

## DIET AND RADIOCARBON DATING OF TOLLUND MAN: NEW ANALYSES OF AN IRON AGE BOG BODY FROM DENMARK

Nina H Nielsen<sup>1\*</sup> • Bente Philippsen<sup>2,3</sup> • Marie Kanstrup<sup>2</sup> • Jesper Olsen<sup>2,3</sup>

<sup>1</sup>Museum Silkeborg, Hovedgårdsvej 7, 8600 Silkeborg, Denmark.

<sup>2</sup>Aarhus AMS Centre, Department of Physics and Astronomy, Aarhus University, Ny Munkegade 120, 8000 Aarhus C, Denmark.

<sup>3</sup>Centre for Urban Network Evolutions (UrbNet), Aarhus University, Moesgård Allé 20, 8270 Højbjerg, Denmark.

**ABSTRACT.** Tollund Man is one of the most famous Iron Age bog bodies due to his well-preserved head. Since he was unearthed in 1950 in Bjældskovdal, Denmark, he has been subjected to several scientific investigations, but until now no attempts to reconstruct his general diet through isotope analyses have been conducted. Furthermore, previous radiocarbon (<sup>14</sup>C) analyses have only been able to date him broadly to the 3rd–4th century BC. In this study, stable isotope measurements ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ) on bone collagen from Tollund Man's femur and rib showed that the diet of Tollund Man was terrestrial-based and that the crops he ate probably were grown on manured fields. Accelerator mass spectrometry (AMS) <sup>14</sup>C dates were obtained on both the <30kDa and >30kDa fractions of ultrafiltered collagen. Results showed that the ultrafiltration removed contamination from older substances from the burial environment. The femur was dated to  $2330 \pm 23$  BP, the rib to  $2322 \pm 30$  BP. These dates statistically agree with a previously published AMS <sup>14</sup>C age on skin. By combining the new dates with the previous date of his skin it was possible to narrow down the age of Tollund Man to the period 405–380 cal BC (95.4% confidence interval).

**KEYWORDS:** bog bodies, collagen extraction, Iron Age, radiocarbon dating, stable isotopes, Tollund Man.

### INTRODUCTION

Tollund Man—especially his head—is among the best-preserved human remains from prehistory. He belongs to the group of so-called “bog bodies,” which are naturally mummified bodies that have been found in acidic, anaerobic peat bogs throughout large parts of northwestern Europe, such as in Britain, Ireland, Denmark, northern Germany, and the Netherlands (e.g. van der Sanden 2013). Although bodies dating to almost all prehistoric and historic periods have been found in bogs, there is a clustering of interesting bog bodies dating to the European Late Iron Age and Roman Period (200 BC–AD 400) (van der Plicht et al. 2004) or in Denmark especially to the so-called Pre-Roman Iron Age (500–1 BC) and the Early Roman Iron Age (AD 1–200) (Mannering et al. 2010). Tollund Man is one of these Iron Age finds.

From the time of his discovery, Tollund Man has attracted much scientific interest. Through the years he has therefore been subjected to a number of forensic and scientific investigations seeking to unravel the secrets of his death (which presumably was part of a ritual sacrifice) and to uncover as much as possible about his life and health and thereby also the living conditions of Iron Age people in general (Helbæk 1950; Fischer 2012; Nielsen 2017, 2018; Zanello et al. 2017). This article presents the results of the newest investigations regarding his diet and date.

### DISCOVERY AND PRESERVATION OF TOLLUND MAN

Tollund Man was found in 1950 in a raised bog in the valley of Bjældskovdal, located about 10 km west of Silkeborg, Central Jutland, Denmark (approx.  $56^{\circ}09'53.3''\text{N}$ ,  $9^{\circ}23'34.7''\text{E}$ ) (Figure 1a) (see Thorvildsen 1951; Glob 1969; Fischer 1980, 2012 for information on the discovery and subsequent investigations of Tollund Man). Today Bjældskovdal leads directly into

\*Corresponding author. Email: nhn@museumsilkeborg.dk.



Figure 1 (a) Maps showing the discovery location of Tollund Man in Bjældskovdal, a narrow valley leading into the basin of Lake Bølling (contains data from The Danish Agency for Data Supply and Efficiency). (b) Tollund Man after excavation at the Danish National Museum (photo by Lennart Larsen). (c) Close-up view of the well-preserved head of Tollund Man (photo by Arne Mikkelsen).

the recreated, 360 ha Lake Bølling, but during the Iron Age the lake was much smaller and the majority of the area was covered by bog (Aaby 2006). The distance from Bjældskovdal to the small freshwater lake would at that time have been ca. 1 km.

In 1927 and 1938, peat cutting in Bjældskovdal revealed two other bog bodies less than 100 m away from the discovery location of Tollund Man: Elling Woman and another body, which was never recovered due to a collapse of the trench cut into the peat. Like these bog bodies, Tollund Man was found by local peat cutters and, based on recommendations from Archaeology Professor P.V. Glob, Aarhus University, was transported in a block of peat to the Danish National Museum where he was excavated under more controlled conditions (Figure 1b). The local museum in Silkeborg was very keen on having the entire body preserved and put on display, but at the National Museum they deemed Tollund Man in his entirety to be too macabre a sight to be exhibited and, furthermore, they thought it would be too difficult to undertake a full-body preservation. It was therefore decided to focus primarily on conserving his well-preserved head (Figure 1c). Although today the conservation of a bog body would be approached completely differently, we are now in the fortunate situation that large parts of Tollund Man have not been treated by any chemicals but only dried. These parts—such as his bones, which despite the acidic bog environment are surprisingly well-preserved—are therefore very suitable for

scientific investigations. The different parts of Tollund Man are all stored at Museum Silkeborg, where also the head is displayed together with a reconstruction of the body.

