPs, the required throughput depends on the monitoring strategy rather than on capacity. A monitoring program would, most likely, focus on tracking the ^{14}C across different organisms and map the spread in both space and time to see variations with weather, seasons, ocean currents and NPP activity. Focus would not necessarily be on achieving the highest possible throughput, but rather for the technique to be available to analyze samples with constant time intervals. Instead, reliability, reproducibility, and the ability to detect small deviations from background levels are more critical. It is still possible that a higher throughput is required to be able to track many different organisms and over a larger space. In case of an incident the throughput could potentially need to be higher as well. A measurement system is also a long-term investment, and future demands can be difficult to evaluate. For this reason, it is not reasonable to aim for a device that, already at the beginning, will operate at its maximum throughput capacity. A device with a higher throughput potential offers flexibility and could enable monitoring of other NPPs as well. With the currently planned expansion of the Swedish nuclear energy industry, the need for using the measurement system could increase, and it is therefore advisable to opt for a system with a margin in throughput. In this context, LSC could be suitable if longer counting times, and therefore a delay in results, are acceptable. However, LSC seems to be the least flexible option in terms of scalability. AMS provides sufficient throughput, especially if using an AMS that handles CO_2 directly. Otherwise, the bottleneck, graphitization, could be scaled up using own methods or investing in additional devices, with the trade off being money and time of labor. SCAR offers clear advantages with rapid turnaround and higher sample volumes considering the low labor needed.

5.5 Staffing expertise

The level of staffing expertise required for a technique is a consideration of big practical importance. The required competence affects the reliability, stability, training needs, staffing costs, and the ability to implement a technique outside specialized research laboratories. Different expertise can be required for operation of the instrument than the maintenance of it.

AMS requires a relatively high level of staffing expertise. Operation and maintenance of an AMS requires knowledge of accelerator systems, vacuum technology, ion beams, high-voltage systems, detectors, gas handling, and isotope analysis. The many different technologies featured in an AMS-system demands personnel with technical or scientific education and experience. In practice, the staff that operates an AMS can be made up of engineers, technicians, and researchers, while optimization, troubleshooting, and method development are performed by staff with more expertise and experience, often with a PhD in physics, chemistry, or similar fields. The sample preparation and graphitization require laboratory experience, particularly when analyzing enriched samples where contamination, waste management and radiological safety needs to be put in practice. Newer systems such as MICADAS and LEA are more automated and user-friendly than older accelerators, for example by not requiring manual tuning. LEA can also analyze gas samples instead of graphite targets, making sample preparation easier. The newer devices still require much expertise for stable operation in the long term and maintenance. To conclude, AMS is generally best suited in dedicated laboratories with specialized staff rather than for industrial environments.

Routine usage of LSC appears to require less specialized expertise than AMS once methods have been established. LSC is a very mature technology that is used by chemists, physicists, environmental scientists alike. NPPs usually have experience with using LSC as well. The standard procedure of using the LSC should be able to be performed by people with technical training, knowledge about chemistry and radiation safety, but, for example, quench correction and background minimization might have to be performed by more experienced staff. At the same time, troubleshooting can be a difficult and tedious process. In addition, the sample preparation chemistry can be complex, especially if benzene synthesis is performed, raising the standards for the staff. The chemicals that are used and products of the process can be hazardous; even the scintillation cocktails that are used can be forbidden to be used for any other purpose than science.

Operation of SCAR is the most automated out of the three techniques which simplifies the daily use. However, the technique is based on advanced laser spectroscopy, gas handling, and spectral fitting, meaning that the staff still require understanding of optical systems. Routine measurements could be performed by personnel with technical background after dedicated instruction, while troubleshooting and maintenance could use more specialized expertise. SCAR is used in both specialized laboratories and industry, indicating its user-friendliness.

Routine measurements for all techniques require staff with technical or scientific backgrounds, with experience working in laboratory environments, with radiation safety and with the different fields of technology featured in the three different techniques.

