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Bacterial tetraether lipids in ancient bones record past climate conditions at the time of disposal

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ABSTRACT

Assessing impacts of climate change on ancient human societies requires accurate reconstructions of regional climate variations. However, due to the scarcity of *in situ* climate indicators in archaeological sites, climate interpretation often relies on indirect, geographically distant data from geological archives such as lake or ocean sediments, ice cores and speleothems. Because many cultural changes occurred abruptly over periods of years to decades, and are regional or even local in scale, correlating societal changes with climate reconstructions from geological archives induces significant uncertainties: factors such as chronological dating inconsistencies and geographic heterogeneity of climate can severely undermine interpretation. Here we show, for the first time, that it is possible to determine past climate change by analyzing bacteria-derived ‘branched glycerol dialkyl glycerol tetraethers’ (br-GDGTs) in ancient bones from archaeological sites. To the best of our knowledge this proxy has never been applied before to bones, nor with the intention of developing the method for application in archaeological research. We demonstrate that these compounds are likely derived from bacterial growth within bones following deposition in the ground, and the potential for their distributions to reflect climate and environmental conditions during the years immediately following deposition when bacteria consume internal substrates. Our preliminary results show that bone samples from different climate zones display distinct br-GDGT distributions. Well-dated late Pleistocene and Holocene bones from Alaska yield reconstructed temperatures consistent with existing climate reconstructions. While further work is necessary to determine how quickly the signal stabilizes in the bones, and to continue ongoing refinement of calibrations for temperature, precipitation, and other influences on br-GDGTs, we propose that br-GDGTs from ancient bones in archaeological sites may be taken as a new, *in situ* archive for reconstructing past climate conditions. This opens new perspectives for assessing connections between climate variations and social transformations in the past.

1. Introduction

Climate change has frequently been proposed as a key factor driving social change in ancient human societies. The socio-political collapse of the Classic-period Maya (Haug et al., 2003; Hodell et al., 1995; Kennett et al., 2012) and the transition of Chinese dynasties (Yancheva et al., 2007; Zhang et al., 2008) have for instance been attributed to major deficits of or abrupt changes in rainfall that fomented widespread famine, social unrest, violence and even warfare. The disappearance of Greenland's Norse colonies has been related to abrupt temperature declines during the Little Ice Age (D'Andrea et al., 2011). Evolution of bipedalism in hominins has also been related to forest contraction and grassland expansion as a result of reduced precipitation in East Africa (Potts, 1998).

In these instances, the evidence for climate change is derived from paleoclimate archives such as sediments, speleothems, and ice cores. While they undoubtedly provide key insights to climate variability, evidence from these sources is hampered by two important complications: (a) the challenge of accurately dating geological archives, and/or (b) the geographic heterogeneity of climate change at regional and

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continental scales. Errors associated with radiocarbon dates of sedimentary records can reach hundreds or even thousands of years, making it difficult to precisely correlate climate reconstructions to cultural events that have been dated separately using archaeological or radiometric methods that have their own dating imprecisions, albeit generally smaller. Social changes may occur over relatively short periods of time of just a few decades or less, which are brief enough to be lost in the variations of most techniques used to date geological archives and archaeological events. Correlating archaeological evidence of social change to proxy climate records is therefore extremely challenging.

Even if dating is accurate, as may often be the case for speleothem-based reconstructions, correlating climate change as recorded at the site of the climate proxy with the specific region where social change is observed from archaeological excavations still assumes uniformity of broader regional climate variations. Continental climate is however well known to be geographically highly variable, and proxy data from a single or limited number of locations are unlikely to offer a solid basis for extrapolating broad climate patterns for any given region. The speleothem evidence for climate change in the Maya lowlands is, for instance, derived from only a handful of places in Belize and the northwestern Yucatan Peninsula (Kennett et al., 2012; Medina-Elizalde et al., 2010; Webster et al., 2007), yet modern annual rainfall in the southern lowlands, which saw the Classic period collapse, varies quite dramatically, ranging from 1000 mm in the east (western Honduras) to 3000 mm in the west (Chiapas) (Scherer and Golden, 2014).

Ideally, climate proxies should be obtained directly from archaeological sites so that evidence of climate and social change can be traced in tandem, without the need for temporal or spatial extrapolation. Unfortunately, most archaeological sites do not possess the necessary climate proxies, whereas material culture collected and documented by archaeologists such as stone tools, architectural features or ceramics are not normally suitable for providing independent climate information.

