ROMAN RUINS AS AN EXPERIMENT FOR RADIOCARBON DATING OF MORTAR

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ABSTRACT. The remains of Vindonissa, the Roman legionary camp in Switzerland, have been the subject of extensive archaeological studies. Knowledge of the building time plays a role in reconstructions of the history of this site. We radiocarbon dated mortar samples selected from one of the Roman monuments (Westtor) as well as a nearby Medieval monastery. ^{14}C ages obtained on the first fraction and second fraction of very short dissolution appear close to the expected Roman age of ~2000 BP, while the monastery is dated to historic times, after AD 1308.

INTRODUCTION

Clay and lime mortars, which are cementation materials used in between bricks, have been produced for the last 10 millennia. Lime mortars are of interest to radiocarbon dating because the CO_2 absorbed by the calcium hydroxide during the binding time is preserved in the form of calcium carbonates and provides material with a potential for dating the time of construction (see the reviews of Hale et al. 2003; Ringbom et al. 2011). Until recently, mostly non-hydraulic lime mortars were used for ^{14}C dating and various methods were developed to obtain accurate ages. Roman mortars (*pozzolana*) are classified as hydraulic type and are considered to be more challenging to ^{14}C date.

Typically, Roman sites are dated quite precisely, often with the help of coins and inscriptions or organic remains such as wood, charcoal, leather, and bones that can also be used for ^{14}C dating if required. Frequently, however, questions concerning the time of construction cannot be resolved by ^{14}C dating of wood or charcoal either due to the lack of those materials or the effect of age inheritance (old-wood effect). Mortars provide a possibility to pin down the time of construction, i.e. the time of binding of slaked lime with the atmospheric CO_2 and building the carbonates that carry ^{14}C signature of the atmosphere of at the time of construction. Attempts to date mortar were conducted in the early days of ^{14}C dating, when grams of material were still needed (Baxter and Walton 1970; Kedar and Mook 1978). In those studies, the limitations but also the potentials of the material were recognized. The main challenge in dating mortars is the separation of carbonates that were formed by absorption of atmospheric CO_2 at the time of construction from the contamination with old carbonates (unburned geologic calcite) and even more difficult from post-construction binding of CO_2 (young contamination to modern) (Lindroos et al. 2011). The separation methods developed during the last decades have focused on dating of non-hydraulic mortars. These methods are based on observation that the carbonates, which were formed during the binding process (i.e. those that incorporated atmospheric CO_2 contemporary to the building time) are amorphous, have small grain size, and dissolve faster than the geological/rock carbonates, thus avoiding contamination from old carbon that was not completely oxidized while the $CaCO_3$ was burnt. In the case of hydraulic and Roman mortars, also the young contaminants must be taken into account. If grain size and dissolution separation is applied, the carbonates younger than the building cannot be effectively separated, although sequential dissolution shows that they react even faster than the real component (Lindroos et al. 2011). Moreover, often the fillers that were added to the Roman mortars carry old carbon (limestone, marble). Additional analyses such as an alkalinity test or cathodoluminescence micros-

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copy (CL) can help judge the potential for ^{14}C dating and/or understand the results of ^{14}C analysis. Alkalinity testing is advised to estimate the possibility of contamination with younger CO_2. Cathodoluminescence microscopy can be conducted to show fractions of amorphous and calcite crystal in the fraction separated for dating (Lindroos et al. 2007; Heinemeier et al. 2010). Here, we present the results of ^{14}C dating of mortar from Roman constructions in Switzerland. The first results show that the narrow age range for the site presents an opportunity and a trial at the same time.

