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RADIOCARBON DATING CREMATED BONE: A CASE STUDY COMPARING LABORATORY METHODS

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ABSTRACT. Radiocarbon (^{14}C) results on cremated bone are frequently published in high-ranking journals, but ^{14}C laboratories employ different pretreatment methods as they have divergent perceptions of what sources of contaminants might be present. We found pretreatment protocols to vary significantly between three laboratories (Brussels [RICH], Kiel [KIA], and Groningen [CIO]), which all have a long history of dating cremated bone. We present a case study of 6 sets of replicate dates, to compare laboratory pretreatment protocols, and a further 16 sets of inter-laboratory replicate measurements, which compare specific steps of the conversion and measuring process. The ^{14}C results showed dates to be reproducible between the laboratories and consistent with the expected archaeological chronology. We found that differences in pretreatment, conversion to CO_2 and accelerator mass spectrometry (AMS) measurement to have no measurable influence on the majority of obtained results, suggesting that any possible diagenesis was probably restricted to the most soluble $\leq$5% of each sample, as this proportion of the sample mass was removed under all laboratory protocols.

KEYWORDS: comparing laboratory methods, cremated bone, radiocarbon dating, replicate measurements.

INTRODUCTION

Radiocarbon (^{14}C) dating hydroxyapatite (or bio-apatite) from archaeological samples of cremated bone (CB) has become a standard procedure since Lanting et al. (2001). Results are frequently published in high-ranking journals, yet there are still unknowns in the carbon pathways during cremation and burial (Van Strydonck et al. 2005, 2010; Zazzo et al. 2009, 2012; Hüls et al. 2010; Snoeck et al. 2014, 2016). The re-crystallization of CB makes it less susceptible to contamination from the burial environment, but perception of what sources of contaminants might still be present, influence the choice of pretreatment protocol (Van Strydonck et al. 2005, 2009; Olsen et al. 2008). Indeed, pretreatment protocols vary significantly between the three ^{14}C laboratories that contributed to this study, which all have a long history of dating CB. They were among the six laboratories which participated in a cremated bone dating intercomparison (Naysmith et al. 2007), in which the analyzed material came from sites in Holland and Belgium, an area of mainly sandy soil with low carbonate levels (Scheele 2016). In that study, which produced uniform results within measurement errors, one laboratory (Kiel) pretreated the material using either an acetic acid treatment (Lanting et al. 2001; Olsen et al. 2008) or a leaching treatment (De Mulder et al. 2007), but the other laboratories used only variations on the first treatment.

We compare methods of pretreatment, conversion to CO_2 and AMS measurement of CB, employed at the Laboratory for Radiocarbon Dating (RICH) in Brussels (Belgium), the Leibniz Laboratory (KIA) in Kiel (Germany), and the Center for Isotope Research (CIO) in Groningen (The Netherlands). The aim is to test whether any differences in methods have a measurable influence on the obtained ^{14}C results.

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Different types of replicate measurements allow comparison of different steps of the laboratory protocols. Given the inter-laboratory differences in pretreatment protocols, we considered both type 1 replicates (independent dating by two laboratories of the same CB fragment ("true replicates") or of different CB fragments from the same burial context, whose ^{14}C ages are expected to be congruent), and type 2 replicates (conversion and measurement at two laboratories of material pretreated by one of them). Measurements of independent type 1 replicates provide an estimate of the interlab reproducibility, whereas replicating type 2 measurements of pretreated material at more laboratories provide insight into specific steps of the conversion and measuring process. A single grave was measured in duplicate within the same laboratory (KIA), and even though the results were congruent, we do not attempt to assess intra-laboratory reproducibility based on such limited data.

MATERIAL

White and fully calcined bone material was selected by visual inspection, following standards described in Olsen et al. (2008). The material came from two archaeological sites ca. 40 km apart in Southern Jutland, Denmark, both on sandy soil with low carbonate levels (Aarre ca. 8°dH, Aarupgaard ca. 10°dH). Both sites are urnfield cemeteries, spanning several phases of the Pre-Roman Iron Age, ca. 500–150 BC. There are only a few absolute dates available from this period and archaeologists instead rely on typo-chronologies (Jensen 2005). The cremated human remains were interred in ceramic urns and covered by small earthen mounds within a circular ditch. A minority of the graves contained metal artifacts, which can be approximately dated by seriation of typological traits.