The most user friendly of the three is the SCAR, which benefits from short and simple sample preparation, automated sample handling and easy-to-use software. On the other hand, both AMS and LSC are much more mature techniques with more users. This makes them robust, as it is known that their lifetimes span across decades, but it also makes sharing of knowledge and help internationally easier. Out of the AMS-systems, LEA appears to be the most user-friendly.

In the context of environmental monitoring near NPPs, staffing requirements are important to consider. NPPs already have staff with expertise in radiation protection, radiochemistry, environmental monitoring. To lower the bar for implementation of ^{14}C environmental monitoring, it would be convenient if the technique could fit into the existing competence profile of the NPP. Considering that the NPPs usually have experience with working with LSC, this would be a practical choice. SCAR could also be a reasonable alternative after short training considering its high degree of automatization and simple sample preparation. AMS, however, would likely require the recruitment of specialized personnel, as the knowledge of running an AMS most likely is not present already at the NPPs. This could act as a barrier to implementation for the ^{14}C environmental monitoring if implemented on site.

5.6 Size and weight

The physical size and weight of the different systems can be of interest when evaluating installation requirements or the infrastructure demands. Large and heavy instruments can require dedicated laboratory space, reinforced floors and specialized infrastructure such as cooling, shielding and high-voltage installations. Smaller systems are generally easier to install, and may fit into existing laboratory space.

AMS systems are by far the largest of the investigated techniques. Traditional tandem AMS systems can occupy very large spaces and weigh many tons, which could require renovated spaces or reinforced floors. The SSAMS used in Lund uses about 30 m^2 of floor area. Modern compact systems such as MICADAS and LEA are significantly smaller than older AMS facilities. MICADAS uses about 8 m^2 of floor space while LEA requires almost half of that. They weigh 4,500 kg and 3,000 kg respectively. In addition to the accelerator itself, separate spaces are needed for sample preparation and graphitization equipment. Overall, AMS systems are stationary laboratory installations that are most realistic to be operated at specialized facilities.

LSC systems are much smaller and lighter than AMS systems. Instruments such as the Quantulus 1220 have a footprint of approximately 1 m^2 and can generally be installed in normal laboratory environments without infrastructure modifications. Despite its small footprint, the lead shielding makes the device heavy, with weights being able to exceed 1,000 kg. Additional laboratory space is needed for chemical sample preparation, especially if benzene synthesis is performed. Compared to AMS, LSC instruments are considerably easier to accommodate and install.

SCAR is much smaller and lighter than an AMS system and can fit within a standard laboratory environment and does not require the extensive infrastructure. The system

has a slightly larger footprint than an LSC instrument, 1-2 m^2, but does not feature parts of such substantial weight as LSC; the first generation SCAR weighed 600 kg and the device has since then been reduced. Sample preparation needs space, but less than for LSC and AMS since it features less steps. To conclude, SCAR systems are relatively compact and could realistically be installed within existing monitoring laboratories.

5.7 Sample size

Sample size is an important factor in ^{14}C analysis because it affects both how practical a method is and the analytical performance. If a method only requires small sample sizes it can be advantageous when limited material is available, or when as little as possible should be destroyed of the sample, such as for radiocarbon dating of archeological samples. Small sample sizes can also be used to pinpoint the activity for specific small parts of a sample, such as an organ in a fish or a tree ring. However, very small samples can also increase the influence of contamination and inhomogeneity, since tiny parts of a material may not fully represent the sample as a whole. Larger sample sizes could improve how representative the results are for the sample as a whole and improve counting statistics, but require more extensive preparation and can limit the types of samples that can be analyzed.

When discussing the sample size needed, the sizes are often provided as a weight unit of pure carbon. A dried organic sample can be assumed to consist of roughly 40-50% carbon. For example, if 1 g of carbon is needed, a sample size of 2 g should be provided of the dried sample. The mass of an organic sample is often made up of water to more than half of the total mass, meaning that, for a dried sample size of 2 g, 5 g or more of sample needs to be collected.

