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Isotope analysis in archaeology grand challenge

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Isotope analysis is now an ever-present methodology applied to the study of past human diets, mobility and environments. At a time where ancient DNA (Orlando et al., 2021) and palaeoproteomics (Hendy et al., 2018; Hendy, 2021) seem to provide ground-breaking insights into human history on a near-monthly basis, isotope analysis was already considered to have reached a certain level of "maturity" in archaeological science over a decade ago (Lee-Thorp, 2008; p. 925). Indeed, thanks to a proliferation in the number of isotope laboratories, reductions in preparation and analytical costs, and a growing familiarization with isotope approaches across the archaeological community (Roberts et al., 2018), it is now rare to see a multidisciplinary archaeological study that *does not* include some form of isotope proxy approach. With this in mind, perhaps the greatest challenge facing the "isotope community" in archaeology is to ensure that this "maturity" does not equate to stasis. Indeed, with the novel applications of tested isotopic approaches to well-designed archaeological questions, continued exploration of isotopic variation in modern ecosystems, and the pushing of isotopic methodologies to the level of individual molecules, I would argue that the potential of this approach to reconstruct what we know about the past is greater than ever.

While the majority of review and perspectives papers will tend to focus on the latest state-of-the-art developments in a given field or methodology, I want to begin by looking back towards the beginning of isotope analysis applications in archaeology. In 1977, John C. Vogel and Nikolaas J. van der Merwe published a paper entitled "Isotopic Evidence for Early Maize Cultivation in New York State" in *American Antiquity* (Vogel and van der Merwe, 1977). They argued that observed differences in the measurable ratios of ^{13}C to ^{12}C between plants photosynthesizing using the "C_3" pathway (the majority of temperate grasses, shrubs, and trees) and those using the "C_4" pathway (many tropical grasses including the crop staple maize) (Craig, 1953; Smith and Epstein, 1971) enabled the stable isotope analysis of preserved human tissues to elucidate past dietary change in temperate northern America, particularly with regards to the arrival of maize. This landmark study started isotope analysis on its pathway to prominence in an archaeological context, being swiftly followed by the joint application of stable carbon and nitrogen isotope analysis to bone collagen to look at changes in food sources and trophic level (Sealy and van der Merwe, 1985; Sealy et al., 1987), and the stable carbon isotope analysis of tooth enamel bioapatite to explore hominin diets and environments in deep time (Lee-Thorp et al., 1989).

What I want to center in on here, however, is the thoughtful way in which Vogel and van der Merwe formulated their pioneering idea in this paper. Their premise read as follows (bullet-points inserted between sentences by myself for emphasis) (Vogel and van der Merwe, 1977; p. 239):

- "*Since both animals and humans ultimately derive their carbon from plants, the carbon isotope ratio [of their tissues] can be used to determine the relative intake of C-3 and C-4 plants at the beginning of their food chain.*
- *Where humans living in a predominantly C-3 plant environment have access to a C-4 cultigen which forms an important dietary staple, the relative importance of such a cultigen in the diet should be measurable through an isotopic study of skeletal remains.*
- *More specifically, it should be possible to detect the presence of maize in the diet of prehistoric peoples in many regions of North America, thus distinguishing peoples of the same regions who subsistence on hunting and the gathering of indigenous C-3 plants.*"

While Vogel and van der Merwe's (1977) paper is now nearly half a century old, these three points clearly express the wonderful potential of isotopic analyses (and indeed archaeological science approaches more generally) in archaeological contexts. In essence, they revolve around the observation of a biological, chemical, or physical phenomenon in the present, that can be applied into the past to explore the worlds and experiences of our ancestors. Yet, what Vogel and van der Merwe also beautifully elucidate, is the necessary inferential pathway researchers have to tread to come up with a successful study that will ultimately advance our knowledge of the past. With the increasingly eager, and perhaps what one might call "stock," application of isotope analysis to a variety of time periods and geographical contexts (Vaiglova et al., 2022), one of the main challenges for isotope analysis in archaeology is to make sure that researchers continue to revisit these three points and think about their implications for their own study in each and every case.

To take the statements/bullet points in turn, firstly, does the tissue I am analyzing actually record the parameter I am interested in, whether it is an aspect of diet, environment, or geographical area of formation? This question necessitates constant evaluation of the connection between the sample and its chemical composition and a given ecological or biological observation of isotopic variability. It is this question which continues to drive assessments of appropriate sample size to study a given population (Roberts et al., 2018), of the possibility of diagenetic alteration of samples to influence measured isotopic values (Beasley et al., 2014), the effectiveness and standardization of pretreatment and analytical protocols to yield values representative of the *in vivo* situation (Pellegrini and Snoeck, 2016; Szpak et al., 2017), and of which material should be sampled. For example, following Vogel and van der Merwe's study it was subsequently shown that low protein dietary components, such as maize, would actually be underestimated in bone collagen $\delta^{13}C$ as a result of metabolic routing (Ambrose and Norr, 1993), meaning that $\delta^{13}C$ analysis of human bioapatite would be needed to more effectively track more subtle appearance of maize in northern American diets (Ambrose et al., 2003).

