INTERCOMPARISON OF ^{14}C DATING OF WOOD SAMPLES AT LUND UNIVERSITY AND ETH-ZURICH AMS FACILITIES: EXTRACTION, GRAPHITIZATION, AND MEASUREMENT

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ABSTRACT. We conducted an interlaboratory comparison between our radiocarbon-related research group at Lund University and the established ETH-Zurich facility to test the quality of the results obtained in Lund and to identify sources of potential background differences and scatter. We did find differences between the 2 laboratories in the contributions of chemical preparation, graphitization, and measurements to the overall background. The resulting overall background is, however, almost similar. Multiple measurements on 2 wood samples of known calendar age yield consistent and accurate ^{14}C ages in both laboratories. However, one of our known samples indicates that IntCal09 is ~38 ± 16 ^{14}C BP too young at 7020 calendar yr BP, which is consistent with one of the raw data sets contributing to IntCal09. Overall, our results show that a systematic approach to compare the different steps involved in ^{14}C age determination is a useful exercise to pinpoint targets for improvement of lab routines and assess interlaboratory differences. These effects do not necessarily become apparent when comparing ^{14}C measurements that integrate over the whole process of preparation and measurement of different laboratories.

INTRODUCTION

Recently, a new radiocarbon research group started operation in parallel to the already existing ^{14}C laboratory in Lund, Sweden (Skog et al. 2010). The group uses its own laboratory facilities and equipment for sample preparation, but measurements are carried out using the same NEC-SSAMS as the commercial laboratory. After an initial phase of optimizing routines and internal quality checks, we conducted an interlaboratory comparison with the ETH-Zurich ^{14}C laboratory to provide quality assurance of our results for future research projects. In addition to the investigation of precision and accuracy, the aim was to determine sources of background contribution in the lab to pinpoint areas for future improvements.

Contrary to the large body of literature examining the effect of i) different chemical pretreatment protocols (e.g. Olsson and Possnert 1992; Santos et al. 2001; Nemec et al. 2010a; Southon and Magana 2010), ii) graphitization catalysts (McNichol et al. 1992; Santos et al. 2007), and iii) measurement techniques (Burleigh et al. 1986; Scott 2003; Sakurai et al. 2004), we tried to synchronize laboratory methods whenever possible. Differences in procedures are mainly a consequence of equipment and infrastructure of the laboratories. This allows us an investigation of mere sample reproducibility between the labs. An exchange of sample material between Lund and Zurich before and after chemical preparation and graphitization facilitates an identification of potential sources of variation between the 2 laboratories in more detail than was possible in the last International Radiocarbon Intercomparison projects (Scott 2003; Scott et al. 2010a,b). To our knowledge, this type of study has not been done before and can potentially contribute to resolving interlaboratory scatter.

METHODS

Sample Material

To assess laboratory backgrounds, we used Lund's in-house wood blank, which is a sample of stratigraphically dated Eemian oak. In addition, background determinations where done on routinely

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used batches of anthracite and coal from both laboratories. For measuring precision and accuracy, 2 samples of dendro-dated wood were chosen. One sample being a single-year pine ring from 435 calendar yr BP (dated at the Laboratory for Dendrochronology, Zurich, Güttler et al. 2013), and 1 decadal pine sample with a central ring dendro-dated to 7021 calendar yr BP (Edvardsson et al. 2012a,b).

Study Design

The raw sample material was chemically extracted to cellulose in both laboratories. Subsequently, aliquots of these cellulose samples were exchanged among Lund and Zurich, and graphitized at both facilities. Finally, also the obtained graphite was exchanged between the laboratories and ^{14}C dated. This procedure allows a direct assessment (e.g. of chemical pretreatment backgrounds) by comparing 2 samples that have been pretreated in either Lund or Zurich, but graphitized and measured exclusively in Zurich. Graphitization background contributions can be determined by analyzing samples that have been, for example, extracted in Lund, graphitized in either Lund or Zurich, and measured in Zurich. Accordingly, machine background differences can also be addressed in the same manner.

