Theses and Papers in
Scientific Archaeology 20

university of
groningen

Stockholm
University

# Dealing With Reservoir Effects in Human and Faunal Skeletal Remains

## Understanding the radiocarbon dating of aquatic samples

Jack Dury

Doctoral Thesis in Scientific Archaeology at Stockholm University, Sweden 2021

# Dealing With Reservoir Effects in Human and Faunal Skeletal Remains

## Understanding the radiocarbon dating of aquatic samples

**Jack Dury**

Academic dissertation for the Degree of Doctor of Philosophy in Scientific Archaeology at Stockholm University to be publicly defended on Thursday 18 March 2021 at 14.30 in Broerstraat 5, 9712 CP, Groningen, Netherlands, online via Zoom, public link is available at the department website.

### Abstract

Archaeology relies on the ordering of past events to study cultural developments. This has traditionally been achieved by looking at the stratigraphic depths of materials relative to one another. In this way, chronologies of past technological progressions and stylistic changes can be built. The introduction of radiocarbon dating in the 1950s revolutionised archaeology, allowing for direct, numerical estimates of a sample's age. This allowed for more detailed past chronologies than was previously possible. Radiocarbon dating utilises the radioactive decay of carbon-14 (radiocarbon, $^{14}$C) to estimate a sample's age with older samples having less $^{14}$C. Shortly after the introduction of radiocarbon dating, however, it was demonstrated that $^{14}$C is not evenly distributed globally. Typically, there is less $^{14}$C in marine (and sometimes freshwater) systems compared to the atmosphere. This results in aquatic samples appearing older than they are, a phenomenon known as a 'reservoir effect'. When radiocarbon dating material from archaeological sites with marine activity, this is an important consideration. With aquatic resources being vital for human populations across the globe and for millennia, the ability to interpret aquatic radiocarbon dates is incredibly important. Making use of radiocarbon dates without properly handling any reservoir effects have proved problematic, sometimes resulting in archaeologically incorrect chronologies being constructed. Reservoir effects can, however, be managed.

This thesis demonstrates how archaeologists should interpret radiocarbon dates from aquatic samples, avoiding erroneously-old age estimates. Through careful sample selection, considering complicated carbon source mixing, measuring the scale and variability of reservoir effects within a single ecosystem and using prior knowledge about a sample's age, the dating of aquatic material can be greatly improved. This thesis also details a novel method of dating teeth, reducing uncertainty, and concomitantly estimating the extent of the reservoir effect. This was achieved by dating dental increments, combined with complex modelling. It is clear that there is no single method of handling reservoir effects, and methods for dealing with reservoir effects will differ depending on the archaeological site and specific research question. In this thesis, novel and existing methods of dealing with reservoir effects are demonstrated by considering five case studies from four archaeological sites:

At the site of Hamanaka 2 (Rebun Island, Japan), it is demonstrated that by carefully selecting samples without reservoir effects, the dating of the stratigraphy of the site can be accurately modelled. Concerning the cemetery site of Rounala (northern Sweden), it is demonstrated that by carefully reconstructing complex human diets, the dating of humans can be modelled to a high resolution. This has implications for the understanding of the Church's relationship with the cemetery. At the site of Ekven (Chukotka, Bering Strait) reservoir effect variability between species is carefully described. A more detailed understanding of regional reservoir effects allows for more accurate dating of human remains from the marine hunting Old Bering Sea culture. More accurate dating of human remains allows for the refining of existing Old Bering Sea culture chronologies. Finally, concerning the material from Resmo (Öland, Sweden), a novel dental wiggle matching model is presented as a possible method for reducing dating uncertainty in individuals with a marine dietary component.

**Keywords:** *Radiocarbon Dating, Reservoir Effects, Bayesian, Modelling, Palaeodiet, Stable Isotopes, Skeletal Remains, Collagen.*

Stockholm 2021
http://urn.kb.se/resolve?urn=urn:nbn:se:su:diva-189708

ISBN 978-91-7911-432-9
ISBN 978-91-7911-433-6
ISSN 1400-7835

Stockholm University

**Department of Archaeology and Classical Studies**

Stockholm University, 106 91 Stockholm

# DEALING WITH RESERVOIR EFFECTS IN HUMAN AND FAUNAL SKELETAL REMAINS

Jack Dury

Stockholm
University

# Dealing With Reservoir Effects in Human and Faunal Skeletal Remains

## Understanding the radiocarbon dating of aquatic samples

Jack Dury

ISBN print 978-91-7911-432-9
ISBN PDF 978-91-7911-433-6
ISSN 1400-7835

The cover photograph shows Kvænangen fjord, Norway. Photograph taken by Jack Dury.

Printed in Sweden by Universitetsservice US-AB, Stockholm 2021

To my colleagues and
friends, who have been
so supportive

# I. Acknowledgements

None of my work would have resulted in a completed PhD were it not for the many people who inspired me, shared with me their knowledge and expertise, trusted me with precious archaeological material, and corrected my mistakes. I am grateful, in particular, to my supervisors at the University of Stockholm and the University of Groningen. Gunilla Eriksson, Kerstin Lidén, Peter Jordan and Hans van der Plicht. Their support has been immeasurable. I am grateful too, to have been part of such inspiring and vibrant academic departments in Stockholm and Groningen, I would like to extend my thanks to my colleagues in both departments who I have been so fortunate to work alongside. My transition to life in Sweden and the Netherlands was made easy thanks to their hospitality. I owe so much to my friends, Maiken Hemme Bro-Jørgensen, Anne-Marijn van Spelde and my office mate, Alison Harris, for their academic and moral support throughout the last four years. I could not wish to have arrived in Stockholm with more wonderful people. I am further grateful to Anne-Marijn van Spelde for finding the time to translate descriptions of my research into Dutch. As a visiting researcher, I was made very welcome at several academic institutions throughout my studies. I would like to thank the staff at the University of York's Bioarchaeology department and Hokkaido University's Ainu Studies Centre. In particular, I would like to thank colleagues at the Russian Institute of Archaeology, the Russian Academy of Sciences for their eager collaboration, readiness to offer me support and inspirational passion for our mutual research. For offering such valuable feedback during my final seminar, I would like to thank Jesper Ohlsen of Aarhus University, his constructive review of my early drafts aided me immensely. I would also like to thank the assessment committee for their kind feedback and helpful corrections. Finally, I would like to thank the wider ArchSci-2020 project organisers and European Union's EU Framework Programme for Research and Innovation Horizon 2020 (Marie Curie Actions Grant Agreement No. 676154) for funding this project. I feel fortunate to have been able to undertake my PhD, belonging to a cohort of such brilliant researchers; Aripekka Junno, Eden Richards-Slidel, Jonas Niemann, Tatiana Feuerborn, Madison Llewellin, Xénia Keighley (Weber), Maiken Hemme Bro-Jørgensen, Anne-Marijn van Spelde, Anne Katrine Runge, Alison Harris, Özge Demirci, Manon Bondetti, Theis Jensen and Mariana Muñoz-Rodriguez. Your friendship and support have made my PhD years a pleasure.

# II. List of Papers

I) Junno, A., **Dury, J.P.R**., Leipe, C., Wagner, M., Tarasov, P.E., Hirasawa, Y., Jordan P.D & Kato, H. In Press. High-resolution chronology for a multi-phase maritime forager settlement – a case study of the Hamanaka 2 site in Rebun Island, Hokkaido (Japan). *Journal of Archaeological Science: Reports.* (Accepted, pending corrections).

II) **Dury, J.P.R**., Eriksson, G., Fjellström, M., Wallerström, T & Lidén, K. 2018. Consideration of Freshwater and Multiple Marine Reservoir Effects: Dating of Individuals with Mixed Diets from Northern Sweden. *Radiocarbon*, 60(5), pp.1561–1585.

III) **Dury, J.P.R**., Eriksson, G., Savinetsky, A., Dobrovolskaya, M., Dneprovsky, K., Harris, A.J.T., van der Plicht, J., Jordan, P.D & Lidén, K. Species specific reservoir effects: A case study of archaeological marine samples from the Bering Strait. *The Holocene* (Accepted, pending corrections).

IV) **Dury, J.P.R**., Eriksson, G., Savinetsky, A., Dobrovolskaya, M., Dneprovsky, K., Harris, A.J.T., van der Plicht, J., Jordan, P.D & Lidén, K. Addressing the Chronology of the Ekven Mortuary Site (Chukotka, Russia). (Manuscript).

V) **Dury, J.P.R**., Lidén, K., Harris, A.J.T & Eriksson, G. Dental wiggle matching: Radiocarbon modelling of micro-sampled archaeological human dentine. *Quaternary International.* (Accepted, pending corrections).

# III. Defensible Propositions

1 The building of culture chronologies by radiocarbon dating is particularly difficult in Arctic contexts. Thought should be given to the old wood problem, recycling of datable materials, a high consumption of aquatic foods and potentially large reservoir effects.

2 This thesis demonstrates, using the Bering Strait as a case study, that reservoir effects within a single environment can be highly variable and should be understood prior to the modelling of reservoir affected human radiocarbon dates.

3 When there is reason to believe foods were consumed from multiple carbon sources, the dietary components of human diets should be estimated prior to the modelling of radiocarbon dates. It may be necessary to understand exactly which species were consumed.

4 Reservoir effects can be estimated in a number of ways. These methods, however, are not universally applicable and each has its own strengths and weaknesses for archaeological research. Researchers must consider which method is most suitable given the samples they have available for analysis.

5 Where possible, Bayesian modelling should be applied for the more accurate dating of reservoir affected samples. Where the dating of aquatic material can result in higher uncertainty, modelling of the $^{14}C$ dates within a wider context is very helpful in reducing this.

6 It has been demonstrated that the principle of 'wiggle matching' can be applied to the radiocarbon dating of teeth. This can help reduce dating uncertainty (particularly for humans with mixed diets) and estimate the size of local reservoir ages.

# Contents

# 1 Introduction

Radiocarbon dating is not a technique that measures the age of a sample, but rather provides information on the abundance of $^{14}C$ in a sample. How this then relates to the age of the sample is often quite complicated involving many affecting factors.

Radiocarbon dating has become an essential tool for archaeological research, being perhaps the most affordable and most accessible absolute dating method available. The fact that almost all organic substances can be radiocarbon dated, up to 55,000 years old, gives radiocarbon dating a ubiquity unmatched by other dating methods. Radiocarbon dating has allowed for the building, testing and refining of archaeological and historical chronologies, many of which have changed our understanding of the past.

Despite the opportunities radiocarbon dating undoubtedly offers archaeological research, some samples must be approached differently for accurate dating. ‘Reservoir effects’ acting on aquatic samples (both marine and freshwater) have the potential to lead to erroneously old radiocarbon dates. When radiocarbon dating material from marine or freshwater systems, there is potential for samples to appear much older than they are. The archaeological implications of reservoir effects are substantial. Reservoir effects must be accounted and corrected for; if they are not identified or if they are misunderstood, erroneous dating can still result. Given the importance of aquatic systems for countless human groups throughout time, reservoir effects must be afforded full consideration. Archaeologists have been aware of these phenomena and have been mitigating their effects for several decades. Reservoir effects, however, too often are seen, not as a fact of the radiocarbon cycle, but as problems of radiocarbon dating. Certainly, poorly understood or unknown reservoir effects have resulted in erroneous dating of archaeological material, however, we need to approach reservoir effects as a phenomenon which needs understanding rather than fixing. In recent years there has been an explosion of interest in working with aquatic archaeological materials, however, much more research still needs to be conducted.

It is important to remember that reservoir effects are not a fringe problem in the radiocarbon dating of material associated with human activity. Humans have, for thousands of years, interacted with and consumed marine resources. Civilisations and settlements alike have grown around coastlines and rivers, a testament to the importance of waterways for transport, trade and food. It is not, however, only communities by shorelines who interact with aquatic material. Aside from the residential mobility of humans, the development of preservation technologies allowed for the transportation of aquatic foods through drying, fermenting and salting. On top of food, aquatic materials have been gathered for other purposes; shells, baleen, whale and fish oils, marine mammal ivory and skins, among many others, have all been collected and widely traded. At any archaeological excavation, regardless of time or location, it is highly likely marine material will be recovered. When radiocarbon dating, the possibility of reservoir effects must always be held in mind.

## 1.1 Research Aims

It is the aim of this thesis to explore the ways that archaeologists currently handle reservoir effects in the radiocarbon dating of samples, but also to demonstrate new techniques in mitigating the dating uncertainty often associated with them. Here, several questions will be addressed:

1 For human populations with complex diets, how can reservoir effects be best managed to increase dating accuracy?

2 How can reservoir effects be estimated?

3 How varied can reservoir effects within a single aquatic system be and how does this affect populations utilising resources from those systems?

4 How can Bayesian modelling be used to mitigate reservoir effects and reduce radiocarbon dating uncertainty?

5 Can reservoir effects be used as a tool to help answer questions in other scientific fields?

## 1.2 Structure of the Thesis

The purpose of this PhD has been to address the questions posed and to discuss broadly reservoir effects and how they affect archaeological research. This thesis consists of nine chapters and five research papers. Chapter 2 lays a foundation for subsequent discussions of reservoir effects by explaining the processes of radiocarbon dating. This allows for thorough discussions of radiocarbon reservoir effects (Chapter 3) and radiocarbon calibration/modelling (Chapter 4). Chapters 5–9 each detail different ways archaeologists can interact or ‘deal’ with reservoir effects. A range of existing methods and new techniques will be described. Finally, five case studies (Papers I–V), from four distinct archaeological sites, will be used to address the questions listed and illustrate some of the methods detailed in Chapters 5 through 9. Each case study is designed to 'deal' with reservoir effects in a distinct fashion.

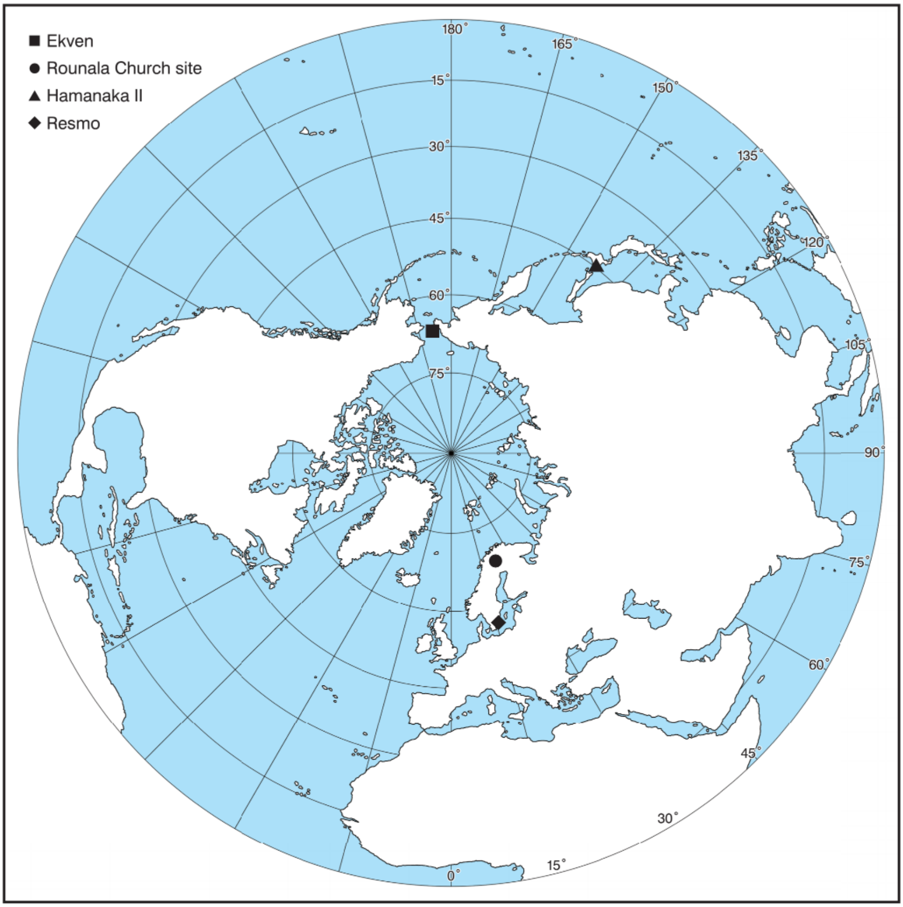


*Figure 1: Map displaying locations of the four case study sites: Ekven (square), Resmo (diamond), Rounala (circle) and Hamanaka 2 (triangle).*

These five case studies, from four sites (figure 1), focus on distinct northern hemisphere populations who have relied heavily on aquatic resources. Though they are spread across a large area, and date to different periods in the past, they demonstrate a need for more refined calibrations of their $^{14}$C dates, to better understand the sites in question. Although the humans at the four sites have all interacted with marine resources, the methods of dealing with their reservoir effects are quite different. These sites are:

1. Hamanaka II (Paper I), an Okhotsk period coastal settlement on Rebun island, off Hokkaido, Japan. The inhabitants fished intensively and processed food in distinctive and ornate pottery. The site is thought to have been in use for some 2000 years. To date, however, there has been no dating of the site's deep stratigraphy, which would aid in refining the dating of the various pottery styles that have developed in this region.

2. Rounala (Paper II), a church and associated cemetery in northern Sweden dating roughly to the 1550s. Though the date of the church's founding is based on strong historical records, it has long been suggested that the cemetery may predate its founding. This has implications for the understanding of the Christianisation of the area. Complex and mixed diets for those buried at the site and the potential for several different reservoir effects pose a challenge to the modelling of human sample radiocarbon dates.

3. Ekven (Papers III-IV), a large burial site on the coast of the easternmost tip of Chukotka, Russia. The site belongs to the Old Bering Sea Culture, a group of sea-mammal hunters, the precursors to the Thule culture. Previous research suggests that there is a very large marine reservoir effect in the Bering strait area. Despite this, the variation in reservoir effects across the species hunted by OBS peoples has not been investigated. Moreover, large scale dating of human remains is required for a more robust culture chronology to be developed.

4. Resmo (Paper V), a megalithic tomb on the Island of Öland, Sweden. The tomb contains the disarticulated remains of many individuals. Stable isotope analysis of their teeth suggests complex and changing diets in their formative years. Applying accurate and specific dating corrections is therefore difficult. Here a novel wiggle match model is tested to help reduce dating uncertainty and to estimate the reservoir effect.

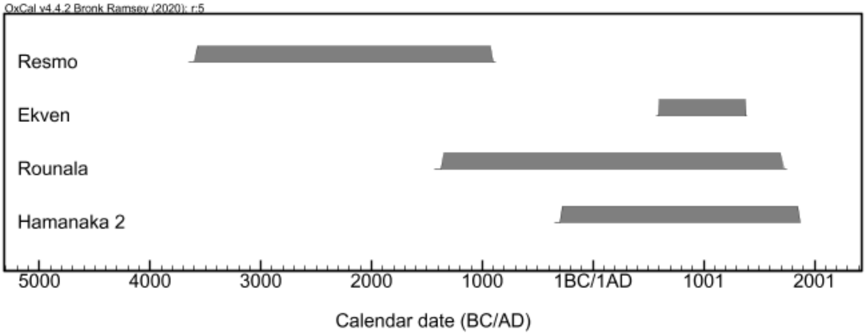


*Figure 2: OxCal Calibrated radiocarbon date estimates for the use of the four case study sites: Ekven, Resmo, Rounala and Hamanaka 2.*

As well as being geographically dispersed, the sites in question also are temporally diverse. Figure 2 displays the best current radiocarbon date estimates for the use of the four sites. Given their geographic and temporal variability, these sites demonstrate the necessity to always consider reservoir effects when investigating those consuming aquatic foods. These case studies will also demonstrate how reservoir effects can be measured and the different ways they can affect humans. These include ensuring that sample selection avoids reservoir effects, accounting for complex and mixed diets of humans, measuring the variability of reservoir effects within a geographic area, using complex modelling to mitigate reservoir-effect related uncertainty, and applying novel techniques to the calibration of reservoir-affected human $^{14}$C dates.

# 2 Radiocarbon Dating

## 2.1 Radiocarbon Decay

In nature, there exists three isotopes of carbon; $^{12}C$, $^{13}C$ (which are both stable isotopes of carbon) and $^{14}C$. All $^{14}C$ is formed in the upper atmosphere when cosmic radiation interacts with $^{14}N$ (figure 3). Radiocarbon dating relies on the fact that $^{14}C$ is an unstable isotope of carbon, and as such may undergo radioactive decay. The utility of the decay of radiocarbon to date organic samples was quickly realised (Libby et al., 1949) becoming an important tool for archaeological research.

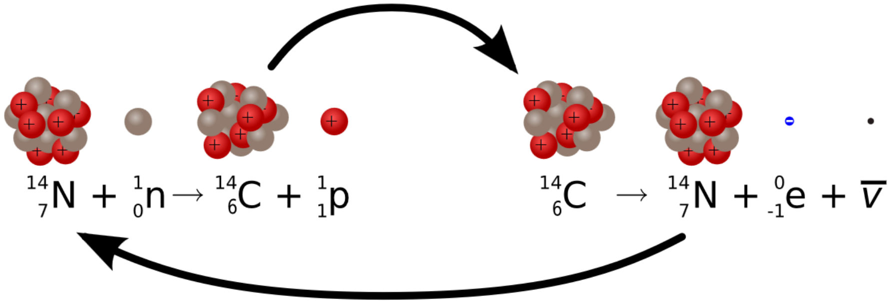


*Figure 3: Formation and decay of radiocarbon* $^{14}_{6}C$. *Superscript denotes mass number, subscript denotes the atomic number. Cosmic rays entering the upper atmosphere produce neutrons which interact with* $^{14}_{7}N$ *to produce radiocarbon and a proton . Radiocarbon is unstable and undergoes β-decay, one of the neutrons decays to a proton forming* $^{14}_{7}N$*, and emitting an electron* ($^{0}_{-1}e$)*and an electron antineutrino* ($\overline{V}$)*.*

Unstable isotopes will ‘decay’ into lighter atoms (sometimes stable, sometimes unstable); $^{14}C$ will decay into stable $^{14}N$, emitting an electron and an electron antineutrino during this process. When considering a single atom of $^{14}C$, radioactive decay is a random phenomenon, there being no way to know when that single atom will decay. When considering the atoms of an entire radioactive sample, however, the rate of decay is more predictable. The half-life of an isotope is a measure of time for half of the atoms in a sample of that isotope to decay. The half-life of radiocarbon has been studied extensively, with the half-life being continuously revised (Kutschera, 2019). The earliest estimate of the half-life of $^{14}C$, 5568±30 (Libby et al., 1949), was later revised to 5730±40 years (Godwin, 1962); more recently a half-life of 5700±30 has been proposed by averaging four half-life measurements (Kutschera, 2013). The half-life of $^{14}C$ has not been 'discovered', but through continued investigation, estimates of increasing accuracy and precision are being proposed. With an estimate of the rate of radiocarbon decay and a measurement of the remaining $^{14}C$ within the sample (see section 4.2), the age of the sample can be easily estimated (figure 4).

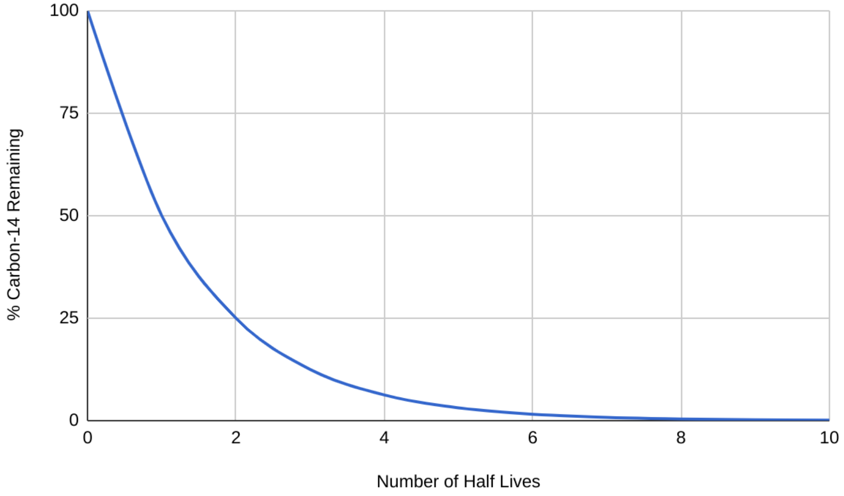


*Figure 4: Remaining $^{14}C$ in a sample (%) against the number of elapsed half-lives (1 half-life = 5568±30).*

## 2.2 Radiocarbon Measurement

To radiocarbon date a sample, the amount of radiocarbon in the sample must be measured. The earliest radiocarbon measurements were made via 'beta counting'. This is based on the detection of β-particles given off as a result of the radioactive decay of $^{14}C$ atoms. Samples with more $^{14}C$ will be more radioactive, emitting more β-particles than samples with less $^{14}C$. By measuring the emission of these β-particles within a given period, the $^{14}C$ concentration of the sample can be estimated. Although useful $^{14}C$ measurements can be made via beta counting, this method has its weaknesses. Firstly, this method of $^{14}C$ measurement is not a direct measurement of $^{14}C$. Secondly, consideration must also be made for the measurement of β-particles given off as a result of background radiation. Thirdly, for accurate measurements, samples must be analysed over a long period. Accurate $^{14}C$ measurements can be made but may take several days to measure, depending on its age. Finally, large sample sizes are needed for analysis,

Samples can also be measured via ‘Accelerator Mass Spectrometry’ (AMS). AMS is the fastest and the most modern development in $^{14}$C analysis, directly measuring the $^{14}$C/$^{12}$C ratio rather than the decay of the sample and can be applied to smaller amounts of carbon. Although β-counting systems can be quite precise (Stuiver et al. 1998, Turney et al, 2010), AMS measurements of the $^{14}$C abundance in a sample are generally more precise. AMS systems ionise the carbon before accelerating those ions to high kinetic energies. Separate isotopes with different masses are separated enabling their measurement.

