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SPECIAL FEATURE: INTRODUCTION

Current developments and future directions in archaeological science

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The field of archaeological science has grown to encompass a wide range of analytical techniques over the past 20 years. The application of methods initially developed and grounded in physics, chemistry, biology, and geology have been brought together to fill in the parts of the human story missing from the traditional archaeological record and added nuance to our understanding of the lived experience of people in the past.

Michael Tite was the first person to hold a chair in archaeological science in the United Kingdom, a role to which he was appointed at Oxford University in 1989. Writing in 1991, he did not consider archaeological science as a discipline all on its own but rather a meeting ground for collaboration; he emphasized the importance of integration of theory and method in both traditional archaeology and various contributing sciences (1). Throughout the 1990s, but particularly the early 2000s, the number of publications in the area of archaeological science skyrocketed (2). Institutional support increased in the United Kingdom but lagged in North America, as did training for graduate students and innovations in archaeological science techniques (3). This trend has largely continued. By 2015, David Killick wrote of the "awkward adolescence" of archaeological science, citing a rapid pace of growth but noting challenges in funding, quality control, and access (4). Most recently, Kate Britton and Michael Richards commented on the acceptance of archaeological science in mainstream archaeology but cautioned that practitioners need to understand the theory behind the techniques they use to ensure proper application, as well as integrate methodology and complex archaeological research questions (5).

In this special feature of PNAS, we recognize several key landmarks in the growth of archaeological science as it continues to move forward as a discipline. We review several developments in the areas of radiometric dating, stable isotope and elemental analysis, and proteomics. We offer insight into the future applications of these methods as analytical techniques continue to be refined and improved. But we agree with our colleagues that multiple challenges remain to be addressed.

In order to fully support the scientific community going forward, data management—and particularly data-sharing standards, quality assurance, and reproducibility—are essential. Beyond a matter of protocol, there is an ethical responsibility that data produced from destructive sampling of irreplaceable, archaeological material are disseminated in an accessible format. Ethics of sampling and stakeholder involvement are also essential, and not supplemental, to the continuing growth of this field. The development of anthropology and archaeology throughout the 19th and 20th centuries took place in a colonialist context, through the lens of Western scientific perspectives (e.g., refs. 6 and 7). Although broader archaeological practice has more recently engaged with decolonization and antiracist efforts, challenges remain (8–12). Likewise, colonialism and global economics have recently been shown to produce biases in the study of deep-time biodiversity; paleontology has favored rich countries over poor ones, and continued to practice "parachute science," sampling poorer countries without including or publishing with local collaborators (13). Archaeological science must be wary of falling into the same patterns of exploitation, especially as the kinds of increasingly sophisticated analytical techniques highlighted in this volume require substantial economic investment, decreasing the likelihood that much of this infrastructure will be broadly available outside of the wealthiest nations and institutions.

Advancement of Techniques

Dating. Since the first application of radiocarbon dating to archaeological samples in the mid-20th century (14, 15) and subsequent technological developments in specialist subfields throughout the latter half of the 20th century, the drive to improve and refine dating methods and chronologies has resulted in a rapid pace of advancement. By the 1980s, the advent of accelerator mass spectrometry (MS) for radiocarbon dating improved accuracy and made it possible to analyze much smaller samples of organic material (16, 17). As methods of purification improved, ultrafiltration became standard for processing bone collagen, and it is now possible to perform compound-specific dating of individual molecules, such as single amino or fatty acids (18, 19). Casanova et al. (20) demonstrate the power of these more refined radiometric approaches by dating fatty acids extracted from residues on ceramics. Not only do these techniques provide a means with which to date the use of the ceramic vessels, but because dairy fats in particular can be targeted, they allow the spread of the use of ruminant milk to be accurately dated. Casanova et al. use this technique to determine that dairying by the *Linearbandkeramik* cultural group in central Europe was

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associated with the earliest settlers in the region, rather than a more gradual diffusion of these practices.

Successive iterations and refinement of the radiocarbon calibration curve have made dating more accurate (21, 22). Through Bayesian statistical modeling, site and regional chronologies are constantly being refined (23), and syntheses of radiocarbon dates have resulted in multiple national and international databases (24). While there is scope for the continued improvement of precision and calibration in radiocarbon dating, allowing for the analysis of ever smaller sample sizes and date ranges, the technique cannot be applied for the whole of human history. For samples older than 50,000 y of age, alternative dating methods have been developed with varying levels of success. Among these are trapped charge and amino acid racemization (AAR), which are reviewed in this issue by Penkman et al. (25). These methods have the potential to expand the reach of scientific dating well beyond the limits of radiocarbon. In the case of AAR, the intracrystalline portion of the protein must be present and cannot have undergone diagenesis, making it possible to date mollusc shell, tooth enamel, eggshell, and bone (26, 27). This technique has been in development almost as long as radiocarbon (28–30), but it has been less systematically applied by archaeologists to date. As Penkman et al. (25) highlight, advances in calibration and cross-checks should advance the use of this valuable alternative dating method in the future.