Chemical Pretreatment

The wood samples have been pretreated to holocellulose following the BABAB protocol as described in Nemec et al. (2010a). Single steps of the method are shown in Figure 1. Anthracite and coal background samples did not undergo any chemical pretreatment.

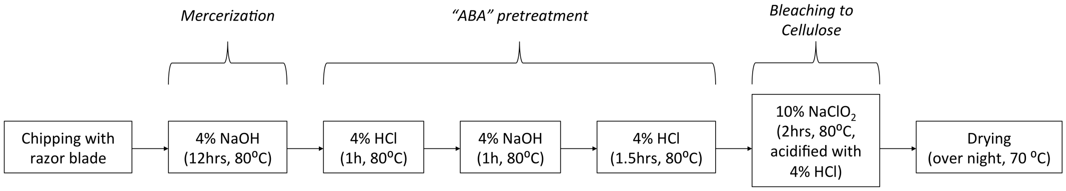

Figure 1 Scheme of the BABAB holocellulose extraction method applied to wood samples. Samples were washed close to neutral in between every step. The last 30 min of the bleaching step were carried out in an ultrasonic bath.

The chemical extractions were carried out by the same person in both labs. Reaction vessels in Lund are Duran glass beakers, which are reused after being cleaned in 4% NaOH (>24 hr), 34% HNO_3 (>24 hr), and deionized water (>24 hr, and flushing multiple times), and dried (>2 hr, 100 °C). Zurich mainly uses plastic centrifuge tubes (Greiner bio-one Polysterene tubes, 15 mL), which are disposed after usage (Güttler et al. 2013). Preparation of chemical solutions and washing of samples is done with deionized water or Milli-Q™ water in Lund and Zurich, respectively.

Graphitization

Before graphitization, the Fe catalyst is cleaned inside the reactors in both laboratories. First, the iron is baked for 3 min in ambient air or 700 mbar O_2 at 500 °C in Zurich and Lund, respectively. Next, the Fe is reduced under H_2 atmosphere (800 mbar, 500 °C, 5 min) twice, followed by a third reduction step (800 mbar H_2, 500 °C, 20 min). The reactors are evacuated in between each of these steps. Zurich uses Alfa Aesar -325 mesh (44 µm), 99% purity Fe, whereas Lund uses Merck 10-µm Fe powder, but recently switched to the Alfa Aesar -325 mesh as well, which was observed to provide a more homogeneous sputter surface throughout the AMS measurement.

Graphitization in Zurich is carried out with the automatic graphitization system "AGE" (Wacker et al. 2010c). The samples are weighed into tin capsules and combusted in an elemental analyzer that transfers solely the CO_2 in helium to the graphitization system. The CO_2 is then trapped on zeolite while the helium carrier gas is removed. The CO_2 is thermally released and transferred to the reactors by gas expansion. The amount of CO_2 is kept constant to provide constant $CO_2/H_2/Fe$ ratios for the graphitization at 580 °C (0.9 mg carbon, 4.2 mg iron, H_2/CO_2 ratio = 2.3). Water formed from the reduction is frozen in a Peltier cooled trap (about –5 °C). The reaction is stopped automatically after 2.2 hr when residual gas pressures are stable. A detailed description of the graphitization conditions in Zurich can be found in Nemec et al. (2010b).

Lund uses the semi-automatic graphitization system "Hamster" (Unkel 2006) and sealed quartz tube combustion. All quartz tubes and the CuO are prebaked, at 850 °C for 3 hr. An aliquot of the sample equal to ~1 mg of carbon is placed in a small quartz glass tube together with 60–100 mg of CuO and a piece of silver wire. This small tube is then placed inside a larger quartz glass tube that is subsequently evacuated and sealed using a torch. The samples are combusted in a muffle oven at 850 °C for 3 hr. Each tube is then cracked inside the graphitization system. The CO_2 is cryogenically purified and loaded onto the reactors. H_2 is added to the reactor to obtain a H_2/CO_2 ratio of 2.3. The graphitization reaction takes place at 580 °C and is stopped when the pressure in the reactor stabilizes after ~4.5 hr. The emerging water vapor from the reduction is captured by a $MgClO_2$ water trap. The different reaction times in Lund and Zurich are caused by the smaller reactor volumes as well as a better gas circulation in Zurich. The latter is achieved by the vertical alignment of the oven underneath the peltier cooling element.