Aarre urnfield cemetery is a large and well-documented site, with originally up to 1000 burials. Only three graves are included in this study, as most of the site was excavated in the 1950s, before cremated human remains were routinely stored by archaeological institutions (Becker 1961; Lorange 2015).

Aarupgaard urnfield cemetery is an even larger site, with originally up to 1500 burials. Although there is no direct stratigraphic relationship between any of the burials, typo-chronology shows that the cemetery extended southwards over time. The site was totally excavated in the early 1970s and the cremated remains are today archived at the Laboratory of Forensic Anthropology at University of Copenhagen (Jørgensen 1975; Terkildsen 2015). Seventeen graves are included in this study, spanning the entire typo-chronological sequence (Jensen 2005). Both sites were sampled by a physical anthropologist, ensuring there were not multiple individuals in the selected cremations urns.

METHODS

To confirm that all bone samples were fully calcined, aliquots of powdered untreated CB samples were analyzed by Fourier-transform infrared spectroscopy (FTIR). The crystallinity index (CI) is a measurement of the re-crystallization in a sample, where high CI values indicate high burning temperatures and thus a higher degree of re-crystallization. CI was estimated as the splitting factor between the two absorption band at ca. 603 and ca. 565 cm^{-1} ($CI = (A_{603} + A_{565})/A_{valley}$) (Olsen et al. 2008; Person et al. 1995).

Overall, 58 graves were dated as part of the first author's doctoral project, of which replicates from 16 graves are included in this study. Three rounds of selected samples from 20 graves were sent to the three laboratories involved in this study (Brussels [RICH], Kiel [KIA], and

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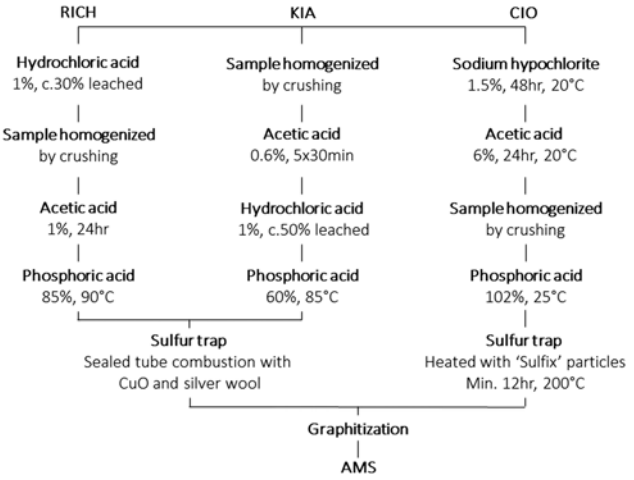

Figure 1 Pretreatment protocols for cremated bone. Laboratory for Radiocarbon Dating (RICH) in Brussels (Belgium), the Leibniz Laboratory (KIA) in Kiel (Germany), and the Center for Isotope Research (CIO) in Groningen (The Netherlands).

Groningen [CIO]), with a few samples replicated between laboratories. CIO uses the traditional acetic acid treatment (Lanting et al. 2001), whereas RICH and KIA use variations of an acid-leaching treatment (Figure 1; De Mulder et al. 2007).

RICH Protocol for Cremated Bone

Initially, ca. 30% by weight of each solid CB sample was leached in 1% hydrochloric acid. The bone was then ground to powder and ca. 1g was treated with 1% acetic acid (24 hr) to remove calcite. CO_2 was extracted from the sample with 85% phosphoric acid (90°C). To remove any sulfur compounds, the CO_2 was heated together with Ag for 30 min at 1000°C. The purified CO_2 was reduced using H_2 and Fe as catalyst and then pressed into targets for AMS measurements (De Mulder et al. 2007; Van Strydonck et al. 2009).