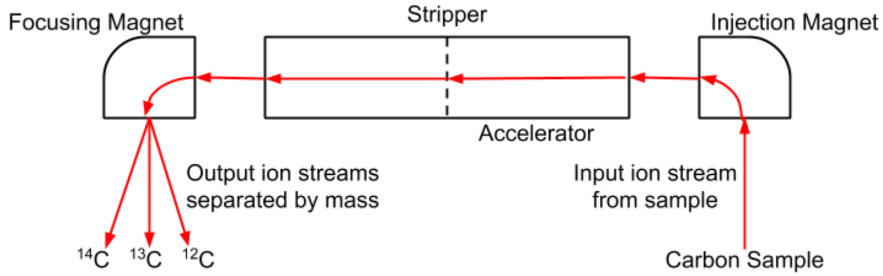


*Figure 5: Accelerator Mass Spectrometer diagram. Carbon samples are negatively ionised and directed towards the accelerator by the injection magnet. Ions inside the accelerator are stripped of electrons, becoming positive, and pushed towards the focusing magnet. This separates different carbon isotopes into separate beams.*

Although any carbon-containing sample can be radiocarbon dated, there is a ‘practical limit’ to the dating of roughly 55,000 years. After this point, any further reduction in the amount of $^{14}$C will be too small to accurately measure. This maximum limit can be pushed back by improvements in AMS dating, however, an ‘absolute limit’ to radiocarbon dating will exist whereby all $^{14}$C in the sample decays. Fossilised material, i.e coal, is radioactively inert, containing no $^{14}$C.

Most organic, carbon-containing substances from the last 50,000 years are, however, suitable for radiocarbon dating. These can be bulk material samples (e.g. wood or charcoal) or carefully extracted compounds, such as lipids (Casanova et al., 2020) or specific amino acids (Nalawade-Chavan et al., 2013). All samples, however, should be free from contamination to achieve accurate and meaningful radiocarbon measurements. Contaminants can come from modern organic material from the handling of samples before analysis, which can result in $^{14}$C dates being erroneously recent. Humic acids, fulvic acids and plant rootlets more recent than the sample can also result in erroneous radiocarbon dates. Careful sampling methods and treatments can be used to mitigate contamination (Brock et al., 2010; Dee et al., 2020; Talamo and Richards, 2011). These pretreatments differ between sample types. The success of pretreatment methods can be tested on samples of known age which are either naturally or artificially contaminated.

# 3 Reservoir Effects

A reservoir effect occurs when samples acquire their carbon from a source other than the terrestrial atmosphere with “older” carbon, resulting in radiocarbon dates which differ from the true age of that sample. With most of the radiocarbon found on earth being formed naturally in the upper atmosphere, organisms acquiring their carbon from the atmosphere are in equilibrium with their contemporary atmosphere. The sourcing and distribution of radiocarbon in freshwater and marine systems, however, is more complex. These systems can contain less $^{14}C$ relative to the atmosphere. Where a system exhibits lower $^{14}C$ activity than the atmosphere, samples from within the $^{14}C$ depleted system are dated as being older than they are. These ‘apparent ages’ are misleading, and the reservoir from which the sample sourced its carbon must be taken into account to correctly model the samples’ actual age.

## 3.1 Marine Reservoir Effects

All radiocarbon is formed in the upper atmosphere, where it is also fairly well mixed. The concentration of $^{14}C$ in the atmosphere, however, is not equal to the $^{14}C$ concentration of the oceans. Oceans are large reservoirs where the surface waters have two sources of radiocarbon, atmospheric radiocarbon and radiocarbon from deep ocean waters. Although these deep ocean waters originally acquired their radiocarbon from surface water, they have residence times of hundreds of years. Due to ocean currents and upwelling in certain areas, the marine reservoir effect is not uniform. The global average marine reservoir age (R) is roughly 500 years, however, sharp changes in the radiocarbon of the atmosphere are smoothed in the marine environment (Heaton et al, 2020). The precise marine reservoir effect of a sample can be stated more accurately by applying a $\Delta R$ value to a marine calibration curve. $\Delta R$ values are therefore correction factors applied to average marine reservoir ages, to date samples more accurately. A negative $\Delta R$ will be applied to samples with marine reservoir ages less than the global average (this is the case for many samples from the Baltic Sea region, e.g. Paper V) and a positive $\Delta R$ value will be applied to samples with marine reservoir ages greater than the global average (this is the case for many arctic samples, e.g. Paper III). The various methods used to calculate ΔR values will be discussed in detail in Chapters 5-9.

## 3.2 Freshwater Reservoir Effects

Freshwater reservoir effects (FRE) are similar to marine reservoir effects, in that they can result in erroneously old dates of material from freshwater systems (Lanting and van der Plicht, 1996; Lanting and van der Plicht, 1998), however, the causes are quite different. The freshwater reservoir effect is mostly due to dissolved inorganic carbon (i.e calcium carbonate, with low $^{14}C$) becoming incorporated into freshwater environments, sometimes referred to as the hard-water effect. This is not the case for all freshwater rivers or lakes. Freshwater reservoir effects are also measured in soft water lakes, caused by slow $CO_2$ exchange between the atmosphere and the lake (a result of large depth-to-surface ratios), wind protection or extended periods of lake ice cover, the addition of waters with old $CO_2$ from glaciers or groundwater and the oxidisation of old organic matter (Philippsen 2013; Boaretto et al. 1998). The freshwater reservoir effect is globally, highly variable, more so than the marine reservoir effect. Depending on the underlying geology, bodies of water can be without the FRE (Svyatko et al., 2017; Paper-II), or exhibit freshwater reservoir ages of up to 4000 years (Philippsen, 2013). Moreover, within the same body of water freshwater reservoir ages can vary up to 2000 years (Philippsen, 2013). Unlike the marine reservoir effect, there is no applicable global average offset that can be applied for FREs, owing to its variability; the size of the reservoir effect can be estimated by comparing the $^{14}C$ age of an aquatic freshwater sample to another estimate of its calendar age.

## 3.3 The Hemisphere Effect

It should be briefly noted that atmospheric $^{14}C$, whilst considerably more homogenous than marine $^{14}C$, is still somewhat variable. The hemisphere effect is a small but well-documented difference between the $^{14}C$ abundances of the atmospheres of the northern and southern hemisphere. This is because the atmospheres of the northern and southern hemisphere circulation systems are mostly independent of each other. There exists a time lag in mixing between the two which has implications for radiocarbon dating. The atmospheric $^{14}C/^{12}C$ ratio is lower in the southern hemisphere compared to the northern hemisphere. Radiocarbon results of terrestrial samples from the southern hemisphere appear roughly 30 years older on average, compared to those from the northern hemisphere. This is due to the larger surface area of the ocean in the southern hemisphere. This has the effect of allowing for more carbon exchanged between the ocean and the atmosphere. Surface ocean waters are depleted in $^{14}C$ because of the marine reservoir effect, in the southern hemisphere there is a net movement of $^{14}C$ from the atmosphere to the oceans at a faster rate compared to the north (Bowman, 1990). The hemisphere effect, however, is small and easily dealt with through the selection of the correct calibration curve (Hogg et al., 2013).

# 4 Radiocarbon Calibration and Modelling

Radiocarbon calibration refers to the process of turning a measurement of $^{14}C$ abundance in a sample to a usable calendar date. The application of prior information, rules and constraints which change the calibrated radiocarbon date is the process of modelling. This additional information, used to construct models, often comes from associated material recovered during excavations, relationships to other radiocarbon-dated materials and historical ‘knowns’ of the sample. Radiocarbon calibration and radiocarbon modelling are both vital aspects in achieving accurate calendar radiocarbon dates.

## 4.1 Purpose of Radiocarbon Calibration

When samples are radiocarbon dated, they are measured and reported as ‘conventional ages’. Radiocarbon dates are reported either as ‘percentage modern carbon' (pMC), ‘fraction modern carbon’ (F14C) or years ‘before present' (BP). pMC and F14C indicate the proportion of radiocarbon atoms in a sample as compared to samples modern in the year 1950. Although expressions of age in $^{14}C$ years BP are derived directly from pMC, they are based on several assumptions.

1 That the ‘present-day’ is defined as 1950 AD.
2 The usage of oxalic acid 1, oxalic acid 2 or another appropriate secondary standard as the modern radiocarbon standard.
3 Correction for sample isotopic fractionation is normalised to a $\delta^{13}C$ value of -25.0‰ relative to the standard VPDB.
4 That the half-life of $^{14}C$ is 5568±30 years, as proposed by Willard Libby.
5 Global $^{14}C$ levels have been constant through time.

These conventions ensure standardised reporting and measurement of $^{14}C$ dates, however, ‘conventional dates’ should not be considered expressions of calendar dates. Calibration of conventional dates is needed to address a number of these assumptions to yield calendar dates suitable for archaeological and historical research purposes:

Firstly, for the practicality of reporting, the year 1950 AD has been taken to represent the 'present'. This year predates the spike in $^{14}C$ production caused by atomic bomb testing (Hua et al., 2013). Assuming this, conventional radiocarbon dates expressed as BP dates will still be accurate in the years post-publication.

Regarding point 2, a sample is needed to define the $^{14}C$ abundance of 'modern' carbon. This has been defined as the radiocarbon activity (measured in 1950) of a sample of wood growing in the northern hemisphere in the year 1890 AD. This date predates the fossil fuel effects of the industrial revolution and atomic bomb testing (Tans et al., 1979; Hua et al., 2013). From laboratory measurements, an oxalic acid standard (HOx1) has a $^{14}C$ activity 0.95 times the activity of the 1890 wood sample. Since HOx1 is almost exhausted, a further has been prepared (HOx2) (van der Plicht and Hogg, 2006), which has a $^{14}C$ activity 1.2933 times HOx1 (Mann, 1983).

Thirdly, it should not be assumed that the measured $^{14}C$ value of a sample has only been affected by radioactive decay. Fractionation of isotopes of carbon can occur during various biological processes. During photosynthesis, for example, lighter isotopes, such as $^{12}C$, are more likely to be involved in biochemical processes than heavier ones, to the extent that both $^{14}C$ and $^{13}C$ are depleted relative to the atmosphere. This fractionation, therefore, has implications for radiocarbon dating as radiocarbon is less likely to be involved in biological processes. If fractionation which results in $^{14}C$ depletion is not recognised, the lower $^{14}C$ abundance will be explained by assuming an increased age of the sample. For accurate radiocarbon dating and the conversion of a $^{14}C$ measurement to a calendar date, it must be understood to what extent this fractionation has occurred in the sampled material. If isotopic fractionation occurs in natural processes, a correction can be made by measuring the $^{13}C/^{12}C$ ratio of a sample being dated. This ratio is expressed as a $\delta^{13}C$ value in parts per thousand (per mil, ‰) relative to the content of the $^{13}C/^{12}C$ of the international standard, VPDB. Radiocarbon laboratories frequently correct for the effects of fractionation in dated samples, with 'normalised dates' being produced.

Fourth, that reported AMS dates are calculated assuming a radiocarbon half-life of 5568±30 years, this is referred to as the 'Libby half-life'. The half-life of $^{14}C$ has since been revised to 5730±40 years; this is known as the 'Cambridge half-life' (Godwin, 1962). Despite the revision of the estimate of the half-life of radiocarbon, for consistency across all published conventional dates, it was agreed at the 1962 Radiocarbon Conference in Cambridge (UK) to continue to use the Libby half-life of 5568±30 years in the calculation of conventional $^{14}C$ dates.

Finally, it is now well known that global $^{14}C$ levels have fluctuated through time. Through the $^{14}C$ dating of samples of known calendar age, such as dendrochronologically dated tree rings, it has been demonstrated that the intensity of $^{14}C$ production has not been constant. Production rates are variable, with factors such as cosmic ray flux (affected by changes in solar winds and the solar magnetic field), and variations in the earth's magnetic field increasing or decreasing production rates. Throughout history, there have been significant moments of radiocarbon production (spikes), recorded in the dendrochronological record (Miyake et al., 2012).

Although conventional radiocarbon dates are based on these conventions, conventional $^{14}C$ dates can be calibrated into calendar dates. Calibration curves, such as IntCal20 (Reimer et al., 2020) are made by plotting the conventional radiocarbon dates of samples against their calendar age. A curve of measured $^{14}C$ conventional dates against calendar dates is constructed. When a sample of unknown calendar age is radiocarbon dated, the conventional age of that sample (despite the assumptions discussed above) can be calibrated against the curve to yield the most appropriate calendar age.

When the date of a sample is quoted, the reader should be aware that if it is an uncalibrated date (a term used for dates given in radiocarbon years) it may differ substantially from the best estimate of the actual calendar date. Laboratory $^{14}C$ dates are not suitable for most archaeological purposes, however, can easily be calibrated to be so.

## 4.2 Construction of Calibration Curves

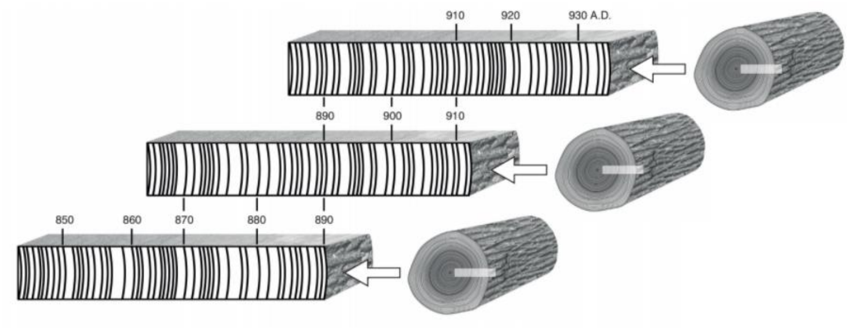


*Figure 6: Representation of overlapping dendrochronological tree ring sequences between 850-930 AD.*

Willard Libby was the first to note that radiocarbon ages diverged from calendar age, publishing a ‘curve of knowns’ (Libby, 1955). Since then, it has been recognised that a model of past $^{14}C/^{12}C$ concentrations has been necessary for the understanding of conventional radiocarbon dates. Dendrochronological records rely on the comparison and overlapping of tree rings. This is illustrated in figure 6. This allows for precise annual measurement of atmospheric $^{14}C$ (by radiocarbon dating individual tree rings) extending back 13 cal kBP (Reimer, Bard, Bayliss, Beck, et al., 2013). By plotting the calendar age of the tree ring against its measured $^{14}C$ value, a calibration curve can be drawn. This allows for samples of unknown calendar age to be $^{14}C$ dated, and their calendar age estimated. Calibration programs such as OxCal, CalPal and Calib take into account $^{14}C$ measurement uncertainty and uncertainty associated with the measurements used to construct the calibration curve, to estimate a probability density for the calendar age of the sample (Figure 7).

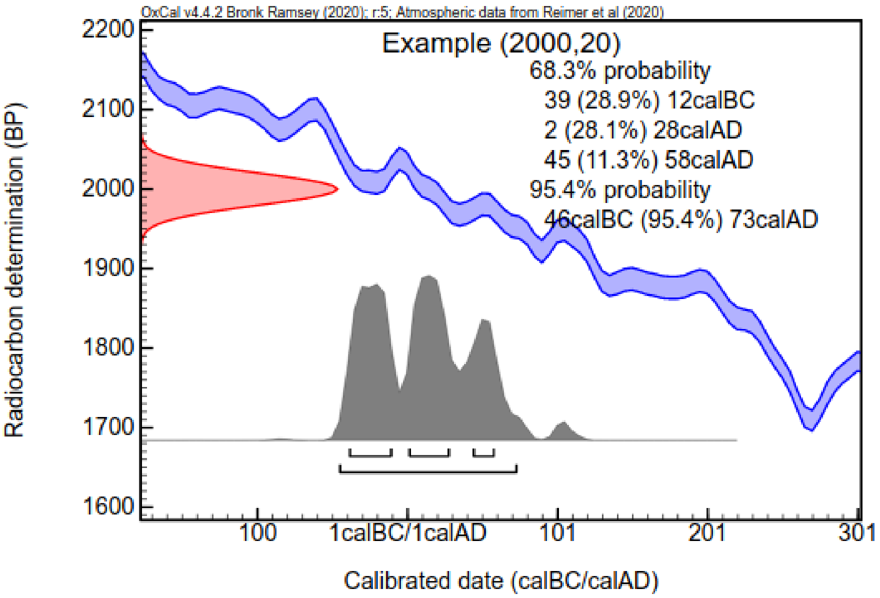


*Figure 7: Sample with $^{14}C$ conventional age 2000±20 BP calibrated against IntCal13 curve in OxCal. Date range estimates of 1sd (68.2% probability) and 2sd (95.4% probability) are shown.*

To date, the most recent iteration of the terrestrial calibration curve is the IntCal20 curve (Reimer et al., 2020). Not all organisms, however, will acquire their $^{14}C$ from the terrestrial atmosphere. This would make calibration of these samples against terrestrial curves unsuitable. For samples whose carbon was acquired from the oceans, a calibration curve specifically for marine samples must be used.

The most recent marine calibration curve is Marine20 (Heaton et al., 2020). The construction of the Marine20 is dependent on more modelling than IntCal20. Between 0-10.5 cal kBP, Marine20 is based on terrestrial data, adjusted using the ocean-atmosphere box diffusion model (Oeschger et al., 1975). Marine calibration, older than 10.5 cal kyr BP, is provided by data from

Cariaco Basin and coral Uranium/Thorium ages (Hughen et al., 2004; Reimer et al., 2009; Reimer, Bard, Bayliss, Warren Beck, et al., 2013). Independently dated marine samples (including marine corals) can be used to create a calibration curve (unless otherwise stated, all ΔR values discussed in the text refer to the Marine20 curve, rather than the Marine 13 curve). A comparison of the last 10,000 years of Marine20 and IntCal20 calibration curves can be seen below in figure 8.

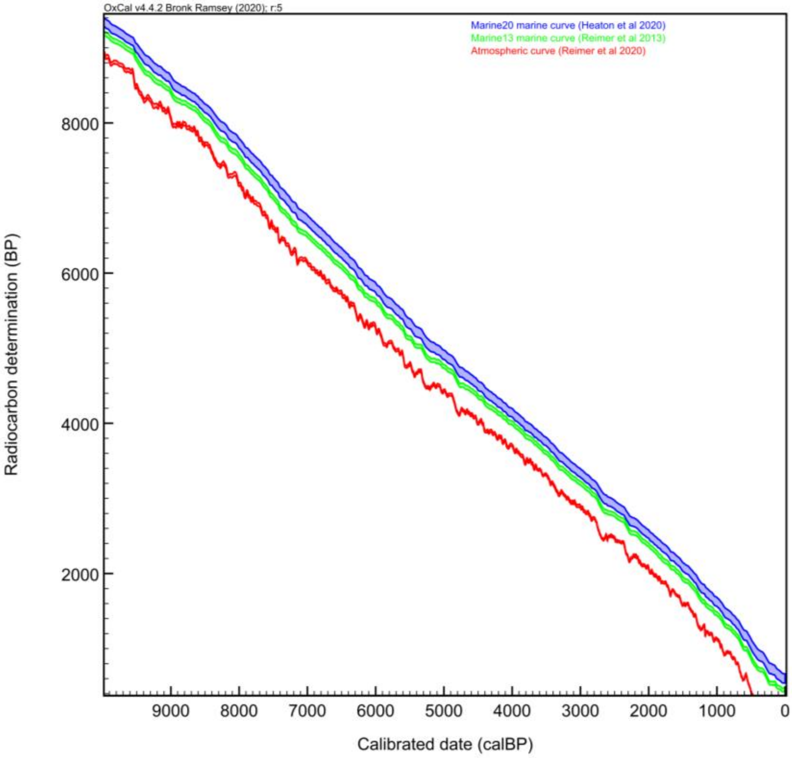


*Figure 8: The last 10,000 years of the IntCal20 terrestrial calibration curve (red), the Marine13 curve (green) and Marine20 marine curve (blue), generated using OxCal.*

Other factors which affect calibration, especially in more recent periods, must be considered. Since the start of the intensification of human fossil fuel use, atmospheric $^{14}$C concentration has been impacted. Fossil fuels, given their geological age, are radioactively inert. Their combustion releases $^{14}$C-depleted $CO_2$, diluting the concentration of $^{14}$C in atmospheric $CO_2$. This effect is referred to as the Fossil Fuel effect or Suess effect (Tans et al., 1979). Because of this effect, radiocarbon dating material from the period c. 1650-1950 is made more challenging.

Understanding the conventional dates of samples which acquired their radiocarbon since atomic bomb testing also requires special consideration. From about 1950 until 1963, above-ground nuclear testing released large numbers of neutrons, increasing the formation of atmospheric $^{14}$C. The amount of atmospheric $^{14}$C was doubled, peaking in the northern hemisphere in 1964 and the southern hemisphere in 1966. This is referred to as the 'bomb pulse', or 'bomb spike', with the $^{14}$C described as 'bomb-carbon'. Since the abandonment of nuclear testing, the atmospheric concentration of $^{14}$C has gently dropped (Hua et al., 2013). When radiocarbon dating material post-1950, other specific datasets are available for calibration (Hua et al., 2013).

## 4.3 Bayesian Modelling

Bayesian statistical models use Bayes' theorem to calculate and update probabilities after obtaining new data. Bayesian statistical models describe the probability of an event, based on data as well as prior information. This makes it very applicable to radiocarbon dating material recovered from archaeological excavations. A sample is radiocarbon datable even without a context, but Bayesian modelling requires further information to achieve more accurate dating. When calibrating $^{14}$C dates, facts about the sample which have a bearing on its dating are relevant. These can be built into models using computer programs such as OxCal. Prior information can include, e.g. sample ordering based on stratigraphy, maximum and minimum dates or a known fixed time interval between samples. Programs such as OxCal are designed to take advantage of this principle and perform several hundreds of thousands of 'runs', finding calibration solutions to the data entered. Bayesian modelling has proved very useful in more accurately dating material for which dating is inherently more uncertain, e.g. in very old samples (Becerra-Valdivia et al., 2018) or material which has been subject to reservoir effects (Krus et al., 2019).

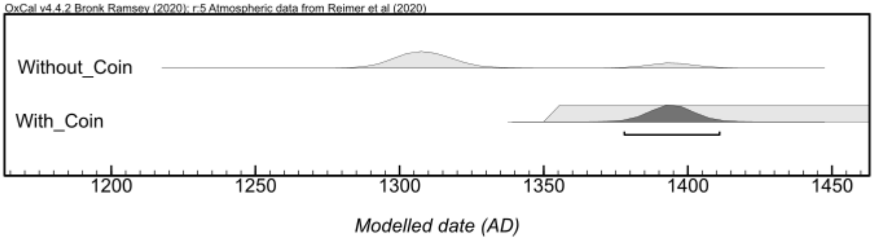


*Figure 9: Two OxCal estimates of a hypothetical human's date of death (using a $^{14}$C value of 700±25 BP plus a tissue-formation to death offset of 20±5 years). The date of death is modelled twice, with and without a coin of known date acting as a prior.*

For example, the figure above (figure 9) demonstrates the calibration of a hypothetical human with a $^{14}$C date of 700±25 BP (plus a tissue-formation to death offset of 20±5 years). This hypothetical individual was excavated with a coin, known to date from around the year 1350 AD. The first distribution 'Without_Coin' calibrates the date of the human remains with no consideration of the associated artefact, and at 95% probability, finds two separate date ranges for the sampled remains. The consideration of the dated coin by applying a prior, stating that the human must date between 1350 and 1900 AD (With_Coin), finds that the earlier distribution is not possible. It may be asked why radiocarbon dating is needed at all in this example if a dated coin was recovered. From the second distribution plot, however, at 95% probability the human dates between 1378 and 1411 cal AD. The human can be more recent than the coin, but not older. This model finds that the human's death dates very closely to the coins minting. Without radiocarbon dating, the coin could only act as a *terminus post quem*, $^{14}$C dating is therefore still applicable

The example in figure 9 is very simple, yet effective in changing our understanding of the dating of a sample. In theory, the more relevant information is built into a Bayesian model in programs like OxCal, the narrower the possible range of dating solutions. This has the effect of reducing calibrated date uncertainty. This will be discussed more fully in Chapter 8. A Bayesian model is, however, subject to the accuracy of the prior information which is built into it. If erroneous information, weak assumptions or tenuous links are modelled as facts, the final date estimates of a radiocarbon-dated sample will suffer. Though Bayesian statistics are incredibly well suited to radiocarbon dating, it challenges researchers to bridge gaps between field archaeology and statistics. In this way, the building of Bayesian models for radiocarbon dating requires archaeological thought and constant assessment of what we can be confident about, what information is relevant to a sample and what the result of the modelling can tell us. Where possible, when referring to the estimated calendar dates of a radiocarbon-dated sample in published works, it should be briefly stated if the dates have been modelled according to any information other than a calibration curve.

