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'Old wood' effect in radiocarbon dating of prehistoric cremated bones?

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ABSTRACT

Numerous reports of successful radiocarbon dating of cremated bones have emerged during the last decade. The success of radiocarbon dating cremated bones depends on the temperature during burning and the degree of recrystallisation of the inorganic bone matrix. During cremation bones undergo major morphological and mineralogical changes which have raised some interesting questions and discussion on the origin of the carbon source in archaeologically cremated bones. Recent laboratory experiments reveal that the properties of the combustion atmosphere play a significant role regarding the source carbon in cremated bones. Thus radiocarbon dating cremated bones is potentially dating the wood used for the cremation fire. Here we compare a high precision radiocarbon dated human bone with an associated dendrochronological age from an oak coffin. We find that the age discrepancy between the dendrochronological age and the cremated bone of 73 ± 26 ^{14}C yr is best accounted for by the so called 'old wood' effect.

1. Introduction

Radiocarbon dating of collagen in well-preserved human bone has routinely been carried out for decades, but cremated bone samples were always excluded because cremation destroys the bone collagen. However, within the last decade successful ^{14}C dating of cremated bones has frequently been reported (e.g. De Mulder et al., 2009, De Mulder et al., 2007, Lanting et al., 2001, Olsen et al., 2011). Furthermore, uniform results of radiocarbon dating of cremated bones have been proven in laboratory intercomparison tests (Naysmith et al., 2007). The intercomparison test was designed to test the dating protocol, i.e. using the same method laboratories get similar ages on the same material within measurement error. Hence problems related to whether or not ^{14}C dating cremated bone yields an estimate of the true calendar age were not tested. Here we present new information on a previously published cremated bone sample found in an oak coffin which has been dated by dendrochronology (Olsen et al., 2008). Our updated results will be discussed in light of new laboratory studies suggesting that ^{14}C dating of cremated bones reflects the burning atmosphere of the cremation fire (e.g. Hüls et al., 2010; Van Strydonck et al., 2010). We believe that our case study may represent an archaeological example supporting the recent laboratory conclusions.

Radiocarbon dating of bio-apatite is possible because of incorporation of carbonate ions into the inorganic bone matrix in living organisms. The carbonate ions originate from the energy production in cells and are substituted with phosphate ions in the bone matrix into the bio-apatite mineral-like bone structure (Krueger, 1991; Lee-Thorp and van der Merwe, 1991; Munro et al., 2007; Newesely, 1988; Pate and Hutton, 1988; Posner, 1969; Saliège et al., 1995; Sandford, 1993; Wright and Schwarcz, 1996).

Radiocarbon dating of the bio-apatite fraction has in general been abandoned decades ago due to incorrect ^{14}C results caused by contamination effects (Berger et al., 1964, Hassan et al., 1977, Stafford et al., 1987). In fossil bones, exchange reactions with the bicarbonate ions dissolved in soil waters lead to ^{14}C contamination (Hassan et al., 1977; Hedges and Millard, 1995; Surovell, 2000; Tamers and Pearson, 1965). Apparently, the exchange reaction with the dissolved bicarbonate ions does not occur for cremated bones and hence the bio-apatite fraction of cremated bone yields reliable ^{14}C results (Lanting et al., 2001, Olsen et al., 2008). This is because heating of bones results in numerous microscopic and macroscopic changes which altogether yield a more robust and inert bio-apatite

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structure as a consequence, i.e. heating results in re-crystallization of the bio-apatite bone matrix into a more robust structure (Newesely, 1988; Stiner et al., 1995; Van Strydonck et al., 2005). Crucial to radiocarbon dating of calcined or burned bones is assurance about the degree of bio-apatite re-crystallisation. As shown characterisation and subsequent careful selection of well cremated bones is essential for reliable ^{14}C age results (Olsen et al., 2008, Van Strydonck et al., 2009). To this end the cremated bones of humans should be characterised by visual inspection, IR spectroscopy (crystallinity index (CI) and the carbonate to phosphate ratio (C/P)), $\delta^{13}C$ of bio-apatite and the carbon weight percentage (Olsen et al., 2008, Thompson et al., 2009).

For radiocarbon dating knowledge of the carbon origin is in general of utmost importance because the carbon source defines the event being dated. The loss of structural carbon, the major morphological and mineralogical changes occurring during the cremation process has raised some interesting questions and discussion regarding the origin of the carbon source in archaeologically cremated bones (e.g. Hüls et al., 2010, Van Strydonck et al., 2010, Zazzo et al., 2009). Put simply, it all boils down to one plain question: What are you dating when radiocarbon dating cremated bones? It is remarkable that the $\delta^{13}C$ of charred and unburned bone apatite change from c. −15‰ to $\delta^{13}C$ values around −23‰ for cremated bones (Lanting et al., 2001, Olsen et al., 2008, Van Strydonck et al., 2005). This has lead to considerations about kinetic fractionation to explain the very depleted $\delta^{13}C$ values of cremated bones as favoured by Zazzo et al. (2009). On the other hand, carbon exchange processes during the fire may potentially explain the remarkable carbon isotope signature of cremated bones. Carbon from atmospheric CO_2, from bone organic matter (collagen) or from CO_2 evolving during combustion may all contribute even in tandem with kinetic isotope fractionation. Recent laboratory experiments by Hüls et al. (2010) and Van Strydonck et al. (2010) has demonstrated that the properties of the burning atmosphere plays a significant role as a carbon source in cremated bones. They found that the exchange processes between produced CO_2 during combustion and bio-apatite control the stable carbon isotope ($\delta^{13}C$) signature and radiocarbon age of cremated bones. Hüls et al. (2010) further argue that kinetic isotope fractionation is needed to fully explain their results, but this process is much less significant than exchange reactions with the burning atmosphere. Thus radiocarbon dating cremated bones is potentially equivalent to dating the wood used for the cremation fire. Despite similar ^{14}C ages has been demonstrated of paired samples of associated context material (mostly pitch and charcoal) and cremated bone samples (Lanting et al., 2001, Olsen et al., 2008, Van Strydonck et al., 2005), this opens the possibility of the 'old wood' effect when radiocarbon dating cremated bones.