ARCHAEOLOGICAL BACKGROUND OF THIS STUDY

The small towns of Brugg and Windisch (Canton Aargau) are situated 40 km west from Zurich. Nearly all the school kids of Zurich and Aargau know about the Roman ruins of ancient Vindonissa, especially that of the famous Amphitheatre that is located on the outskirts of Windisch. Archaeological finds and systematic excavations were made there since the end of the 19th century. Recent modern excavations document remains of the roman legionary camp and the surrounding civil settlement that was constructed at the beginning of the 1st century AD to be the home basis of the Roman legions XIII Gemina, XXI Rapax, and XI Claudia Pia Fidelis. Among recent discoveries are remains of a Roman road that connected Vindonissa and Augusta Raurica, baths with fragments of wall paintings, a water supply system and plumbing system, as well as ruins of an amphitheatre. Dating of the site is mostly based on artifacts (coins, inscriptions, and ceramics). The object that we attempt to date is the foundation of the main gate of the legionary camp, the *porta principalis dextra* or today known simply as the Westtor of which the construction time is not yet clear (Figure 1).

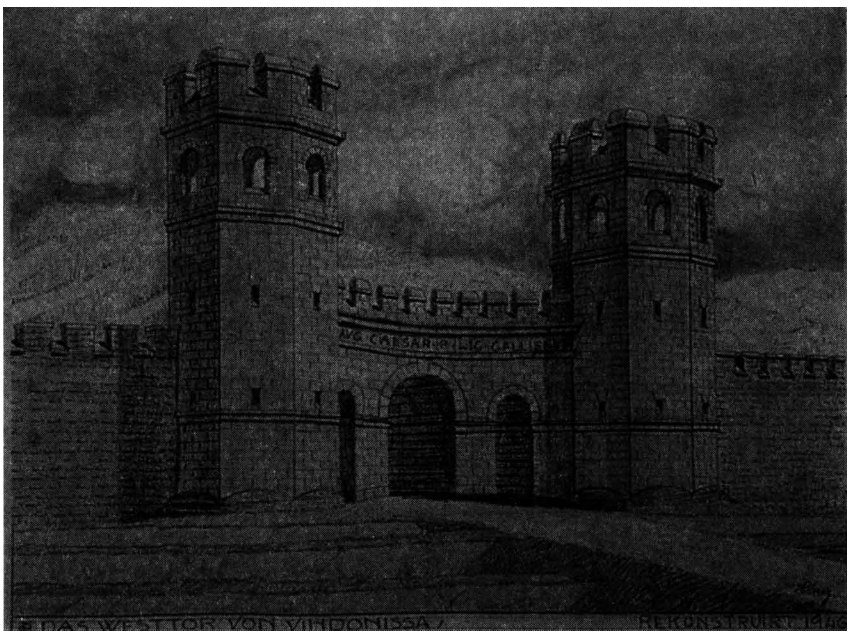

Figure 1 Drawing showing the hypothetical reconstruction of the Westtor of Vindonissa, after Jahresbericht Ges. Pro Vindonissa 1946. [R] Kantonsarchäologie Aargau. Reprinted with permission.

Because of the unusual plan and the exceptional dimensions of the gate, the working hypothesis in 20th century archaeology was that this gate does not belong to the legionary camp of the 1st century, but rather that this gate belongs to a fortified civil settlement of the 3rd century AD (Trumm 2010).

Dating mortar is the only possible way to answer this question, but the challenge is significant for 2 reasons. First, can we really separate the carbonate that was formed while Romans where walking the streets of Vindonissa? And if we can do this, is this sufficient to resolve the age difference between the 1st and 3rd centuries? The archaeology team selected mortar material from the site (Figure 2) and included 2 control samples from the Medieval monastery of Königsfelden, which was built nearby after AD 1308.

Figure 2 Remains of the Westtor of Vindonissa sampled for ^{14}C dating of the mortar samples. Sample FK20 (monastery) is shown and the line of stars represents the samples from the foundation of Westtor (underground, intact mortar). ® Kantonsarchäologie Aargau. Reprinted with permission.