Zurich uses prebaked (500 °C, 3 hr) Duran glass tubes as reaction vessels that are disposed after usage. In Lund, the reaction is conducted in reusable quartz glass tubes inside which the Fe powder is placed on small quartz glass ships to avoid electrostatic dispersion of the catalyst. Before usage, the ships and reactor tubes are cleaned along with regular glassware (see above) and baked at 850 °C for 3 hr.

Measurement

Lund performs ^{14}C measurements on a NEC-SSAMS system. The ion source has been modified from the original NEC design in 2009 following Southon and Santos (2007) and now incorporates a new Cs oven, a spherical ionizer, a cathode immersion lens, and a new extractor. Details on the performance and instrumentation can be found in Skog (2007) and Skog et al. (2010). In October 2011, we changed the stripper gas from N_2 to He (Schulze-König et al. 2011). Zurich uses the MICADAS tandem accelerator for measurement (Synal et al. 2007). A recent description of its performance can be found in Wacker et al. (2010b).

Both facilities use on-line measurements of the $^{13}C/^{12}C$ ratio for normalization and fractionation correction. For AMS data reduction and ^{14}C age calculation, the Bats software (Wacker et al. 2010a) is used. Lund has to correct $^{13}C/^{12}C$ ratios for beam current dependencies in the NEC-SSAMS system (Freeman et al. 2008; Skog et al. 2010) prior to loading the data into Bats.

Statistics

To summarize blank values from multiple measurements, we use the error-weighted mean. However, since blank scatter is often beyond the statistical uncertainty, we use a bootstrapping method (Efron 1979) to infer uncertainties and confidence intervals of the weighted mean. That is, each sample group is randomly resampled with replacement to a group of similar population size and the error-weighted mean is calculated. This procedure is repeated 10,000 times and the standard deviation of the obtained error-weighted means taken at 1σ uncertainty. If the sample variance of the population is solely controlled by normally distributed measurement errors, the obtained standard deviation will match the standard error of the weighted mean. If, however, non-random fluctuations increase the scatter, the "bootstrapped error" will be larger. Confidence intervals around the error-weighted mean can be inferred in a similar way.

Prior to this analysis, the results of the measurements of each cellulose extract were summarized to their respective mean values ±1σ uncertainty using the procedure described above. This is done to avoid biases in the analysis arising from the different number of targets produced from each cellulose extract. Multiple measurements of wood samples of finite ^{14}C ages are reported as error weighted means ±1σ standard error. The reduced χ^2 statistic was used to describe the consistency of multiple ^{14}C age determinations and single measurements within quoted uncertainties.

RESULTS AND DISCUSSION

Overall Background

We find average backgrounds of 45,300 ± 530 BP (0.34 ± 0.07 pMC) and 46,550 ± 590 BP (0.29 ± 0.06 pMC) for wood blank cellulose that has been entirely prepared and measured in Lund and Zurich, respectively. The background measurements in Zurich are in good agreement with earlier studies (Wacker et al. 2010b). Backgrounds achieved in Lund are slightly higher than in Zurich but lower than reported previously for processed anthracite (Skog et al. 2010). The background for the processed blank is also in agreement with Fallon et al. (2010) who use the same NEC-SSAMS. However, Freeman et al. (2008) reach significantly lower background employing the same type of accelerator. In the following sections, we deploy our study design to further investigate potential sources of background contribution.

AMS Machine Background Differences

To assess differences in machine background, we use a single blank cellulose extracted from Eemian oak, prepared and graphitized in Zurich. Measuring 2 targets in Lund and 5 in Zurich we find a lower machine background by 0.14 ± 0.04 pMC in Zurich than in Lund. Comparing Lund and Zurich AMS measurements on anthracite blanks that have been graphitized in Zurich yields a similar difference of 0.08 ± 0.03 pMC. This consistent and significant (99%) difference in AMS machine background can partly be explained by the empirical isobar correction done for the MICADAS (Synal et al. 2007). This correction cannot be performed at the NEC-SSAMS due to the absence of an offset Faraday cup to measure the breakup of ^{13}CH molecules after the high-energy bending magnet.