AMS systems generally require very small sample sizes compared to other ^{14}C techniques. Standard procedures for graphite targets require 1 mg of carbon or slightly less than that, while specialized labs could reduce this even further, to tens or hundreds of μg. For modern systems, like MICADAS and LEA, it is possible to do analysis on gas targets which only requires sample sizes of tens of μg. This is possible because AMS directly counts individual ^{14}C atoms rather than radioactive decays. The small sample size requirement is one of the major advantages of AMS and explains its usage for archeological samples for which the sample is supposed to be preserved. However, the small sample masses also make AMS more sensitive to contamination and sample inhomogeneity, as was clearly shown in the case of, for example, the sample FS_Bua shown in figure 18. It is difficult to do representative sampling and homogenize the sample very well before measurement when the sample size required is as small as in the case for AMS.

LSC generally requires much larger sample sizes than AMS. Since the method relies on detecting radioactive decay events, enough carbon mass is needed to produce well counting statistics. Depending on the preparation method, the required carbon amount is typically on the order of a few grams instead of milligrams or micrograms. The size also varies depending on the activity of the sample; an enriched sample may

give enough counts even for a smaller sample size than a less active sample. Benzene synthesis in particular uses larger sample sizes than direct absorption methods. The larger sample size can be advantageous for heterogeneous environmental samples because the analyzed material becomes more representative of the sample as a whole. The samples that were sent for analysis at CARER contained roughly 15-25 g of dried samples.

SCAR, just like AMS, requires small sample sizes. Because the method measures $^{14}CO_2$ directly using laser spectroscopy, it does not require the large carbon masses typical for LSC, but it generally requires somewhat larger samples than AMS. Typical SCAR measurements use carbon masses on the order of one or a few milligrams. The second generation of the ppqSense C14-SCAR can measure samples with 0.6 mg of carbon.

Overall, AMS requires the smallest sample sizes, followed by SCAR, while LSC requires by far the largest amounts of material. The small sample size of AMS is highly regarded for specialized applications where sample material is limited, but it also increases sensitivity to contamination and inhomogeneity. AMS or SCAR, could, however, be used to study the fine ^{14}C distribution within a sample, which cannot be done with LSC. LSC benefits from larger and potentially more representative sample masses, something that is indicated by the results for sample FS_Bua in figure 18, but not confirmed for the other samples, such as FV_Lys in figure 19b.

For routine environmental monitoring near NPPs, determining a "reasonable" sample size is difficult. One one hand, the incredibly small sample sizes required by AMS and somewhat SCAR may be advantegous because it minimizes the effects on the local ecosystem, by not requiring the harvest of large amounts of sample. Both AMS and SCAR could also be used to study the small variations of ^{14}C activity within a sample, and therefore track what the heterogeneity of the sample looks like, although this might be of more interest for academia than for a NPP. The larger sample requirements of LSC could naturally average out inhomogeneity of a sample. At the same time, to extract a few grams of pure carbon requires the harvesting of large fresh samples, which, in the long term, for routine monitoring, can turn out to be destructive and ecologically unsustainable.

5.8 Degree of automation

Automation is an important factor when evaluating the different analytical techniques, since it very strongly affects other points of evaluation, such as the throughput, the turnaround time, staffing requirements, precision and the cost of analysis. A high degree of automation can reduce manual labor, minimize human errors and make the workflow more standardized, thus improving the precision. On the other hand, it could also be more expensive to invest in an automated device, and it could alter the flexibility of the workflow.