AMS ^{14}C dating was conducted using a Micadas (195.5 kV) AMS system (Boudin et al. 2015). The resulting ^{14}C-content was corrected for fractionation using the simultaneously AMS-measured $^{14}C/^{12}C$ and $^{13}C/^{12}C$ isotope ratios (Stuiver and Polach 1977).

KIA Protocol for Cremated Bone

A 1.5g piece of CB was first crushed and washed 5 × 30 min in 0.6% (0.1M) acetic acid at room temperature to remove any calcite. After repeated rinsing in demineralized water, ca. 50% of the solid sample was leached in hydrochloric acid (10 mL 1% HCl 1h + 1.6mL HCl conc. until pH <1). After washing, drying and weighing, the sample was reacted with 60% phosphoric acid (85°C) to produce CO_2. To remove any sulfur compounds, the CO_2 was sealed in a quartz tube with CuO and silver wool and heated for 4 hr at 900°C. The purified CO_2 was reduced at 600°C using H_2 and iron powder as catalyst and then pressed into targets for AMS measurements (Hüls et al. 2010).

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AMS ^{14}C dating was conducted using a HVE 3MV Tandetron 4130 AMS system (Nadeau et al. 1997). The resulting ^{14}C-content was corrected for fractionation using the simultaneously AMS-measured $^{14}C/^{12}C$ and $^{13}C/^{12}C$ isotope ratios (Stuiver and Polach 1977).

Some samples were pretreated in Groningen following the CIO protocol, but were subsequently converted to CO_2 and dated in Kiel. They followed the KIA protocol from the phosphoric acid step onwards.

CIO Protocol for Cremated Bone

1.5% sodium hypochlorite is used to remove organic material from the entire CB sample (48 hr, 20°C), which was then rinsed with decarbonized water. 6% (1M) acetic acid was added to remove readily soluble calcite, absorbed carbonates and the less crystalline fractions of apatite (24 hr, 20°C) (Lanting et al. 2001). The apatite was rinsed with decarbonized water to neutral pH, dried and crushed (~ 2 × 2 mm). CO_2 was extracted from ca. 1800 mg apatite with 102% (1.89 kg/m^3) phosphoric acid (24 hr, 25°C). To remove any sulfur compounds, the CO_2 was heated with "Sulfix" particles (containing Ag_2O and Co_3O_4; WAKO, mesh 8~20; min. 12 hr, 200°C). The purified CO_2 was graphitized with iron as a catalyst and pressed into targets for AMS measurement.

AMS ^{14}C dating was conducted using the previous HVEE 4130 2.5 MV Tandetron AMS system (Wijma et al. 1996) and the present Micadas (180 kV) AMS system, which is in operation since 2017 (Aerts-Bijma et al., in prep.). The resulting ^{14}C-content was corrected for fractionation using the simultaneously AMS-measured $^{14}C/^{12}C$ and $^{13}C/^{12}C$ isotope ratios (Stuiver and Polach 1977).

RESULTS

Initially, CIO results were often significantly younger than results from RICH and KIA, and inconsistent with the expected chronology of the Aarupgaard cemetery. Results from RICH and KIA were in good agreement with each other, and with the expected chronology, thus an anomaly was suspected in the CIO dating process, leading to offsets of 100–300 years. The anomaly was apparently unrelated to AMS measurement, as it occurred in measurements performed on both the previous HVEE AMS and the present Micadas system. Through laboratory testing at CIO and KIA, the source of the anomaly was identified as a contaminated batch of "Sulfix" in the period April 2017–March 2018 (see supplemental Appendix 1). The CIO laboratory protocol was updated following the test results, and this paper only reports results obtained using the updated protocol.

A total of 43 ^{14}C results on 20 samples are reported in this paper. Summary results on type 1 replicates are given in Table 1 and replicate sets of ^{14}C measurements are shown in Figure 2. Summary results on type 2 replicates are given in Table 2 and replicate pairs of ^{14}C measurements are shown in Figure 3 (see supplemental Appendix 2 for full details).