METHODS

In order to obtain the desired ^{14}C age of ~2000 BP, we applied the separation method designed and applied successfully by Heinemeier et al. (2010) and Ringbom et al. (2011). Detailed microscopic, X-ray diffraction, and thermal analyses of 51 samples from Roman mortar from Vindonissa showed the presence of quartz, dolomite, muscovite, and other minerals. Based on the results of X-ray investigations, the fine fraction consists mainly of ~15–50% quartz (SiO_2) and ~25–75% calcite ($CaCO_3$) (Jacobs, forthcoming). All samples taken from Westtor were hydraulic mortar, i.e. with visible additions of rubbed roof tiles. Samples were wet-sieved through various sieves and the smallest fraction (<32 μm) was then used. No additional analyses such as cathodoluminescence microscopy were performed at that point.

Sieved carbonate samples (~50 mg) and acid (85% H_3PO_4) were placed in separated chambers of the glass vessel (Hajdas et al. 2004) and evacuated. Using a flexible metal connector, we mixed the sample with acid and froze the first (1st fraction) and eventually second fraction (2nd fraction) and graphitized those. The remaining material was later (1 hr) transferred for graphitization. Graphite samples were then analyzed for ^{14}C content using the MICADAS system (Synal et al. 2007).

The first dating attempt was made using CO_2 trapped after 10 seconds (s) of reaction time. A sufficient amount of C (~1 mg) was collected for each sample, but the results were clearly too old for this Roman monument. Therefore, a second attempt at dating was realized by sequential dissolution that involved catching CO_2 released during only the first 3 s. In addition, a 2nd fraction of 3-s dissolution was also trapped for graphitization, and a 3rd fraction comprising the remaining material collected after 1 hr was also analyzed.

RESULTS AND DISCUSSION

The results of both dating approaches are summarized in Tables 1 and 2. With the exception of Medieval sample FK20, the first 10-s dissolution fraction for all samples was too old and with a clear limestone δ^{13}C signature of the remaining fraction for all the Roman age samples. This useful indicator of fossil carbon contaminant (Van Strydonck et al. 1986) could not be applied to the results from the second dating campaign due to the clear fractionation effect observed when a very short (3 s) collection time is applied (Table 2). The δ^{13}C values measured on the graphite of the 1st and 2nd fraction might be fractionated due to the freezing of CO_2 during dissolution of the sample and due to incomplete graphitization of those typically very small samples. The first 3-s fraction provided very small (50–100 μg C) samples that were nevertheless graphitized and measured, and the correction for fractionation was applied to the final ^{14}C age using the measured δ^{13}C. In the future, this problem of fractionation due to freezing out of CO_2 at the time of rapid dissolution might be resolved when the method described by Hodgins et al. (2011) is applied. This involves dissolution of the required portion of mortars by adding a specific amount of acid and the collection of a sufficient amount of C (>100 μg C).

Table 1 Results of the first dating approach with a longer dissolution time (10 s) of the first phase. Samples FK20 and FK21 are from the Medieval monastery; all others are from the Roman monument of Westtor ("rest" = material remaining).

Lab nr	Sample code	Fraction	^{14}C age BP	±1σ	δ^{13}C (‰)
ETH-38805	FK 20	1st fraction, 10 s	800	30	–14.2
	FK 20	2nd fraction, rest	1465	30	–6.2
ETH-38806	FK 21	10 s+	1340	35	–11.5
	FK 21	1st fraction, 10 s	965	30	–16.1
	FK 21	2nd fraction, rest	2080	30	–8.6
ETH-38807	FK 22	1st fraction, 10 s	2340	35	–11.8
	FK 22	2nd fraction, rest	3835	35	–4.4
ETH-38808	FK 27	1st fraction, 10 s	3325	35	–19.5
	FK 27	2nd fraction, rest	6990	35	–8.6
ETH-38809	FK 33	1st fraction, 10 s	2680	35	–23.3
	FK 33	2nd fraction, rest	3735	35	–8.2
ETH-38810	FK 37	1st fraction, 10 s	2330	30	–19.5
	FK 37	2nd fraction, rest	3270	35	–6.3
ETH-38811	FK 46	1st fraction, 10 s	2715	35	–30.3
	FK 46	2nd fraction, rest	5090	35	–12.3