## DIET OF TOLLUND MAN

How prehistoric people lived and what foods they ate represent fundamental aspects of culture that must be known if one wants to understand prehistoric societies. Analyzing the physical remains of prehistoric people is a way of obtaining this type of information. Based on analyses of the stomach contents of Tollund Man, we know that his last meal consisted of a kind of gruel primarily made up of hulled barley (*Hordeum vulgare* var. *vulgare*), flax (*Linum usitatissimum*), and seeds from *Camelina* and pale persicaria/curlytop knotweed (*Polygonum lapathifolium*/*Persicaria lapathifolia*), but which also contained the remains of a number of other plant species (Helbæk 1950). However, until now, no attempts have been made to obtain information on his long-term diet. Stable isotope analyses of carbon and nitrogen in human bone collagen are standard practice in the study of past diet. The isotopic composition ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) provides information about long-term average diet and of certain dietary patterns. Unless the diet is extremely protein-deficient, bone collagen is largely produced from ingested protein. Collagen  $\delta^{13}\text{C}$  values predominantly reflect the dietary protein.  $\delta^{15}\text{N}$  values reflect only the dietary protein, as there is no nitrogen in the other nutrients (fats and carbohydrates). Thus, the combined measurement of carbon and nitrogen isotope ratios in bone collagen is very suitable for identifying dietary resources in human diets (e.g. Ambrose 1993; Bonsall et al. 2004; Eriksson 2004; Fischer et al. 2007; Kanstrup 2008; Philippsen 2013). Information on any possible marine or freshwater influence in the diet is, apart from contributing to the understanding of the lifestyle of ancient people, also vital for obtaining correct  $^{14}\text{C}$  dates as such influences require a reservoir correction of the radiocarbon ( $^{14}\text{C}$ ) results (e.g. Olsen and Heinemeier 2009; Fernandes et al. 2015).

Since agriculture generally formed the basis of Iron Age societies, we would expect Tollund Man to show evidence of a terrestrial-based diet. However, as he was found in the lake district of Denmark it is possible that a certain amount of freshwater fish was included in the diet. Furthermore, preliminary results of a strontium isotope analysis of his femur indicate that within the last ten years prior to his death, Tollund Man, who died approximately at age 40, had been living at least 40 km away (Frei 2017; Nielsen 2018). Based on this analysis, it cannot be ruled out that Tollund Man had spent some time at the coast, which—even though a strong marine signal would never be expected—makes it even more relevant to measure the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  on Tollund Man. It was therefore decided to undertake stable isotope analyses on samples from a rib and femur. The collagen in the femur reflects the average diet during adolescence and adulthood (Hedges et al. 2007). We expect the rib to have a shorter turnover time than the femur, as it is less compact. Therefore, the isotope values of the rib collagen should be closer to those of the diet during adulthood and the last years of Tollund Man's life.

## DATE OF TOLLUND MAN

One of the most pressing questions regarding Tollund Man is the date of his death. P.V. Glob assumed that Tollund Man died around the birth of Christ (Glob 1969), but scientific dating was not conducted until 1977, when conventional  $^{14}\text{C}$  analyses of two samples of muscle tissue produced four dates ranging from  $2260 \pm 75$  BP to  $2110 \pm 75$  BP (K-2814A and B, Tauber 1980). In 1997 and 2000, new AMS dates were obtained on a rib ( $2345 \pm 40$  BP, AAR-3328) and a piece of skin ( $2290 \pm 30$  BP, GrA-14179) by Aarhus and Groningen, respectively (van der Plicht et al. 2004). Based on all the dates of Tollund Man (summarized in van der Plicht et al. 2004), the former director of Museum Silkeborg wrote in 2012 about Tollund Man that “*we are*

now confident that his time of death lies between 375 and 210 BC, but further minor changes must be expected” (Fischer 2012:76). Although some uncertainty in  $^{14}\text{C}$  age determinations must sometimes be accepted when studying prehistoric objects, a desire to obtain a more precise date resulted in the decision to also  $^{14}\text{C}$  date the bones that were already planned to be analyzed for stable isotopes. Furthermore, the previous  $^{14}\text{C}$  analysis was conducted before the widespread introduction of ultrafiltration as the standard pretreatment protocol in many radiocarbon laboratories (e.g. Bronk Ramsey et al. 2004). Ultrafiltration removes the smaller molecular weight proteins and has been shown to improve the accuracy of bone  $^{14}\text{C}$  age determinations (e.g. Bronk Ramsey et al. 2004; Brock et al. 2007). By analyzing different skeletal elements (i.e. rib and femur), we wish to address some considerations of the bioarcheology and remodeling of the different bones in order to see whether this can better resolve the timing of Tollund Man’s death.

## METHODS

Samples were obtained from a rib and a femur, respectively, which are among the best preserved bones of Tollund Man. We followed a modified Longin procedure with ultrafiltration (Longin 1971; Brown et al. 1988; Brock et al. 2013). The surface of the bone pieces was cleaned with a scalpel, and bone minerals were dissolved in hydrochloric acid at  $4^\circ\text{C}$  for several days. The acid was renewed regularly until the mineral fraction was removed completely. Humic substances were removed using 0.2M NaOH at  $4^\circ\text{C}$ , with frequent changes of the NaOH, each step lasting several hours, until the solution stayed clear. The samples were subsequently rinsed with 0.01M HCl and gelatinized in 0.01M HCl at  $58^\circ\text{C}$  overnight, with an additional 3-day gelatinization afterwards. The extracted “collagen” was ultrafiltered in pre-cleaned Amicon Ultra Centrifugal Filters Ultracel – 30K, made of regenerated cellulose (REF UFC803096, LOT R4CA36137). The  $<30\text{kDa}$  fraction is usually discarded, but was dated here in addition to the  $>30\text{kDa}$  fraction (fresh type I collagen has molecular masses between 95 and 102 kDa). Both collagen fractions were weighed after ultrafiltration and freeze-drying.

The collagen was converted to  $\text{CO}_2$  by combustion in sealed evacuated quartz tubes with 200 mg CuO. The  $\text{CO}_2$  was reduced to graphite by the  $\text{H}_2$  reduction method using an iron catalyst and  $\text{MgClO}_4$  to remove the water (Vogel et al. 1984; Santos et al. 2007). The samples were  $^{14}\text{C}$  dated using the HVE 1MV tandemron accelerator AMS system at the Aarhus AMS Centre (Olsen et al. 2016).  $^{14}\text{C}$  dates are reported as uncalibrated  $^{14}\text{C}$  ages BP normalized to  $-25\text{‰}$  according to international convention using online  $^{13}\text{C}/^{12}\text{C}$  ratios (Stuiver and Polach 1977). Dates in the article have been calibrated with OxCal v.4.3 (Bronk Ramsey 2009) using the calibration curve IntCal13 (Reimer et al. 2013) and are reported as calibrated ages BC.