## 4.4 Identifying Outliers

Often, archaeological research depends on several different samples being radiocarbon dated. Complex Bayesian models can be built, grouping samples which share the same context (i.e. material from the same grave, or material from the same stratigraphic layer) or separate materials which do not (i.e. by ordering materials in a stratigraphic sequence). Bayesian models are constructed to find the most appropriate calibration solutions, by applying rules (algorithms). It is often the case, however, that to 'obey the rules' of a model, the $^{14}$C date of a sample is not just constrained slightly, but shifted more extremely. Samples which do not 'fit', or require too much manipulation to fit, are typically considered outliers. A $^{14}C$ date within a Bayesian model could be considered as an outlier for several reasons:

- The random error should be understood from the published date range. At 2σ, 1 in 20 samples will have a true date that falls outside the 95% range.
- There may be an error in $^{14}C$ measurement. This could be due to machine error or contamination of the sample.
- The sample could be misidentified or have been improperly curated. In such a case it may be possible the sample is calibrated against the wrong curve or inappropriate corrections are applied.
- The $^{14}C$ date of the sample may not fit the priors of the model, for example, not adhering to a stratigraphic sequence due to bioturbation, post-depositional interference or animal action.
- The prior information built into the model which constrains the conventional $^{14}C$ dates is incorrect.

It must always be remembered, however, that it is not the case that samples which are highlighted as outliers by Bayesian models must be removed from further analysis. A sample which does not fall foul of any of the points listed above may still be considered an outlier. It may be the case that within a group of samples, $^{14}C$ variability is high. If a phase is represented by only a few samples, the model may incorrectly identify ‘outlying’ samples. Researchers must give careful thought to how models are constructed, balancing the purpose of the model (to reduce dating uncertainty) with a need for flexibility within the model (so as not to highlight material with reasonable $^{14}C$ dates for removal).

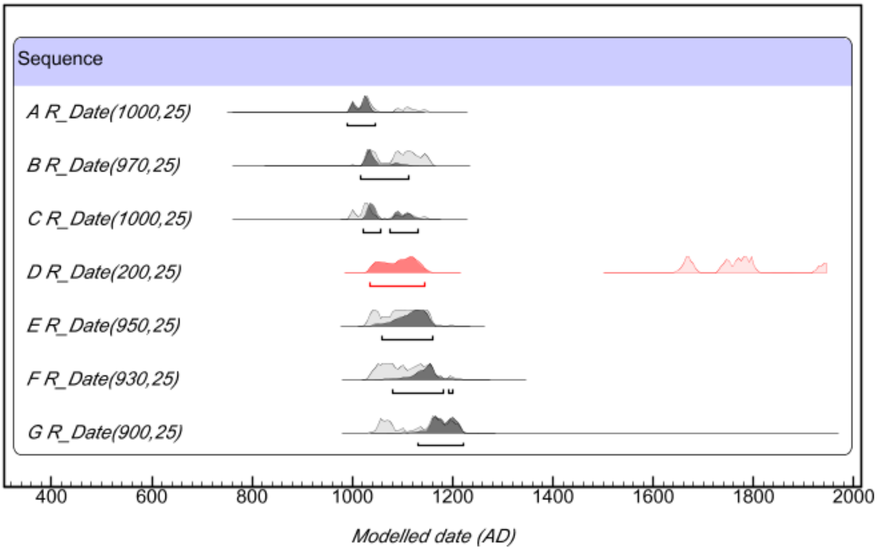


*Figure 10: An OxCal modelled sequence of hypothetical $^{14}$C dates with an outlier highlighted in red.*

Figure 10 illustrates how a sequence of seven hypothetical $^{14}$C dates might be modelled and how an outlier might be affected by this modelling. The strict parameters of the model have forced the sample ‘D’ to calibrate to a date much earlier than an unmodelled calibration would. A visual inspection of the data would lead most to conclude that ‘D’ is indeed an outlier, however, not all outliers will stand out quite as clearly. Moreover, an inspection of the data in this way can be quite subjective. The modelled and un-modelled probability distributions of sample ‘D’ in this example, do not overlap. If they were to overlap, its classification as an outlier would be more difficult. More objective methods of outlier identification exist to overcome this. OxCal models can be constructed to identify outliers on a statistical basis; one such method of outlier removal is outlined by (Bronk Ramsey, 2009). Here, of any samples failing to reach a 60% agreement threshold (calibrated date distribution compared to modelled date distribution) the sample with the lowest agreement is removed. The OxCal model is then re-run. This process is continued until all samples’ agreement indexes are more than 60%. A more strict method of sample removal is OxCal’s outlier analysis function; typically samples outside the 95% confidence interval are highlighted as outliers (Bronk Ramsey, 2009).

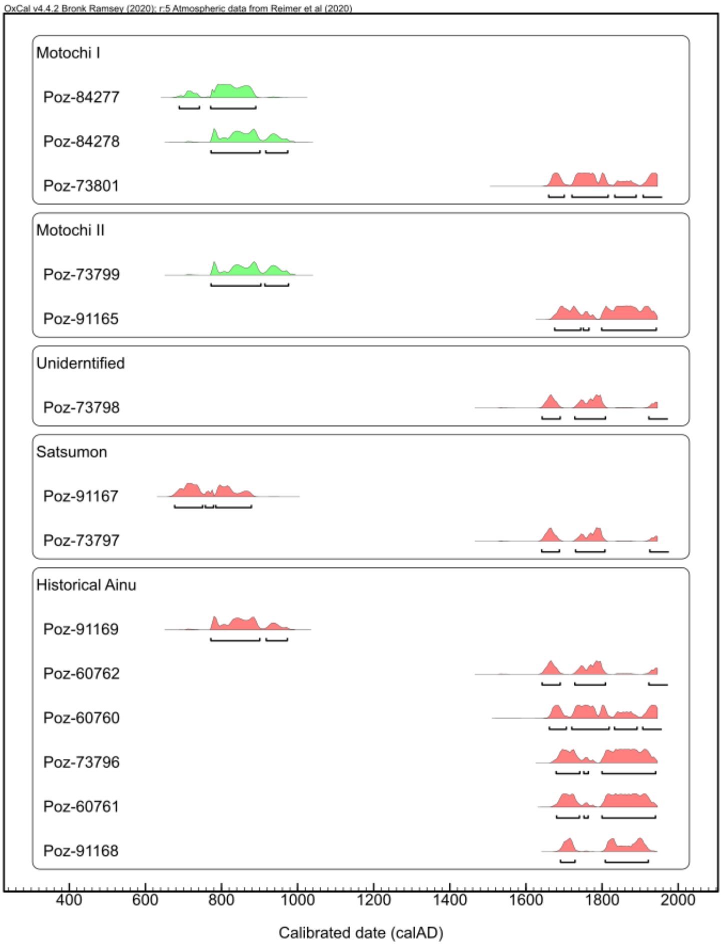


*Figure 11: Unmodelled $^{14}$C dates from Motochi I to Historical Ainu layers at Hamanaka 2 (Paper I). Distributions highlighted red do not match existing historical chronologies. Generated using OxCal.*

The removal of samples from a dataset after running a calibration model must be done with caution. The specific context from which the samples came must be considered. The model above (figure 11) displays unmodelled $^{14}$C dates from seeds from the upper 5 layers (of 13 layers) from the site of Hamanaka 2 (Rebun island, Japan). The earliest samples from the Motochi I phase are in perfect agreement with existing chronologies of the region, as is the sample Poz-73799 from the layer Motochi II (these are highlighted in green). The other $^{14}$C dates samples, however, have calibrated dates which are too old or too recent (these are highlighted in red). Running these data in a Bayesian model which utilises the stratigraphic sequence as a prior, the model highlights the samples Poz-73799, Poz-91167 and Poz-91169 as being outliers. The modelling solution which requires the least manipulation of the $^{14}C$ data is to remove the fewer earlier dates and retain the later dates. This, however, has the effect of extending the phase Motochi I up to the year 1600 and excludes a perfectly reasonable date for the Motochi II phase (Poz-73799). This demonstrates the need to consider suggested outliers within an archaeological context. Here the question must be asked, is it more likely that bioturbation of the upper layers has resulted in the introduction of more recent seed samples, or should the chronology of the site be reorganised to fall in line with this data? In this case, there is a plausible explanation for the recent $^{14}C$ dates in the sequence, however, the modelling solution which requires the least manipulation is not archaeologically acceptable.

Even when a sample fails outlier checks, it is important for researchers not to remove the sample from the model without first considering reasonable reasons why its fit is poor. This can be the result of human error (i.e. a mislabeled bone in an archive or a sample becoming dislodged from a section profile) or natural processes (i.e bioturbation or animal action). Contaminated samples, although perhaps from the correct context, should also be considered outliers. Researchers should also consider the construction of their model as the cause of outlying dates. It is expected that contemporaneous samples will exhibit variation in their $^{14}C$ dates; this will be due to natural variation and measurement error. A model must be robust, accounting for this variability but not so strict that samples which deviate from its priors, even slightly, will be omitted.

Outlier analysis is particularly useful when considering materials which may have been affected by reservoir effects. If a model is constructed which considers samples' carbon sourcing appropriately and no samples are highlighted as outliers, it can be said with more confidence that the reservoir effect has been accounted for properly. In this way, the dating of more 'problematic' aquatic material can use less 'problematic' terrestrial material as a check to ensure that similar calendar dates are being modelled. If either is highlighted as an outlier, it may be that reservoir effects have not been properly mitigated.

The modelling of radiocarbon dates is a different process to calibration. Whereas the purpose of calibration is to allow for conventional dates to be tuned into calendar dates, modelling manipulates calibration based on prior knowledge about the sample. Prior knowledge can include the position of the sample within a sequence of other dates, minimum and maximum dates, or mixing of calibration curves. Calibration solutions which do not adhere to the model's priors are discounted. This typically results in reduced uncertainty of the modelled date, relative to the un-modelled (calibrated) date. The priors applied, however, may reveal a $^{14}C$ date to be inconsistent with the model. Samples may be identified as outliers by their agreement index when their modelled dates differ too far from their un-modelled dates. Outlier analysis can also highlight samples which differ from similar samples in the model. Statistical outlier analysis functions in OxCal is a powerful tool for identifying material with a more complex depositional history. Similarly, outlier analysis functions can highlight material with different carbon sourcing i.e. marine or freshwater material among terrestrial samples. It should be remembered, however, that OxCal models are built, and refuge cannot be taken in the apparent certainty of statistical models. Being constructed, we should be aware of the limitations of any Bayesian model. Archaeological knowledge should not be abandoned when reviewing the results of modelled radiocarbon data. Concerning reservoir effects, calibration is essential, and modelling of marine/freshwater $^{14}C$ dates can greatly aid in reducing overall uncertainty. Outlier tests are also a valuable check of whether reservoir effect corrections have been appropriately applied.

# 5 Avoiding Reservoir Effects

When radiocarbon dating archaeological material from cultures which utilised aquatic resources, it may seem that a full understanding of local reservoir effects is necessary to interpret any radiocarbon dates. In most cases, however, researchers will have a choice in the materials they choose to radiocarbon date and terrestrial samples are likely to be available. Reservoir effects, if poorly understood or unrecognised, have the potential to cause large inaccuracies in modelled dates. The freshwater reservoir effect can be as large as 4000 years (Philippsen, 2013) and the marine reservoir effect can be highly variable (Russell et al., 2011). Further difficulties arise in the interpretation of carbon source mixing of humans with complex diets (Paper-II) and pottery with mixed contents (Casanova, Knowles, Ford, et al., 2020). These sources of uncertainty contribute to uncertainty in the calibration and modelling of $^{14}$C dates. The dating of certain materials to answer some archaeological questions may require a high level of dating resolution. This may not be achieved through the consideration of marine resources.

## 5.1 Sample Selection

Possibly the best way to ‘deal’ with reservoir effects, both marine and freshwater, is to avoid the sampling of any aquatic material entirely. If all aquatic samples are excluded, any problems which may arise from reservoir effects are negated. Although this may seem the most simple approach to radiocarbon dating in contexts where there may be reservoir effects, care must still be taken. Firstly, it must be fully understood what material is being dated; the sample must be known to be terrestrial. Radiocarbon dating an unknown fragment of bone, for example, would leave open the possibility the sample may carry a reservoir effect. Zoological investigation, stable isotope analysis, ZooMS or aDNA analysis may be useful in such a circumstance to identify the taxa being sampled. Secondly, it must be known what ‘event’ is being considered. Wood is one of the most commonly radiocarbon dated materials. Its carbon coming entirely from the atmosphere, there is no possibility of reservoir effects compromising calibration. Where aquatic and terrestrial samples are recovered from a closed context, the calibrated date of the wooden sample is often taken to be the date of the aquatic sample by association. When radiocarbon dating wood, assuming the outer rings have been sampled, the ‘event’ being dated is the felling of the tree, i.e its death, when it ceased to exchange carbon with the atmosphere. In many circumstances, however, wood is not utilised immediately upon felling. For buildings with uncertain construction dates, for example, it must be remembered that wood is often left to age to avoid warping, often for several decades. The dating of wood in this case would not represent the building event, but the felling event. Similarly, driftwood may be decades old before it is worked or burned. Thirdly, it should be recognized that objects and materials have complex histories. Objects can be made of old materials, perhaps reworked from other objects and raw materials unused for long periods. An object, such as an antler figurine, may have been made from an earlier antler tool. Once created, the figurine may have been kept for several generations before its eventual deposition. The dating of the sample will not reflect the creation of the artefact, but the death of the animal (Pitulko, 2000). Neither will the dating reflect the deposition event.

Finally, the recovery of a terrestrial sample from a context or stratigraphic layer does not mean that it ‘belongs’ to that context or layer. Historical human action (such as backfilling or intentional manipulation), animal action and bioturbation all have the potential to disturb stratigraphies, moving samples into new positions. Within a stratigraphic sequence, a small bone fragment, for example, may (through the mechanisms suggested) move between stratigraphic phases. The assumption that the calibrated date of this sample should reflect the date of the stratigraphic layer will not be sound.

For the reasons outlined, even though terrestrial materials do not carry $^{14}$C reservoir effects, this does not mean that the interpretation of their $^{14}$C data is straight forward. The most secure terrestrial samples to date will not have been reused, reworked or stored for long periods. The $^{14}$C dates of charred seeds, twigs and faunal refuse are likely to represent a date very close to the use/deposition events. Several radiocarbon dates should be taken and Bayesian analysis performed to ensure the sample is stratigraphically secure. It should also be remembered, if aquatic objects were to be radiocarbon dated directly, some of the problems discussed here may still apply in addition to any reservoir effects.

## 5.2 Application

*Figure 12: The 2016 Hamanaka 2 excavations, Rebun Island (image courtesy of Ari Junno).*

Though avoiding reservoir effects would seem like the simplest approach in dealing with reservoir effects, as previously discussed, this approach is not without its pitfalls. Several considerations must still be made to achieve accuracy in radiocarbon dating. This is well demonstrated by considering the site of Hamanka 2 (Paper I), an Okhotsk period site on Rebun island (off the coast of northwest Hokkaido, Japan). The dating of the site's deep stratigraphy (figure 12) needed evaluating to better understand the evolution of the region's complex pottery traditions. Much of the material at the site, however, was marine in origin, owing to the maritime cultures who occupied the area. Given the precision needed to understand these shifts, however, radiocarbon dating marine samples or the pottery itself was judged not to be appropriate. By carefully considering which samples might have been subject to the MRE and excluding all marine samples from analysis, reservoir complexities were negated and led to more accurate radiocarbon dating. Only terrestrial seeds, charcoal and twigs were considered for dating.

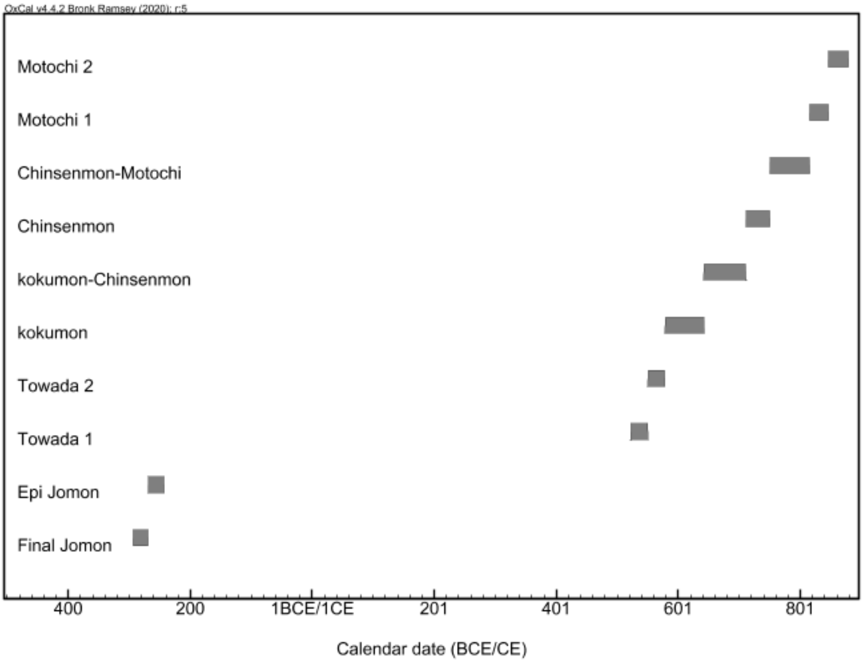


*Figure 13: Mean modelled date ranges for the stratigraphic layers of Hamanaka 2, generated using OxCal.*

As a result of omitting marine samples from analysis, a chronological sequence of the site's stratigraphic layers (named after the pottery traditions of the region) could be dated with some accuracy (figure 13). This may not have been possible if marine material was prioritised for analysis. This case study demonstrates that, in some instances, the best way of dealing with reservoir effects is to avoid them entirely. It may be that now this chronology of radiocarbon-dated stratigraphic layers has been developed for the site of Hamanaka 2, other researchers may wish to incorporate marine $^{14}$C dated material into the model. This may help in reducing the uncertainty of marine $^{14}$C date calibrations or testing if the reservoir age has been suitably calculated.

All archaeological material may be subject to complex depositional histories. Raw materials and artefacts can be worked, reworked, and stored. Unless careful thought is given to what a $^{14}$C date represents, and the 'event' being dated, erroneous conclusions could be reached. This is true for all archaeological material, regardless of its carbon source. Marine samples, however, are susceptible to both these depositional uncertainties and any reservoir effects. In certain circumstances, where high dating resolution is required, it may be that marine samples will not be suitable to answer archaeological questions. The case study of Hammanka II, for example, illustrates how when presented with samples from a maritime adapted culture, complex reservoir effects can be avoided by sampling small terrestrial samples. This is, of course, not applicable in all circumstances, and not to radiocarbon date aquatic material deprives researchers of very important and interesting sources of information. Given the importance of marine and freshwater resources throughout history, avoiding the dating of these materials cannot be a general solution to the challenges of dealing with reservoir effects. Other methods of handling reservoir effects must be utilised.

# 6 Measuring Reservoir Effects

A more direct approach to dealing with reservoir effects is to attempt to measure and quantify the scale of the reservoir effect. The benefit of such an approach is that researchers are not limited to the dating of terrestrial material. Reservoir effects, however, can be highly variable and rarely a single measurement or value is enough to define the reservoir effect in a way which would be suitable for accurate radiocarbon dating. There is variability within ecosystems and especially across species (Ascough et al., 2005; Petchey et al., 2008; Russell et al., 2011). The variability of reservoir effects due to geographic location (Lougheed et al., 2013) and time period (Björck et al., 2003) have been well documented. It is, therefore, appropriate to explore the range of reservoir values at a given site (Russell et al., 2011). When measuring reservoir effects, three main points should be considered by the researcher:

1. The location which the measured reservoir effect will represent.
2. The kind of sample being measured.
3. The strengths and weaknesses of the techniques used to estimate reservoir effect.

How these points are considered and addressed will vary depending on the specific project, funding and material available for analysis. These points will be discussed in turn.

## 6.1 Reservoir Effects and Geographic Variation

Reservoir effects can be highly variable, and one source of variability is the geographic origin of a sample. Since marine reservoir effects are expressed as ΔR values, relative to the marine calibration curve, it should not be assumed that the marine curve alone will suffice for accurate calibration. The carbon of the oceans is much less homogenous than that of the atmosphere. In marine contexts, different bodies of water have different overall $^{14}C/^{12}C$ ratios. Factors such as marine currents (Petchey et al., 2008), deep water upwelling (Macario et al., 2016) and input from freshwater systems (Lougheed et al., 2013) will affect the ΔR of a given body of water. Across different freshwater systems, reservoir effects can be even more variable than those of marine systems (Philippsen, 2013). Freshwater reservoir effects are affected by the kind of freshwater system it is (lake or river), the size of the system and the underlying geology. Freshwater reservoir effects can be very large (Ascough et al., 2012), or completely absent (Paper-II; Svyatko et al., 2017). When selecting samples for measuring a reservoir effect, effort should be made to select material from a location-specific to the needs of the research.

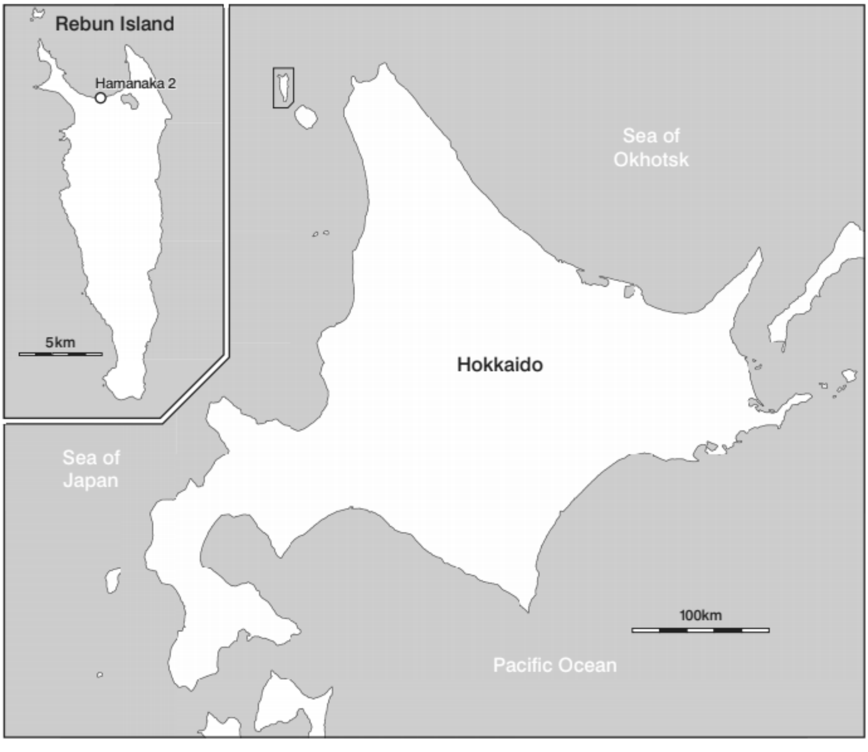


*Figure 14: The position of Hamanaka 2 relative to the Sea of Japan and the Sea of Okhotsk, redrawn from (Naito et al., 2010).*

The waters around Rebun Island off the coast of Hokkaido, Japan, perfectly illustrate the need to consider geographical reservoir effect variability. Situated on the north of Rebun island is the site of Hamanaka 2 (Paper I), an Okhotsk period site with a long history of coastal adapted cultures utilising marine food sources (Junno et al., 2020; Naito et al., 2010). It is therefore important to consider the variability of reservoir effects in this area before attempts are made to calibrate sampled human $^{14}C$ dates. The position of the site, however, means the groups who utilised marine resources would have been exposed to two very different marine reservoirs. This presents a problem for radiocarbon dating marine resources.

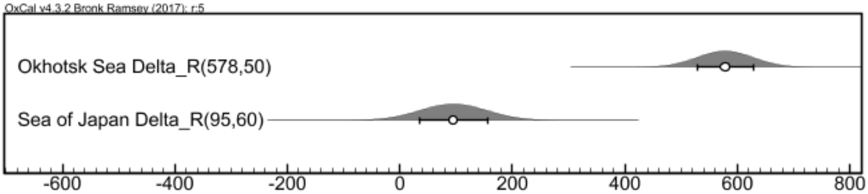
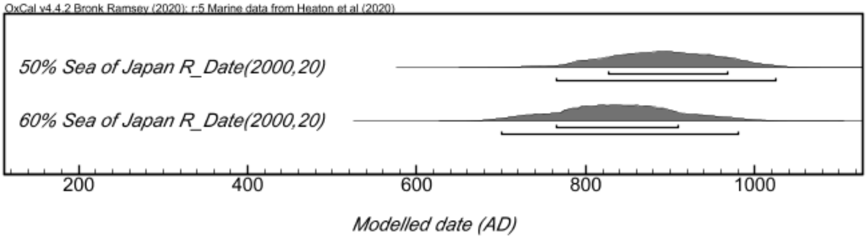


*Figure 15: OxCal probability distributions, mean value and standard deviations of ΔR values of the Okhotsk Sea and the Sea of Japan (Kuzmin et al., 2007).*

Figure 15 illustrates the difference between the two reservoir effects of the Seas of Japan and Okhotsk, as measured by Kuzmin et al (2007). The difference between the two ΔR values is roughly 500 years. Such a difference makes radiocarbon dating marine samples from the site of Hamanaka 2 difficult. Unless it can be known precisely how a marine sample relates to the two seas, large dating discrepancies can result. A spotted seal, for example, may have spent time feeding in both bodies of water, exposed to both reservoir effects.