Stable Isotopes. The applications of isotope ratio MS to archaeological remains have grown as methods have become streamlined and more automated, concomitant with decreased costs. Some of the early analyses in archaeology sought to address questions of paleodiet by targeting the stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) in modern animal tissues and bone collagen (31–33). This was accompanied by the development of oxygen isotope ($\delta^{18}O$) analysis in marine molluscs to understand seasonality (34–36). In subsequent years, isotopic analysis has broadened to include its application to a wide range of questions, from engaging with paleoclimate and paleoenvironmental reconstruction, mobility, point of origin, and life history reconstruction (e.g., refs. 37 and 38). The exploration of novel isotope systems offers the possibility of broadening the range of insights that can be obtained from geologically old materials, specifically those dating beyond the range of collagen preservation. This is exemplified in the contribution from Jaouen et al. (39), which uses analysis of zinc isotopes in Neanderthals and associated fauna from a Middle Paleolithic site over 50,000 y old in Iberia to detect evidence for a high degree of carnivory. While the use of nitrogen isotopes has previously been used to suggest high levels of meat consumption in Neanderthals, the authors combine zinc and strontium, carbon, oxygen isotopes, and trace element analysis from tooth enamel to parse out potential metabolic versus dietary effects to support this hypothesis. In so doing, they provide a precedent for the application of zinc isotope analysis in cases where nitrogen analysis cannot be performed due to poor collagen preservation.

Trace Element Analysis. Alongside isotopic analysis, the quantification of trace elements has also become increasingly common in archaeology, and the use of X-ray fluorescence (XRF) and inductively coupled plasma optical emission spectrometry have grown considerably in the field (40, 41), as have studies employing scanning electron microscopy and Fourier-transform infrared spectroscopy in elemental analysis. These techniques are often combined to maximize applications, ranging from studying surface modifications of archaeological ceramics to identifying waterlogged wood anatomy, to identifying the earliest uses of lacquer (42–44). The development of relatively inexpensive portable XRF (pXRF) units has facilitated archaeologists' capacity to nondestructively generate elemental data from artifacts and raw materials in a unique way. Frahm and Carolus (45) analyzed a large number of obsidian artifacts from Neolithic sites in the Near East using pXRF to determine the geological origin of the raw materials. Their findings shed important light on how connections among communities intensified in the region over time, as evidenced by increasing diversity in obsidian sources. Using other archaeological data, the authors link these patterns to increasing population density. This kind of study is facilitated by the nondestructive nature and high throughput capacity of the pXRF technique.

Proteins and Proteomics. The characterization of proteins from ancient samples has seen tremendous development in recent decades. Initial forays into this area used immunoassay techniques, often on tool surfaces, in hopes of identifying the animal taxa that were processed; these studies were fraught with a host of problems, notably false positives and a lack of reliability (46). The development of high-sensitivity MS techniques, including matrix-assisted laser desorption/ionization coupled to time of flight MS (MALDI-TOF) and liquid chromatography with tandem MS, has catalyzed rapid growth in the field of paleoproteomics, especially since the turn of the millennium (47, 48). These techniques have been utilized to address a broad range of questions related to species identification, phylogeny, ancient human diet, protein degradation, and ancient diseases (49–51).

Richter et al. (52) review the application of peptide mass fingerprinting, also known as "zooarchaeology by mass spectrometry" or "ZooMS," to the identification of archaeological bones and discuss the potential for its expansion to noncollagenous proteins and application to nonmammalian fauna. They note that one crucial limiting factor in the widespread use of ZooMS is not its technical advancement, but rather the accessibility of established collagen spectra for comparison across laboratories. The development of open access databases containing reference spectra will be essential in the continued development of ZooMS.

The study of ancient proteins extends beyond the sequencing of collagen peptides from bones and teeth. Demarchi et al. (53) apply a paleoproteomic approach to identify the avian taxon responsible for Late Pleistocene eggs that were exploited by Indigenous people in Australia around 50,000 y ago, resolving a decades-long controversy. These specimens yielded no endogenous DNA, but protein sequences were able to identify that the eggshell fragments belonged to the extinct, giant flightless bird *Genyornis newtoni* through the exclusion of other possible bird taxa. The taxonomic identity of these eggshell samples is consequential because they bear evidence of cooking and therefore demonstrate direct interactions between humans and *Genyornis*. There is currently no skeletal evidence for the exploitation of *Genyornis* at archaeological sites in Australia; therefore, these cooked eggshells demonstrate that *Genyornis* was not extinct before humans arrived on the continent and that humans made use of this resource.

Data Sharing and Accessibility

Alongside technical advancements in analytical techniques, quality control, data accessibility, and data reproducibility are central to the practice of archaeological science. Standards for data citation and data sharing (54, 55) and terminology and reporting (55) have been proposed but are not uniformly applied. Studies explicitly focusing on the development and application of various quality-control approaches have been limited (but see refs. 56 and 57 for examples in stable isotope analysis). The existence of databases beyond the project level, including within subfields and in varying formats, testifies the importance and utility of data aggregation, but there is uncertainty about long-term sustainability of many databases. In addition, data reporting standards and vocabularies vary across disciplinary boundaries, creating an additional challenge in data synthesis and integration (58). Several of the grand challenges listed in Kintigh et al.'s (59) landmark PNAS paper in 2014 on the "grand challenges for archaeology" included the synthesis of legacy data in addition to new data generation; this arguably remains a grand challenge for archaeology in and of itself.

Practicing FAIR principles (Findability, Accessibility, Interoperability, and Reuse) (60) has the potential to transform the current rugged archaeological science data landscape. In this issue, Kansa and Kansa (61) offer solutions at the level of data creation with the use of unique, persistent identifiers, which allow the reuse of data beyond the original analysis and support quality control (61). Moving forward, it will be critical to incorporate these practices as standard throughout the archaeological science research process.

Conclusion

As the field of archaeological science matures, several key issues remain. The need for care and maintenance of the physical collections that are sampled cannot be overstated (62). Likewise, there is a need for improvement in bridging archaeological science and local stakeholders, including indigenous knowledge and participation, from theoretical inception alongside methodological development (63–65). The advancement and application of new techniques may be paramount, but must not surpass concomitant efforts in data management, stewardship of physical collections, and increase in equity of practice: namely, an ethics of practice that is adhered to by all engaged in the discipline.

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