Replicate measurements have been tested for consistency and weighted means calculated as described by Ward and Wilson (1978) using the R_Combine function in OxCal (Bronk Ramsey 1995). The function can be used where dates arise from the same event (within the resolution of the calibration curve, <5 yr), rather than just from the same sample. This certainly applies to the type 2 replicate measurements on apatite pretreated by CIO and type 1 "true

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Table 1 ^{14}C results, summary statistics and χ^2 test results on type 1 replicates from Aarupgaard.

Sample ID	KIA	RICH	CIO	Absolute difference (yr)	Sigma difference[1]	Weighted mean (BP)	χ^2test
Grave 3869	2456 ± 25*	2504 ± 26	2540 ± 20*	—	—	2507 ± 14	6.9[2]
Grave 3330	2503 ± 27	—	2535 ± 20[3]	32 ± 34	0.9	2524 ± 17	0.9[4]
Grave 51	2448 ± 26*	—	2445 ± 20*	3 ± 33	0.1	2446 ± 16	0.0[4]
Grave 1076	—	2198 ± 27	2260 ± 20	62 ± 34	1.8	2246 ± 14	4.0[4]
Grave 1847	2228 ± 25*	—	2255 ± 20*	27 ± 32	0.8	2244 ± 16	0.7[4]
Grave 1791	2167 ± 25*	—	2230 ± 25*	63 ± 35	1.8	2199 ± 18	3.2[4]

*"True replicates."
[1]The absolute difference (yr) divided by its uncertainty.
[2]T'(5%) = 6.0, df = 2.
[3]Date used as both type 1 and 2 replicate.
[4]T'(5%) = 3.8, df = 1.

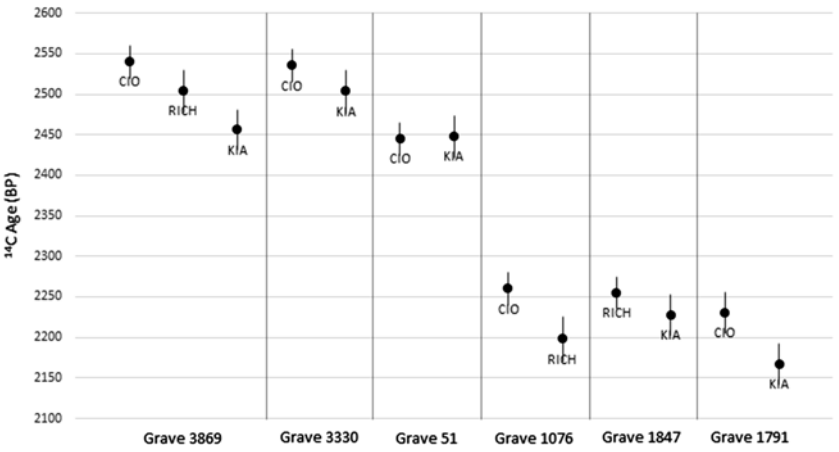

Figure 2 ^{14}C results (±1σ) on type 1 replicate dates from Aarupgaard.

replicates," and we argue that it also applies to the remaining type 1 replicate measurements, as we expect different CB fragments from the same burial context to have congruent ^{14}C ages.

Four out of six pairs of results on type 1 replicates pass the Ward and Wilson (1978) χ^2 test, i.e. are statistically consistent at the 5% significance level. CIO results are on average slightly older than RICH and KIA results, as are RICH results compared to KIA results. Grave 1076 results narrowly fail the χ^2 test (T= 4.0, T'(5%)=3.8, ν=1), but would be regarded as consistent based on the traditional formula whereby their difference is less than 2σ (twice the uncertainty in the

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Table 2 ^{14}C results, summary statistics and χ^2 test results on type 2 replicates.