Historically, the sample preparation for AMS has involved many manual steps such as chemical purification, combustion, and graphitization, making the procedure quite

labor-intensive. Some laboratories still perform the sample preparation without only using automated devices, such as Atomki in Debrecen. While the manual sample preparation occupies more time from the workers, it is also reported to add flexibility to the facility due to the ability to scale up or down the sample preparation, making it less of bottleneck. Modern laboratories increasingly turn to systems like EA and AGE for automated combustion and graphitization. After the sample preparation, modern AMS systems have a very high degree of measurement automation. Once sample targets are loaded into the instrument, and potentially after some manual ion source tuning, measurements are taken care of by the software, which automatically for example optimizes the beam, switches between samples and compile the data. AMS measurements usually even run overnight. Modern AMS systems can also handle gas samples, making it possible to couple an EA directly to the device, decreasing manual handling and making the process more automated.

For LSC measurement is highly automated but sample preparation is still heavily manual. Modern devices such as the Quantulus 1220 instrument have mechanical sample changers that can hold many samples, often a few dozens. The instrument can autonomously do sequential counting cycles, identify the samples, and perform long measurements without user intervention. However, the sample preparation is still very manual. Operators must combust the sample, for example by using a partly automated pyrolyser, and then synthesize the carbon into a suitable liquid form such as benzene and mix it with a liquid scintillation cocktail in vials. The high degree of manual work makes the workflow more prone to human error, quenching, prolongs the time before counting can begin.

SCAR has a high degree of automation which is largely due to the sample preparation skipping steps, essentially only requiring sample combustion in an EA that is directly coupled to the SCAR device. During measurement the system automatically, for example, regulates cavity temperature, doses precise pressures of sample gas into the optical cavity, and dynamically scans over the frequencies for the transition of interest, requiring minimal spectroscopic expertise from the operator.

Out of the three techniques, it can be concluded that SCAR has the highest degree of automation. AMS has become more automated with the introductions of EA and AGE. Some laboratories still rely on manual sample preparation, and it can still provide a useful complement to the automated sample preparation, since it can reduce bottlenecks. For LSC, whose sample preparation still requires much manual handling, the lack of automation can become the bottleneck.

5.9 Turnaround time

Turnaround time is a practical parameter that can be useful to evaluate the different techniques. The turnaround time represents the time that passes from start to finish, meaning that it includes both sample preparation and measurement times. Every laboratory working with any of the three techniques have special work flows and conditions, making turnaround times even for the same device in different laboratories unique.

The turnaround time for a sample in AMS is largely made up by sample preparation rather than measurement itself. Graphitization, that could be performed with own methods or by automated devices such as the AGE, typically requires several hours and is carried out in batches. Some AMS-devices do not use graphite targets, and avoids the time for graphitization as they measure CO_2 instead. The actual measurement in the AMS-system is relatively fast, usually on the order of a few minutes per sample per run, and the total time depends on how many runs per sample is deemed necessary. For high-activity samples, shorter or fewer runs could be performed, while for age determination of archeological samples, measurements can carry on for longer. Since the AMS-devices have room for many samples, often 40 or more, there is usually a period of waiting for enough samples having been graphitized until the AMS can fill up and start, adding to the total time for one sample. The turnaround time for a single sample is in practice on the order of a few days but depends greatly on the workflow of the laboratory, the degree of automatization and often the waiting time. For example, the turnaround time for sample at the SSAMS laboratory in Lund is weeks to months, which largely is described to long queues, as the actual measurements only take 2.5-3 days. The results from Atomki in Debrecen had a turnaround time of a few weeks.

For LSC, turnaround time depends on both sample preparation and counting duration. Preparation usually takes several hours; for example, benzene synthesis can be estimated to take a full day. Samples are then left to cool in the dark for fluorescence to decay, at least overnight. The largest contributing factor is, however, the counting time itself: to achieve good precision at environmental ^{14}C-levels, samples are often measured for many hours. Background samples require as much as 50 hours to complete while other samples can be sufficiently measured overnight or in a working day. The counting time is decided by the activity and sample size. Many instruments have slots for multiple samples that can be switched automatically by the machine, but this does not reduce the elapsed time per sample. As a result, the turnaround time for a single LSC measurement is typically on the order of days or weeks, particularly for low-activity samples where longer counting times are required. The turnaround time for the samples sent to CARER in Mangalore was 4-5 weeks after the samples reached the institute.