Graphitization Background Differences

The assessment of background differences induced by the graphitization equipment only is more ambiguous. In a first experiment, blank cellulose samples were prepared in Lund and aliquots of each were graphitized in Lund and Zurich. Subsequently, all obtained targets were pressed and measured in Zurich. The targets graphitized in Lund gave consistently very low beam currents and 2 out of 3 samples had to be removed from the measurement. The low ion beam currents are probably related to the target pressing and, thus, different properties of the targets during sputtering. Lund and Zurich use Fe powder of different grain size (10 and 44 μm, respectively) and Zurich's target pressing method is optimized for their individual iron powder. Hence, we can only compare 1 graphite target from Lund to the mean of 5 targets graphitized in Zurich, indicating lower graphitization backgrounds in Zurich (0.04 ± 0.01 pMC).

A second attempt to derive graphitization background differences was performed by measuring aliquots of 1 cellulose extract prepared in Zurich, graphitized in Zurich and Lund, and measured in Lund. Again, we found large differences in the obtained beam currents from targets graphitized in either Lund or Zurich. However, this time higher beam currents were obtained from targets graphitized in Lund. We can exclude problems with pressing for these targets and assign the observed effect to different sputtering properties due to the grain sizes of the graphitization catalysts. To circumvent this problem, we measured the samples from Lund and Zurich in separate AMS runs and adjusted the ion source in between, so that final beam currents were in the same range for both runs. Comparison of the 2 targets graphitized in Lund to the 2 targets graphitized in Zurich indicated lower graphitization backgrounds in Lund by 0.06 ± 0.02 pMC.

Regarding the complications in both experiments, we cannot quantify the differences in graphitization backgrounds with certainty. Besides the differences in beam currents, a potential bias in the experiment may be introduced by the transport of graphite from one lab to the other, increasing the chance of contamination. In addition, we found that the use of $MgClO_2$ in the Lund graphitization system can introduce a contamination of up to 2 μg Modern carbon if not exchanged frequently. This has been reported earlier, even though the observed contamination was less pronounced (Santos et al. 2004). In between the 2 graphitization background experiments, we started trying to minimize this problem by exchanging the drying agent for every graphitization. Thus, the $MgClO_2$ might be a source of contamination for the first experiment (Zurich measurements) but not for the second experiment (Lund measurements).

Extraction Background Differences

For determination of differences in the background contributions derived from chemical pretreatment, we conducted 2 experiments of similar kind. First, we extracted cellulose from blank wood samples in Lund (6 extractions) and Zurich (5 extractions). These cellulose samples were then graphitized and measured in Zurich multiple times (11 and 22 targets from Lund and Zurich cellulose, respectively). To infer the differences in extraction backgrounds, we first averaged multiple measurements of single extractions by the bootstrapping method described previously. This reduces the impact of single extractions that have been measured more often than others in the outcome. We find lower background contributions arising from chemical pretreatment in Lund than in Zurich (0.08 ± 0.02 pMC).

For the second experiment, again cellulose was extracted in both laboratories but graphitized and measured in Lund. The experiment comprised 7 and 2 extractions, measured as 11 and 4 targets from Lund and Zurich, respectively. Consistent with the first experiment, we find lower extraction backgrounds in Lund (0.05 ± 0.02 pMC).

The reason for the slightly lower extraction backgrounds in Lund is speculative. We find 2 extractions from Zurich containing ~0.1 pMC more than other extractions from Zurich, which yield almost similar backgrounds as cellulose prepared in Lund. These 2 extractions were used in both experiments, and thus may bias the analysis. However, the source and nature of this apparent contamination is unclear, which is why they have not been excluded from this analysis. A removal of these 2 extractions from the analysis still yields slightly lower extraction backgrounds in Lund (0.07 ± 0.04 pMC) for the first experiment.