Sample ID	CIO	KIA	Absolute difference (yr)	Sigma difference[1]	Weighted mean (BP)	χ^2 test[2]
Aarre, grave A394, x785	2465 ± 18	2422 ± 20	46 ± 37	1.2	2446 ± 14	2.6
Aarre, grave A117, x762	2445 ± 20	2416 ± 20	29 ± 28	1.0	2431 ± 15	1.1
Aarre, grave A281, x484	2320 ± 20	2271 ± 20	49 ± 28	1.8	2296 ± 15	3.0
Aarupgaard, grave 3330	2535 ± 20[2]	2471 ± 26	64 ± 33	1.9	2511 ± 16	3.8
Aarupgaard, grave 83	2485 ± 30	2409 ± 27	76 ± 40	1.9	2443 ± 21	3.5
Aarupgaard, grave 1186	2465 ± 20	2408 ± 20	57 ± 28	2.0	2437 ± 15	4.1
Aarupgaard, grave 280	2405 ± 20	2402 ± 22	3 ± 30	0.1	2404 ± 15	0.0
Aarupgaard, grave 81	2425 ± 30	2379 ± 22	46 ± 37	1.2	2395 ± 18	1.5
Aarupgaard, grave 230	2546 ± 19	2362 ± 25	184 ± 31	5.9	2480 ± 16	34.1
Aarupgaard, grave 766	2285 ± 20	2252 ± 23	33 ± 30	1.1	2271 ± 16	1.2
Aarupgaard, grave 681	2310 ± 19	2305 ± 20	5 ± 28	0.2	2308 ± 14	0.0
Aarupgaard, grave 1001	2235 ± 20	2253 ± 21	18 ± 29	0.6	2244 ± 15	0.4
Aarupgaard, grave 382	2280 ± 20	2229 ± 25	51 ± 32	1.6	2260 ± 16	2.5
Aarupgaard, grave 1076	2260 ± 20[2]	2199 ± 29	61 ± 35	1.7	2240 ± 17	3.0
Aarupgaard, grave 1363	2225 ± 20	2195 ± 24	30 ± 31	1.0	2213 ± 16	0.9
Aarupgaard, grave 2262	2255 ± 25	2214 ± 28	41 ± 38	1.1	2237 ± 19	1.2

[1]The absolute difference (yr) divided by its uncertainty.
[2]T'(5%) = 3.8, df = 1.

difference). A slight discrepancy like this might possibly reflect an inhomogeneity of ^{14}C ages in the two dated CB fragments, caused by an uneven influence of "old-wood" from the pyre-fuel (Olsen et al. 2013). Grave 3869 results narrowly fail the χ^2 test (T= 6.9, T'(5%)=6.0, ν=2), whereas if the test is limited to results from either KIA and RICH (T= 1.8, T'(5%)=3.8, ν=1) or RICH and CIO (T= 1.8, T'(5%)=3.8, ν=1) it is acceptable.

Fifteen out of 16 pairs of results on type 2 replicates are also statistically consistent at the 5% significance level. One or both dates on grave 230 must however be an outlier (T= 34.1, T'(5%)=3.8, ν=1, difference= 5.9σ), which cannot be explained by differences in wood-age offset, as the sample was homogenized by crushing before being split between laboratories.

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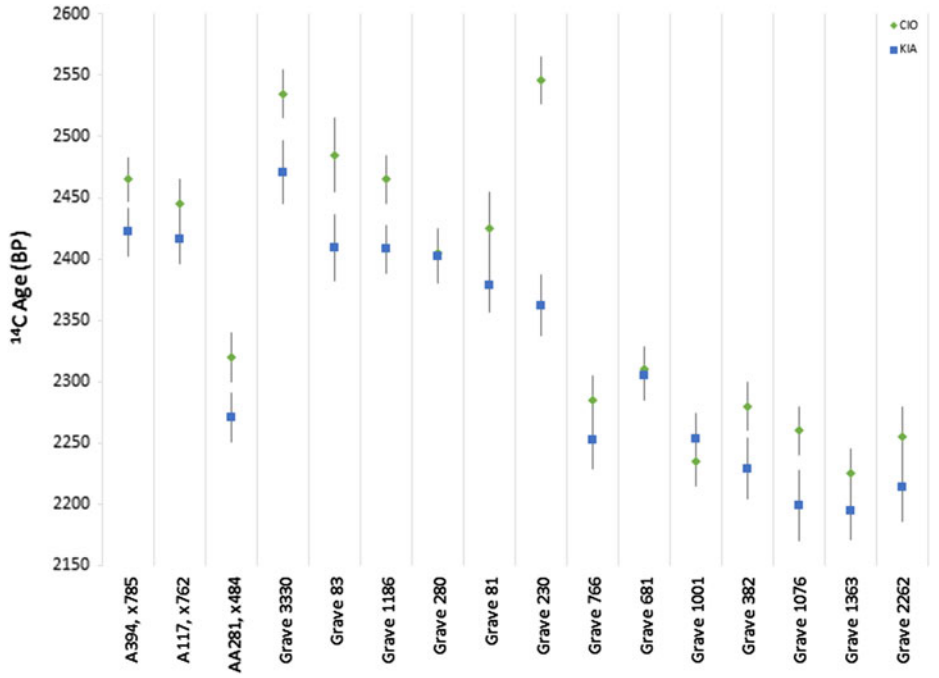