SCAR offers significantly shorter turnaround times. This reduction is due to minimal sample preparation and short analysis times. The technique operates directly on CO_2 gas and preparation mainly involves combustion and gas purification, which is completed quickly and automatically in less than an hour. The measurement itself is fast, less than one hour, with most of the samples in this project being measured for 30 minutes. The short measuring times allows results to be obtained almost immediately after sample preparation. In practice, the total turnaround time for a SCAR measurement can be on the order of a few hours. Samples analyzed by RISE and ppqSense using the SCAR had their results reported back in a matter of a couple of days.

The three methods differ substantially in turnaround time. AMS usually requires more extensive preparation although measurements are fairly quick. LSC, is usually limited by both sample preparation and measurement times. SCAR, in contrast, benefits from both faster preparation and more rapid measurement, resulting in significantly shorter turnaround times. SCAR is clearly the fastest technique, while AMS and LSC are comparable but limited by different stages of the workflow. AMS that avoids graphite

targets could cut turnaround time significantly.

In the context of environmental monitoring near NPPs, turnaround time requirements depend on the scenario. For routine measurements, the turnaround time does not have to be the quickest possible. In the routine cases, the information of interest is the trend of ^{14}C variations in time, and a rapid result is not needed. In case of an incident, such as a suspected leak, faster turnaround times could be critical, if decision-making relies on it. In such cases, the SCAR has clear advantages over the other techniques, but for routine measurements, graphite AMS and LSC can be considered to have sufficient turnaround times.

5.10 Comparative table

Table 2: Comparative table of analytical techniques for environmental ^{14}C monitoring.

Parameter	**SSAMS**	**MICADAS & LEA**	**LSC**	**SCAR**
	Lund	Debrecen	Mangalore	Borås & Florence
Precision	High (∼0.3–0.5%)	Very high (<0.2%)	Low–Medium (∼0.8–1.4%)	Low–High (∼0.45–2%)
Detection limit and range	∼50,000 years, medium range	>50,000 years, high range	∼65,000 under good circumstances, high range	0.01–1,000 $F^{14}C$
Investment cost	Very High (∼20,000,000 SEK)	Very High (∼20,000,000–22,000,000 SEK)	Low–medium (∼2,500,000) without extras	Medium (∼7,000,000 SEK)
Cost of analysis	∼4,000 SEK	∼4,000 SEK	∼2,400 SEK	∼3,500–5,500 SEK, has potential of being lower
Throughput	800–900 per year	4,500–5,000 per year	600–800 per year at almost full capacity	100–200 per year, but they have a much higher capacity
Sample preparation	Medium	Low if using gas targets, medium for using automated graphitization, high if using own methods for graphitization	Medium–high	Low
Staffing expertise	High	Medium	Medium	Low
Size and weight	Very large and heavy, 35 m^2	Small–medium size (4.5–8 m^2). 3,000–4,500 kg	Small (1 m^2) and comparatively heavy (1,000 kg)	Small (1-2 m^2) and comparatively light (<600 kg)
Sample size	Low (∼1.0 mg C)	Low (1 μg–1 mg C)	High (1–5 g C)	Low (∼1.0 mg C)
Degree of automation	Low–medium	Low if own graphitization, medium for using automated graphitization, high if using gas targets	Low–medium	High
Turnaround time	Days	Hours (if using gas samples)–Days	Days	Hours

5.11 Is there a need for environmental control?

Before a conclusion can be drawn about whether environmental control with any of the described methods should be implemented, it first needs to be clear whether environmental control of ^{14}C is needed or desired.

The arguments against the environmental control of ^{14}C could be of many different natures. It could be argued that ^{14}C already is measured sufficiently. The overwhelming majority, 99.5% of all releases from NPPs, are airborne. From a purely statistical viewpoint, it can seem fruitless to focus on the waterborne releases.