Recent discovery of a class of ubiquitous bacterial lipids called branched GDGTs (br-GDGTs) in soils (Sinninghe Damsté et al., 2000; Weijers et al., 2006a) may, however, offer an opportunity to reconstruct climate conditions using material from archaeological sites. These lipids have been shown to vary systematically in structure with temperature and soil pH change across large gradients over the globe (De Jonge et al., 2014; Naafs et al., 2017; Peterse et al., 2012; Weijers et al., 2007a, 2006b). They are highly stable and can preserve past climate information over geological time scales (Gao et al., 2012; Peterse et al., 2011; Weijers et al., 2007b), and are thought to be produced by heterotrophic acidobacteria yet to be isolated (Weijers et al., 2006b; Sinninghe Damsté, 2016). The basis for geological and archaeological applications of GDGTs as chemical biomarkers is the assumption that variation in climatic variables, including mean annual air temperature (MAAT), soil pH, and precipitation, are reflected in the composition and distribution of these compounds, where GDGTs can vary in the number of methyl branches and the number of cyclopentane moieties (Sinninghe Damsté et al., 2000; Weijers et al., 2006b; Dang et al., 2016). Two global soil surveys, including 134 archive soils (Weijers et al., 2007a) and an expanded survey of 278 soils (Peterse et al., 2012), illustrate that the distribution of different GDGTs is correlated with climatic and environmental conditions, predominately mean annual air temperature (MAAT) and soil pH. The number of methyl branches was found to be closely related to MAAT and to a lesser extent to soil pH, while the number of cyclopentane moieties is related to soil pH (Weijers et al., 2007a). Cyclisation of Branch Tetraethers (CBT) and Methylation of Branched Tetraethers (MBT) indices were developed to quantify these changes. These indexes can then be expressed as functions of soil pH and MAAT based on GDGT distributions in global soil calibration datasets.

In practice, there may be additional factors influencing the relationship between the environmental variables (e.g., temperature or pH) and abundances of GDGTs. This has been reflected in relatively larger errors of the MBT-CBT paleotemperature proxy. However, natural soil samples with known temperature and pH gradients have illustrated the general reliability and potential of MBT-CBT-derived indices calculated using GDGT distributions, as well as the need to refine the calibration function (Peterse et al., 2012). The MBT-CBT proxy and its derivatives have been successfully used to infer past changes in continental temperature and pH from ocean margin sediments (Hopmans et al., 2004; Schouten et al., 2007; Weijers et al., 2007b), lake sediments (Blaga et al., 2010; Loomis et al., 2012; Tierney et al., 2012; Wu et al., 2013; Shanahan et al., 2013), paleosols and loess-paleosol sequences (Gao et al., 2012; Peterse et al., 2012; Yang et al., 2014); and peats (Zheng et al., 2015).

Faunal remains such as bones, whether from wild or domestic animals, are routinely encountered in substantial numbers at archaeological sites. The organic components of bone may offer excellent substrates for br-GDGT-producing heterotrophic bacteria, and if consumed relatively quickly, the resulting br-GDGTs inside bones would capture a snap-shot of the climate conditions soon after the bones' disposal. The physical structures of bone are moreover likely to isolate the br-GDGTs produced and prevent later modifications once available organic components inside bones are depleted. We therefore hypothesize that br-GDGTs from faunal remains may provide *in situ* paleoclimate proxies for archaeological sites on archaeologically relevant time scales.

The objectives of the present study are to: (a) demonstrate that br-GDGTs are indeed a common component in bones from archaeological sites; (b) obtain initial data on the rate of accumulation of br-GDGTs in animal bone from experimentally deposited bone at a controlled study site; (c) show that recently deposited bones from different regions display different compound distributions that reflect regional climate and environmental conditions; (d) demonstrate that br-GDGTs can be retrieved from ancient bones and matched to existing climate reconstructions, using two well-dated bone assemblages from Alaska, spanning respectively the past 30 thousand and ~1500 years.

2. Experimental

2.1. Samples

A total of 91 well-preserved animal bone samples from cultural management resource surveys and museum collections (Table 1; Figure S1) and 9 samples from a decomposition research facility (Forensic Anthropology Center at Texas State, Texas State University, San Marcos, TX) with documented artificial exposure times (Table 2) were analyzed to assess the reliability and sensitivity of the proxy across modern climate gradients. Samples represent a broad chronological range; while most date from the last 1500 years, those from the North Slope, Alaska, extend back 30,000 years. Bones were selected opportunistically, irrespective of animal types although no teeth were used (see Supplement for sample recommendations and below for discussion of bone types); no human bones were used.

Bones from recent archaeological excavation from 15 states across the U.S. roughly represent three climatic regimes, namely the northeastern and southeastern U.S. (moist regions), Arizona and New Mexico (arid region), and Alaska (cold region). All sites were open air, no samples were from caves sites without soil matrix. All bones have been stored in museum and cultural heritage collection archives. Unless otherwise noted, all bones were found disarticulated, with no indications of use and processing (burning, boiling, or cutting) or heavy post-depositional wear (gnawing or dramatic weathering).

One suite of bone samples was analyzed from sites excavated during the 1950s and early 1960s by J. Louis Giddings and Douglas Anderson of Brown University at Cape Krusenstern, on the Chukchi Sea coast, north of Kotzebue, Alaska (Giddings and Anderson, 1986). Cape Krusenstern has been occupied for the past 4200 years, since the cape