Stable isotopes  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  were measured using a continuous-flow IsoPrime IRMS coupled to an Elementar PyroCube elemental analyser at the Aarhus AMS Centre (AARAMS), Aarhus University, Denmark. An in-house standard Gel-A was used as primary standard yielding  $\pm 0.2\text{‰}$  and  $\pm 0.3\text{‰}$  for carbon and nitrogen analysis respectively. Further, secondary in-house and international standard were used to check the normalization to the VPDB and AIR scale.

FTIR spectra were obtained using an Agilent Technologies Cary 630 ATR-FTIR instrument. Scans were performed in the range from  $4000\text{--}650\text{ cm}^{-1}$  with a resolution of  $2\text{ cm}^{-1}$ . Each spectrum is an average of 64 scans. The spectra are baseline corrected and C/P and N (Amide I)/P ratios are calculated using the peak heights of  $1630\text{ cm}^{-1}$  (“N”, Amide I),  $1450\text{ cm}^{-1}$  (“C”,  $\text{CO}_3^{2-}$ ) and  $1030\text{ cm}^{-1}$  (“P”,  $\text{PO}_4^{3-}$ ).

Table 1 Results of the measurements on previous and this study's samples of Tollund Man. The extracted bone collagen of the new samples (AAR-26386 and AAR-26387) was ultrafiltered. Both the >30kDa and the <30kDa fractions were analyzed. The >30kDa fraction of AAR-26386 was dated twice (1a and 1b), and the weighted mean value is presented under AAR-26386.1. For combined results a reduced  $\chi^2$  test (95.4% confidence level) is given as  $X \leq Y$ . The  $\chi^2$  is passed if and only if  $X \leq Y$  (Bevington and Robinson 2003).

Lab no.	Description	Collagen yield (%)	C (%)	N (%)	C:N (atomic)	$\delta^{13}\text{C}$ (‰) VDPB	$\delta^{15}\text{N}$ (‰) AIR	$^{14}\text{C}$ age (BP)	$\delta^{13}\text{C}_{\text{AMS}}$ (‰) VDPB	Calibrated age (95.4% confidence interval)	FTIR: C/P peak ratio	FTIR: N/P peak ratio
GrA-14179	Skin (ABA)	n/a	49.0	n/a	n/a	-23.56*	n/a	2290 ± 30		405 BC (70.9%) 353 BC 292 BC (24.5%) 231 BC		
AAR-3328	Rib (collagen)	14.4	17.8	n/a	n/a	-21.10*	n/a	2345 ± 40		728 BC (0.8%) 716 BC 707 BC (0.9%) 694 BC 542 BC (92.5%) 358 BC 274 BC (1.1%) 259 BC		
AAR-26386.1a	Femur (>30kDa)	20.1	43.1	13.6	3.7	-21.50	10.3	2342 ± 33	-18			
AAR-26386.1b	Femur (>30kDa)	20.1	n/a	n/a	n/a	n/a	n/a	2320 ± 34	-24			
AAR-26386.1	Femur (>30kDa), $\chi^2$ test: 0.2 ≤ 3.84	20.1	43.1	13.6	3.7	-21.50	10.3	2330 ± 23			2.8	6.0
AAR-26386.2	Femur (<30kDa)	12.1	31.0	10.0	3.6	-20.20	10.0	2391 ± 28	-19		2.7	5.1
AAR-26387.1	Rib (>30kDa)	21.2	42.4	13.4	3.7	-21.00	10.0	2322 ± 30	-18		2.9	6.1
AAR-26387.2	Rib (<30kDa)	12.1	31.0	10.5	3.5	-20.50	9.7	2377 ± 29	-17		2.7	5.7
Combined (>30kDa), $\chi^2$ test: 0.04 ≤ 3.84			42.7	13.5	3.7	-21.30	10.2	2327 ± 18		407 BC (95.4%) 380 BC		
Combined (<30kDa), $\chi^2$ test: 0.12 ≤ 3.84			31.0	10.3	3.6	-20.40	9.9	2384 ± 20				
Difference (>30kDa - <30kDa)			11.7	3.2	0.1	-0.90	0.3	-57 ± 27				
Combined (>30kDa & GrA-14179), $\chi^2$ test: 0.58 ≤ 3.00								2318 ± 15		405 BC (95.4%) 380 BC		

\*Dual inlet analysis, uncertainty ±0.05‰. n/a: information not available.

## RESULTS AND DISCUSSION

The results obtained on Tollund Man's bones are listed in Table 1. Here, we will start by discussing the quality criteria for bone collagen and the effects of ultrafiltration. This is followed by a discussion of the results of the stable isotope analysis and the  $^{14}\text{C}$  analysis.

### Pretreatment

Ultrafiltration separates the gelatinized collagen into two fractions determined by the cut-off rigidity (here 30 kDa) of the applied ultrafilter and was introduced by Brown et al. (1988). The <30 kDa fraction is normally disregarded as it is more likely to contain possible contaminants such as short-chain proteins from degraded collagen, salt products, fulvic acids and plant- or soil-derived amino acids (Brock et al. 2007). Contaminating substances such as humic or fulvic acids can attach to the ends of the collagen fibers and larger proportions of contamination are therefore expected for the smaller molecules (cf. van der Plicht et al. 2004). The higher molecular weight protein is on the other hand hoped to only represent collagen originating from the bone sample itself (Brown et al. 1988; Bronk Ramsey et al. 2004). Thus, in most cases only the high molecular weight fraction (>30kDa) is used for  $^{14}\text{C}$  analysis, whereas the low molecular weight fraction is discarded.