Sum of Background Differences

Integrating over all the experiments on background contribution differences described in the previous sections, we find lower backgrounds in Zurich than in Lund by 0.04 ± 0.03 pMC. This is consistent with the determination of the experimental overall background and serves as an internal quality check of the study design presented here. Moreover, assuming that the determinations of the experimental overall background and the inferred differences in machine and extraction backgrounds are correct, we can deduce that graphitization background differences are negligible.

Precision

Figures 2 and 3 show the results of ^{14}C determinations of the known-age wood samples as derived from the sample exchange experiments. The precision for single measurements is consistently better in Zurich, mainly resulting from better counting statistics. To minimize the described dependencies of ^{13}C/^{12}C ratios on ^{12}C currents in the NEC-SSAMS (Skog et al. 2010), Lund measures generally with $^{12}C^{+}$ currents of ~8 µA compared to about 15 µA at ETHZ. For higher measurement precision, we compensate this deficiency by increasing the measurement time, which, however, was not done for all samples in this study. In addition, long-term evaluation of reproducibility indicates a limit to achievable precision on single targets in Lund to ~3‰ so far. For all sample exchange experiments, the standard deviations of multiple measurements are in good agreement with measurement uncertainties as indicated by the reduced χ^2 statistic. For the pine sample dated to 7021 ± 5 cal BP, the scatter of ^{14}C dates on samples entirely prepared and measured in Zurich is slightly elevated (Figure 3). However, we do not observe the same effect for the wood sample from 435 cal BP, where standard deviations are consistently lower than statistically expected for all sample exchange experiments (Figure 2).

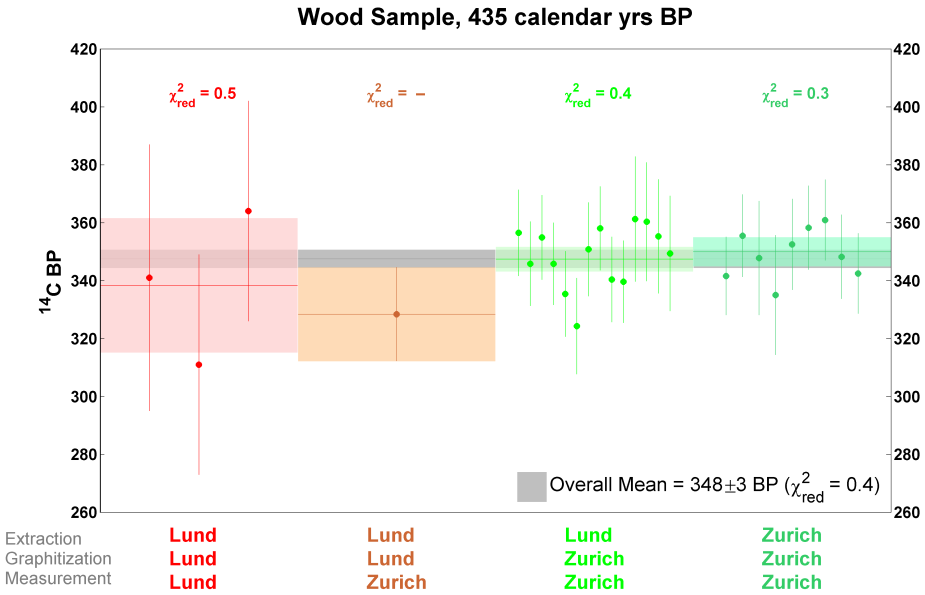

Figure 2 ^{14}C ages of wood samples of known calendar age (435 calendar yr BP). The place of extraction, graphitization, and measurement is indicated on the *x* axis. Colored lines and shaded areas show error-weighted means and ±1σ standard errors. The gray bar represents the weighted mean (1σ) of all measurements. The reduced χ^2 statistic is also given.