Figure 3 ^{14}C results (± 1σ) on type 2 replicate dates from Aarre and Aarupgaard.

The KIA date fits the expected age range, whereas the CIO date is well back in the Late Bronze Age and can thus be rejected, based on the archaeological information. We do however have no technical explanation for this difference, with δ^{13}C and %C values from both laboratories within the normal ranges.

Stable isotope values on δ^{13}C (measured by AMS) are plotted against ^{14}C ages in Figure 4. CIO has a slight tendency towards lower δ^{13}C values compared to KIA, which could indicate incomplete conversion, but here probably reflects use of different AMS systems and does not account for the pattern of slightly higher ^{14}C ages at CIO. %C results are plotted against ^{14}C ages in Figure 5. Overall CIO has lower %C values (mean= 0.09%) than KIA (mean= 0.23%) and RICH values fall in between (mean= 0.16%, n= 2). Differences in %C do however not appear to be correlated with ^{14}C ages.

DISCUSSION

The reported results from Aarupgaard, with the CIO date for grave 230 as an exception, are consistent with the typo-chronological phasing of the burials, based on an overall Bayesian model of the site chronology (Rose, dissertation in prep.). Results on the three graves from Aarre are consistent with the expected ages of the metal artifacts from these burials. Small wood-age offsets affecting the ^{14}C ages of CB cannot be excluded based on typo-chronology alone.

The independent replicate pretreatment and measurement of the same CB fragment or of different CB fragments from the same burial context by two laboratories (type 1 replicates) should allow us to test whether differences in pretreatment have a measurable influence on

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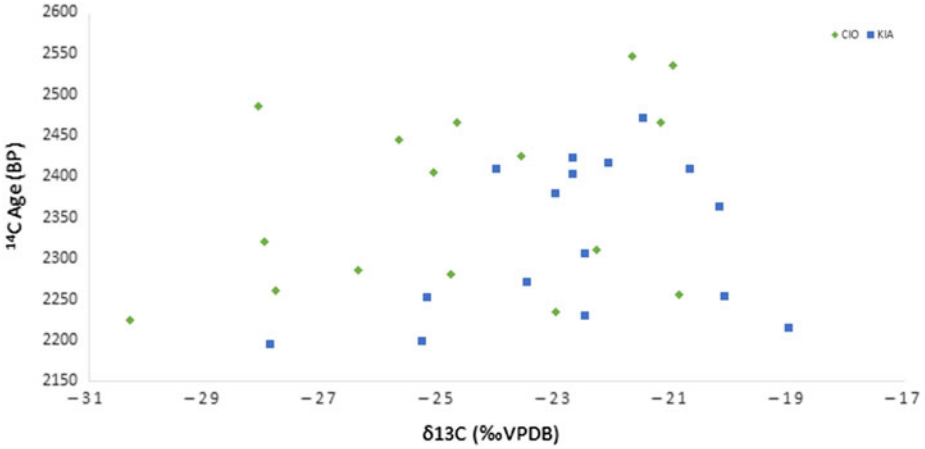

Figure 4 $\delta^{13}C$ (AMS) values and ^{14}C results on type 2 replicate dates from Aarre and Aarupgaard.