Apart from the fact that the releases to water are so small in comparison to the air, the ^{14}C releases themselves are so low-energy that the risk to be affected by ionizing radiation is almost non existent when it is external, and the internalization rate of liquid releases of ^{14}C could be assumed to be limited.

From this thesis it is also evident that the investment cost for a facility to analyze the ^{14}C-content in a sample is high. It also requires time and working hours which would lower the incentive for the nuclear power industry to, on their own initiative, invest in such a facility. It is likely that such a decision would have to be made due to regulations imposed by the Swedish Radiation Safety Authority. Whether such a regulation is imposed or not, it can be argued that the cost does not reflect the benefit of the investment as it is difficult to evidently support the idea that the lack of monitoring of water-based ^{14}C-releases leads to losses for the public in terms of health or economics.

Given these considerations, it might appear straightforward to conclude that monitoring liquid ^{14}C releases has low priority, and that resources such as time and financial capital would be more effectively allocated on high-energy fission products or gamma emitters. However, this perspective ignores critical ecological and regulatory frameworks that make a compelling counter-argument.

Despite being a low energy emitter, ^{14}C is consistently the radioisotope delivering the largest radiation dose to the public, although the delivered dose is well below dose limits and restrictions. This demonstrates that the potential hazard associated with ^{14}C is not a function of its decay energy, but instead its abundance, the high mobility, and high potential for internalization. While the releases to water are far less than the releases to air, the quantity remains large. By not routinely monitoring the liquid releases of the very radioisotope responsible for the highest delivered dose, a logical inconsistency arises. This runs counter to the fundamental safety purpose described by the International Atomic Energy Agency (IAEA) in Safety Guide No. GSG-8, which dictates that people and the environment, both present and future, must be protected against radiation risks [69]. This inconsequence also has the potential of opening up a discussion about whether monitoring any radioisotope at all is necessary, since the others, while their decay energy is larger, do not deliver as large doses as ^{14}C.

Because carbon is so efficiently cycled through the environment, the local releases from NPPs can lead to "hotspots" of higher specific activity in nearby food products. This exposure pathway is particularly critical from a regulatory standpoint; as stated in

IAEA Safety Guide No. RS-G-1.8, persons consuming substantial amounts of locally produced food comprise the most exposed group of a population with regard to radiation exposure via the ingestion pathway [70]. Monitoring the liquid releases, and components of the marine ecosystem since they constitute the exposure pathways to humans, is therefore necessary to validate the dose assessments and confirm that they are not breached.

Apart from being a question of only radiation safety, this problem ultimately becomes a question of equity and institutional trust. The IAEA guidelines state that in scenarios where public radiation doses are relatively low, the justification process must go beyond traditional radiation protection to include, for example, societal and ethical aspects [69]. The public has a fundamental right to know whether local marine food products, such as fish or seaweed, show activities several times higher than regional background levels. The public also has to be given the chance to trust both the nuclear power industry, and the radiation safety authorities, and trust that the necessary regulations are put into place and followed. This trust would be much needed in the current polarized discussion about the nuclear future in Sweden, and could reward all parts; the public, the nuclear power industry and the authorities.

By monitoring the release of ^{14}C, nuclear facilities fulfill the duty they have of environmental consciousness, preventing the unnecessary accumulation of a isotope that, once released, can never be removed from the ecosystems until the decay of the isotope. This proactive management is necessary to minimize the long-term radiological footprint of nuclear energy on the global population and the wider ecosystem.

Fundamental to international radiation safety is a three-tiered system of protection: justification, optimization, and dose limitation.

Optimization is governed by the principle of ALARA (As Low As Reasonably Achievable). ALARA does not dictate that radiation exposures must be reduced to zero or as low as technically possible, but it mandates that dose reductions must be balanced against economic and social factors. Despite liquid ^{14}C releases delivering low absolute doses, ^{14}C still represent the largest contribution to the public dose. By omitting the measurement of the liquid ^{14}C releases, the NPPs limit their ability to transparently showcase that they comply with the optimization process.