We do not see consistent and significant differences in the means that can be attributed to place of extraction, graphitization, or measurement, confirming the good sample reproducibility between the 2 laboratories in general; nor do we find significant offsets between the laboratories. There seems to

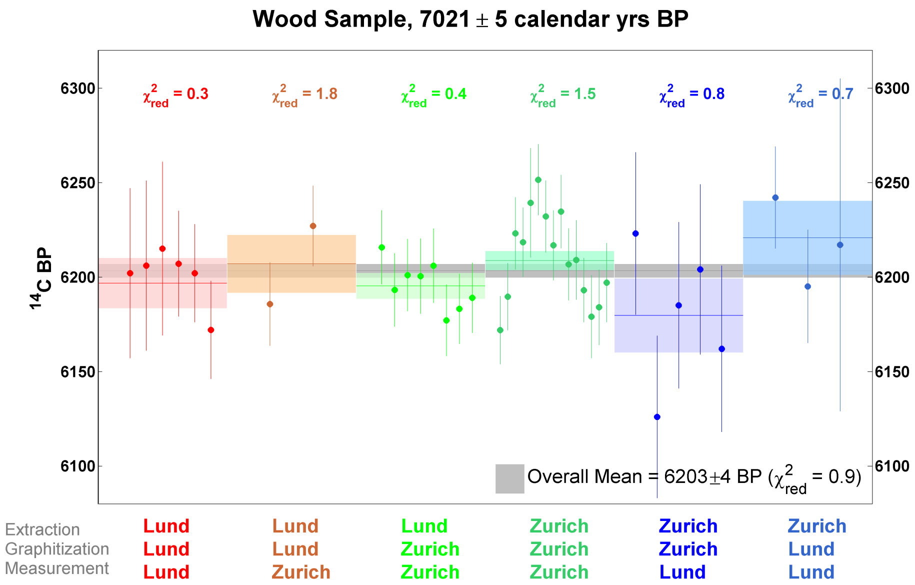

Figure 3 ^{14}C ages of wood samples of known calendar age (7021 ± 5 calendar yr BP). The place of extraction, graphitization, and measurement is indicated on the *x* axis. Colored lines and shaded areas show error weighted means and ±1σ standard error. The gray bar represents the weighted mean (±1σ) of all measurements. For all means, the reduced χ^2 statistic is given.

be a systematic difference between 7021 ± 5 cal BP pine samples extracted in either Lund or Zurich and graphitized and measured in Zurich (Figure 3, green). However, this difference does not reach the 95% confidence level and is not observed for the 435 cal BP sample (Figure 2, green). In addition, the reduced χ^2 statistic on all measured samples indicates consistency of the overall scatter with quoted measurement uncertainties.

Accuracy

To assess the accuracy of our results, we use data from IntCal09 (Reimer et al. 2009) and the underlying raw data sets (see Table 1) for comparison. Our results for the single-year pine ring sample from 435 BP are in good agreement with the single-year data from the University of Washington (Stuiver and Braziunas 1993; Stuiver et al. 1998).

Measurements on the decadal wood sample from 7021 ± 5 cal BP are on average about 38 ± 16 ^{14}C yr older than the corresponding ^{14}C age of IntCal09. However, our results are consistent with the decadal data set from the University of Washington (Stuiver et al. 1998), which is, among others, used for the construction of IntCal09 (see Figure 4). In fact, Stuiver et al. (1998) recognized systematic differences between the data sets from Washington, Belfast, and Heidelberg (Kromer et al. 1986; Pearson et al. 1993; Stuiver et al. 1998, respectively). However, for the 8–7 ka BP interval, the Washington data set was found to be on average 34 ± 3 and 56 ± 9 ^{14}C BP younger than the Belfast and Heidelberg data, respectively. Since we find an offset towards older ages for our data and the Washington data from IntCal09, this is unlikely due to a laboratory offset. In addition, the magnitude of this offset to IntCal09 is consistent in the Washington, Zurich, and Lund data. Thus, our results may rather indicate a small bias towards younger ages in the IntCal09 record at this time, rather than a systematic error in our measurements, and the Stuiver et al. (1998) data set.

Table 1 Weighted averages of multiple AMS ^{14}C measurements of the 2 known-age wood samples in Lund and Zurich in comparison to reference values cited.