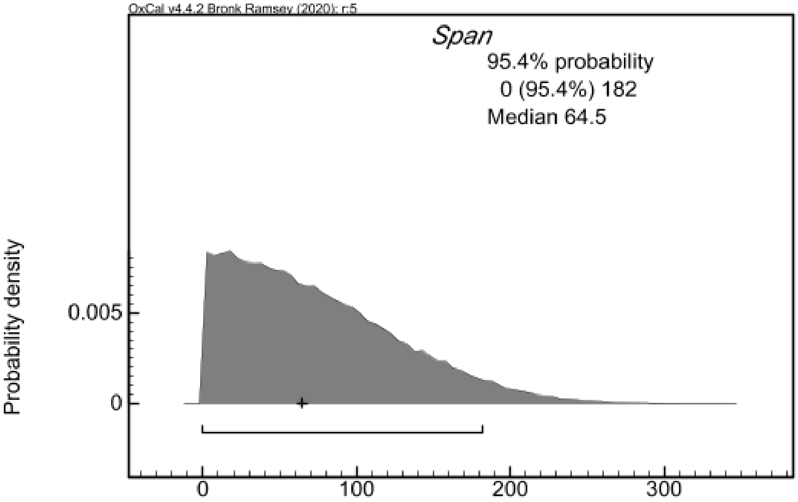


*Figure 16: Above, two identical $^{14}$C Dates (2000±20 BP) modelled assuming marine dietary input from the Sea of Japan (ΔR=95±60) of 50% and 60% respectively, the remaining dietary input coming from the Sea of Okhotsk (578±50). The 95.4% and 68.2% modelled date estimates are displayed as well as the median (+) and 95.4% difference (span) between the two estimates. Generated using OxCal.*

Figure 16 shows how a 10% discrepancy in $^{14}$C carbon sourcing can affect a single $^{14}$C date. A 10% difference in radiocarbon sourcing between the two dates, assuming no error, yields a difference between calibrated ages of up to 182 years (at 95.4% confidence), and a median difference of 65 years. Moreover, the model above assumes that there is no error attached to the marine contribution estimates. The compounding errors associated with marine and terrestrial mixing result in larger dating uncertainties, the error associated in mixing two very different marine carbon sources would be even larger.

The example of Rebun Island, positioned between two such different marine bodies, demonstrates the importance of considering geographic reservoir effect variation. Unless both seas are considered, the calibration of radiocarbon dates from humans and marine fauna from the site will suffer.

## 6.2 Reservoir Effects and Ecological Niches

The site of Hamanaka 2 illustrates how consideration of geographical ΔR variability could be important for accurate calibration of $^{14}$C dates. There are, however, other sources of reservoir effect variability, one of the most important for archaeological consideration being the species sampled. Different species interact with their environment in different ways. In many freshwater systems, there are aquatic species whose main dietary input has a terrestrial carbon source, such as frogs which mainly consume terrestrial insects. The size of their reservoir offset will differ, depending on the behaviour of the freshwater species. This is true for marine species also. Marine waters are not evenly mixed, so feeding depth can have a large impact on the ΔR of a marine species. For example, it may be expected that shellfish by the immediate coastline will have a smaller ΔR than shellfish of the same species in deeper ocean waters. Moreover, the openness of marine environments allows marine species lots of mobility. It would be reasonable to hypothesise that a marine organism moving between two different marine systems might have a different ΔR value to a more stationary species.

The reservoir effect variability that can exist between species is demonstrated by the radiocarbon dating of marine organisms from the archaeological site of Ekven in the Bering Strait (Paper III). The region is biodiverse and supports large populations of many different sea mammals, fish, shellfish and marine bird species. Ekven is a large burial site dating to the OBS period. A number of the excavated burials have yielded contemporaneous marine and terrestrial fauna. This was utilised for radiocarbon dating to determine their specific ΔR values.

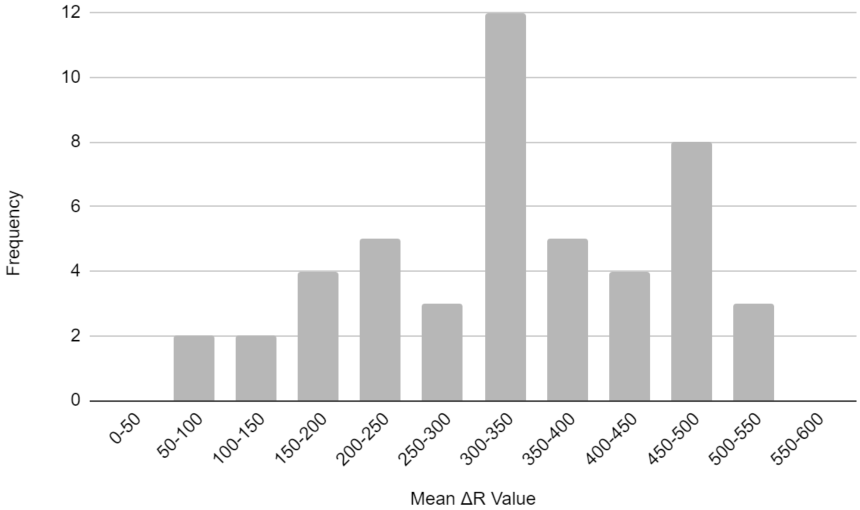


*Figure 17: Histogram showing ΔR frequency of all samples of known species from the Bering Strait (Paper III).*

Even with all species coming from the same body of water it should not be assumed that their ΔR values will be similar. The mobility of species between the Bering and Chukchi Seas and beyond, as well as different feeding habits, leads to a range in measured ΔR values. This is demonstrated in figure 17. Here the mean ΔR values have been organised into groups of 50-year intervals. Though this histogram considers all ΔR values regardless of species, the histogram may still be affected by sampling bias which may distort the spread of the data. The dataset, for example, considers more marine mammals than fish and shellfish. A Kolmogorov-Smirnov test yielded a test statistic of 0.08746 and a p-value of 0.82509. This indicated that the spread of the mean ΔR values does not differ significantly from a normally distributed dataset. This statistical test of course only considers the mean ΔR values.

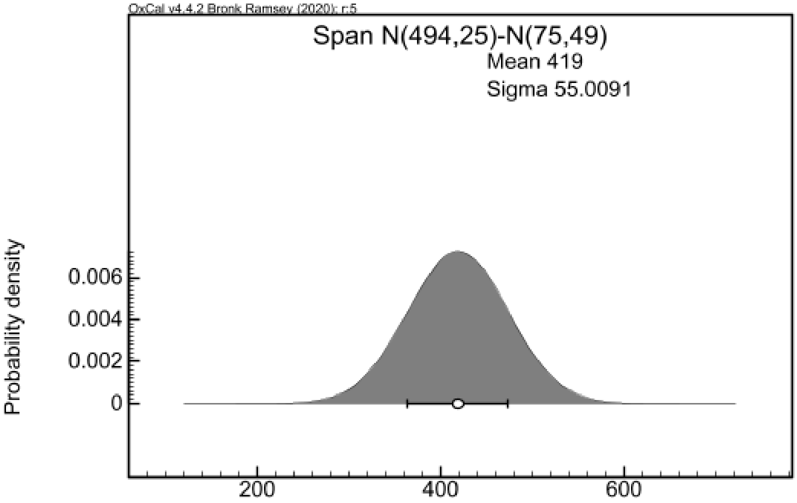


*Figure 18: The span of all known species ΔR values from the Bering Strait region, generated using OxCal.*

Figure 18, above, shows the span of the ΔR values from these known species from the Bering Strait region. The mean range of the ΔR values is 419±55 years. This is quite a high spread of ΔR estimates and highlights the importance of considering ΔR variability within a single body of water. Given the biodiversity of the region and the many factors that contribute to ΔR variability, where possible, the full range of ΔR values should be explored.

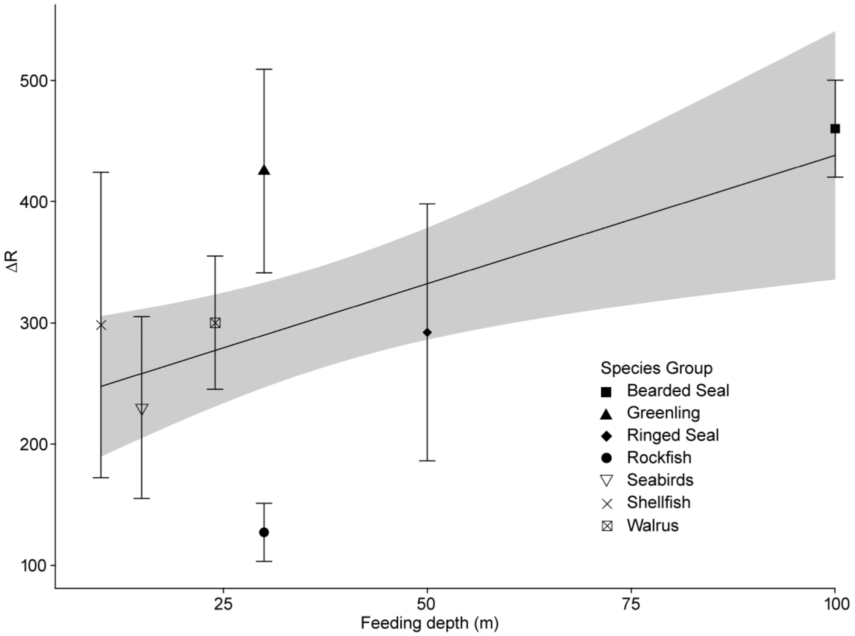


*Figure 19: ΔR values of Bering Strait species against feeding depth. The error bars and the shaded area around the regression line display uncertainties of 1 standard deviation (Paper III).*

The figure above (figure 19) compares the ΔR values of each marine species from the Bering Strait against their estimated feeding depth. An $R^2$ value of the regression line reveals that feeding depth alone can only explain around 40% of the ΔR variance, though there is a notable relationship between the two. Other factors will likely be influencing the ΔR of the sampled species, such as their migratory behaviour and the species they feed on. This case study demonstrates well the importance of considering which species should be used to calculate a reservoir effect. The calibration of seal-hunting humans, for example, would require an estimate of the scale and range of reservoir effects of local seals. Calibrating sampled human $^{14}C$ dates against local shoreline shellfish may not suffice. Where possible, researchers should attempt to source aquatic species that suits their specific research needs.

## 6.3 Methods of Estimation

There is no single method of calculating a reservoir effect in a sample. Most methods, however, rely on the comparison between the $^{14}$C date of an aquatic sample, and a second estimate of the sample's age. Different techniques, however, have different strengths and inherent weaknesses. The measurement of reservoir effects can be conducted in the following ways:

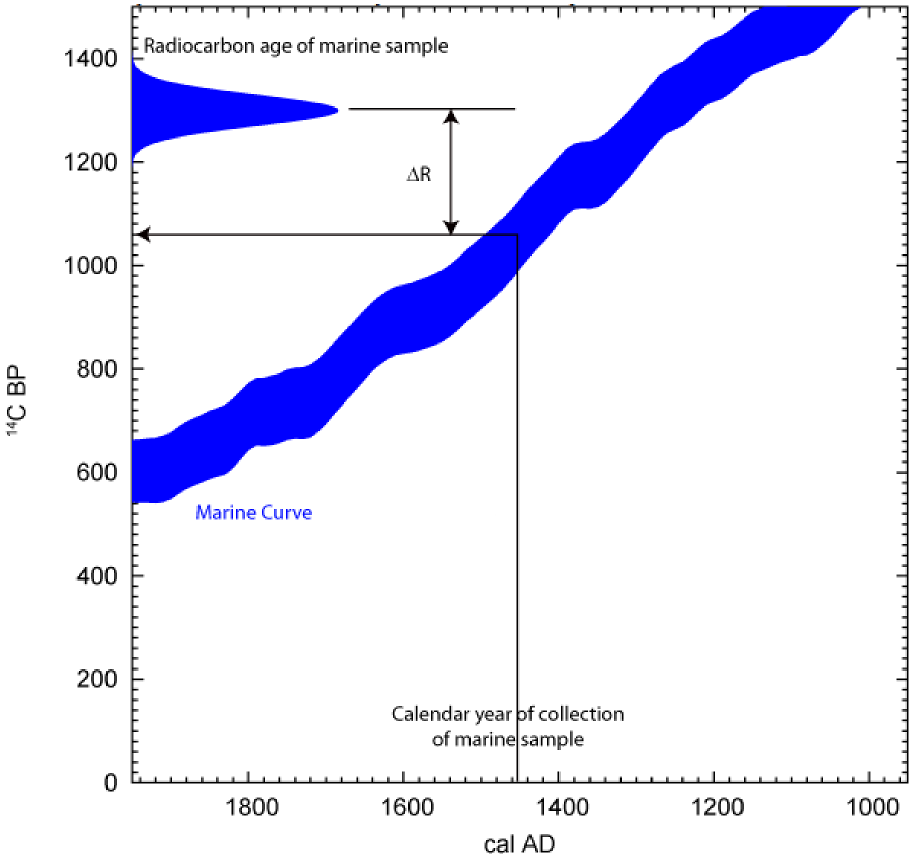


*Figure 20: ΔR calculation, using a marine sample of known date (14chrono Dataset) (Stuiver et al., 2015).*

One of the most frequently employed methods for measuring reservoir effects is to radiocarbon date aquatic samples of known age. This method of using samples with known collection dates results in accurate measurements of reservoir effects for that specific period. In figure 20, the difference between the $^{14}$C dates that would be expected of a sample of known age, and the measured $^{14}$C value, is the ΔR value. Freshwater reservoir effects can be calculated by comparison with the terrestrial curve. The radiocarbon dating of samples with known age is utilised in the $^{14}$Chrono database (Reimer and Reimer 2001) which calculates geographically specific ΔR values using marine samples of known age. As well as a recorded collection, the age of a sample may be known because it corresponds to a recorded historical event. The offset between the radiocarbon age of human remains in Herculaneum and the 79 AD eruption, for example, allowed for the calculation of the scale of their marine reservoir ages (Craig et al., 2013). This method of utilising known dates, however, does have its limitations. Firstly, this method relies somewhat on luck, that a past researcher or enthusiast, before atomic bomb testing around 1950, collected the sample and it was subsequently curated, or that an appropriate sample from a dated event is available for analysis. Secondly, that such a sample exists for the specific research area. Thirdly, that the sample is of a relevant species. Finally that the sample is contemporaneous to the period of interest (this is not likely for most archaeological purposes). The 'known age' method is best employed as a tool for calculating indicative reservoir effects. This may suffice for many research purposes.

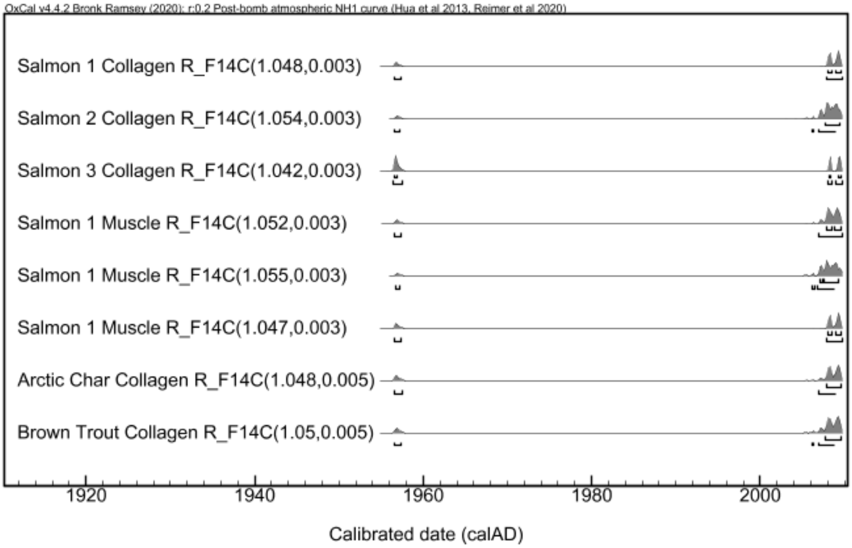


*Figure 21: OxCal Calibration of freshwater fish and salmon samples against the Bomb13-NH1 curve.*

To combat these weaknesses, modern samples can also be radiocarbon dated to calculate the scale of contemporary reservoir effects, a method with several advantages. Researchers can pick precise species for analysis from precise locations. Moreover, different tissues can be dated which might not survive in the archaeological record. Finally, samples do not suffer from poor preservation. These measurements can still be useful for archaeological purposes if it can be reasonably assumed that reservoir effects have not changed greatly over time in that area. Figure 21 shows the calibrated $^{14}$C dates of modern freshwater fishbone and muscle from sites in northern Fennoscandia (Paper-II). From these data, it is apparent that there is no reservoir effect in sampled material. The majority of the calibrated confidence intervals (at 95.4% and 68.2%) fall in a contemporary period when calibrated against the Bomb13-NH1 curve (Hua et al., 2013). Some of the calibrated dates fall before 1960, before the peak of the bomb spike. The samples in this example highlight an important point, that not all aquatic samples will carry a reservoir effect.

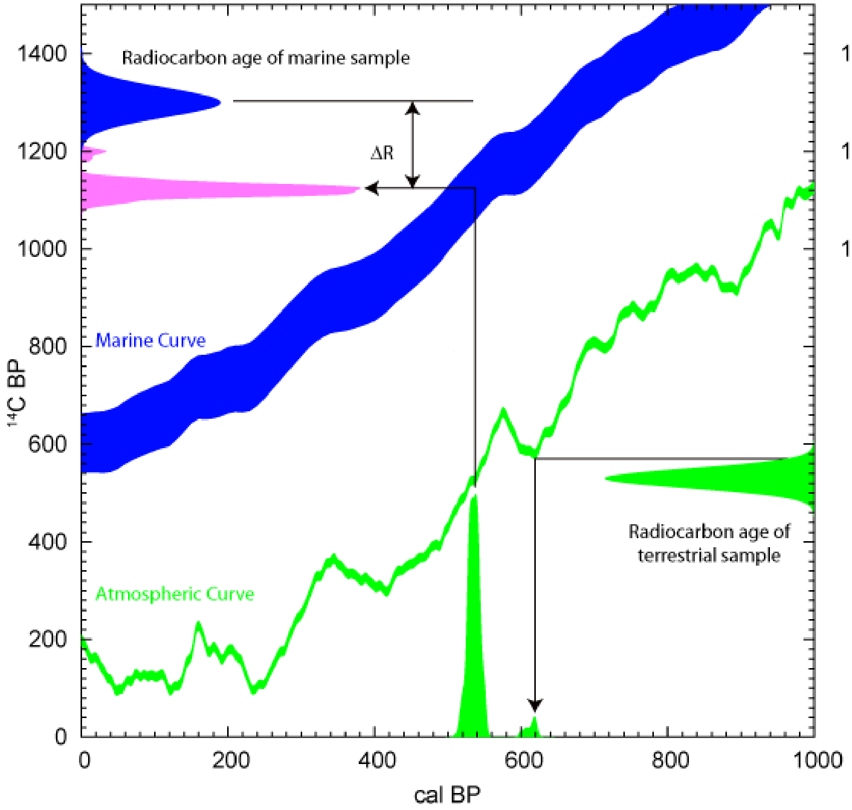


*Figure 22: ΔR calculation, using contemporaneous terrestrial dated sample (14Chrono dataset) (Stuiver et al., 2015).*

Similar to the ‘known-age method’, reservoir effects can be calculated by comparing terrestrial and aquatic samples of the same age. This method relies on securely closed contexts to ensure that samples are contemporaneous. Where this can be established, the calibrated age of the terrestrial sample is calibrated back to yield an expected $^{14}$C date for the aquatic sample. In figure 22 the difference between the expected and measured $^{14}$C value is taken to be the ΔR value. The measurement of freshwater reservoir effects would utilise the terrestrial curve to calculate the total reservoir effect. This method is limited compared to the ‘known age’ method because it relies on a calibrated terrestrial date defining the ‘true age’ of the sample. This being the case, more uncertainty is accrued (calibrated ages are probability distributions of possible ages, whereas collection year is a single value without error). This leads to a larger error calculating reservoir effects. Moreover, the reservoir effect calculated is only accurate if the contemporaneity of the aquatic and terrestrial samples is secure. Despite these limitations, this method is flexible. It allows for the calculation of temporal and species-specific reservoir effects in locations where no aquatic samples of known age have been curated. For this reason, it is the most commonly employed method of calculating reservoir effects in archaeological contexts.

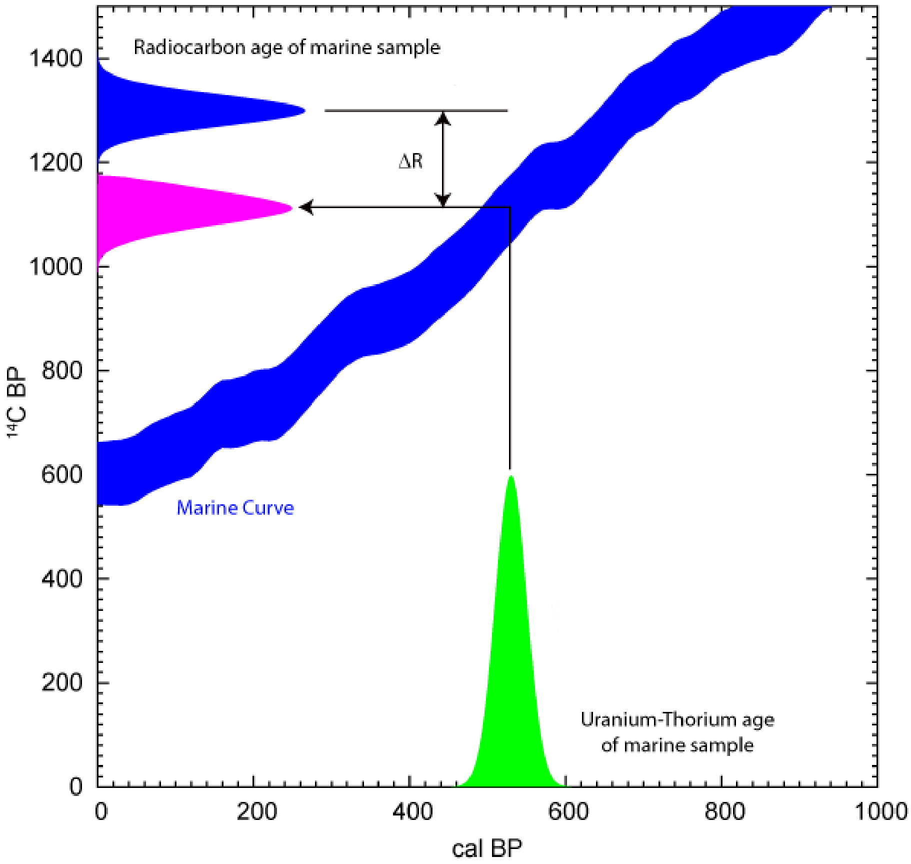


*Figure 23: ΔR calculation, through comparison of measured $^{14}$C date and Uranium-Throium date of a marine sample (14Chrono dataset) (Stuiver et al., 2015).*

Samples of interest will not, however, always come from closed or secure contexts and other methods of estimating reservoir effects have therefore been utilised. Reservoir effects can also be measured in a sample by comparing the measured radiocarbon date of the sample to the sample's calendar date, as measured by another independent direct dating method. The rationale of this method is that a second independent dating mechanism, which does not look at isotopes of carbon, will not be subject to the reservoir effects. This method has been utilised to allow for a reservoir effect to be calculated for a sample from any time period or geographical region, without having to resort to sample pairing (Hall and Henderson, 2001). This method, however, is subject to the accuracy and precision of the second independent dating method and any other weaknesses of the second dating method i.e. contamination and leaching (Alcaraz Pelegrina and Martínez-Aguirre, 2005) will affect the estimation of the reservoir effect.

These methods have been previously demonstrated to yield useful estimates of radiocarbon reservoir effects. This thesis will also present a novel method of estimating reservoir effects through dental wiggle matching. Wiggle matching is a kind of radiocarbon date modelling which uses known relationships between a series of $^{14}$C dates, matching them to the 'wiggles' of a calibration curve. Wiggle match models have been successfully used to reduce dating uncertainty of dendrochronological tree ring dates, sub-sampled marine shells and sediment cores. The known relationships between dated sub-sampled dental increments allow for a wiggle match model to be developed. For individuals with a marine (or freshwater) dietary input, it is hypothesised that there will be a narrow range of reservoir ages that will allow for the calibration of the specific sequence of dental sub-samples. This novel method of reservoir effect estimation was tested on a population from the Resmo tomb (Öland, Sweden) (Paper-V). The reservoir effect of the region had previously been estimated through more traditional sample pairing, yielding a ΔR value of -238±30 years. The model was built according to the following specifications to see if a similar reservoir effect could be determined.

1. All $^{14}$C dates were calibrated according to the estimated proportion of 'marine carbon' in the sample. All samples were calibrated against the IntCal20 curve and the Marine20 curve. An open ΔR correction was applied across all individuals in the model which assumed a ΔR value between -400 and +400 years.

2. For each individual, the date of death was estimated. The date of death was estimated by a date function, which is offset from the most recently formed dental increment. The offset is defined as the osteological age-at-death estimate (based on dental attrition and eruption), minus the formation age of the last increment. To give the model archaeological relevance, it was important to design the wiggle-match model to estimate the date of death, rather than the date the individual's dentine was formed.

3. The default resolution of an OxCal model is 5 years. The IntCal20 curve has a 1 year resolution between 0-5000 Cal BP. Due to the resolution the model demands, the resolution of the model was set to 1 year:

```
Options()
{
Resolution=1;
};
```

This has the effect of 'smoothing' the calibration curve so a resolution higher than the curve would otherwise allow can be considered.