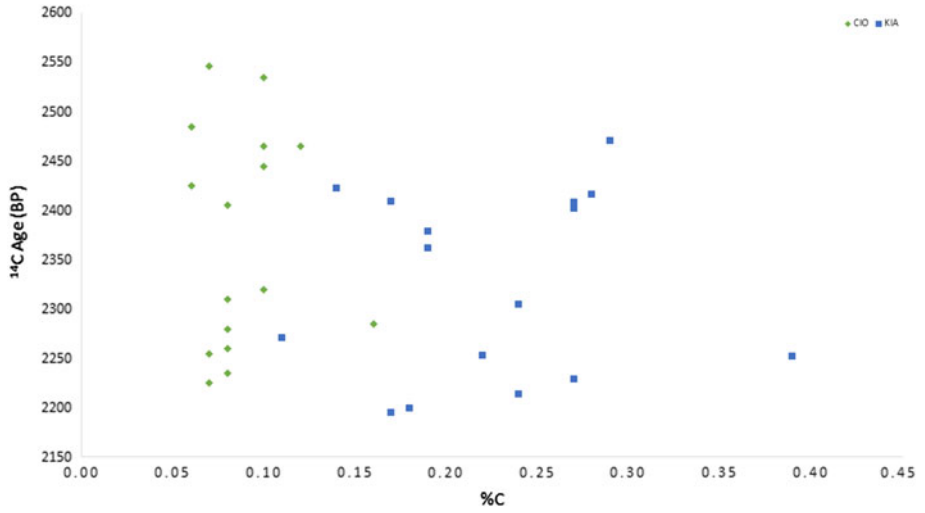

Figure 5 %C values and ^{14}C results on type 2 replicate dates from Aarre and Aarupgaard.

the ^{14}C results. Grave 3869 is the only one to be dated by all three laboratories, but while the three dates are not statistically consistent with a single ^{14}C age, only the KIA and CIO measurements are significantly different to each other. The chi-test statistic is slightly too high for the three results to be considered accurate measurements of the same ^{14}C age, but whether this means that the errors are slightly underestimated or whether there is a real difference between the ^{14}C ages of the extracts (e.g. due to differences in pretreatment) is hard to answer. It is difficult to discern a clear pattern from a single case, but we find it interesting that CIO, which removed only 3% of the sample mass during pretreatment, produced the oldest date, RICH removed 36.5% and produced a younger date, whereas KIA, which removed 41.2%, produced the youngest date, which incidentally also fits the archaeological chronology best (Rose, dissertation in prep.). There is no clear linear relationship between % material removed and ^{14}C ages and we do not know if even younger results would be obtained if

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even more material was removed. Applying the logic of mortar dating, the youngest (KIA) date would be the most reliable, but we do not know if this is the true date of the sample as we have too few measurements to observe the necessary plateau of ^{14}C ages (Lindroos et al. 2007). Results on the remaining five type 1 replicates are consistent (differences less than 2σ) and demonstrate ^{14}C results on CB to be reproducible between the laboratories. Yet, we note that ^{14}C ages appear to fit the same order of laboratories as in grave 3869, and we therefore cannot rule out that some post-depositional contamination was not completely removed under all laboratory protocols, but to investigate this further would necessitate a pretreatment protocol with stepwise etching (Van Strydonck et al. 2009).

CIO assumes apatite to be resistant to contamination and uses the least aggressive pretreatment method, removing calcite, absorbed carbonates and the less crystalline fractions of apatite (Van Strydonck et al. 2005, 2009; Olsen et al. 2008). KIA and RICH assume the apatite might also be diagenetically altered and consequently etch the outer 30–50% of the samples. The assumption is that any diagenetic carbon substitution must be greater near the surface of a bone. Our results cannot be used to test whether apatite can indeed be contaminated, but it is possible that any diagenesis was mainly restricted to the most soluble $\leq$5% of the sample, as this proportion of the samples mass was removed under all protocols. If diagenesis went beyond this $\leq$5%, we would expect the RICH and KIA dates to fit the archaeological chronology better than CIO dates, which is not the case (Rose, dissertation in prep.). The material comes from low carbonate burial environments, but we expect material from carbonate-rich environments might need different pretreatment (Van Strydonck et al. 2009).