The act of measuring liquid ^{14}C effluents does not enforce dose reductions, and its primary goal is not inherently to reduce releases that are already securely within safe thresholds. Instead, environmental monitoring of nearby marine organisms provides data that can be used to demonstrate and confirm compliance with the limits. Without tests under real environmental conditions, safety assessments rely only on theoretical transport modeling.

5.12 Conclusions

This thesis evaluated the technical and operational feasibility of using AMS, LSC, and SCAR for potential routine environmental monitoring of ^{14}C in the vicinity of nuclear power plants. Based on the experimental results and the evaluation of their

performance across several parameters, some definitive conclusions can be drawn.

First, relying only on measurements of airborne releases creates an error in the public dose assessments. Although liquid releases only amount to a very small fraction, approximately 0.5%, of total NPP ^{14}C releases, their local radiological impact, especially in marine environments, is profound. The highly effective accumulation observed in especially higher-trophic marine organisms shows that waterborne releases create distinct local peaks of ^{14}C specific activities. Implementing environmental monitoring of liquid releases is therefore justified to guarantee public safety and trust.

The comparison between AMS, LSC and SCAR shows that all three techniques are capable of measuring ^{14}C in environmental samples, but that they differ substantially in performance, practicality and suitability. AMS is the most established and highest-performing method overall. The technique provides excellent precision, very low detection limits, high accuracy, and strong reproducibility. Among the AMS systems studied, MICADAS and LEA demonstrated the best precision, while the SSAMS still showed highly reliable performance despite being an older system. The main disadvantages of AMS are the very high investment costs, infrastructure requirements, relatively complicated sample preparation and the need for skilled personnel. Throughput can be high if sample preparation is not a bottleneck. The sample preparation can become more flexible by having a manual method for graphitization that can be used when there is high demand. Sample preparation can be made easier and more automated by choosing an AMS that allows for gas targets as well, a feature in both MICADAS and LEA. For the purpose of environmental monitoring, AMS is likely too expensive, and the incredibly high precision and low detection limits, that justify the price, are not strictly necessary for this context. AMS remains best suited for specialized laboratories, not for industry. If AMS is to be used by NPPs, it should be contracted use at an already established AMS laboratory

LSC represents the most economical alternative. The instrumentation is comparatively inexpensive, and requires less advanced infrastructure than AMS. The method requires large sample sizes, which on one hand could reduce the impact of sample inhomogeneity, but on the other hand making it logistically impractical and unsustainable for long-term monitoring. LSC has a lower precision than both AMS and SCAR, mainly because the method relies on radioactive decay counting. Long counting times limit throughput and turnaround time. While LSC can be sufficient for basic routine monitoring of samples, its limitations become significant. The precision is likely too low for the purpose, and the turnaround times too long.

SCAR occupies a middle position between AMS and LSC in many categories. The technique combines relatively high sensitivity with rapid analysis, minimal sample preparation and a high degree of automation. SCAR showed significantly faster turnaround times and higher potential throughput than both AMS and LSC, while requiring less laboratory infrastructure and lower investment costs than AMS. The discussion also showed that modern SCAR systems can achieve precisions approaching those of some AMS systems. At the same time, SCAR is still less established than AMS, and it is difficult to predict the lifetime and maintenance needs in the long-term. Nevertheless, the results indicate that SCAR is highly promising for environmental monitoring applications, particularly when rapid response and operational

simplicity are important.

In summary, while AMS remains irreplaceable for certain applications, such as radiocarbon dating of archeological samples, and LSC is the cheapest alternative, second-generation SCAR devices present a quick, simple, highly automated, economically accessible and the most reasonable alternative for routine environmental monitoring and the measurement of ^{14}C releases from nuclear facilities.

6 Reference list

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7 Appendix A

This appendix features all of the measurement results from this thesis project. Table A1 feature the first four samples, in order from closest to furthest from the outlet of Ringhals NPP. Table A2 feature the other four samples. All of the different methods of analysis are included in the tables.