4 From each tooth, the overall period of development was defined. For example, the $M_1$ molar of individual R20, the formation period of the tooth's growth would be defined as being 7±1 years using the following line of code:

```
Difference("Overlap", "R20M1four", "R20M1one", N(7,1));
```

The formation period of the tooth took into account the average biological growth period of the tooth in question (AlQahtani et al., 2010) and was adjusted for incomplete development or significant dental wear (resulting in individual R20 having a dental $M_1$ tooth representing about 7 years of growth). An example of this individual's significant dental wear can be seen in figure 24.

*Figure 24: Mandible of individual R20 from Resmo (Paper V), dentition showing signs of significant attrition.*

5 For certain individuals, multiple teeth were sub-sampled for dating. The order of the sub-samples can be modelled, as well as the relative order of the formation of the teeth. This could not be modelled in a simple sequence, however (e.g. molar $M_2$ does not start to form only when molar $M_1$ has finished forming). Different dental sequences

could be modelled as ‘overlapping sequences’. However, this allows for the possibility that the sequences overlap in such a way that molar $M_2$ would start forming before molar $M_1$, which is not the case (figure 25).

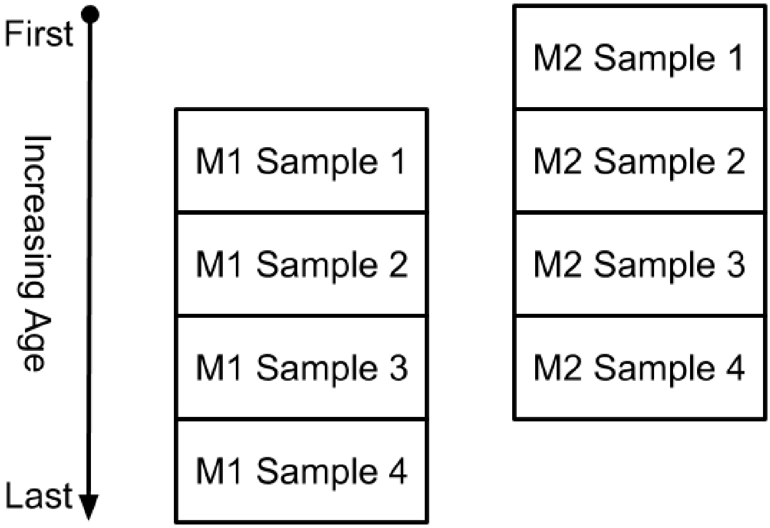


*Figure 25: Two overlapping dental sequences without priors that organise them into the correct order, resulting in erroneous sequences.*

Simply stating that the $M_1$ and $M_2$ sequences must overlap does not inform the direction of overlap. Such a parameter would allow for the $M_2$ tooth to appear to form before the $M_1$ tooth (as the figure above illustrates). Different ways of modelling the order of the teeth in such a way that the dates were consistent with dental formation were investigated. Two possibilities for modelling this specific overlap were tested. The first solution is the application of a ‘difference’ function which defines the direction of overlap of the start and end of two phases:

*Difference("Overlap", "Start M2", "End M1", U(0,1000));*

Such a line of code would ensure that the start of the formation of the $M_2$ molar must predate or equal the end of the formation of the $M_1$ molar (but not predate more than 1000 years). Several priors, such as these, successfully order the dental sequences.

A second solution (and arguably less complicated) is to build a ‘reference sequence’ within the OxCal model. The ‘start’ and ‘end’ boundaries of each dental sequence are built into this ‘reference sequence’ and as such must obey the rules of the sequence:

```
Sequence("Reference 1")
{
Date("=Start M1");
Date("=Start M2");
Date("=Start M3");
};
Sequence("Reference 2")
{
Date("=Start M2");
Date("=End M1");
};
Sequence("Reference 3")
{
Date("=Start M3");
Date("=End M2");
};
Sequence("Reference 4")
{
Date("=End M1");
Date("=End M2");
Date("=End M3");
};
```

The above code ensures that the calibration of the $^{14}$C dates obeys the rules of dental formation, that $M_1$ dentine begins to be formed before $M_2$ dentine and $M_3$ dentine, and that the $M_1$ molar will have finished forming before the $M_2$ molar. Although both methods work perfectly well, the reference sequence method was selected to keep the model and script as simple as possible. Moreover, upon visual inspection, it is easier to understand and edit the priors which have been applied to the $^{14}$C data. A series of shorter reference sequences were scripted instead of a single reference sequence, as shown below:

```
Sequence("Reference 1")
{
Date("=Start M1");
Date("=Start M2");
Date("=Start M3");
Date("=End M1");
Date("=End M2");
Date("=ENd M3");
```

*};*

This reference sequence (above) forces there to be an overlap in the formation period of the $M_1$ and the $M_3$ molars. To allow more flexibility in the calibration of the sub-sampled dental $^{14}C$ dates, this single reference sequence was not built into the model. The separate reference sequences allows for an overlap in the formation period of $M_1$ and $M_3$ molars but does not require there to be an overlap.

6 An outlier model was applied to all samples to identify any samples with a poor statistical fit within the model concerning the modelled priors.

A visual schematic of the OxCal model can be found below:

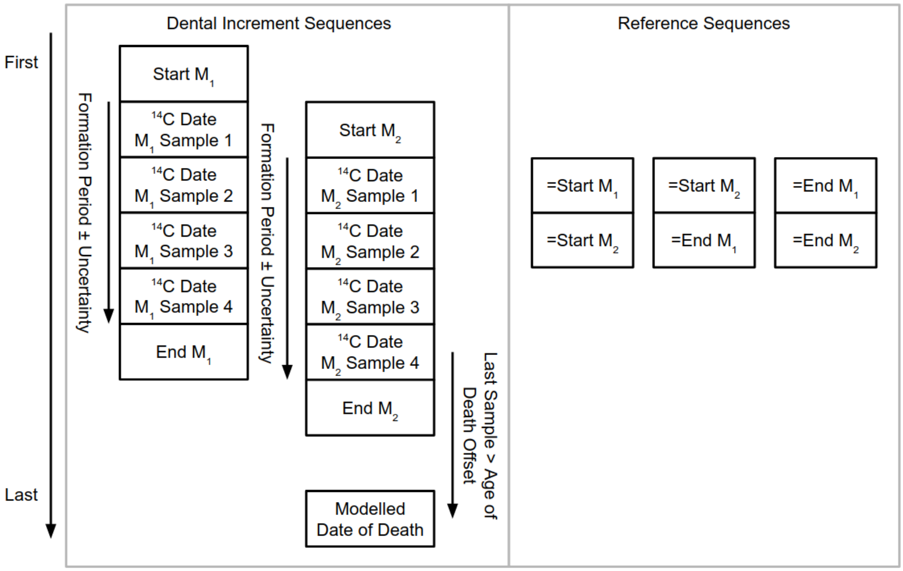


*Figure 26: Schematic representation of the dental wiggle-match calibration model for an individual with sampled M1 and M2 molars.*

The full code for the model can be found in the appendix of Paper-V. The measured reservoir effect of the Baltic is 162±30 years (Paper V) when comparing Gotland humans to terrestrial fauna (their % marine contribution being taken into account). This is equivalent to a ΔR value of -238±30 years. When the $^{14}$C dates are modelled in one phase, together, sharing a single ΔR function, the modelled value (-275±29) is very close to the measured reservoir effect (figure 27). This demonstrates that there may be some utility for the model to estimate reservoir effects acting on individuals where no marine or terrestrial samples are available for comparison. This will be further examined in future research with individuals with a known date of death, or narrow date of death range.

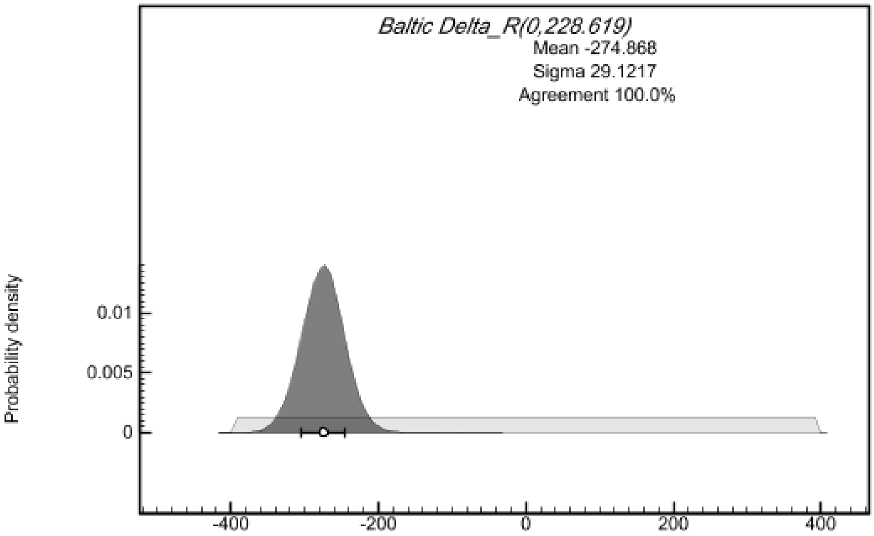


*Figure 27: Baltic ΔR mean and standard deviation as calculated by the OxCal dental wiggle match model.*

## 6.4 Reservoir Effects in the Bering Strait

As discussed in this chapter, there are several sources of reservoir effect variability. Moreover, the method of measuring reservoir effects can affect the calculated value. When measuring a reservoir effect, researchers need to ask what it is they are measuring and for what purposes. Thought needs to be given to the kind of samples being selected for analysis and the location from which those samples came. Furthermore, the relative strengths and weaknesses of each method of estimating the ΔR value of a sample should be considered before proceeding with sampling.

*Figure 28: Summary of published ΔR values from the Bering Strait region (data from Mason and Rasic 2019). Shaded box represents 1σ, whisker represents 2σ, the central line represents mean ΔR value.*

The data in figure 28 are taken from Mason and Rasic (2019) and demonstrate how different researchers have utilised different methods to calculate the ΔR of the Bering Strait region. It demonstrates that, from a single area, very different ΔR values can be calculated to define a given region's reservoir age.

The authors listed have employed different sampling strategies, where there are differences in the comparative terrestrial material used to calculate ΔR values, what the authors consider appropriate 'contemporaneity' between samples, and the marine species considered. Given these different approaches, the ΔR estimates vary in terms of both their mean estimates and the standard deviations around those mean estimates. This highlights the importance of considering how exactly a ΔR value is being calculated, and for what purpose it is being calculated. Slight deviations in the methods employed can lead to very different estimates. Measuring reservoir effects is not a question of finding the true reservoir effect of an area, or of a species, but rather of calculating values specific to the aims of the research

# 7 Reservoir Effects and Complex Diets

Where a plant will acquire all of its $^{14}C$ from $CO_2$, all animals acquire their $^{14}C$ from the foods consumed. A seal, for example, will only consume marine resources, so its $^{14}C/^{12}C$ ratios will reflect this. A deer will eat terrestrial plants, so all of its carbon will be acquired indirectly from the atmosphere. Diets and radiocarbon dating must therefore be considered together. Humans, and some other animals, can have quite complicated diets, often utilising resources from freshwater, marine and terrestrial systems. Moreover, humans transport and trade foods, meaning their dietary inputs can be more unpredictable. With complex mixed diets, carbon could be sourced from several different geographic systems. This is important for properly understanding and modelling radiocarbon dates. The marine, freshwater and terrestrial pools of radiocarbon can be quite different in their $^{14}C/^{12}C$ ratios. Because a consumer's tissues carbon comes from the foods consumed, the proportions of foods from different reservoirs must be calculated. This must be taken into account when interpreting their $^{14}C$ dates.

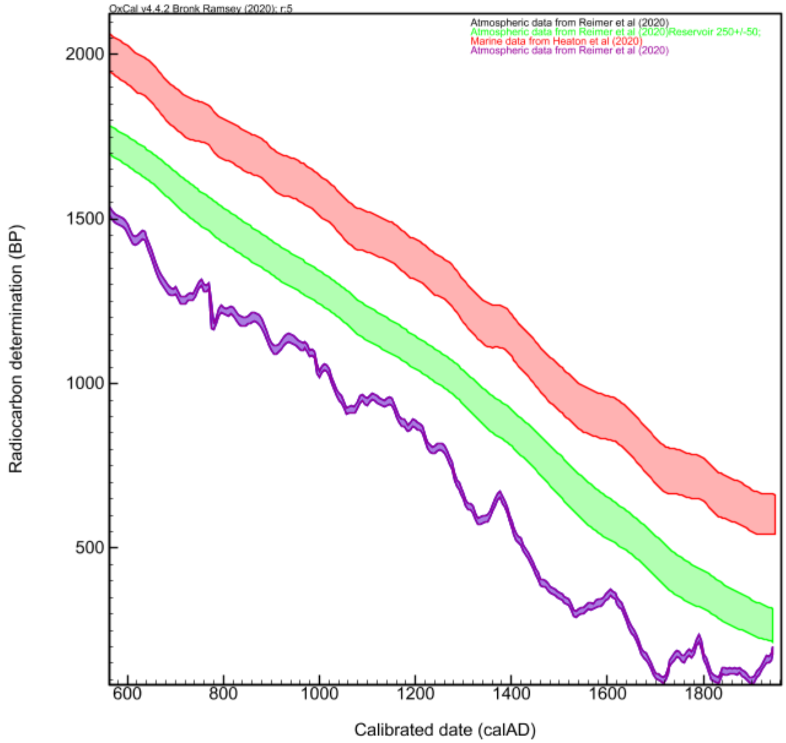


*Figure 29: Hypothetical calibration curves for terrestrial carbon (Purple, IntCal20), freshwater carbon (Green, IntCal20 + 250±50) and Marine carbon (Red, Marine20).*

The importance of this is illustrated in figure 29 above, where marine, terrestrial and a hypothetical freshwater reservoir-adjusted terrestrial curves have been plotted. A complex mixed diet incorporating foods from multiple carbon sources would need careful modelling, with a mixed curve calibration of their $^{14}C$ dates. Diet should be considered to understand radiocarbon dates, however, radiocarbon is not a particularly good isotope for dietary reconstruction as it is unstable. Time elapsed since death and diet both influence $^{14}C/^{12}C$ ratios. It is difficult to tell based on $^{14}C$ alone if a sampled consumer with a mixed marine/terrestrial diet, for example, has a low $^{14}C/^{12}C$ ratio because the sample is old or because their marine dietary component is high. Therefore, other dietary indicators must be used to estimate the sources of $^{14}C$.

## 7.1 Stable Isotope Analysis

Because they do not decay, measurements of stable isotopes are reliable indicators of what was consumed, and by extension, from which sources a consumer's tissues radiocarbon originated. Several samples can be considered for analysis including bone collagen, dental apatite (Lee-Thorp et al., 1989), dentine collagen (van der Sluis et al., 2015) as well as nail and hair keratin (Cameron et al., 2017; O'Connell et al., 2001). Most commonly, for analysis of this material, measurements of $\delta^{13}$C, $\delta^{15}$N and $\delta^{34}$S isotopes are used to indicate the source of dietary protein sources (marine versus terrestrial versus freshwater). Other isotopic measurements, such as δD (Reynard and Hedges, 2008) are occasionally used. Stable isotopic values are expressed as *δ* values, expressing the sample's measured ratio of two isotopes relative to a standard ratio, for example:

$$\delta^{13}C = \frac{(^{13}C\,/\,^{12}C)\ Sample}{(^{13}C\,/\,^{12}C)\ Standard} - 1\ (\times\ 1000‰)$$

$$\delta^{15}N = \frac{(^{15}N\,/\,^{14}N)\ Sample}{(^{15}N\,/\,^{14}N)\ Standard} - 1\ (\times\ 1000‰)$$

$$\delta^{34}S = \frac{(^{34}S\,/\,^{32}S)\ Sample}{(^{34}S\,/\,^{32}S)\ Standard} - 1\ (\times\ 1000‰)$$

The principle utilised by stable isotopic dietary reconstructions is that a consumer's tissue isotopic signal will reflect the average isotopic signal of the organisms they consumed plus a fractionation offset. Fractionation offsets occur between food and consumer. This is because lighter isotopes are more likely to be involved in biological processes, leading to an accumulation of heavier isotopes up the food chain. For dietary modelling, these offsets often describe the difference between the same tissue of prey and consumer. For example, the $\delta^{13}$C offset between the collagen of prey and the collagen of a consumer is typically 1-2‰ (Schoeninger and DeNiro, 1984; Lee-Thorp et al., 1989; Ambrose and Norr, 1993; Fernandes et al., 2012) and 3-5‰ for $\delta^{15}$N (O'Connell et al., 2001). The best dietary resolution can be achieved by considering multiple isotopes of many samples (Britton et al., 2018; Bonsall et al., 2015; Hesslein et al., 1993; Sayle et al., 2014).

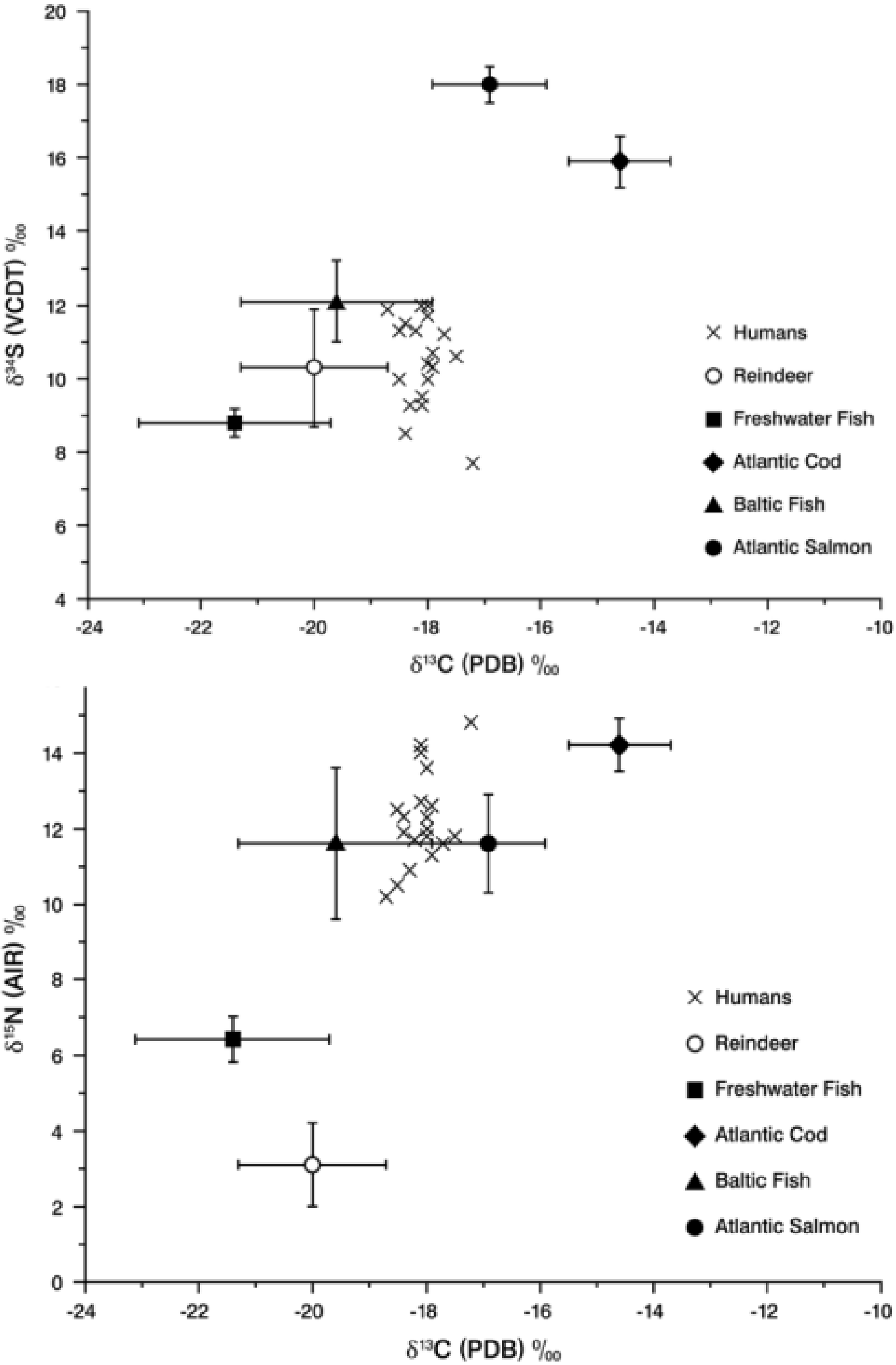


*Figure 30: Probability distribution plots summarising modelled Atlantic marine and Baltic marine contributions to the diets of the Rounala individuals (Paper-II) based on $\delta^{34}$S, $\delta^{13}$C and $\delta^{15}$N values.*

Figure 30 shows stable carbon, nitrogen and sulphur isotope measurements from human and faunal remains from the cemetery at Rounala church (Paper-II). Understanding that isotopic fractionation occurs, it can be seen from figure 30 that the humans are consuming a mix of protein sources. Their isotopic measurements do not demonstrate that one food group was dominant. If these humans were to be properly radiocarbon dated, their $^{14}C$ dates would therefore be calibrated against both marine and terrestrial calibration curves, as no reservoir age was measured in the freshwater samples (Paper-II). Accurate modelling, however, demands a numerical estimate of this dietary mixing. This is difficult to achieve by visually inspecting the graphs above.

A linear endpoint model measures the isotopic contribution of two different dietary sources to a consumer's tissues. A linear endpoint model is appropriate when there are only two main dietary sources, e.g. $C_3$ vs. $C_4$ protein or marine vs. terrestrial protein. Linear endpoint models can focus on several different dietary isotopic indicators. However, when considering diets for radiocarbon calibration, assuming there is no dietary input from $C_4$ plants, $\delta^{13}C$ isotopes are most appropriate. A hypothetical 100% marine diet and a hypothetical 100% terrestrial diet are estimated. These are calculated by establishing $\delta^{13}C$ values for local marine and terrestrial fauna with the addition of a dietary fractionation offset. The percentage marine contribution can be calculated using the formula below, this formula can be adapted for $^{15}N$ isotopes or different dietary contributions:

$$\%\ \text{Marine Protein Contribution} = \left( \frac{(\delta^{13}C_{\text{Terrestrial}} + \text{Offset}) - (\delta^{13}C_{\text{Sample}})}{\delta^{13}C_{\text{Marine}} + \text{Offset}) - (\delta^{13}C_{\text{Terrestrial}} + \text{Offset})} \right) \times 100$$

Linear endpoint models are best considered visually. Figure 31 displays the intersection between a hypothetical consumer's isotopic value ($\delta^{13}C = -17‰$), and the regression line between the two dietary endpoints (terrestrial $\delta^{13}C = -22‰$ and marine $\delta^{13}C = -13‰$). This results in an estimate of the individual's marine protein contribution. The linear endpoint model below, figure 31, is designed to highlight the importance of considering error in dietary estimates. Error is compounded and associated with the sample measurement, the fractionation offset uncertainty and the standard deviation of the two dietary end members. Here, considering all sources of error, the 1σ marine protein contribution range is between 40-73%. The probability distribution of the marine dietary contribution estimate, however, cannot be calculated in a simple linear endpoint model.

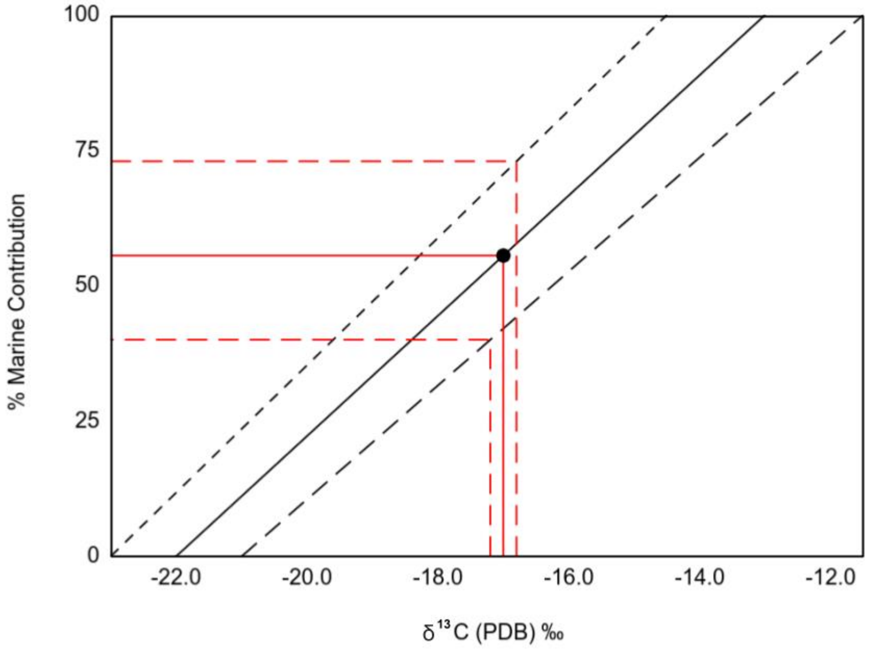
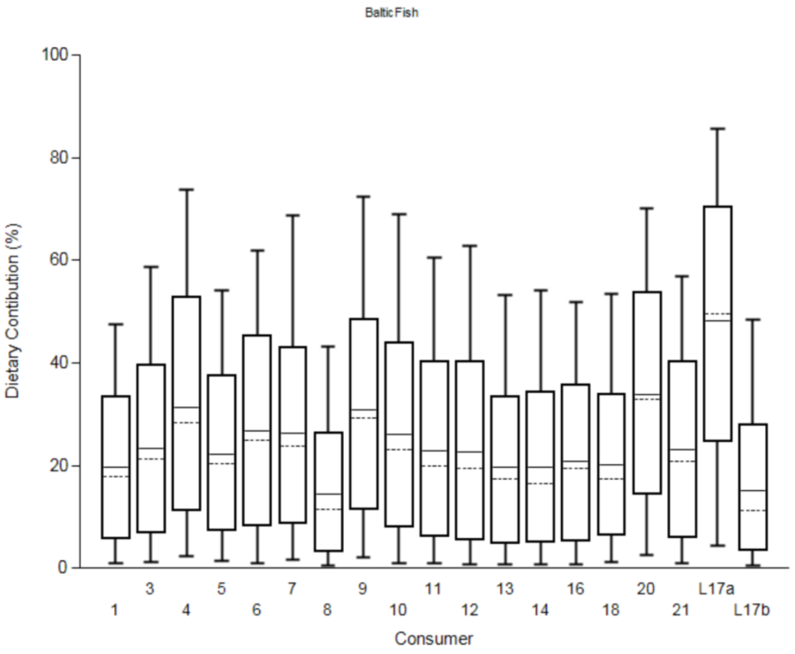


*Figure 31: Linear regression line (black line) between 100% marine and 100% terrestrial diets. The red line represents a consumer's δ$^{13}$C value. The point which they intersect (black circle) indicates the consumer's marine dietary contribution. Dashed lines either side of the regression line (black) and consumer line (red) represent 1sd.*

Such a linear endpoint model may not be appropriate for estimating the marine dietary input for the Rounala humans (fig 30). This is because estimates for freshwater, Baltic and Atlantic fish (all carrying different reservoir ages) would have to be calculated. A 2 point linear endpoint model would not provide this dietary resolution. Multi-source mixing models, such as FRUITS (Fernandes et al., 2014) can be used to help model past diets, taking into account several sources of error. FRUITS, and other mixing models, such as SIAR (Parnell and Jackson, 2013), can also consider several isotopic indicators, and more than two dietary sources.