All three laboratories use acetic acid to dissolve calcite, but add it at different points in the process and at different concentrations, temperatures and reaction times. After an initial sodium hypochlorite treatment, CIO treats the solid bone with acetic acid for 24 hr, thus targeting the surface of the sample along with the surface in the voids of the CB. KIA starts the pretreatment with acetic acid, but only washes the crushed bone for 5×30 min. RICH on the other hand uses acetic acid for 24 hr, but only after first leaching and grinding the sample. These differences in method result in varying weight losses: KIA removed less than 0.5% of the starting weights, CIO removed $\leq$5% and RICH removed ca. 15% (see supplemental Appendix 2 for full details). The comparatively high removal percentage by RICH, even though CIO uses a higher concentration of acetic acid, suggests that the increased surface area due to grinding is the decisive factor. These differences also suggest that much of the material dissolved during the 24 hr acetic acid treatments at CIO and RICH was apatite, not calcite.

The overall consistent results on type 1 replicate dates from the three laboratories indicate that CIO's bleaching and acetic acid pretreatment was probably sufficient in most cases. The results do not show whether the weaker acetic acid wash in KIA would also be adequate in this case. Also, the opposite order of the acetic acid and the hydrochloric acid steps between KIA and RICH appears to have no influence on the results. Both outcomes would be expected, of course, if the samples were only contaminated by secondary calcite, without diagenetic alteration of apatite (or if diagenetically altered apatite was soluble in 6% acetic acid).

Type 2 replicate measurements were introduced to the study when anomalously younger results from Groningen were observed. Apatite pretreated by CIO was sent to KIA for CO_2 extraction and AMS dating, allowing us to focus specifically on differences in the conversion and measuring processes between CIO and KIA. The results show that the differences in these processes between the Groningen and Kiel laboratories did not significantly affect the ^{14}C

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results. CIO results are on average slightly older than KIA results, but differences in blank correction would account for most of this pattern; the KIA results are calculated using KIA's blank correction, but as these samples were pretreated at CIO, the CIO blank correction is perhaps more appropriate.

There are no agreed indicators to assess sample quality when ^{14}C dating CB, but quality nevertheless needs to be considered when judging if results are reliable. It is already common to test whether CB is fully calcined, but we suggest also reporting type of burial environment along with any other risk of contamination (ex. chalk manuring). Experimental studies have shown values of $\delta^{13}C$ to be highly influenced, although to variable degrees, by fuel source, cremation temperature and duration (Van Strydonck et al. 2005; Zazzo et al. 2009, 2012; Hüls et al. 2010; Snoeck et al. 2016), but we expect $\delta^{13}C$ values from a single site, or from sites that used similar cremation techniques, to cluster. A greater scatter may be expected for results measured by AMS rather than IRMS, but clear outliers will need further investigation. Comparing infrared spectroscopy (FTIR) and x-ray diffractometry (XRD) spectra at different stages in pretreatment, and giving more weight to results from samples whose spectra do not change much between pretreatment steps might here prove useful, but it needs to be further investigated. Inter-laboratory replication may be a useful approach to detecting contamination problems, particularly when there are significant differences in laboratory protocols; the consistency of results based on more or less aggressive pretreatments helps to validate the dates obtained.

CONCLUSION

We have replicated ^{14}C dates of CB at three laboratories and shown differences in pretreatment, conversion to CO_2 and AMS measurement to have no measurable influence on the majority of obtained results. The material comes from low carbonate burial environments and except for material from a single context any possible diagenesis was probably restricted to the more soluble $\leq 5\%$ of the samples, as this proportion of the samples mass was removed under all laboratory protocols. The ^{14}C results are reproducible and consistent with the expected archaeological chronology.

SUPPLEMENTARY MATERIAL

To view supplementary material for this article, please visit https://doi.org/10.1017/RDC.2019.70

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