The >30kDa collagen yield of Tollund Man's bones (Table 1) is very similar to that of fresh bone at 22 wt % collagen (van Klinken 1999). This is much higher compared with bones from different Danish Roman Iron Age Burial sites where yields in the range 0.4–13% have been reported (Figure 2; Jørkov 2007). We suspect that part of the minerogenic fraction of the bones was dissolved in the acidic bog environment, thereby inflating the apparent collagen yield. The carbon (42.7%, >30 kDa) and nitrogen (13.5%, >30 kDa) weight percent fall within the expected range for good quality collagen (van Klinken 1999). The carbon weight percentage is

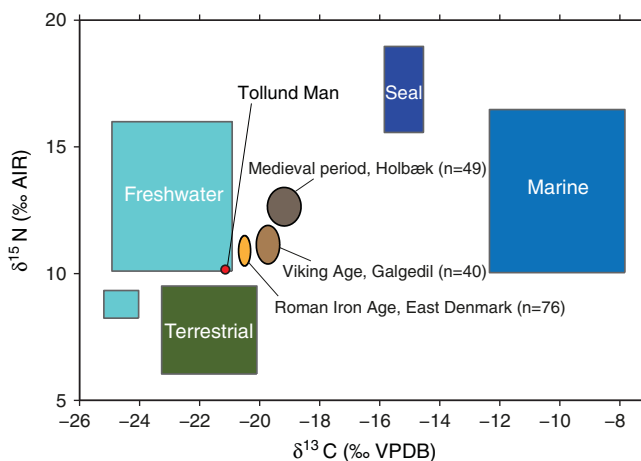


Figure 2 The Tollund Man isotope data compared with other published Danish isotope studies. The food item boxes are from different Mesolithic and Neolithic sites (Fischer et al. 2007). Isotopic values on human bones from the Galgedil site on Funen (Kanstrup 2008), east Danish Roman Iron Age sites (Jørkov 2007) and from a medieval cemetery in Holbæk on Zealand (Jørkov 2007) are also plotted for reference. The ellipse axis denotes  $\pm 1\sigma$  and the center value is average of all humans for each site respectively.

fairly close the expected collagen carbon content of fresh bones at 47%, whereas the nitrogen weight percentage is much lower than would be expected from fresh bones with a collagen weight percentage of 17% (Ambrose and Krigbaum 2003). The average C/N ratio of the >30 kDa fractions is 3.7, falling just outside the range between 2.8 and 3.6, which is considered to reflect well-preserved collagen (DeNiro 1985). The higher C/N ratios can be explained as being caused by either additional carbon in the samples (i.e. contamination) or a preferential removal of nitrogen. We speculate that microorganisms in the nutrient-poor bog fed on the nitrogen from the bones. This could increase the  $\delta^{15}\text{N}$  values slightly, as microbes preferentially break the weaker bonds containing the lighter isotopes leaving behind an enriched  $^{15}\text{N}$  residual, i.e. bone collagen (Talbot 2001).

FTIR spectra suggest that both fractions contain collagen (Figure 3b). However, the carbon and nitrogen weight percent values of the <30kDa fractions are much smaller than those of the >30kDa fractions, 31.0% and 10.3% for carbon and nitrogen respectively. These weight percentage values indicate poorly preserved collagen (van Klinken 1999). Furthermore, the yield for the <30 kDa fraction is 12.1%. The <30 kDa fraction C/N ratios of 3.6 are compatible with a protein origin and the lower pretreatment yield indicates the gelatinized collagen contains only a minor fraction of low molecular weight proteins. The FTIR inferred average C/P and N/P ratios for the >30 kDa fraction are  $2.83 \pm 0.02$  and  $6.04 \pm 0.04$  respectively, whereas the low molecular weight fraction yields an average C/P ratio of  $2.72 \pm 0.03$  and an average N/P ratio of  $5.4 \pm 0.4$  (Table 1). Even though these differences in ratios are small it appears that the high molecular weight fraction contains less phosphate and/or higher carbonate and nitrogen (Amide I). Phosphate is dominantly derived from the bio-apatite fraction of the bone and hence a low phosphate content is preferred in the extracted collagen fraction as this would indicate better removal of the inorganic bone fraction. The lower carbon and nitrogen weight percentages together with the C/P and N/P ratios, however, suggest that the low molecular weight fraction is constitutionally different from the collagen fraction contained in the >30 kDa

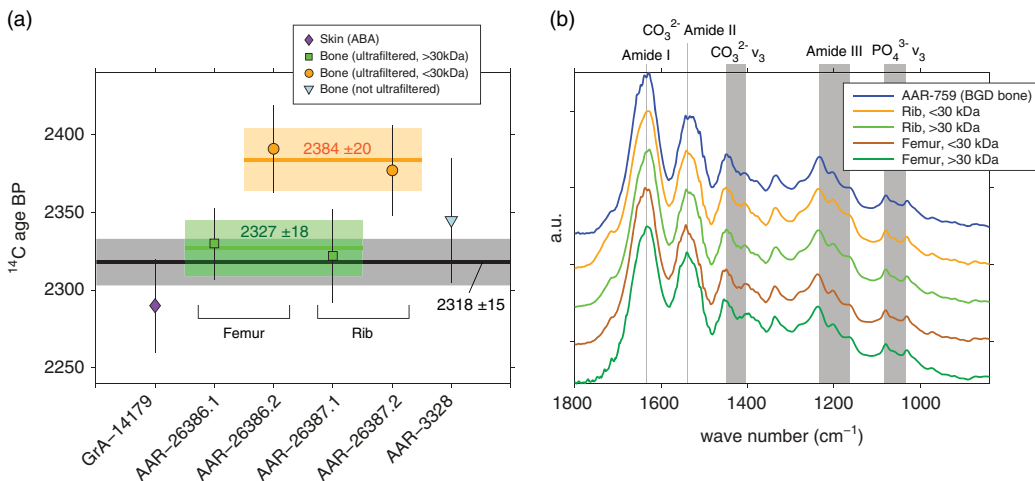


Figure 3 (a)  $^{14}\text{C}$  ages of the femur and rib samples together with previously published skin and bone AMS  $^{14}\text{C}$  ages. The weighted average  $^{14}\text{C}$  ages for the >30kDa fractions ( $2327 \pm 18$  BP) and <30kDa fractions ( $2384 \pm 20$  BP), respectively, are also shown together with the weighted average of the skin  $^{14}\text{C}$  age (GrA-14179) and the >30kDa  $^{14}\text{C}$  ages (AAR-26386.1 and AAR-26387.1) yielding  $2318 \pm 15$  BP. (b) FTIR absorbance spectra for the >30kDa and <30kDa fractions. For reference the >30kDa laboratory background whale bone FTIR spectrum is also shown (BGD bone).

fraction (Table 1). As the <30 kDa fraction usually is discarded, it is difficult to find references for comparison.