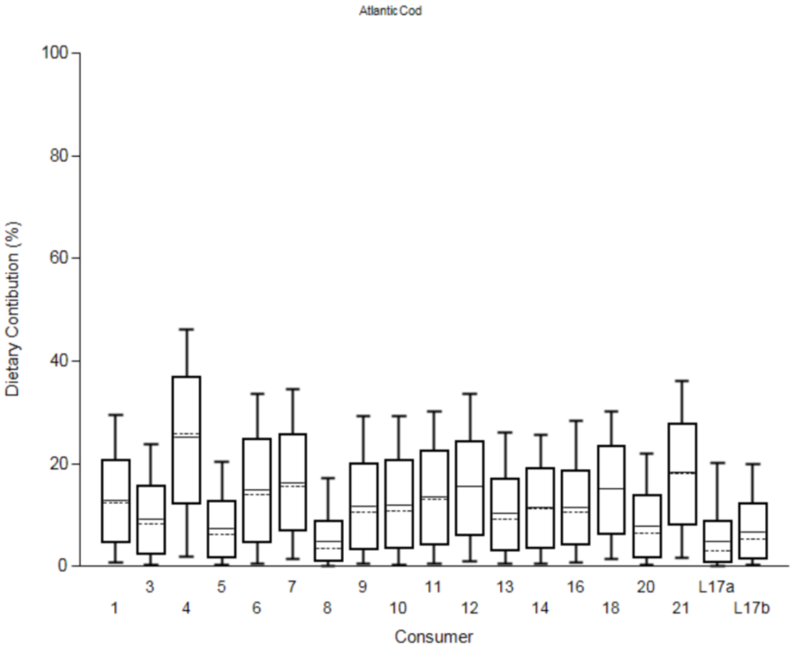


*Figure 32: Probability distribution plots summarising modelled Atlantic marine and Baltic marine contributions to the diets of the Rounala individuals (Paper-II) based on $\delta^{34}S$, $\delta^{13}C$ and $\delta^{15}N$ values.*

By considering all isotopic signals available ($\delta^{34}S$, $\delta^{13}C$ and $\delta^{15}N$) the possible contributions of the different food sources to the Rounala human's diets were calculated (figure 32). These contribution estimates have a lower uncertainty than if $\delta^{13}C$ and $\delta^{15}N$ were considered without $\delta^{34}S$. Given the number of different aquatic food sources being considered, the resolution added by this third isotope is useful. The uncertainty of the estimates for Atlantic cod consumption is reasonably small. Estimates for the consumption of Baltic resources, however, are much larger. Based on the isotopic data available, and the complex diets of the people of northern Fennoscandia, this is the best possible numerical estimate of dietary contributions available.

## 7.2 Dating Implications

The modelling of a consumer's $^{14}C$ dates is tied very closely with the diet of that individual. Depending on the proportions of foods carrying a reservoir age, there could be large differences in the modelled age of that sample. With modelled estimates of a human's diet, it is then possible to calculate to what extent a reservoir effect should be applied when modelling $^{14}C$ dates.

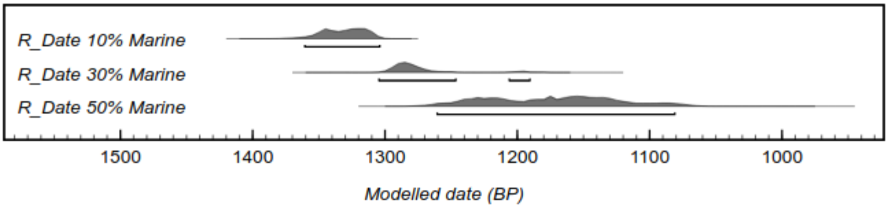


*Figure 33: OxCal calibration of three samples with conventional radiocarbon dates of 1500±15 BP assuming marine carbon inputs of 10, 30 and 50% and assuming a ΔR value of 0±0.*

The example (figure 33) demonstrates the importance of understanding dietary inputs when calibrating conventional radiocarbon dates. Considering a single conventional date (1500±15 BP) calibrated multiple times (assuming 10%, 30% and 50% marine carbon inputs, respectively) reveals large differences in modelled date ranges. The calibrated dates of the 10% and 50% marine dates do not overlap at 95.4% probability. This particular example assumes that the marine reservoir effect matches the Marine20 curve (a ΔR value of 0). The Rounala data illustrate a difficult case where calculating what was consumed was key because the different food groups consumed carry very different reservoir ages. Estimates for two different marine food sources (Baltic and Atlantic fish) were calculated.

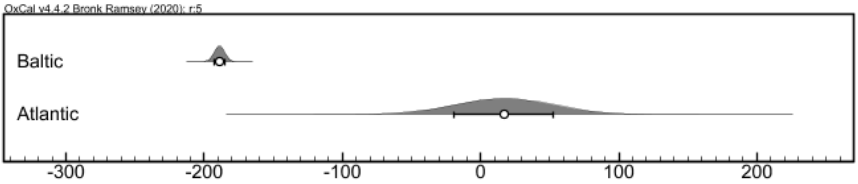


*Figure 34: Baltic and Atlantic ΔR values, as calculated using the Chrono marine $^{14}$C database.*

The estimated ΔR of samples from these two bodies of water were calculated based on data from the $^{14}$Chrono database (Reimer and Reimer 2001). The modelling of the Rounala human's mixed marine diets (figure 34), however, allows for the calculation of an individual's specific reservoir age.

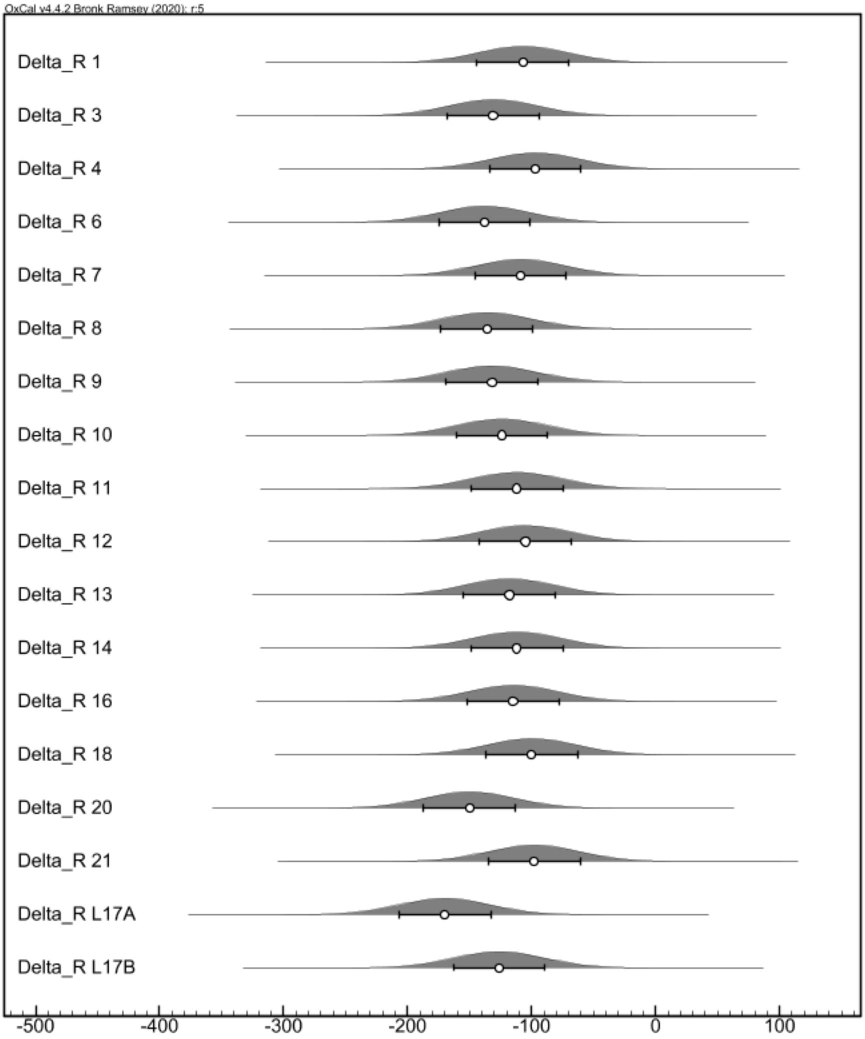


*Figure 35: Weighted ΔR (Marine13) values for the 18 Rounala humans.*

The ΔR values for each Rounala consumer are expressed in figure 35. The ΔR values have been calculated by weighting the two different ΔR values of the Baltic and Atlantic (figure 35) according to the individual's dietary inputs. These ΔR values allowed for the accurate modelling of these sampled humans' $^{14}$C dates. The case study of the Rounala humans is particularly difficult because the uncertainty of their calibrated dates was compounded by error in the $^{14}$C measurement, the dietary estimate and the ΔR estimates. This, however, is preferable to calibrating the sampled human $^{14}$C dates against only a Baltic adjusted marine curve, or an Atlantic adjusted marine curve.

Understanding the dietary complexities allows for the best possible estimate of a sample's date of death.

When a sample is known to have sourced its carbon from a single source, i.e the ocean or atmosphere, the calibration of that sample's $^{14}C$ date is straightforward. Not all samples' carbon, however, come from a single source. Pottery lipids and food crusts, for example, may reflect the many different foodstuffs (marine and terrestrial) cooked or held in that pot. Other substances, such as glues, lamp oils and pigments may have been made from materials routing their carbon from different reservoirs. Archaeologists must give this careful consideration. Most frequently, the topic of carbon source mixing is discussed concerning humans' diets. For thousands of years, humans have utilised marine and freshwater resources, several populations being reliant, either entirely or in part, on aquatic foods. The utilisation of different foods from different resources adds to societal stability and reduces the chances of food shortages. The effect of consuming a mix of foods with different carbon sources, however, is the acquiring of a reservoir age. This reservoir age will be a fraction, or average, of the dietary resources consumed. This requires an understanding of the reservoir ages which could affect a consumer and the dietary choices of that consumer. The tool most frequently employed by archaeologists attempting to estimate dietary contributions is stable isotope analysis. This particular method of dietary reconstruction lends itself well to radiocarbon dating as $^{14}C$ is routed to a consumer's tissues from their dietary inputs. The Rounala project demonstrates a method of dealing with reservoir effects in humans with mixed diets, utilising multiple isotopic signals of diet. This has previously been recognised as an important factor in the accurate calibration of other humans with complex mixed diets (Sayle et al., 2016). By considering stable isotopic indicators of dietary mixing, an estimate of bone collagen carbon sourcing was estimated. This illustrates the need for researchers to consider carefully food procurement of any humans being investigated. Though fractionation of $^{14}C$, relative to $^{12}C$ and $^{13}C$, is accounted for at the measurement of a conventional radiocarbon age, the calibration and modelling of that conventional date does require an understanding of diet. Humans trade food over long distances and are themselves quite mobile. Many radiocarbon laboratories now operate dual AMS, IRMS systems, routinely offering IRMS measurements of $^{13}C$ and $^{15}N$ (Aerts-Bijma et al., 2001). This must be considered before the sample is discussed in terms of its calibrated age.

# 8 Reducing Reservoir-Effect Related Uncertainty

Typically, when defining reservoir effects there is an error associated with that estimate. Concerning human's diets, there will be further errors associated with the estimate of the proportion of aquatic foods consumed. These, combined with the $^{14}C$ date error and calibration uncertainty, can lead to less precise date ranges in marine samples than terrestrial samples. In certain circumstances, the reduction of that compounded uncertainty is very important. Fortunately, the use of Bayesian statistics in radiocarbon dating has been used to reduce uncertainty greatly. When a sample is recovered from an archaeological context, its relationship with other samples can be used as prior factors to constrain possible calibration solutions and reduce overall dating error. Calibration software, such as OxCal, was designed for this purpose (Bronk Ramsey, 1995). Moreover, modelling large numbers of radiocarbon dates together can help reduce dating uncertainty. The dating of marine and terrestrial material together, from closed contexts, benefits greatly from this approach.

Sample ordering is the most simple and effective application of Bayesian modelling. Basic stratigraphy, for example, can provide useful information for reducing the uncertainty of calibrated radiocarbon dates. In figure 36, three hypothetical sample's $^{14}C$ dates have been calibrated with their relative stratigraphic order modelled. The samples have been labelled in terms of their depth '2 m, 1.5 m and 1 m', describing their stratigraphic position. In this example, the unmodelled sample from the bottom of the stratigraphic sequences (2 m) appears to date to a more recent period than the central sample (1.5 m). The stratigraphic sequence should not allow this. The modelled distributions (dark grey) take this prior information into account, correctly ordering the dates and reducing uncertainty.

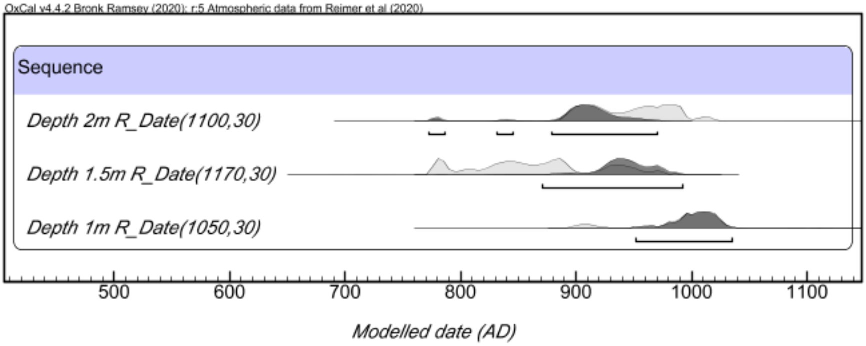


*Figure 36: OxCal calibrated hypothetical dates for three samples (named according to their depths) modelled in sequence (light grey probability distribution representing the calibrated un-modelled date, the dark grey distribution representing the calibrated modelled date).*

Bayesian radiocarbon models, in many forms, have proven to be useful. Long and complex stratigraphic sequences can be built, considering samples from several distinct stratigraphic layers (Batt et al., 2015; Sayle et al., 2016; Schmid et al., 2018), and complex relationships between sub-samples of a single sample can also be used to reduce overall dating accuracy (Helama and Hood, 2011). These models can be used in circumstances where greater dating resolution is required than would otherwise be achieved by calibrating individual samples. The need for greater dating resolution was identified for the radiocarbon dating of human remains from the Ekven burial site (paper IV). The site belongs to the old Bering Sea (OBS) culture, ancestral to the Thule culture. The origins of the OBS have remained somewhat vague, however, with the precise timing cultural shifts around the Bering Strait being debated. The application of a complex Bayesian model to this problem is a good illustration of the power of modelling to reduce dating uncertainty.

In the calibration of the human samples $^{14}$C dates from Ekven, large uncertainties were expected. This was because of the large reservoir ages of the marine fauna at the site, and its variability (Paper III). The OBS reliance on marine resources meant it was expected that the sampled human's $^{14}$C dates would be calibrated almost entirely against a marine curve. When dealing with humans who have consumed aquatic resources yielding large reservoir ages, it is very important to accurately estimate the proportion of dietary carbon from different sources. The average ΔR of the marine mammals they most likely consumed has been measured as 303±98.

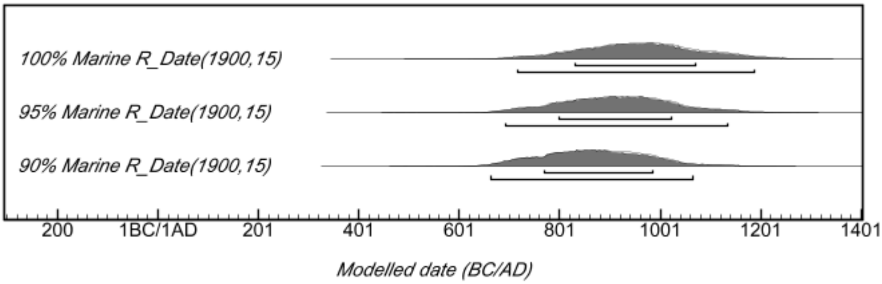


*Figure 37: A $^{14}$C date of 1900±15 BP calibrated assuming a marine carbon contribution of 100, 95 and 90% and a ΔR value of 303±98.*

Making sure that the dietary estimates of the humans are accurate was very important in this study. With the ΔR value being so high, slight differences in the dietary estimate could result in large discrepancies in the calibrated dates of the consumers. Figure 37 shows a single $^{14}$C date of 1900±15 modelled assuming a marine dietary contribution of 90%, 95% and 100%. The difference between these diets is only 10%, but the mean span of these estimates is 134 years (figure 38).

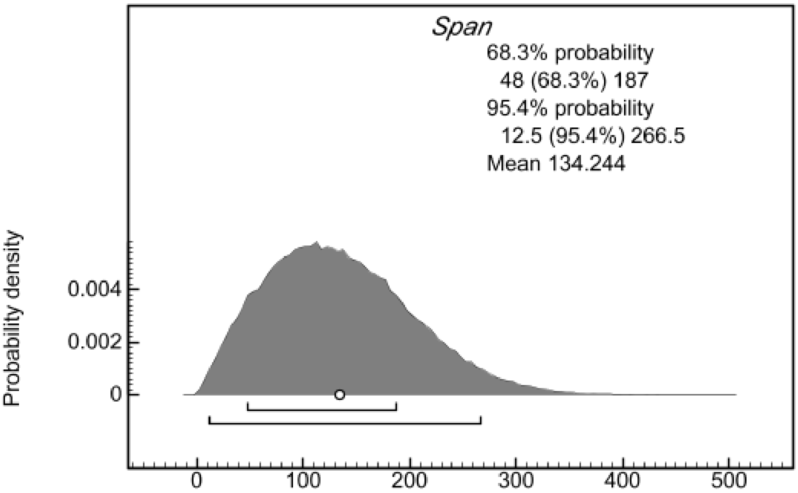


*Figure 38: OxCal Probability distribution estimating the span of the dates in years (100%, 95% and 90%).*

Stable isotope analysis of the human remains indicate that their diet was entirely marine, however, the terrestrial faunal remains at the site remind us that foods other than marine foods were consumed. FRUITS modelling was used to estimate the dietary inputs of the human samples from the site, with all being modelled as having a very large marine input. The precise estimates were incorporated into an OxCal model. To reduce the uncertainty of the humans' calibrated dates as much as possible, further priors were added to constrain the modelled dates. Any stratigraphic information available was incorporated into the model. Any graves which overlapped, or cut into adjacent graves, were modelled as sequences. This provided a clear order of burial, the humans in the disturbed graves must be older than the humans in the undisturbed graves. The calibration of the human's $^{14}$C dates from these grave contexts can be seen in figure 39. Stratigraphic priors have been useful in reducing calibration uncertainty in these individuals. Further such priors would be useful for other individuals in other burial contexts.

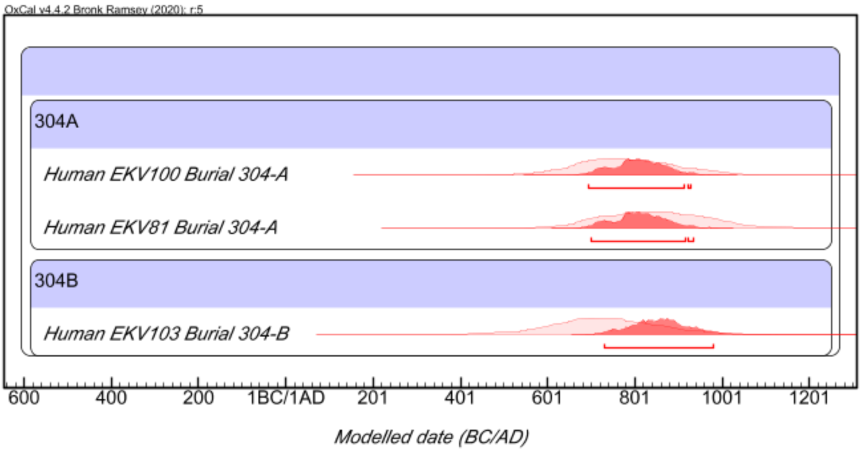


*Figure 39: OxCal calibration of Ekven sampled human $^{14}$C dates from burials 304-A and 304-B, where stratigraphic priors are available. EKV100 is a R_Combine of two $^{14}$C dates.*

Uncertainty has also been successfully reduced by considering the samples' $^{14}$C dates within overlapping 'phases'. The phases (preceded by a 'start' and 'end' boundary function) are formed of samples which share a relationship, in this case, coming from the same grave context. The figure below (figure 40) demonstrates how, by considering samples within a single phase, a mixed consumers' modelled calibrated date range can be altered relative to the $^{14}$C date's unmodelled calibration. When calibrated within the context of a model, sharing a phase with terrestrial samples, the modelled ages of the two human samples are greatly reduced relative to their unmodelled dates. The utility of such a function can be seen across many of the burials at Ekven. It is clear that the human samples calibrated with other fauna, marine and terrestrial, have benefited from being considered within a single 'phase'.

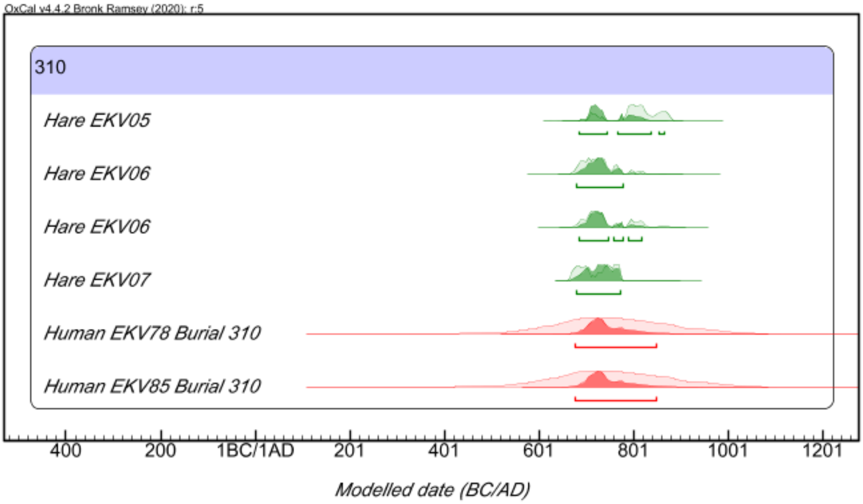


*Figure 40: OxCal calibration of $^{14}$C dates from Ekven burial 310.*

The radiocarbon dating of material carrying a reservoir age results in added uncertainty. If a marine sample (for example) is calibrated entirely against the Marine20 calibration curve, its calibrated date range is likely to be wider than that of a contemporaneous terrestrial sample. This is because the Marine20 curve is a modelled curve, based on the terrestrial curve, and carries with it further uncertainty. Even more uncertainty is compounded if a sample has sourced its carbon from multiple sources. The larger uncertainties associated with dating reservoir affected material can, however, be mitigated. By considering reservoir affected $^{14}$C dates within the context of Bayesian models, uncertainty can be reduced. This can come in the form of sample pairing or stratigraphic sequencing. The Ekven dataset demonstrates the utility of large Bayesian modelling of $^{14}$C dates to better understand the use of a site. Sampled human radiocarbon dates are constrained, allowing for more archaeologically useful estimates of the dates of the burial events. Samples which are found in isolation will usually not be able to benefit from the application of such models, however, where sample contexts were recorded during excavation, and the funds are available to sample several different remains, the construction of Bayesian dating models is useful. If outlier analysis functions are paid attention to, these models also have the added benefit of informing the researcher if estimates of the reservoir age applied to the model are correct. When data are considered together, greater overall resolution can be achieved and archaeological conclusions are more secure.