Sample	Nominal (^{14}C BP)	Reference	Lab	Nr of measurements	Mean ± standard error (^{14}C BP)	χ^2_{red}
Wood (435 BP)	355 ± 17	Stuiver et al. 1998	Lund Zurich	3 26	338 ± 23 348 ± 3	0.5 0.4
Wood (7021 BP)	6165 ± 16	Reimer et al. 2009	Lund Zurich	14 29	6198 ± 10 6204 ± 4	0.6 1.2

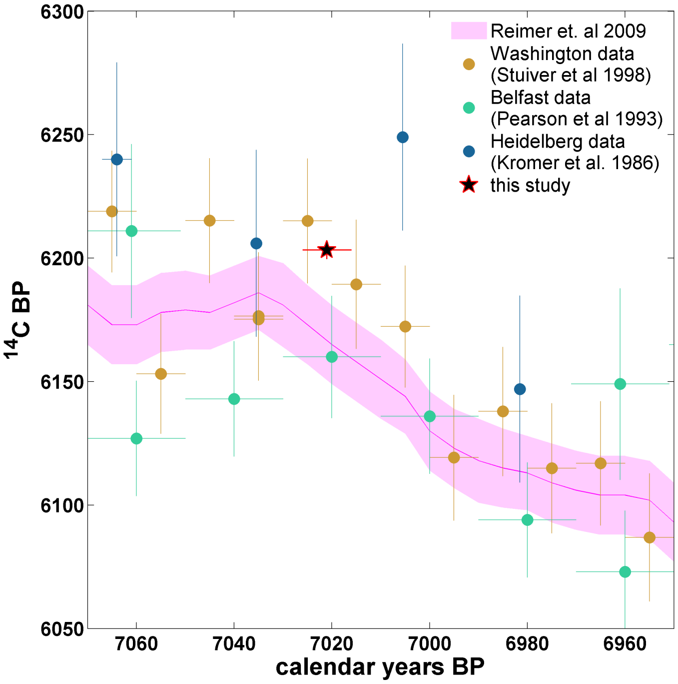

Figure 4 Error-weighted mean (±1σ) of all ^{14}C age determinations on our wood sample from 7021 calendar yr BP, IntCal09 (pink curve, Reimer et al. 2009), and the underlying data sets. The error shown bars in the *x* axis direction indicate the age covered by the measured samples (i.e. ring span).

CONCLUSION

We applied a systematic approach to compare the contributions of extraction, graphitization, and measurement to the overall background at 2 AMS ^{14}C facilities. Synchronization of laboratory protocols allows an assessment of interlaboratory differences with relatively few variables. The additional use of samples of known calendar age also ensures precision and accuracy. This method is a helpful tool to identify targets for improvement in each laboratory. It allows identification of interlaboratory differences that may not become apparent when comparing results that integrate over the complete process of preparation and measurement. Thus, this type of intercomparison can aid in the investigation of laboratory offsets, which would be a useful exercise for laboratories contributing data to the ^{14}C dating calibration record. Especially towards very old ^{14}C ages, a precise knowledge of background contributions of each step involved in ^{14}C dating becomes crucial.

The results presented here provide a quality assurance of the laboratory procedures and measurements performed by the new ^{14}C research group in Lund. We did see significant differences in the individual background contributions between Lund and Zurich, which, however, result in almost similar total backgrounds. Nevertheless, Lund will aim to further reduce the amount of contamination introduced by our sample combustion and/or graphitization. So far, Lund measures with consistently lower precision on single targets than Zurich due to lower ^{14}C counting rates. In our experience, the NEC-SSAMS sets upper limits to reliably measurable beam currents, as described by Freeman et al. (2008). Whereas Freeman et al. (2010) propose to overcome this problem by not injecting $^{12}C^-$ into the stripper channel, Lund increases the measurement times when necessary. This has the advantage that on-line ^{13}C/^{12}C measurements can be used to correct for fractionation during the measurement. However, despite better counting statistics, long-term reproducibility presently indicates a limit to reliably achievable precision to ~3‰ on single targets in Lund.

A comparison to reference data from IntCal09 and the underlying data sets (Reimer et al. 2009) underpins that the results presented here are also accurate within the quoted precision. Interestingly, it appears that IntCal09 itself may be about 38 ± 16 ^{14}C BP too young at about 7020 calendar yr BP.

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