Table 1 Results of the first dating approach with a longer dissolution time (10 s) of the first phase. Samples FK20 and FK21 are from the Medieval monastery; all others are from the Roman monument of Westtor ("rest" = material remaining). *(Continued)*

Lab nr	Sample code	Fraction	^{14}C age BP	±1σ	$\delta^{13}C$ (‰)
ETH-38812	FK 64	1st fraction, 10 s	2055	35	–15.0
	FK 64	2nd fraction, rest	3020	35	–6.3
ETH-38813	FK 66	1st fraction, 10 s	3510	35	–17.0
	FK 66	2nd fraction, rest	6985	40	–2.2
ETH-38815	FK 92	1st fraction 10 s	3040	35	–23.0
	FK 92	2nd fraction, rest	6670	35	–7.2

Table 2 Results of the second dating approach with a 3-s dissolution time of the first phase. Sample FK20 is from the Medieval monastery, the others from Roman Westtor ("rest" = material remaining).

Lab nr	Sample code	Fraction	^{14}C age BP	±1σ	$\delta^{13}C$ (‰)
ETH-38805	FK 20	1st fraction, 3 s	700	50	–47.2
	FK 20	2nd fraction, 3 s	725	35	–30.6
	FK 20	3rd fraction, rest	1155	35	–12.2
ETH-38807	FK 22	1st fraction, 3 s	2040	70	–55.2
	FK 22	2nd fraction, 3 s	1940	40	–30.4
	FK 22	3rd fraction, rest	3405	40	–5.1
ETH-38808	FK 27	1st fraction, 3 s	2735	40	–42.4
	FK 27	2nd fraction, 3 s	2925	35	–14.8
	FK 27	3rd fraction, rest	5565	40	–13.7
ETH-38809	FK 33	1st fraction, 3 s	2390	45	–46.9
	FK 33	2nd fraction, 3 s	2520	35	–25.8
	FK 33	3rd fraction, rest	3405	35	–4.7
ETH-38810	FK 37	1st fraction, 3 s	2110	45	–41.5
	FK 37	2nd fraction, 3 s	2055	35	–28.4
	FK 37	3rd fraction, rest	2625	35	–7.6
ETH-38811	FK 46	1st fraction, 3 s	2570	35	–37.3
	FK 46	2nd fraction, 3 s	2555	35	–24.4
	FK 46	3rd fraction, rest	4065	35	0.4
ETH-38812	FK 64	1st fraction, 3 s	2070	65	–58.7
	FK 64	2nd fraction, 3 s	1895	35	–38.7
	FK 64	3rd fraction, rest	2805	35	–2.4
ETH-38813	FK 66	1st fraction, 3 s	2690	35	–37.5
	FK 66	2nd fraction, 3 s	2790	35	–31.3
	FK 66	3rd fraction, rest	4390	35	0.8

Our results summarized in Table 2 show that we partially succeeded in obtaining ages close to 2000 BP. Samples FK 22, FK37, and FK 64 all show the 2nd fraction (3 s) being slightly younger than the first fraction. Such a distribution of ages is rather unique, but ages of the 2nd fractions of 2 samples (FK22 and FK64) are close the expected age for these samples (1940 ± 40 and 1895 ± 35 BP, respectively). Also, the 2nd fraction of sample FK37 is close to 2000 BP but a bit older, i.e. 2055 ± 35 BP. Still, not all of the samples were successfully dated to Roman times. It is worth noting that those are the samples (FK 46 and 66) showing the oldest ages for the remaining fraction in the first attempt (10 s and rest). In the case of these samples, for which the CO_2 was collected after dis-