2. Method

Sample preparation follows procedures described in Olsen et al. (2008): Cremated bone samples (2 g) are soaked in a 1.5% sodium hypochlorite solution to dissolve remaining organic material (48 h, 20 °C). The sample is then washed and submerged in 1 M acetic acid to remove post-depositional carbonates as well as less crystalline, soluble fractions of bio-apatite (24 h, 20 °C). Next the sample is washed and dried (12 h, 80 °C) with a bio-apatite yield of approximately 96%. The pre-treated sample is crushed and 1.5 g is treated with 100% de-hydrated phosphoric acid (8 h, 25 °C) to liberate CO_2 from which sulphur impurities are removed prior to conversion to graphite for AMS targets (Lanting et al., 2001). Part of the resulting CO_2 gas was used for $\delta^{13}C$ analysis on a GV Instruments Isoprime stable isotope mass spectrometer to a precision of 0.15‰, while the rest was converted to graphite for AMS ^{14}C measurements via reduction with H_2 using cobalt as a catalyst (Vogel et al., 1984). The AMS ^{14}C measurements were carried out using the EN tandem accelerator at Aarhus University (Denmark). The dating results are reported as conventional ^{14}C dates in ^{14}C yr BP based on the measured $^{14}C/^{13}C$ ratio corrected for the natural isotopic fractionation by normalising the result to the standard $\delta^{13}C$ value of −25‰ PDB (Andersen et al., 1989).

The samples have been visually inspected for surface and interior colour and burn cracks and IR-spectroscopy was performed on powdered pretreated sample material, i.e. bio-apatite. The sample material was mixed with KBr and hydraulically pressed into pellets prior to measurement of infrared spectra with a Perkin Elmer FTIR spectrometer (PARAGON 1000). The spectrum of KBr was automatically subtracted by an online computer. IR spectra on the bio-apatite bone fraction provide information on the crystallinity index (CI) and carbon to phosphor ratio (C/P) (Garvie-Lok et al., 2004, Olsen et al., 2008).

3. Results and discussion

A well-preserved coffin from Egtved, Denmark, consisting of a hollowed-out oak trunk was excavated in 1921 by the Danish National Museum. It contained the famous Egtved girl, dressed in full costume covered with a woollen blanket and wrapped in a cow skin (Thomsen, 1929; Alexandersen et al., 1983; Aner and Kersten, 1990, No.4357A). The grave goods consisted of a belt-plate, a small bronze earring, two arm rings, an awl in a wooden handle, and a horn comb. The archaeological date is the Bronze Age, period II (1500–1300 BC, Randsborg, 2006). At her feet there was a bucket of bark, which contained residues from honey sweetened beer, and at her left leg a bundle of cloth with the cremated bones of a child. There was another bucket of bark at her head also with a few cremated bones, the mentioned awl and remains of a hair net (Fig. 1). Consistent with the archaeological finds, the coffin has been dated to 1370 BC by dendrochronology (Christensen, 2006). The investigation carried out by Kjeld Christensen showed that the lower part as well as the lid was well preserved. 110 tree rings were preserved and 9 of these were sapwood rings. Moreover, the preserved bark ring consisted of early wood as well as a very narrow zone of latewood indicating that the tree presumably was felled in July or August prior to the end of the growth season (Christensen, 2006). All Danish dendrochronological dates of oak coffins resulted in a master curve comprising 419 years, and this curve was anchored to a German reference chronology (Christensen, 2006).

The human remains of the young (16–18 years old) woman in the coffin were rather poor due to the humid and acid peat bog environmental conditions from which she was retrieved. Only the woman's hair, brain, teeth, nails, and parts of her skin were preserved, but no bones at all (Thomsen, 1929; Alexandersen et al., 1983; Aner and Kersten, 1990, No.4357A). In contrast, the cremated bones found at the young woman's head and left leg appeared well preserved (Fig. 1, Thomsen, 1929; Alexandersen et al., 1983; Hvass, 2000). The cremated bones are most likely from the same individual, as fragments from the two sets of bones proved to fit precisely and represent a 5–6 year old child (Alexandersen et al., 1983).

Because of the age difference between the two individuals which excludes a mother–child relationship, it has without any evidence been suggested that the child was a sacrifice (Thomsen, 1929; Alexandersen et al., 1983; Jensen, 1997). It appears that the cremated bones correspond to regular cremated bone samples, i.e. colour, structure, fragmentation and form (Alexandersen et al., 1983; Olsen et al., 2008). One could imagine, in case of ritual deposition of the cremated bones (e.g. ancestral bones) that a number of years elapsed from cremation to deposition in the coffin. There are, however, remains of the funeral pyre among the