# 9 Making Use of Reservoir Effects

In most circumstances, reservoir effects are considered a hindrance to accurate radiocarbon dating. At worst, reservoir effects are described as a 'problem' associated with radiocarbon dating. Because $^{14}C/^{12}C$ ratios are not homogenous globally and differ between environments, reservoir effects should be accepted as a fact of radiocarbon dating. However, there are cases where the reservoir effect itself is the tool/phenomenon needed to answer other archaeological questions. Whereas most research is made more complex by the consideration of reservoir effects, the existence of reservoir effects has allowed other research to take place. The difference in the radiocarbon abundances allowing for archaeological questions to be answered. 'Dealing with reservoir effects' need not mean mitigating their effects, but could involve their utility.

Reservoir effects have occasionally been used to answer questions of historical diets. For example, a hypothetical human with a $^{14}C$ date of 2000±15 BP has a known date of 400 AD. The human' $^{14}C$ date is modelled against the terrestrial IntCal20 curve and the Marine20 curve, the mixing between the two is stated to be any value between 0-100%. Because the date of death is fixed, the only flexibility the model allows is in the mixing of the two curves. The figure below (41) shows that the mean % marine contribution is 69±9%. This example assumes that there is no $\Delta R$ correction to the marine curve. Such a correction, which would carry uncertainty, would likely result in a wider uncertainty of the marine contribution estimate. The resolution of this as a dating estimate may be improved by considering other isotopic indicators which could apply a further before the modelled curve mixing.

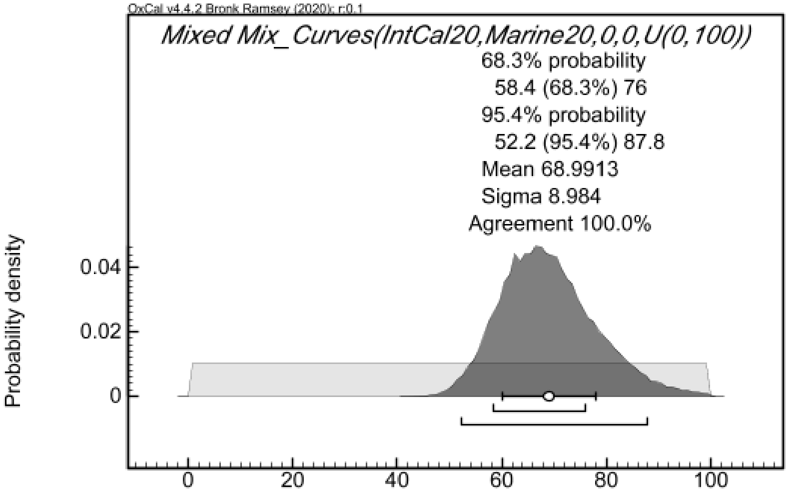


*Figure 41: OxCal estimate of the percentage contribution of marine carbon to a hypothetical human calculated using their known date of death of 400 AD and a $^{14}C$ date of 2000±15 BP.*

Though this application, described above, is limited to samples with a known date of death, there have been some instances where it has been applied. The offset between the radiocarbon age of a sampling of the human remains at Herculaneum and the known date, 79 AD, of the eruption event, allowed for the calculation of the scale of marine reservoir ages of the sampled individuals, and also the marine contribution towards their diets (Craig et al., 2013). This led to the conclusion that for every 1‰ increase in $\delta^{13}C$ and $\delta^{15}N$ values, 56 years and 34 years, respectively, were added to the sample's radiocarbon age.

The utility of $^{14}C$ as a dietary indicator, however, is not limited to individuals with known dates of death. Associated terrestrial material can act as an indicator of the individual's calendar date of death. This, of course, carries more uncertainty than if the precise date of death was known. The difference between the two dates can help identify individuals who have consumed resources carrying a reservoir age. This principle was used to identify freshwater resource consumption among ancient individuals in Germany (Fernandes et al., 2016) where traditional stable isotopic methods had underestimated their consumption.

Outside of archaeology, the variability in $^{14}C$ measurement across different sources has also allowed for the examination of ocean cycling (Guilderson et al., 2000; Levin and Hesshaimer, 2000), using radiocarbon as a tracer to differentiate between different bodies of water. Radiocarbon reservoir effects have also been utilised as a natural tracer of ancient $^{14}C$ depleted permafrost melting across the arctic (Hugelius et al., 2010; Walter Anthony et al., 2018;

Wild et al., 2019). Differences in radiocarbon age have also allowed for the study of soil turnover (Braakhekke et al., 2014). In forensic medicine, the bomb pulse has been a useful indicator of sample age (Buchholz and Spalding, 2010; Kondo-Nakamura et al., 2011; Taylor et al., 1989).

There are a small number of instances where radiocarbon reservoir effects may be of use to researchers. These are mainly to identify aquatic resource consumption and learn more about the dietary routing of isotopes. Sample pairing can, however, be used when precise dates of death are elusive. Although this use of radiocarbon dating is not applicable in all situations, it should be remembered that reservoir effects (although problematic in some instances) are not inherently, or by definition, a problem. They are a fact of environmental radiocarbon distribution and can be used to the advantage of the researcher. This is clear when we consider their use in investigating ocean cycling.

# 10 Conclusion

Archaeology is about understanding human diversity and change throughout time. To do this, researchers need accurate chronological frameworks. As a result, radiocarbon dating is perhaps the most important scientific development in the field of archaeology. The emergence of radiocarbon dating as a research tool has allowed researchers to build accurate absolute chronologies and to order events and processes. When reflecting on the development of archaeology as a discipline, it is commonly referred to as the radiocarbon revolution. Radiocarbon dating has been instrumental in allowing for the understanding of key archaeological transitions, such as the end of the Last Glacial Period, the peopling of the Americas, as well as the onset and spread of the Neolithic from the Levant. For the first time, previously impossible research about cultural changes being correlated with climate shifts, and comparisons of technological revolutions between disparate global societies could be had. As a technique, it has become increasingly cost-effective and can be applied to a plethora of organic samples. In recent years there have been promising developments allowing for increased confidence in the dating of older material; AMS machines are allowing the for improved dating of older samples with lower $^{14}$C concentrations (Ding et al., 2019) and the IntCal20 and Marine20 curves have been extended from 50ka (Reimer et al., 2013) to 55ka (Heaton et al., 2020; Reimer et al., 2020). There is also an increasingly powerful case to be made for the radiocarbon dating of early modern material (Thompson et al., 2019) as the process of modelling allows for increasingly useful date estimates. The discovery that radiocarbon in some aquatic systems was not equivalent to terrestrial systems prompted a significant amount of research. We now have a much better understanding of carbon cycles, radiocarbon formation and improved computer modelling, allowing for a much better understanding of $^{14}$C dates, particularly $^{14}$C dates from aquatic samples.

In section 1.1, the aims of this thesis were outlined, designed to explain the current state of archaeological understanding of reservoir effects and how best they can be mitigated for the benefit of a more accurate building of past chronologies.

The first question asked how reservoir effects best be managed to increase dating accuracy for human populations with complex diets. This thesis has made clear the strong relationships between diet and the radiocarbon content of a human's tissues. With the potential for large differences between the

$^{14}C/^{12}C$ ratios of aquatic and terrestrial foodstuff, having accurate estimates of what was consumed is key. This can be provided by the archaeological record, with zoological and archaeobotanical data providing clues. There are, however, uncertainties associated with the archaeological record and what was consumed. Many foods will not survive as well in the archaeological record, for instance plant food. Moreover, the processing of some food items away from a site may leave their consumption archaeologically invisible. Finally, the links between what foods were consumed and the specific nutrients becoming incorporated into the bodies tissues are complex. Chapter 7 discusses in detail how stable isotope analysis can be used to address point 1. The isotopic ratios of different elements in a consumer's body tissues are a time capsule, allowing for dietary reconstructions. The case study of the Rounala humans illustrates the need for complex diets to be explored so accurate radiocarbon dating is possible.

The second question asked how reservoir effects can be estimated. There is no single way of defining the reservoir age of a sample, though most methods rely on comparing a radiocarbon date of a marine sample, with a second estimate of the sample's age (using a method where reservoir effects are not an issue). Various methods of estimating reservoir effects have been detailed in Chapter 6. Each method has its limitations. Sample pairing, though it is one of the most applied methods, is subject to the assumed contemporaneity of the terrestrial and aquatic samples being archaeologically solid. As demonstrated in Paper III, outlier analysis can be used to identify samples which may not be suitable for sample pairing and reservoir effect calculations. This relies on several radiocarbon dates being considered together and may also have the effect of omitting samples which have reservoir ages which naturally differ from the rest of a group. Other dating methods could be applied to a sample (e.g. U-Th dating) but higher uncertainties associated with other methods of dating would be applied to the calculation of radiocarbon reservoir effects. Modern samples, or samples with known collection dates, yield the most accurate reservoir effect estimates. Archaeologists, however, often require more geographically, temporally and taxonomically specific reservoir effects than historical prebomb collections can offer. Chapter 5 and Paper V also detail a novel method of reservoir-effect estimation using radiocarbon wiggle matching of subsampled dentine. Archaeologists need to think carefully about which method to employ to best suit their research purposes.

Question three asked how varied can reservoir effects within a single aquatic system be and how does this affect populations utilising resources from those systems. This thesis also demonstrates the importance of considering the variation in reservoir effects from a single environment. This has been previously illustrated across temporarily, geographically and ecologically diverse cases (Russell et al., 2011; Hua et al., 2015; Lougheed et al., 2013). Paper IV illustrates how the species available to marine hunters at Ekven can vary in terms of their marine reservoir effects. Any measurement of any single species would not yield a reservoir age sufficient to calibrate the $^{14}$C dates of sampled humans remains from the site. Researchers attempting to understand the $^{14}$C dates of humans should consider the reservoir effects of the precise species consumed to yield the most accurate modelled dates. At Ekven, stable isotope analysis did not provide the resolution required to suggest which particular marine species may have been the most important to the humans' diets. However, archaeological evidence points towards a culture whose economy was focused on pinniped hunting. Though the modelling of radiocarbon dates requires statistical thought, the power of archaeological data to help focus Bayesian dating models, however, should not be forgotten.

The fourth question asked how Bayesian modelling could be used to mitigate the consequences of reservoir effects and reduce radiocarbon dating uncertainty. Bayesian modelling has proven to be an ideal statistical tool for radiocarbon dating. Complex models can account for many sources of error in radiocarbon dating, but also prior information concerning samples which can help reduce this uncertainty. Chapter 8 discusses how such models can be built, bringing together archaeological knowledge concerning the samples and their $^{14}$C dates. Papers I, IV and V show how complex relationships between samples can be built into OxCal models as priors. Calibration solutions which do not adhere to these priors are discounted, and in theory, overall dating uncertainty reduced.

Finally, the fifth question asked how reservoir effects could be used as a tool to help answer other research questions. Reservoir effects are a fact of carbon cycling which does not undermine the principle of radiocarbon dating. To describe reservoir effects as a 'problem' with radiocarbon dating is a misunderstanding. Consideration of reservoir effects does, however, generally increase dating uncertainty. As described in this thesis, however, it is quite possible to measure and mitigate reservoir effects. Radiocarbon is not distributed evenly globally, and though this can be an added dating consideration, it can also be a tool for certain research questions. In limited circumstances, radiocarbon can be used as a dietary proxy or used to investigate global ocean circulation.

The seas, lakes and rivers have allowed for travel and, perhaps most importantly, the provision of food. Entire cultures have depended on the nutrition aquatic foods provide. With the trade of these foods and the mobility of people, it is possible to find reservoir effects in human remains and other materials at archaeological sites, regardless of their geographic origin. This being the case the radiocarbon dating and research into aquatic material will continue. The five papers which make up this PhD demonstrate the variability of reservoir effects, illustrate how Bayesian modelling can be utilised to reduce dating uncertainty, and describe the relationships between complex human interactions with aquatic systems. These papers act as a summary of approaches to 'deal' effectively with reservoir effects in archaeological contexts.

# 11 Sammanfattning

I kapitel 1 presenteras syftet med den här avhandlingen, att belysa de utmaningar som arkeologer ställs inför vid $^{14}$C-datering av prover påverkade av reservoareffekter från havs- och sötvattensmiljöer. Variationen och komplexiteten hos reservoareffekter kan leda till ökad osäkerhet i $^{14}$C-dateringarna, eller i värsta fall helt felaktiga tolkningar. Trots de potentiella problem som reservoareffekter kan orsaka, är det fortfarande viktigt för arkeologer att kunna datera prover från både marina och sötvattensmiljöer. Vatten är livsviktigt för människan; hav, sjöar och vattendrag tillhandahåller mat och råvaror, och möjliggör handel och transport. På grund av människors mobilitet och handel är akvatiska resurser viktiga också för dem som inte lever i direkt anslutning till en strand. Det bör därför aldrig förutsättas att det inte finns akvatiskt påverkade prover på en arkeologisk lokal. Även om reservoareffekter kan leda till felaktiga dateringar, eller dateringar med större osäkerhet, så finns det sätt att minska den negativa påverkan. Denna avhandling tar upp befintliga och nya metoder för att hantera och korrigera för reservoareffekter genom följande frågeställningar:

1. För mänskliga populationer med komplexa dieter, hur kan reservoareffekter bäst hanteras för att öka noggrannheten i dateringen?

2. Hur uppskattas och beräknas storleken på reservoareffekter?

3. Hur varierande kan reservoareffekter inom enskilda vattensystem vara och hur påverkar detta populationer som använder resurser från dessa system?

4. Hur kan bayesiansk modellering användas för att korrigera för reservoareffekter och minska osäkerheten i $^{14}$C-dateringar?

5. Kan reservoareffekter användas som ett verktyg för att svara på frågor inom andra vetenskapsområden?

I kapitel 2 förklaras de grundläggande principerna för $^{14}$C-datering. Eftersom $^{14}$C är en instabil (radioaktiv) isotop av kol, med känd halveringstid, kan den användas för att beräkna ålder på ett prov. $^{14}$C-datering är allmänt tillämpligt som dateringsmetod eftersom alla levande organismer innehåller kol. Med en praktisk dateringsgräns på 55.000 år täcker denna dateringsmetod dessutom hela människans historia och stora delar av förhistorien.

$^{14}$C-datering är dock en process snarare än bara en mätning. För att omvandla en mätning av $^{14}$C-halten till ett årtal måste $^{14}$C-dateringen kalibreras. Under denna process justeras mätningar av $^{14}$C mot en kalibreringskurva och detaljerna i denna process beskrivs i kapitel 4. Kalibreringskurvor konstrueras med hjälp av $^{14}$C-dateringar av material vars kalenderålder är känd eller mycket exakt modellerad. Kalibreringsprocessen är nödvändig eftersom mängden $^{14}$C som produceras i atmosfären har fluktuerat över tiden. Det är nödvändigt att veta hur mycket $^{14}$C som ursprungligen fanns i ett prov.

Val av lämplig kalibreringskurva är viktigt, eftersom $^{14}$C inte är jämnt fördelat över jordklotet. På grund av att kol kan lagras i djupa hav under lång tid, finns det vanligtvis mindre $^{14}$C i haven än i atmosfären (där $^{14}$C produceras). På samma sätt kan det i en del sötvattensmiljöer finnas en lägre halt $^{14}$C än i atmosfären, på grund av införlivandet av äldre kol från olika delar av avrinningsområdet. I kapitel 3 diskuteras dessa fenomen, kända som reservoareffekter. Det första steget för att hantera reservoareffekter är att förstå att de finns och att välja en lämplig kalibreringskurva. $^{14}$C-datering av marint material kräver en marin kalibreringskurva och kalibrering av sötvattensmaterial kan kräva en reservoarjusterad atmosfärisk kalibreringskurva (det finns ingen specifik kalibreringskurva för sötvatten). På grund av variationen i reservoareffekter kanske valet av en lämplig kurva inte är tillräckligt; mer arbete krävs för att säkerställa korrekt $^{14}$C-datering. Det går att hantera reservoareffekter på flera olika sätt. Exempel på olika metoder som arkeologer kan använda för att arbeta med reservoareffekter beskrivs i kapitel 5-9.

Det enklaste sättet att hantera reservoareffekter och att helt undvika påverkan från dem, är att inte datera akvatiska prover. Om den arkeologiska frågeställningen kan besvaras genom att ta prover från terrestriskt material, kan det vara det lättaste sättet. Denna strategi tillämpades i Paper I vid lokalen Hamanaka 2 (Rebun Island, Japan), tillhörig sen jomonkultur. Ön Rebun är omgiven av två olika hav, med två olika reservoareffekter, vilket utgör ett problem för få en korrekt $^{14}$C-datering av marint material. Om den specifika arkeologiska frågeställningen inte kräver $^{14}$C-datering av marint material och kan besvaras via $^{14}$C-datering av terrestriskt material (som inte påverkats av någon reservoareffekt) är det lämpligt att undvika datering av marint material. Vid Hamanaka 2 var målet att datera de olika kulturlagren närmare genom $^{14}$C-datering. Detta uppnåddes mer precist genom datering av fossila växtrester än genom datering av marina material. Som förklaras i kapitel 5 måste dock arkeologerna vara säkra på att det de daterar faktiskt är terrestriskt. Ett oidentifierat skelettben kan till exempel bära på en reservoareffekt.

Det mest omedelbara sättet att hantera reservoareffekter på är att mäta reservoaråldern så exakt som möjligt. En korrekt uppskattning av storleken på en reservoarålder möjliggör bästa möjliga kalibrering av andra marina material. Kapitel 6 diskuterar olika metoder för att uppskatta reservoaråldern hos ett prov. Den vanligaste metoden för uppskattning av reservoaråldern är genom parvis datering av två samtida prov – ett terrestriskt och ett marint. Dateringen av terrestriska prover kräver ingen korrigering för reservoareffekter. Om ett terrestriskt och ett marint prov kan visas komma från samma slutna kontext, kan de paras ihop och den kalibrerade uppskattningen av dateringen för det terrestriska provet anses utgöra kalenderåldern för det marina provet. Denna metod användes på lokalen Ekven på Tjuktjerhalvön (Chukotka) i Ryssland vid Berings sund (Paper III). Denna uppskattning av kalenderåldern kan användas för att beräkna storleksordningen på reservoaråldern. Om det terrestriska provet är olämpligt för $^{14}$C-datering, kan en annan dateringsmetod (t.ex. U-Th-datering) användas för att uppskatta provets kalenderålder. Noggrannheten i dessa uppskattningar beror på upplösningen hos den andra dateringsmetoden. De mest exakta mätningarna av reservoarålder kommer från marint material med känd ålder. Om ett prov från en levande organism samlats in och året registrerats, eller om provet kommer från en historisk kontext med känd ålder, kommer den enda källan till mätosäkerhet i reservoaråldern från $^{14}$C-dateringen. I denna avhandling presenteras också en ny metod för att uppskatta reservoareffekter med hjälp av en modell för *wiggle match*-datering av tänder, från gånggriften i Resmo (Öland, Sverige) (Paper V). Genom att dela upp tänderna i mindre delar och sedan $^{14}$C-datera varje delprov, gick det att få fram en noggrann uppskattning av den lokala reservoareffekten i denna del av Östersjön under yngre stenålder. Denna modell har stor potential att utvecklas till ett mer rutinmässigt redskap för att uppskatta reservoareffekten.

Det är också viktigt att mäta hur mycket reservoareffekten varierar inom en och samma kontext.. Beräkningen av en enskild reservoarålder genom parvis datering (eller annan metod) avslöjar kanske inte hela intervallet av reservoaråldrar som kan ha påverkat ett prov. Från Ekven parades marina och terrestriska prover från slutna gravkontexter ihop för $^{14}$C-datering (Paper III). Eftersom de var från samma grav kunde proverna antas ha samma kalenderålder. Här påvisades att reservoareffekten varierade stort mellan marina arter från samma havsområde. Det var ett viktigt resultat, eftersom det sannolikt inte är tillräckligt med ett enda värde på reservoaråldern, baserat på en art, för att modellera $^{14}$C-dateringar på människor med komplexa marina dieter. Olika faktorer såsom artens ekologiska nisch (kapitel 6.2) och den exakta geografiska platsen för ett prov (kapitel 6.1) kan påverka storleken på reservoaråldern. Att förstå hur reservoareffekter varierar är således ett bra sätt att hantera dem.

Det är ofta nödvändigt för arkeologer att hantera reservoareffekter genom att analysera människors komplexa dieter. När terrestrisk föda konsumeras tillsammans med en eller flera akvatiska födoämnen är det viktigt att förstå från vilka olika källor $^{14}$C härrör; vikten av detta förklaras i kapitel 7. Stabila isotopanalyser av kollagen i humana ben (och andra vävnader) kan användas för att dra slutsatser om vad en människa kan ha konsumerat. Beräkningar av andelen sötvattens- och marina proteinkällor användes för att $^{14}$C-datera mänskliga kvarlevor från kyrkogården vid Rounala kyrka (Lappland, Sverige) (Paper II). Det påvisades att kyrkogården troligen var i bruk redan innan kyrkan uppfördes. Dessa slutsatser var endast möjliga att dra genom att först analysera människornas diet och sedan modellera $^{14}$C-dateringarna utifrån de reservoareffekter som påverkades av dieten.

Även om man beräknar andelen marin kost och mäter variationen i lokala reservoareffekter kan det ändå vara svårt att få en precis $^{14}$C-datering. I kapitel 8 förklaras hur bayesiansk modellering kan användas för att minska osäkerheten i beräkningen av reservoareffekter. Provernas arkeologiska kontext och deras inbördes stratigrafiska relationer kan användas för att avgränsa dateringarna. Detta är principen bakom modelleringsverktyg som OxCal. Endast kalibreringslösningar som överensstämmer med den arkeologiska kontexten beaktas i modellerade dateringsintervall. På detta sätt kunde de beräknade dateringarna av gravarna i Ekven avgränsas (Paper IV). Information om gravarnas stratigrafiska relationer och om vilka individer som begravts tillsammans i samma grav införlivades i modellen. Detta minskade kraftigt osäkerheten i de beräknade dateringarna jämfört med icke-modellerade beräkningar. Användandet av OxCal och andra modelleringsprogram är utmärkta sätt att hantera reservoareffekter när det finns kontextuell information tillgänglig.

Som framgår av kapitel 9, är det viktigt att komma ihåg att reservoareffekter inte i sig är att betrakta som ett problem med $^{14}$C-datering. Även om de kan orsaka problem för arkeologer som hoppas kunna få exakta dateringar, är de bara ett resultat av kolets kretslopp. Reservoareffekter kan ibland användas till arkeologens fördel, till exempel genom att skatta den marina andelen av dieten hos människor med känt dödsdatum. Reservoareffekter har också använts för att studera oceancirkulation samt glaciär- och permafrostavsmältning.

I kapitel 10 dras slutsatsen att reservoareffekter inte är att betrakta som ett marginellt problem vid $^{14}$C-datering av material förknippat med mänskliga aktiviteter. Människor har interagerat med och utnyttjat marina resurser i tusentals år. Civilisationer och bosättningar har vuxit längs kustlinjer och floder, ett bevis på vattenvägarnas betydelse för transport, handel och mat. Det är dock inte bara strandnära samhällen som interagerar med akvatiska resurser. Förutom människors mobilitet har även utvecklingen av olika konserveringsmetoder möjliggjort transport av akvatiska födoämnen, genom torkning, jäsning och saltning. Akvatiska resurser har utöver mat även samlats in för andra ändamål; skal, barder, val- och fiskoljor, elfenben och skinn, bland mycket annat, har samlats in och bedrivits handel med i stor utsträckning. Vid varje arkeologisk utgrävning, oavsett tid eller plats, är det därför mycket troligt att marint eller annat akvatiskt material kommer att påträffas. Vid $^{14}$C-datering måste alltid eventuella reservoareffekter has i åtanke. Det är därför viktigt för arkeologer att hantera reservoareffekter, vilket kan göras på flera sätt. Reservoareffekter kan undvikas, direkt mätas och korrigeras för genom bayesiansk modellering. Denna avhandling presenterar också fem fallstudier av fyra olika populationer på norra halvklotet som har utnyttjat akvatiska resurser i stor utsträckning. Även om de är spridda över ett stort geografiskt område och är från olika kronologiska perioder, visar de på behovet av mer förfinade kalibreringar av $^{14}$C-dateringar för att bättre förstå platserna i fråga. Dessa olika fallstudier belyser hur komplexa reservoareffekter är, och att de bör hanteras på olika sätt beroende på de specifika omständigheterna på lokalen.