Table A1: Measurement results in $F^{14}C$ for four of the analyzed samples including uncertainties. w/o: without; w.: with; hom.: homogeneity; unc.: uncorrected; corr.: corrected.

Method	**ME_Out_1** $[\overline{F}^{14}C]$	σ	**ME_Out_2** $[\overline{F}^{14}C]$	σ	**FS_Bua** $[F^{\overline{14}}C]$	σ	**FV_Sär** $[F^{\overline{14}}C]$	σ
SSAMS w/o pretreatment	2.812	0.010	2.795	0.010	1.253	0.005	-	-
SSAMS w pretreatment	-	-	-	-	1.235	0.005	1.048	0.006
SSAMS hom. test 1	-	-	2.510	0.008	-	-	-	-
SSAMS hom. test 2	-	-	2.755	0.010	-	-	-	-
MICADAS sealed tube 1	-	-	-	-	1.128	0.003	-	-
MICADAS sealed tube 2	-	-	-	-	1.170	0.003	-	-
MICADAS AGE	-	-	-	-	1.223	0.003	-	-
LEA sealed tube 1	-	-	-	-	1.136	0.002	-	-
LEA sealed tube 2	-	-	-	-	1.242	0.002	-	-
LEA AGE	-	-	-	-	1.178	0.003	-	-
LSC	-	-	-	-	1.190	0.010	-	-
SCAR RISE unc.	-	-	-	-	1.21	0.02	-	-
SCAR RISE corr. w. AMS-$\delta^{13}C$	-	-	-	-	1.19	0.02	-	-
SCAR ppqSense unc.	3.012	0.015	-	-	1.196	0.005	1.048	0.005
SCAR ppqSense corr. w. other $\delta^{13}C$	2.994	0.015	-	-	1.172	0.005	-	-
SCAR ppqSense corr. w. own $\delta^{13}C$	-	-	-	-	-	-	1.038	0.005

Table A2: Measurement results in $F^{14}C$ for four of the analyzed samples including uncertainties. w/o: without; w.: with; hom.: homogeneity; unc.: uncorrected; corr.: corrected.

Method	**FV_Lys** $[F^{\overline{14}}C]$	σ	**ME_Båt_1** $[\overline{F}^{14}C]$	σ	**ME_Båt_2** $[\overline{F}^{14}C]$	σ	**PP_Kat** $[F^{\overline{14}}C]$	σ
SSAMS w/o pretreatment	1.052	0.005	1.056	0.005	1.059	0.005	-	-
SSAMS w pretreatment	1.068	0.006	-	-	-	-	-	-
SSAMS hom. test 1	-	-	-	-	-	-	1.106	0.005
SSAMS hom. test 2	-	-	-	-	-	-	1.107	0.005
MICADAS sealed tube 1	1.062	0.002	-	-	-	-	1.112	0.002
MICADAS sealed tube 2	1.065	0.003	-	-	-	-	1.109	0.002
MICADAS AGE	1.064	0.002	-	-	-	-	1.110	0.003
LEA sealed tube 1	1.058	0.002	-	-	-	-	1.104	0.002
LEA sealed tube 2	1.054	0.002	-	-	-	-	1.100	0.002
LEA AGE	-	-	-	-	-	-	1.101	0.002
LSC	1.066	0.015	-	-	-	-	1.095	0.015
SCAR RISE unc.	1.07	0.02	-	-	-	-	1.06	0.02
SCAR RISE corr. w. AMS-$\delta^{13}C$	1.05	0.02	-	-	-	-	1.05	0.02
SCAR ppqSense unc.	1.058	0.005	1.052	0.005	1.044	0.005	1.100	0.005
SCAR ppqSense corr. w. AMS-$\delta^{13}C$	1.037	0.005	-	-	-	-	1.087	0.005
SCAR ppqSense corr. w. own $\delta^{13}C$	-	-	1.049	0.005	1.042	0.005	-	-