Interestingly, the isotopic signature of the low molecular weight fraction (<30 kDa) appears to be similar to bone collagen (>30 kDa) isotopic values (Table 1). If anything, the  $\delta^{13}\text{C}$  values of the <30 kDa fraction are slightly higher by ca. 0.9‰ than the values of the >30 kDa fraction, while the  $\delta^{15}\text{N}$  values of the <30 kDa fraction are lower by c. 0.3‰ than those of the >30 kDa fractions. Given the analytical precision of  $\pm 0.2\text{‰}$  the  $\delta^{13}\text{C}$  difference of 0.9‰ is probably significant and suggests the low molecular weight carbon to be slightly enriched in  $^{13}\text{C}$ . As humic and fulvic acids have an isotopic  $\delta^{13}\text{C}$  signature closely resembling the plants from which they are derived, it would be expected that terrestrially derived humic and fulvic acids have  $\delta^{13}\text{C}$  values lower than  $-25\text{‰}$ . Thus, it is unlikely that the low molecular weight fraction is contaminated by humic or fulvic acids. Furthermore, humic and fulvic acids would probably have increased the carbon weight percentage if they were present in significant amounts. Likewise, a large amount of terrestrially derived plant amino acids in the low molecular weight fraction would probably have lowered the  $\delta^{15}\text{N}$  values contrary to what is observed. Aquatic plants, on the other hand, may assimilate bicarbonate, which would be  $^{13}\text{C}$  enriched and therefore amino acids derived from bicarbonate assimilating plants would be a probable source to the low molecular weight fraction.  $\delta^{15}\text{N}$  values in aquatic plants are very variable and are not unlikely to have values around 10‰ (Talbot 2001). Of course, it is also possible the low molecular weight protein is dominantly derived from the bone collagen and that the breaking to smaller molecules is caused by microbial activity. The subtle difference in the  $\delta^{13}\text{C}$  values would then have been caused by microbial metabolic fractionation, though it appears peculiar if microbial activity would leave the  $^{15}\text{N}$  values unchanged. However, the data presented here are too few to make any definitive conclusion on the origin.

The  $^{14}\text{C}$  ages of the <30 kDa fraction are older than the >30 kDa fractions (Table 1). The weighted average of the femur and rib <30 kDa fraction is  $2384 \pm 20$  BP and for the >30 kDa fraction the weighted average is  $2327 \pm 18$  BP. Thus, the  $^{14}\text{C}$  age difference between the two fractions can be calculated to  $57 \pm 27$   $^{14}\text{C}$  years (Figure 3a). This corresponds to 2.1 standard deviations away from a 0 year difference, in other words, there is less than 5% chance that the two fractions are equal. Even though this difference is small, it suggests that the low molecular weight fraction contain foreign protein of a slightly different age than the bone collagen contained in the >30 kDa fraction.

The isotope data demonstrate the possibility of contamination of the <30 kDa fraction and the  $^{14}\text{C}$  ages suggests that this might result in older dates. Thus the low molecular weight fraction (<30 kDa) appears different from the collagen fraction (>30 kDa) we will refrain from using the <30 kDa fraction in any further analysis. Furthermore, this may require caution when using previously published  $^{14}\text{C}$  age where the pretreatment methods may have included the low molecular weight fraction. However, due to the small  $^{14}\text{C}$  age difference between the two fractions this effect will be difficult to detect statistically.

### Stable Isotope Analysis

The  $\delta^{13}\text{C}$  values of the bone collagen (>30 kDa) from the femur and the rib are  $-21.5 \pm 0.2\text{‰}$  and  $-21.0 \pm 0.2\text{‰}$ , respectively (see Table 1). The  $\delta^{15}\text{N}$  values are  $10.3 \pm 0.2 \text{‰}$  for the femur and  $10.0 \pm 0.2 \text{‰}$  for the rib. Because the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values agree within uncertainties we cannot determine a significant shift in average diet during adolescence and adulthood of Tollund Man. The  $\delta^{15}\text{N}$  value of the femur is only 0.25‰ higher than that of the rib. Generally, a



higher  $\delta^{15}\text{N}$  value would indicate a higher trophic level, e.g. increased meat or aquatic diets. However, the difference in  $\delta^{15}\text{N}$  values in the rib and femur is considered too small to indicate significant changes in the diet; usually, the shift is about 3–5‰ from one trophic level to the next (cf. Schoeninger and DeNiro 1984; Ambrose 2000). We can therefore conclude that the average diet reflected in the femur is the same as that reflected in the rib.

The  $\delta^{13}\text{C}$  values of ca.  $-21\text{‰}$  in Tollund Man indicate a purely terrestrial diet. Thus, nothing suggests that Tollund Man exploited marine resources during the last years of his life, in contrast to the Viking Age and Medieval individuals that are included as references in this study (Figure 2). From the Viking Age and Medieval periods, fish consumption is attested from various sources, and this fish intake results in a greater variation in  $\delta^{13}\text{C}$  values from individuals of this time (Jørkov 2007; Price et al. 2014). The  $\delta^{15}\text{N}$  values of ca. 10‰ in Tollund Man are higher than in Neolithic individuals (ca. 9.6‰, Fischer et al. 2007) who relied on agricultural products, but lower than in inland Maglemosian (Mesolithic) individuals (ca. 10.7‰, Fischer et al. 2007) who utilized freshwater fish. Although the diet of Tollund Man may have included some freshwater fish, the slightly enhanced  $\delta^{15}\text{N}$  values compared to Neolithic individuals are probably best explained as being caused by the consumption of crops grown on manured fields (cf. Bogaard et al. 2013). This interpretation would be in accordance with the evidence of manuring that we find in the field systems and in isotope values of carbonized grain from this period (Kanstrup et al. 2014; Nielsen and Dalsgaard 2017). The fact that the  $\delta^{15}\text{N}$  values of Tollund Man are lower than those in the samples from later periods is probably partly because