Svensk översättning: Gunilla Eriksson

# 12 Samenvatting

Archeologie is afhankelijk van het ordenen van gebeurtenissen uit het verleden voor het bestuderen van culturele ontwikkelingen. Dit werd traditioneel gedaan door te kijken naar de stratigrafische positie van objecten ten opzichte van elkaar. Op deze manier kunnen chronologieën/ tijdlijnen van technologische vooruitgang en stilistische veranderingen uit het verleden worden opgesteld. De introductie van koolstofdatering zorgde voor een revolutie in de archeologie, waardoor directe, getalsmatige, schattingen van de ouderdom van een sample mogelijk werden gemaakt. Dit zorgde ervoor dat meer gedetailleerde tijdslijnen van het verleden konden worden gemaakt dan voorheen mogelijk was. Koolstofdatering maakt gebruik van het radioactieve verval van koolstof-14 om de ouderdom van een sample te schatten, waarbij het principe geldt dat oudere samples minder koolstof-14 bevatten dan nieuwe. Kort na de opkomst van koolstofdatering werd echter aangetoond dat koolstof-14 niet gelijkmatig over de hele wereld is verdeeld. Doorgaans is er minder koolstof-14 in zee- en zoetwater systemen in vergelijking met de atmosfeer. Als gevolg hiervan lijken samples uit zee- en zoetwater contexten ouder dan ze daadwerkelijk zijn, een fenomeen dat bekend staat als een ‘reservoireffect’. Deze reservoireffecten bleken lastige fenomenen, wat met regelmaat heeft geresulteerd in de samenstelling van onjuiste archeologische chronologieën / tijdlijnen. Het is echter mogelijk om met reservoireffecten om te leren gaan. Dit proefschrift demonstreert hoe archeologen koolstofdateringen van aquatische monsters zouden moeten interpreteren om onjuiste schattingen van de ouderdom van samples te vermijden. Door een zorgvuldige steekproefselectie, het rekening houden met menging van koolstofbronnen, het meten van de omvang en variatie van reservoireffecten binnen één enkel ecosysteem, en het toepassen van voorkennis over de ouderdom van een monster, kan de datering van aquatisch materiaal aanzienlijk worden verbeterd. Dit proefschrift beschrijft tevens een nieuwe methode om tanden met grotere precisie te dateren. Omdat aquatische middelen van groot belang zijn voor menselijke populaties over de hele wereld en door de tijd heen, is en blijft het vermogen om koolstofdateringen uit aquatische context correct te interpreteren ongelooflijk belangrijk.

Nederlandse vertaling: Anne-Marijn van Spelde

# 13 References

Arts-Bijma, A.T., van der Plicht, J. and Meijer, H.A.J., 2001. Automatic eAMS sample combustion and CO2 collection. *Radiocarbon*, 43(2A), pp.293–298.

Alcaraz Pelegrina, J.M. and Martínez-Aguirre, A., 2005. Isotopic fractionation during leaching of impure carbonates and their effect on uranium series dating. *Quaternary science reviews*, 24(23), pp.2584–2593.

AlQahtani, S.J., Hector, M.P. and Liversidge, H.M., 2010. Brief communication: The London atlas of human tooth development and eruption. *American journal of physical anthropology*, 142(3), pp.481–490.

Ambrose, S.H. and Norr, L., 1993. Experimental evidence for the relationship of the carbon isotope ratios of whole diet and dietary protein to those of bone collagen and carbonate. In: J.B. Lambert and G. Grupe, eds. *Prehistoric Human Bone: Archaeology at the Molecular Level*. Berlin, Heidelberg: Springer Berlin Heidelberg, pp.1–37.

Ascough, P., Cook, G. and Dugmore, A., 2005. Methodological approaches to determining the marine radiocarbon reservoir effect. *Progress in Physical Geography: Earth and Environment*, 29(4), pp.532–547.

Ascough, P.L., Church, M.J., Cook, G.T., Dunbar, E., Gestsdóttir, H., McGovern, T.H., Dugmore, A.J., Friðriksson, A. and Edwards, K.J., 2012. Radiocarbon reservoir effects in human bone collagen from northern Iceland. *Journal of archaeological science*, 39(7), pp.2261–2271.

Batt, C.M., Schmid, M.M.E. and Vésteinsson, O., 2015. Constructing chronologies in Viking Age Iceland: Increasing dating resolution using Bayesian approaches. *Journal of archaeological science*, 62, pp.161–174.

Becerra-Valdivia, L., Waters, M.R., Stafford, T.W., Jr, Anzick, S.L., Comeskey, D., Devièse, T. and Higham, T., 2018. Reassessing the chronology of the archaeological site of Anzick. *Proceedings of the National Academy of Sciences of the United States of America*, 115(27), pp.7000–7003.

Björck, S., Koç, N. and Skog, G., 2003. Consistently large marine reservoir ages in the Norwegian Sea during the Last Deglaciation. *Quaternary science reviews*, 22(5), pp.429–435.

Bonsall, C., Cook, G., Pickard, C., McSweeney, K., Sayle, K., Bartosiewicz, L., Radovanović, I., Higham, T., Soficaru, A. and Boroneanţ, A., 2015. Food for thought: re-assessing Mesolithic diets in the Iron Gates. *Radiocarbon*, 57(4), pp.1–11.

Boaretto, E., Thorling, L., Sveinbjörnsdottir, A.E., Yechieli, Y. and Heinemeier, J., 1998. Study of the effect of fossil organic carbon on 14C in groundwater from Hvinningdal, Denmark. *Radiocarbon*, 40(2), pp.915-920.

Bowman, S., 1990. *Radiocarbon Dating*. Blum, Fred H. University of California Press.

Braakhekke, M.C., Beer, C., Schrumpf, M., Ekici, A., Ahrens, B., Hoosbeek, M.R., Kruijt, B., Kabat, P. and Reichstein, M., 2014. The use of radiocarbon to constrain current and future soil organic matter turnover and transport in a temperate forest. *Journal of Geophysical Research: Biogeosciences*, 119(3), pp.372–391.

Britton, K., McManus-Fry, E., Nehlich, O., Richards, M., Ledger, P.M. and Knecht, R., 2018. Stable carbon, nitrogen and sulphur isotope analysis of permafrost preserved human hair from rescue excavations (2009, 2010) at the precontact site of Nunalleq, Alaska. *Journal of Archaeological Science: Reports*, 17, pp.950–963.

Brock, F., Higham, T., Ditchfield, P. and Ramsey, C.B., 2010. Current pretreatment methods for AMS radiocarbon dating at the Oxford radiocarbon accelerator unit (ORAU). *Radiocarbon*, 52(1), pp.103–112.

Bronk Ramsey, C., 1995. Radiocarbon calibration and analysis of stratigraphy; the OxCal program. *Radiocarbon*, 37(2), pp.425–430.

Bronk Ramsey, C., 2009. Dealing with outliers and offsets in radiocarbon dating. *Radiocarbon*, 51(3), pp.1023–1045.

Buchholz, B.A. and Spalding, K.L., 2010. Year of birth determination using radiocarbon dating of dental enamel. *Surface and interface analysis: SIA*, 42(5), pp.398–401.

Cameron, J., Devièse, T. and Green, F., 2017. Scientific analysis of a preserved head of hair at Romsey Abbey, UK. *Journal of Archaeological Science: Reports*, 13, pp.265–271.

Casanova, E., Knowles, T.D.J., Bayliss, A., Dunne, J., Barański, M.Z., Denaire, A., Lefranc, P., di Lernia, S., Roffet-Salque, M., Smyth, J., Barclay, A., Gillard, T., Claßen, E., Coles, B., Ilett, M., Jeunesse, C., Krueger, M., Marciniak, A., Minnitt, S., Rotunno, R., van de Velde, P., van Wijk, I., Cotton, J., Daykin, A. and Evershed, R.P., 2020. Accurate compound-specific 14C dating of archaeological pottery vessels. *Nature*, 580(7804), pp.506–510.

Casanova, E., Knowles, T.D.J., Ford, C., Cramp, L.J.E., Sharples, N. and Evershed, R.P., 2020. Compound-specific radiocarbon, stable carbon isotope and biomarker analysis of mixed marine/terrestrial lipids preserved in archaeological pottery vessels. *Radiocarbon*, pp.1–19.

Craig, O.E., Bondioli, L., Fattore, L., Higham, T. and Hedges, R., 2013. Evaluating marine diets through radiocarbon dating and stable isotope analysis of victims of the AD79 eruption of Vesuvius. *American journal of physical anthropology*, 152(3), pp.345–352.

Dee, M.W., Palstra, S.W.L., Th Aerts-Bijma, A., Bleeker, M.O., de Bruijn, S., Ghebru, F., Jansen, H.G., Kuitems, M., Paul, D., Richie, R.R., Spriensma, J.J., Scifo, A., van Zonneveld, D., B M A, Wietzes-Land, P. and Meijer, H.A.J., 2020. Radiocarbon dating at Groningen: New and updated chemical pretreatment procedures. *Radiocarbon*, 62(1), pp.63–74.

Ding, X.F., Fu, D.P., Gao, P., Zhou, L.P. and Liu, K.X., 2019. AMS radiocarbon measurement with high accuracy. *Nuclear instruments & methods in* *physics research. Section B, Beam interactions with materials and atoms*, 439, pp.80–83.

Fernandes, R., Millard, A.R., Brabec, M., Nadeau, M.-J. and Grootes, P., 2014. Food reconstruction using isotopic transferred signals (FRUITS): a Bayesian model for diet reconstruction. *PloS one*, 9(2), p.e87436.

Fernandes, R., Nadeau, M.-J. and Grootes, P.M., 2012. Macronutrient-based model for dietary carbon routing in bone collagen and bioapatite. *Archaeological and anthropological sciences*, 4(4), pp.291–301.

Fernandes, R., Rinne, C., Nadeau, M.-J. and Grootes, P., 2016. Towards the use of radiocarbon as a dietary proxy: Establishing a first wide-ranging radiocarbon reservoir effects baseline for Germany. *Environmental Archaeology*, 21(3), pp.285–294.Godwin, H., 1962. Half-life of Radiocarbon. *Nature*, 195(4845), pp.984–984.

Guilderson, T.P., Caldeira, K. and Duffy, P.B., 2000. Radiocarbon as a diagnostic tracer in ocean and carbon cycle modeling. *Global biogeochemical cycles*, 14(3), pp.887–902.

Hall, B.L. and Henderson, G.M., 2001. Use of uranium–thorium dating to determine past 14C reservoir effects in lakes: examples from Antarctica. *Earth and planetary science letters*, 193(3), pp.565–577.

Heaton, T.J., Köhler, P., Butzin, M., Bard, E., Reimer, R.W., Austin, W.E.N., Ramsey, C.B., Grootes, P.M., Hughen, K.A., Kromer, B., Reimer, P.J., Adkins, J., Burke, A., Cook, M.S., Olsen, J. and Skinner, L.C., 2020. MARINE20—The marine radiocarbon age calibration curve (0–55,000 CAL BP). *Radiocarbon*, 62(4), pp.779–820.

Helama, S. and Hood, B.C., 2011. Stone Age midden deposition assessed by bivalve sclerochronology and radiocarbon wiggle-matching of Arctica islandica shell increments. *Journal of archaeological science*, 38(2), pp.452–460.

Hesslein, R.H., Hallard, K.A. and Ramlal, P., 1993. Replacement of sulfur, carbon, and nitrogen in tissue of growing broad whitefish (Coregonus nasus) in response to a change in diet traced by δ34S, δ13C, and δ15N. *Canadian Journal of Fisheries and Aquatic Sciences*, 50, pp.2070–2076.

Hogg, A.G., Hua, Q., Blackwell, P.G., Niu, M., Buck, C.E., Guilderson, T.P., Heaton, T.J., Palmer, J.G., Reimer, P.J., Reimer, R.W., Turney, C.S.M. and Zimmerman, S.R.H., 2013. SHCal13 Southern hemisphere calibration, 0–50,000 years cal BP. *Radiocarbon*, 55(4), pp.1889–1903.

Hua, Q., Barbetti, M. and Rakowski, A.Z., 2013. Atmospheric radiocarbon for the period 1950–2010. *Radiocarbon*, 55(4), pp.2059–2072.

Hua, Q., Webb, G.E., Zhao, J.-X., Nothdurft, L.D., Lybolt, M., Price, G.J. and Opdyke, B.N., 2015. Large variations in the Holocene marine radiocarbon reservoir effect reflect ocean circulation and climatic changes. *Earth and planetary science letters*, 422, pp.33–44.

Hugelius, G., Kuhry, P., Tarnocai, C. and Virtanen, T., 2010. Soil organic carbon pools in a periglacial landscape: a case study from the central

Canadian Arctic: Soil Organic Carbon in the Central Canadian Arctic. *Permafrost and Periglacial Processes*, 21(1), pp.16–29.

Hughen, K.A., Baillie, M.G.L., Bard, E., Warren Beck, J., Bertrand, C.J.H., Blackwell, P.G., Buck, C.E., Burr, G.S., Cutler, K.B., Damon, P.E., Edwards, R.L., Fairbanks, R.G., Friedrich, M., Guilderson, T.P., Kromer, B., McCormac, G., Manning, S., Ramsey, C.B., Reimer, P.J., Reimer, R.W., Remmele, S., Southon, J.R., Stuiver, M., Talamo, S., Taylor, F.W., van der Plicht, J. and Weyhenmeyer, C.E., 2004. Marine04 marine radiocarbon age calibration, 0-26 cal kyr BP. *Radiocarbon*, 46(3), pp.1059–1086.

Junno, A., Isaksson, S., Hirasawa, Y., Kato, H. and Jordan, P.D., 2020. Evidence of increasing functional differentiation in pottery use among Late Holocene maritime foragers in northern Japan. *Archaeological Research in Asia* [Online], 22, pp.100194.

Kondo-Nakamura, M., Fukui, K., Matsu'ura, S., Kondo, M. and Iwadate, K., 2011. Single tooth tells us the date of birth. *International journal of legal medicine*, 125(6), pp.873–877.

Krus, A.M., Jensen, A.M., Hamilton, W.D. and Sayle, K., 2019. A context-appropriate approach to marine 14C calibration: ΔR and bayesian framework for the Nuvuk cemetery, Point Barrow, Alaska. *Radiocarbon*, 61(3), pp.733–747.

Kutschera, W., 2013. Applications of accelerator mass spectrometry. *International journal of mass spectrometry*, 349-350, pp.203–218.

Kutschera, W., 2019. The half-life of 14C—Why is it so long? *Radiocarbon*, 61(5), pp.1135–1142.

Kuzmin, Y.V., Burr, G.S., Gorbunov, S.V., Rakov, V.A. and Razjigaeva, N.G., 2007. A tale of two seas: Reservoir age correction values (R, ΔR) for the Sakhalin Island (Sea of Japan and Okhotsk Sea). *Nuclear instruments & methods in physics research. Section B, Beam interactions with materials and atoms*, 259(1), pp.460–462.

Lanting, J.N. and van der Plicht, J., 1996. Wat hebben Floris V, skelet Swifterbant S2 en visotters gemeen? *Palaeohistoria*, pp.491–519.

Lanting, J.N. and Van Der Plicht, J., 1998. Reservoir effects and apparent 14C ages. *The Journal of Irish Archaeology*, 9, pp.151–165.

Lee-Thorp, J.A., Sealy, J.C. and van der Merwe, N.J., 1989. Stable carbon isotope ratio differences between bone collagen and bone apatite, and their relationship to diet. *Journal of archaeological science*, 16(6), pp.585–599.

Levin, I. and Hesshaimer, V., 2000. Radiocarbon: A unique tracer of global carbon cycle dynamics. *Radiocarbon*, 42(1), pp.69–80.

Libby, W.F., 1955. *Radiocarbon Dating*. Chicago: University of Chicago Press.

Libby, W.F., Anderson, E.C. and Arnold, J.R., 1949. Age determination by radiocarbon content: World-wide assay of natural radiocarbon. *Science*, 109(2827), pp.227–228.

Lougheed, B.C., Filipsson, H.L. and Snowball, I., 2013. Large spatial variations in coastal $^{14}$C reservoir age – a case study from the Baltic Sea. *Climate of the Past*, 9(3), pp.1015–1028.

Macario, K.D., Alves, E.Q., Chanca, I.S., Oliveira, F.M., Carvalho, C., Souza, R., Aguilera, O., Tenório, M.C., Rapagnã, L.C., Douka, K. and Silva, E., 2016. The Usiminas shellmound on the Cabo Frio Island: Marine reservoir effect in an upwelling region on the coast of Brazil. *Quaternary geochronology*, 35, pp.36–42.

Mann, W.B., 1983. An international reference material for radiocarbon dating. *Radiocarbon*, 25(2), pp.519–527.

Miyake, F., Nagaya, K., Masuda, K. and Nakamura, T., 2012. A signature of cosmic-ray increase in AD 774-775 from tree rings in Japan. *Nature*, 486, pp.240–242.

Naito, Y.I., Chikaraishi, Y., Ohkouchi, N., Mukai, H., Shibata, Y., Honch, N.V., Dodo, Y., Ishida, H., Amano, T., Ono, H. and Yoneda, M., 2010. Dietary reconstruction of the Okhotsk culture of Hokkaido, Japan, based on nitrogen composition of amino acids: Implications for correction of 14C marine reservoir effects on human bones. *Radiocarbon*, 52(2), pp.671–681.

Nalawade-Chavan, S., McCullagh, J., Hedges, R., Bonsall, C., Boroneant, A., Ramsey, C.B. and Higham, T., 2013. Compound specific radiocarbon dating of essential and non-essential amino acids: Towards determination of dietary reservoir effects in humans. *Radiocarbon*, 55(2–3), pp.709–719.

O'Connell, T.C., Hedges, R.E.M., Healey, M.A. and Simpson, A.H.R.W., 2001. Isotopic comparison of hair, nail and bone: Modern analyses. *Journal of archaeological science*, 28(11), pp.1247–1255.

Oeschger, H., Siegenthaler, U., Schotterer, U. and Gugelmann, A., 1975. A box diffusion mode*l to study* the carbon dioxide exchange in nature. *Tell'Us*, 27(2), pp.168–192.

Parnell, A. and Jackson, A., 2013. *SIAR: Stable isotope analysis in R. R package version 4.2* (v.4.).

Petchey, F., Anderson, A., Hogg, A. and Zondervan, A., 2008. The marine reservoir effect in the Southern Ocean: An evaluation of extant and new ΔR values and their application to archaeological chronologies. *Journal of the Royal Society of New Zealand*, 38(4), pp.243–262.

Philippsen, B., 2013. The freshwater reservoir effect in radiocarbon dating. *Heritage Science*, 1(24), pp.1–19.

Pitulko, V.V., 2000. Another life of bones: the use of pleistocene faunal remains in the post-pleistocene site of arctic Siberia. In: Lars Larsson, Hans Knutsson, David Loeffler, Agneta Åkerlund, ed. *Mesolithic on the move*. Oxbow Books, pp.680–693.

van der Plicht, J. and Hogg, A., 2006. A note on reporting radiocarbon. *Quaternary geochronology*, 1(4), pp.237–240.

Reimer, P.J., Austin, W.E.N., Bard, E., Bayliss, A., Blackwell, P.G., Bronk Ramsey, C., Butzin, M., Cheng, M., Edwards, R.L., Friedrich, M., Grootes, P.M., Guilderson, T.P., Hajdas, I., Heaton, T.J., Hogg, A.G., Hughen, K.A., Kromer, B.K., Manning, S.W., Muscheler, R., Palmer, J.G., Pearson, C., van der Plicht, J., Reimer, R., Richards, D.A., Scott, E.M., Southon, J.R., Turney, C.S.M., Wacker, L., Adolphi, F., Büntgen, U., Capano, M., Fahrni, S., Fogtmann-Schulz, A., Friedrich, R., Miyake, F., Olsen, J., Reinig, F., Sakamoto, M., Sookdeo, A. And Talamo, S., 2020. The IntCal20 northern hemisphere radiocarbon age calibration curve (0-55 kcal BP). Radiocarbon, 62(4), pp.725–757.

Reimer, P.J., Baillie, M.G.L., Bard, E., Bayliss, A., Beck, J.W., Blackwell, P.G., Bronk Ramsey, C., Buck, C.E., Burr, G.S., Edwards, R.L., Friedrich, M., Grootes, P.M., Guilderson, T.P., Hajdas, I., Heaton, T.J., Hogg, A.G., Hughen, K.A., Kaiser, K.F., Kromer, B., McCormac, F.G., Manning, S.W., Reimer, R.W., Richards, D.A., Southon, J.R., Talamo, S., Turney, C.S.M., van der Plicht, J. and Weyhenmeyer, C.E., 2009. IntCal09 and Marine09 radiocarbon age calibration curves, 0–50,000 years cal BP. *Radiocarbon*, 51(4), pp.1111–1150.

Reimer, P.J., Bard, E., Bayliss, A., Beck, J.W., Blackwell, P.G., Bronk Ramsey, C., Buck, C.E., Cheng, H., Edwards, R.L., Friedrich, M. and Others, 2013. IntCal13 and Marine13 radiocarbon age calibration curves 0-50,000 years cal BP. *Radiocarbon*, 55(4), pp.1869–1887.

Reimer, P.J., Bard, E., Bayliss, A., Warren Beck, J., Blackwell, P.G., Bronk Ramsey, C., Buck, C.E., Cheng, H., Lawrence Edwards, R., Friedrich, M., Haflidason, H., Heaton, T.J., Hoffmann, D.L., Reimer, R.W. and van der Plicht, C.S.M.T.J., 2013. INTCAL13 and MARINE13 radiocarbon age calibration curves. *Radiocarbon*, 55(4), pp.1869–1887.

Reimer, P.J., and Reimer, R. W., 2001. A marine reservoir correction database and on-line interface. *Radiocarbon*, 43(2A), pp.461–463.

Reynard, L.M. and Hedges, R.E.M., 2008. Stable hydrogen isotopes of bone collagen in palaeodietary and palaeoenvironmental reconstruction. *Journal of archaeological science*, 35(7), pp.1934–1942.

Russell, N., Cook, G.T., Ascough, P.L., Scott, E.M. and Dugmore, A.J., 2011. Examining the inherent variability in ΔR: New methods of presenting ΔR values and implications for MRE studies. *Radiocarbon*, 53(2), pp.277–288.

Sayle, K.L., Cook, G.T., Ascough, P.L., Gestsdóttir, H., Derek Hamilton, W. and McGovern, T.H., 2014. Utilization of δ13C, δ15N, and δ34S analyses to understand 14C dating anomalies within a late Viking Age community in Northeast Iceland. *Radiocarbon*, 56(2), pp.811–821.

Sayle, K.L., Hamilton, W.D., Gestsdóttir, H. and Cook, G.T., 2016. Modelling Lake Mývatn's freshwater reservoir effect: Utilisation of the statistical program FRUITS to assist in the re-interpretation of radiocarbon dates from a cemetery at Hofstaðir, north-east Iceland. *Quaternary geochronology*, 36(Supplement C), pp.1–11.

Schmid, M.M.E., Zori, D., Erlendsson, E., Batt, C., Damiata, B.N. and Byock, J., 2018. A Bayesian approach to linking archaeological, paleoenvironmental and documentary datasets relating to the settlement of Iceland (Landnám). *Holocene*, 28(1), pp.19–33.

Schoeninger, M.J. and DeNiro, M.J., 1984. Nitrogen and carbon isotopic composition of bone collagen from marine and terrestrial animals. *Geochimica et cosmochimica acta*, 48(4), pp.625–639.

van der Sluis, L.G., Reimer, P.J. and Lynnerup, N., 2015. Investigating intra-individual dietary changes and 14C ages using high-resolution δ13C and δ15N isotope ratios and 14C ages obtained from dentine increments. *Radiocarbon*, 57(4), pp.665–677.

Stuiver, M., Reimer, P.J., and Braziunas, T.F., 1998. High precision radiocarbon age calibration for terrestrial and marine samples. *Radiocarbon*, 40, pp.1127–1151.

Stuiver, M., Reimer, P. and Reimer, R., 2015. Marine reservoir correction database, The 14CHRONO Centre, Queen's University Belfast, Belfast. *URL: http://calib. qub. ac. UK/marine*.

Svyatko, S.V., Schulting, R., Poliakov, A., Ogle, N. and Reimer, P.J., 2017. A lack of freshwater reservoir effects in human radiocarbon dates in the Eneolithic to Iron Age in the Minusinsk Basin. *Archaeological and anthropological sciences*, 9(7), pp.1379–1388.

Talamo, S. and Richards, M., 2011. A comparison of bone pretreatment methods for AMS dating of samples >30,000 BP. *Radiocarbon*, 53(3), pp.443–449.

Tans, P.P., de Jong, A.F.M. and Mook, W.G., 1979. Natural atmospheric 14C variation and the Suess effect. *Nature*, 280(5725), p.826.

Taylor, R.E., Suchey, J.M., Payen, L.A. and Slota, P.J., Jr, 1989. The use of radiocarbon (14C) to identify human skeletal materials of forensic science interest. *Journal of forensic sciences*, 34(5), pp.1196–1205.

Thompson, V.D., Jefferies, R.W. and Moore, C.R., 2019. The case for radiocarbon dating and Bayesian analysis in historical archaeology. *Historical archaeology*, 53(1), pp.181–192.

Turney, C.M.S., Fifield, L.K., Hogg, A.G., Palmer, J.G., Hughen, K., Baillie, M.G.L., Galbraith, R., Ogden, J., Lorrey, A., Tims, S.G. and Jones, R.T., 2010. The potential of NewZealand kauri (Agathis australis) for testing the synchronicity of abrupt climate change during the last glacial Interval (60,000–11,700 years ago). *Quaternary science reviews,* 29(27–28), pp.3677–3682.

Walter Anthony, K., Schneider von Deimling, T., Nitze, I., Frolking, S., Emond, A., Daanen, R., Anthony, P., Lindgren, P., Jones, B. and Grosse, G., 2018. 21st-century modeled permafrost carbon emissions accelerated by abrupt thaw beneath lakes. *Nature communications*, 9(1), p.3262.

Wild, B., Andersson, A., Bröder, L., Vonk, J., Hugelius, G., McClelland, J.W., Song, W., Raymond, P.A. and Gustafsson, Ö., 2019. Rivers across the Siberian Arctic unearth the patterns of carbon release from thawing permafrost. *Proceedings of the National Academy of Sciences of the United States of America*, 116(21), pp.10280–10285.