Secondly, does the dietary or environmental phenomenon I am interested in exploring actually have a clear isotopic signature that can be used to formulate a testable null hypothesis? For example, had Vogel and van der Merwe (1977) sought to track the arrival of maize into an environment with natural C_4 vegetation, their results and interpretations would have been far more complex. Similarly, it was recognized early on in the applications of isotope analyses that where C_4 environments and marine ecosystems overlapped in potential importance to human diets, then stable carbon isotope analysis of bulk tissues would face interpretive issues. This issue is also behind concerns over baseline variability across space and time. In a palaeoenvironmental context, for example, in the tropics, $\delta^{13}C$ variability in sediments and animal tissues has commonly been used to study the dominance of C_3 (primarily forest, woodland and shrubland) and C_4 (tropical grassland) environments. Such shifts could be linked to climatic changes in temperature and aridity, but they can also be influenced by CO_2 concentration and pCO_2 has also been found to influence $\delta^{13}C$ values (Hare et al., 2018). This question also underlies the need for further ecological baseline studies [e.g., bioavailable strontium (Montgomery, 2010; Snoeck et al., 2020), environmental transects (Vogel et al., 1978; Smith et al., 2002)] and feeding studies to characterize habitat- or species-specific isotopic responses (Webb et al., 2017).

Thirdly, and finally, if I am able to detect the phenomenon of interest in my selected sample, be it an environmental factor or a change in diet, is it, in fact, of archaeological interest? While this is the last point in the progression, it should arguably also be the first. For an isotopic study to be successful it depends, inherently, on knowledge of the archaeological context. Whether this is achieved by a researcher balancing knowledge of chemistry and archaeology and anthropology, or through the ongoing communication between specialists within a given team, it is essential, from the beginning, to ask if isotope analysis can actually add something meaningful to study of a particular archaeological site or research topic. These questions and contexts may even change over time. For example, since Vogel and van der Merwe (1977) were writing it has been acknowledged that Indigenous populations may have been cultivating wild C_3 plants long before the arrival of maize (Smith, 1989, 2006). As a result, the arrival of maize would be linked to a different type of cultivation and food source, rather than the onset of cultivation entirely.

These three questions not only pose challenges to the effective application of isotope analysis in archaeology, they also continue to push the boundaries of the field, leading to novel methodological approaches and interpretive possibilities. For example, a desire to determine the degree to which archaeological samples can be used to determine dietary or environmental parameters has resulted in major advances in single compound isotope analysis. Stable carbon and nitrogen isotope analysis of amino acids is yielding more resolved insights into food sources and more baseline-independent results of trophic level and aquatic food consumption, in contexts ranging from Neanderthal hunting strategies to dietary variation in historical periods (Jaouen et al., 2018; Soncin et al., 2021). In a similar manner, stable carbon and hydrogen isotope analysis of plant waxes extracted from archaeological sediments can provide more precise environmental insights into vegetation communities and hydrology than was possible from "bulk" soil organic matter measurements (Collins et al., 2017; Patalano et al., 2021). The recent ground-breaking stable nitrogen isotope analysis of protein in tooth enamel as a measure of trophic level in "deep time" represents a further exciting development (Leichliter et al., 2021).

Similarly, a desire to understand the degree to which a particular object of study (e.g., mobility, diet or environment) is reflected in a measured isotopic signal is seeing the diligent applications of environmental transect studies and modern ecological studies. In the case of strontium isotope analysis as a tool for exploring past mobility, there are increasingly intensive discussions about the "best" type of material to sample and ways to best map bioavailable strontium in various contexts and what this means for interpreting movement in the past (Bataille et al., 2020; Holt et al., 2021). Meanwhile, although sometimes criticized (Schulting et al., 2022), Bayesian models, which utilize contextual insights and multi-isotope parameters to try and "rule out" certain interpretive scenarios, are arguably providing more precise estimates of dietary composition and human origins than were possible in the past (Fernandes et al., 2014; Cheung and Szpak, 2022). Modern feeding studies and ecological monitoring of isotopic variability are also showing how other non-traditional isotope systems may provide useful, novel avenues for exploration, including zinc isotope analysis of tooth enamel as a proxy for trophic level (Jaouen et al., 2016; Bourgon et al., 2021).

Finally, there continue to be a number of isotope-based papers released every year that seek to advance the state of our knowledge about the human past, testing well-formulated archaeological questions. This can take the form of applying tried and tested methodologies to a part of the world that has been relatively little-explored from an isotopic perspective (Hermenegildo et al., 2017; Louys and Roberts, 2020; Peralta et al., 2022; Skippington et al., 2022) or the use of a new isotopic method in the reconstruction of a novel aspect of past human lives (Bourgon et al., 2021). It can also involve the application of isotope methodologies to new human-associated samples. For example, the isotopic analysis of commensal and domesticated animals can provide important insights into nutrient flows and relationships with humans (Guiry, 2012; Swift et al., 2018). Significantly, the anthropological context of working with human remains is also being acutely recognized in isotope studies, with researchers seeking to align their approach with guidelines as to the ethical treatment of human remains (Squires et al., 2019;) or recognition of the processes of colonialism that have long impacted these and other approaches to archaeological science (Nayak et al., 2021) and the necessity for collaboration with Indigenous communities and repatriation exercises.

The "maturity" of isotope analysis in archaeology should not then be taken as an indicator that it is not continuing to provide novel, significant insights into the human past. As I hope to have shown, since the beginning of isotope analytical applications in archaeology, there have been the seeds of a "self-consciousness" and rigor that must continue to set the tone for novel isotope applications in palaeoanthropological, archaeological, and historical contexts to this day. Revisiting these principles and questions will not only continue to drive the development of effective hypotheses, pretreatment and sample screening approaches, interpretations firmly grounded in chemical, ecological, and environmental theory and observations, and close integration with questions rooted in the social sciences. It will also offer the foundation for the "jumping off" of state-of-the-art isotope methodologies that continue to highlight why this area of research remains one of the most dynamic, widely-applied, and resilient methodologies in the toolkit of archaeological and palaeoecological science.

Author contributions

The author confirms being the sole contributor of this work and has approved it for publication.

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Conflict of interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Publisher's note

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