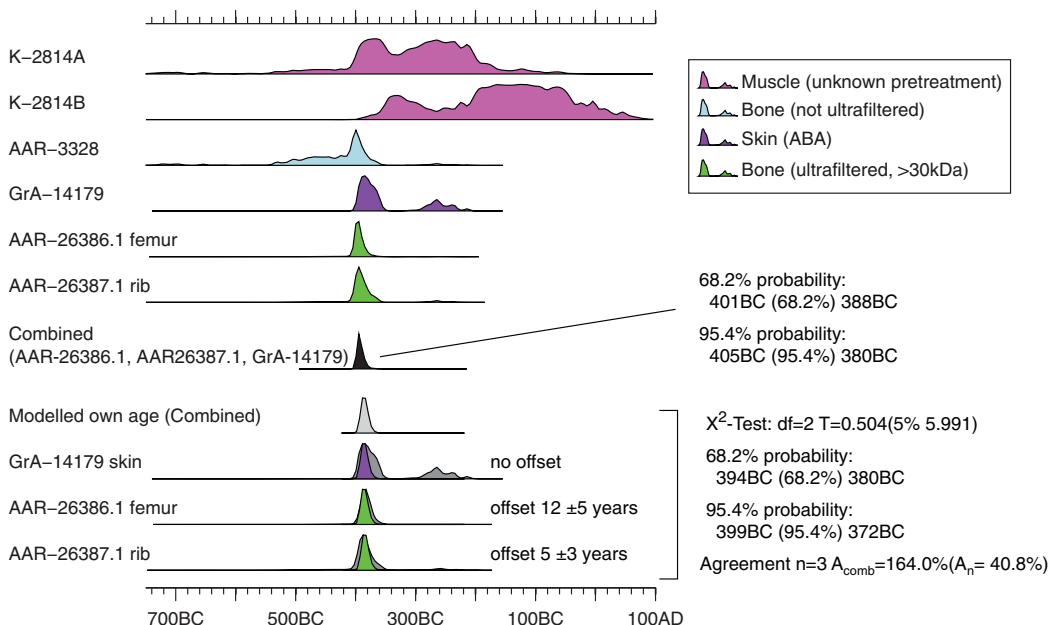


Figure 4 Calibrated probability distributions for selected previous published  $^{14}\text{C}$  ages together with femur and rib (>30kDa) samples from this study. The calibrated probability distribution of the weighted average is also shown as this is considered to be the best age estimate for Tollund Man. The modeled own age calibrated probability distribution is corrected for the bones' inherent age. We assume that the carbon in the ribs is on average  $5 \pm 3$  calendar years old and in the femur  $12 \pm 5$  calendar years (statistics are taken from the OxCal combine function output). See text for details. ABA denotes acid-base-acid pretreatment.

of even more intensive manuring practices in these periods as indicated by higher phosphate levels in the arable soils (e.g. Christensen 1997; Dalsgaard et al. 2000).

### **<sup>14</sup>C Dating**

In an attempt to narrow down the date of the death of Tollund Man, we decided to evaluate whether some of the previous <sup>14</sup>C dates could be combined with the new ones. Since conventional <sup>14</sup>C dates are less precise than AMS dates, and since the sample treatment and possible contaminations are not known in detail, the conventional <sup>14</sup>C dates obtained in 1977 were excluded (K-2812A and K-2814B, Figure 4). When taking a closer look at the four AMS dates (GrA-14179, AAR-3328, AAR-26386.1, and AAR-26387.1 in Table 1 and Figure 4), it becomes evident that the old date on the rib sample (AAR-3328), obtained without ultra-filtration, is about one standard deviation older than the newer AMS dates. This could possibly indicate contamination with older substances, as the indicated by the <30kDa <sup>14</sup>C ages presented here. Therefore, we also excluded AAR-3328 from further analysis. The <sup>14</sup>C age of the skin sample should be more indicative of time of death due to the lower turnover rate (GrA-14179). This date appears reliable and was therefore included in the final calculation of the <sup>14</sup>C age of Tollund Man.

The three AMS dates (GrA-14179, AAR-26386.1, and AAR-26387.1) agree well, and the weighted mean is therefore calculated to  $2318 \pm 15$  BP (Table 1, Figure 4). This narrowed the date of Tollund Man's death to the period 405–380 cal BC at the 95.4% confidence interval (reduced  $\chi^2$  test:  $0.58 \leq 3.00$ , Table 1). Such a precise date, covering an interval of only 25 years, is extremely rare when studying prehistoric human remains. It should be noted that including AAR-3328 in the weighted average yields a <sup>14</sup>C age of  $2320 \pm 15$  BP (reduced  $\chi^2$  test:  $0.53 \leq 2.60$ ) corresponding to a negligible change of the result.

Because the carbon turnover rate in bones is much lower than skin, we tested the effect of the bones having an age offset compared to the skin. As an example, we assume that the carbon in the femur is on average  $12 \pm 5$  calendar years old and the carbon in the ribs  $5 \pm 3$  calendar years based on e.g. the work by Hedges et al. (2007). This was implemented using OxCal's offset function, e.g.:

```
R_Date("AAR-26386_1 femur", 2330, 23)
{
Offset(12,5);
};
```

As a result, the average shifted towards a slightly younger age (Figure 4). However, the shift is only very small. Including the uncertainties associated with collagen-turnover-corrections of the dates, we consider the 405–380 cal BC date to be the most reliable date of Tollund Man's death. Thus, instead of applying an uncertain, speculative correction that almost does not change the date, we choose to report the date without the correction for collagen turnover.

### **Future Perspectives**

The new date of Tollund Man makes one wonder whether new <sup>14</sup>C analyses of Elling Woman, found in the same bog as Tollund Man, could also refine the date of this bog body. Elling Woman is not as well-preserved as Tollund Man and so far AMS <sup>14</sup>C dates have only been undertaken on her fur cape. The cape has been dated to  $2195 \pm 40$  BP (AAR-3415) and

2210 ± 30 BP (GrA14315), i.e. 362–201 cal BC (95.4% confidence interval) when using a weighted mean of 2205 ± 24 BP ( $\chi^2$  test:  $0.09 \leq 3.84$ ). Thus, Elling Woman seems to have been placed in the bog slightly later than Tollund Man. However, a date of 2350 ± 50 BP (GrA-15637), calibrated to 746–232 cal BC (95.4% confidence interval), has also been obtained (van der Plicht et al. 2004). It would therefore be interesting to redate Elling Woman in order to find out whether she was contemporary with Tollund Man, whether the memory of the sacrifice of Tollund Man still existed when Elling Woman was executed, or whether the two sacrifices were performed several generations apart. Ideally, new dates would be conducted on her hair or other body parts that have not received conservation treatment (with among other things organic oils) as the cape has. If Elling Woman is in fact younger than Tollund Man, however, we may not be able to narrow the date of her death any further due to the shape of the calibration curve.