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solution was completed and fractionation effects were not as obvious as for the 3-s fractions, the $\delta^{13}C$ values are high, suggesting a fossil carbonate component. The combined age of the 1st and 2nd fractions for sample FK 20 (only this sample was repeated with shorter dissolution times) results in a ^{14}C age of 717 ± 29 BP. Calibration with OxCal v 3.10 (Bronk Ramsey 1995, 2001) using the IntCal09 curve (Reimer et al. 2009) shows that our dating is very close to the expected age: the monastery was built shortly after AD 1308, but still on the older side (Figure 3).

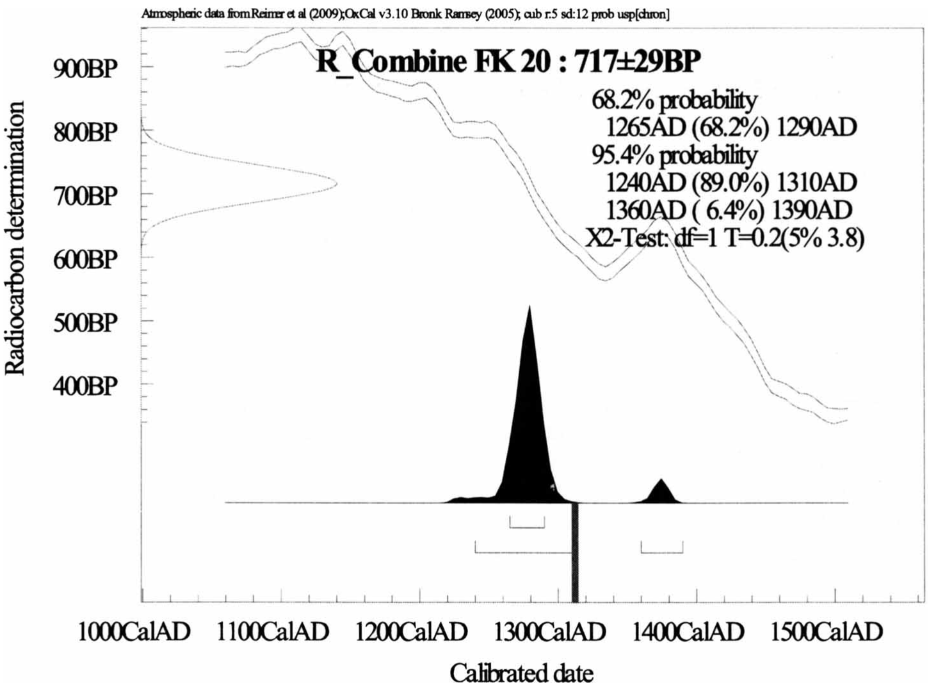

Figure 3 Timing for foundation of the Medieval monastery based on combined ^{14}C ages of the 1st and 2nd fractions of mortar, each collected for 3 s. The historical date is known to be after AD 1308 (vertical line), when King Albert I of Habsburg was murdered and the abbey was founded at the site.

CONCLUSIONS

Our second test for the reliability of ages resulted in ^{14}C ages of the 1st and the 2nd fractions being close to 2000 BP. The results obtained for samples FK22, FK37, and FK64 are still older than the previously postulated 3rd century AD. The early and active phase of the Vindonissa for the construction of the gate seems to be an acceptable explanation. However, before we can definitely exclude a later construction phase, additional analyses are planned on the same samples, including dosing of phosphoric and hydrochloric acids as well as different size fractions. Moreover, we will include cathodoluminescence microscopy as a control for the "quality" of the sieved samples. In addition to our conclusion on the timing of the studied monuments, we have learned that at the time of building the gate the Romans had no fixed procedure for making mortar. They mixed quicklime with gravel/stones from the rivers and chunks of limestone that were in ample supply due to the proximity to the Jura Mountains.

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