## CONCLUSION

The new  $^{14}\text{C}$  dates of Tollund Man have greatly increased the precision of the calibrated age. This illustrates that it is worthwhile to  $^{14}\text{C}$  date old finds again, especially if laboratory processes have been improved. In this case, ultrafiltration is a useful step in the pretreatment sequence as it appears to remove  $^{14}\text{C}$ -depleted contamination. The precise calibrated age of 405–380 cal BC (95.4% confidence interval), an interval of only 25 calendar years, was possible thanks to a combination of laboratory pretreatment, machine precision, averaging three  $^{14}\text{C}$  samples, and encountering a favorable interval on the calibration curve.

## ACKNOWLEDGMENTS

Conservator Lars Vig Jensen is thanked for extracting the bone samples from the femur and rib of Tollund Man. We would also like to thank Ken Ritchie for improving the English of an earlier version of this article and the anonymous reviewers and the editors for their valuable comments. Bente Philippsen was supported by AU STAR – Science and Technology in Archaeological Research and by the Danish National Research Foundation under the grant D NRF119 – Centre of Excellence for Urban Network Evolutions (UrbNet).

## REFERENCES

- Ambrose SH. 1993. Isotopic analysis of paleodiets: methodological and interpretive considerations. In: Sanford MK, editor. *Investigations of Ancient Human Tissue*. Philadelphia: Gordon and Breach Science Publishers. p 59–130.
- Ambrose SH. 2000. Controlled diet and climate experiments on nitrogen isotope ratios of rats. In: Ambrose, SH, Katzenberg, MA. editors. *Biogeochemical Approaches in Paleodietary Analysis*. New York: Kluwer Academic Publishers/Plenum Publishers. p 243–59.
- Ambrose SH, Krigbaum J. 2003. Bone chemistry and bioarchaeology. *Journal of Anthropological Archaeology* 22:193–9.
- Aaby B 2006. Landskab og Vegetation omkring Bølling Sø siden sidste istid. In: BV Eriksen, editor. *Stenalderstudier. Tidligt mesolitiske jæger og samlere i Sydsandinavien*. Aarhus: Aarhus Universitetsforlag. p 161–74.
- Bevington PR, Robinson DK. 2003. *Data reduction and error analysis for the physical sciences*. 3rd ed. Boston, MA: McGraw-Hill.
- Bogaard A, Fraser R, Heaton THE, Wallace M, Vaiglova P, Charles M, Jones G, Evershed RP, Styring AK, Andersen NH, Arbogast R-M, Bartosiewicz L, Gardeisen A, Kanstrup M, Maier U, Marinova E, Ninov L, Schäfer M, Stephan E. 2013. Crop manuring and intensive land management by Europe's first farmers. *PNAS* 110 (31):12589–94.
- Bonsall C, Cook G, Hedges REM, Higham TFG, Pickard C, Radovanovic I. 2004. Radiocarbon and stable isotope evidence of dietary change from the mesolithic to the middle ages in the iron gates:

- New results from Lepenski Vir. *Radiocarbon* 46 (1):293–300.
- Brock F, Bronk Ramsey C, Higham T. 2007. Quality assurance of ultrafiltered bone dating. *Radiocarbon* 49(2):187–92.
- Brock F, Geoghegan V, Thomas B, Jurkschat K, Higham TFG. 2013. Analysis of bone “collagen” extraction products for radiocarbon dating. *Radiocarbon* 55(2):445–63.
- Bronk Ramsey C. 2009. Bayesian analysis of radiocarbon dates. *Radiocarbon* 51(1):337–60.
- Bronk Ramsey C, Hgman T, Bowles A, Hedges REM. 2004. Improvements to the pretreatment of bone at Oxford. *Radiocarbon* 46 (1):155–63.
- Brown TA, Nelson DE, Vogel JC, Southon JR. 1988. Improved collagen extraction by improved Longin method. *Radiocarbon* 30(2):171–7.
- Christensen PM. 1997. Agerbrug i vikingetiden – et studie af fosfor i jorden. *GeologiskNyt* 4:12–5.
- Dalsgaard K, Karlsen AD, Larsen L. 2000. Bonden og agerjorden. In: Dalsgaard K, Eriksen P, Jensen JV, Rømer JR, editors. *Mellem hav og hede. Landskab og bebyggelse i Ulfjord herred indtil 1700*. Aarhus: Aarhus Universitetsforlag, p. 87–103.
- DeNiro MJ. 1985. Postmortem preservation and alteration of *in vivo* bone collagen isotope ratios in relation to palaeodietary reconstruction. *Nature* 317:806–9.
- Eriksson G. 2004. Part-time farmers or hard-core sealers? Västerbjers studied by means of stable isotope analysis. *Journal of Anthropological Archaeology* 23(2):135–62.
- Fernandes R, Grootes P, Nadeau M-J, Nehlich O. 2015. Quantitative diet reconstruction of a Neolithic population using a Bayesian mixing model (FRUITS): The case study of Ostorf (Germany). *American Journal of Physical Anthropology* 158 (2):325–40.
- Fischer A, Olsen J, Richards M, Heinemeier J, Sveinbjörnsdóttir ÁE, Bennike P. 2007. Coast-inland mobility and diet in the Danish Mesolithic and Neolithic: evidence from stable isotope values of humans and dogs. *Journal of Archaeological Science* 34:2125–50.
- Fischer C. 1980. Moseligene fra Bjældskovdal. *KUML. Årbog for Jysk Arkæologisk Selskab* 1979:7–44.
- Fischer C. 2012. *Tollund Man. Gift to the Gods*. Stroud: The History Press.
- Frei KM. 2017. Preliminary Strontium report: Tollund Man’s last years and months [unpublished report]. National Museum of Denmark.
- Glob PV. 1969. *The Bog People: Iron-Age Man Preserved*. London: Faber and Faber.
- Hedges RE, Clement JG, Thomas CDL, O’Connell TC. 2007. Collagen turnover in the adult femoral mid-shaft: Modeled from anthropogenic radiocarbon tracer measurements. *American Journal of Physical Anthropology* 133(2):808–16.
- Helbæk H. 1950. Botanical study of the stomach contents of the Tollund Man. *Aarbøger for Nordisk Oldkyndighed og Historie* 1950:329–41.
- Jørkov MLS. 2007. Drinking with the rich and dining with the poor in Roman Iron Age Denmark: A dietary and methodological study based on stable isotope analysis [unpublished PhD thesis]. Copenhagen University.
- Kanstrup M. 2008. Gastronomy – norm and variation. Dietary studies based on isotope analyses of skeletal remains from Viking Age graves at Galgedil on Northern Funen [unpublished MA dissertation]. Aarhus University.
- Kanstrup M, Holst MK, Jensen PM, Thomsen IK, Christensen BT. 2014. Searching for long-term trends in prehistoric manuring practice.  $\delta^{15}\text{N}$  analyses of charred cereal grains from the 4th to the 1st millennium BC. *Journal of Archaeological Science* 51:115–25.
- Longin R. 1971. New method of collagen extraction for radiocarbon dating. *Nature* 230:241–2.
- Mannering U, Possnert G, Heinemeier J, Gleba M. 2010. Dating Danish textiles and skins from bog finds by means of  $^{14}\text{C}$  AMS. *Journal of Archaeological Science* 37:261–8.
- Nielsen NH. 2017. Øget fokus på forskning inden for arkæologien. *Museum Silkeborg Årsskrift* 2016:29–34.
- Nielsen NH. 2018. Nyt fra den arkæologiske forskning 2017. *Museum Silkeborg Årsskrift* 2017:39–46.
- Nielsen NH, Dalsgaard K. 2017. Dynamics of Celtic fields – a geoarchaeological investigation of Øster Lem Hede, western Jutland, Denmark. *Geoarchaeology – An International Journal* 32(3):414–34.
- Olsen J, Heinemeier J. 2009. AMS dating of human bone from the Ostorf cemetery in the light of new information on dietary habits and freshwater reservoir effects. *Berichte der RGK* 88:309–22.
- Olsen J, Tikhomirov D, Grosen C, Heinemeier J, Klein M. 2016. Radiocarbon analysis on the new AARAMS 1MV Tandetron. *Radiocarbon* 59 (3):905–13.
- Philippsen B. 2013. The freshwater reservoir effect in radiocarbon dating. *Heritage Science* 1(24). Available at <http://www.heritagesciencejournal.com/content/1/1/24>
- Price TD, Prangsgaard K, Kanstrup M, Bennike P, Frei KM. 2014. Galgedil: isotopic studies of a Viking cemetery on the Danish island of Funen, AD 800–1050. *Danish Journal of Archaeology* 3 (2):129–44.
- Reimer PJ, Bard E, Bayliss A, Beck JW, Blackwell PG, Bronk Ramsey C, Buck CE, Cheng H, Edwards RL, Friedrich M, Grootes PM, Guilderson TP, Hafliðason H, Hajdas I, Hatté C, Heaton TJ, Hoffmann DL, Hogg AG, Hughen KA, Kaiser KF, Kromer B, Manning SW, Niu M, Reimer RW, Richards DA, Scott EM, Southon JR, Staff RA, Turney CSM, van der Plicht J. 2013. IntCal13 and Marine13 radiocarbon age

- calibration curves 0–50,000 years cal BP. *Radiocarbon* 55(4):1869–87.
- Santos GM, Southon JR, Griffin S, Beupre SR, Druffel ERM. 2007. Ultra small-mass AMS  $^{14}\text{C}$  sample preparation and analyses at KCCAMS/UCI Facility. *Nuclear Instruments & Methods in Physics Research B* 259:293–302.
- Schoeninger M, DeNiro MJ. 1984. Nitrogen and carbon isotopic composition of bone collagen from marine and terrestrial animals. *Geochimica et Cosmochimica Acta* 48:625–39.
- Stuiver M, Polach HA. 1977. Discussion: reporting of  $^{14}\text{C}$  data. *Radiocarbon* 19(3):355–63.
- Talbot MR. 2001. Nitrogen isotopes in palaeolimnology. In: Last WM, Smol JP, editors. *Tracking Environmental Change Using Lake Sediments*. Dordrecht: Kluwer Academic Publishers. p 401–39.
- Tauber H. 1980. Kulstof-14 datering af moselig. *KUML. Årbog for Jysk Arkaologisk Selskab* 1979:73–8.
- Thorvildsen K. 1951. Moseliget fra Tollund. *Årbøger for nordisk oldkyndighed og historie* 1950:302–10.
- van der Plicht J, van der Sanden WAB, Aerts AT, Streuerman HJ. 2004. Dating bog bodies by means of  $^{14}\text{C}$ -AMS. *Journal of Archaeological Science* 31:471–91.
- van der Sanden WAB. 2013. Bog bodies: underwater burials, sacrifices and executions. In: Menotti F, O’Sullivan A, editors. *The Oxford Handbook of Wetland Archaeology*. Oxford: Oxford University Press. p 401–16.
- van Klinken GJ. 1999. Bone collagen quality indicators for palaeodietary and radiocarbon measurements. *Journal of Archaeological Science* 26:687–95.
- Vogel JS, Southon JR, Nelson DE, Brown TA. 1984. Performance of catalytically condensed carbon for use in accelerator mass spectrometry. *Nuclear Instruments and Methods in Physics Research B* 5(2):289–93.
- Zanello M, Pallud J, Jacqueline S, Augias A, Varlet P, Devaux B, Nielsen O, Abi Haidar D, Charlier P. 2017. Endogenous fluorescence analysis: preliminary study revealing the potential of this non-invasive method to study mummified samples. *International Journal of Osteoarchaeology* 27:598–605.