Pb and Sr isotope composition of human dental enamel: an Indicator of Pacific Islander population dynamics

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Introduction

Of the three geo-cultural groups defined by Domeny de Rienzi, (Dumont-D'Urville 1832), the most cohesive and perhaps the most appropriate grouping, is that of the "distinct race" of Polynesia (Thomas 1989). These peoples inhabit the islands of the Pacific within the near-triangle formed by Hawaii, Rapanui (Easter Island) and Aotearoa (NewZealand). The similarity of physical appearance and social customs across the region allow for the unambiguous identification of these people to geographic location. It is generally accepted that Polynesians settled their islands and started to become a distinct group after 3200BP on the islands of Fiji, Tonga and Samoa. Their ancestors had traveled from Island Southeast Asia or Melanesia in the West (Smith 1995). Within the other Pacific regions of Melanesia and Micronesia, the island societies are considerably more diverse with many separate languages and cultural traditions. Also identified by archaeologists, anthropologists and linguists in these regions are traits associated with Polynesia. Perhaps the best known of these are what have been termed Polynesian Outliers (cf. Terrell (1986) who labels them Melanesian and Micronesian Outliers due to their physical location).

The Polynesian Outliers fall outside of the triangle formed by the Polynesian islands, but these island societies, eighteen in all, speak Polynesian languages. They have been variously considered as either relic populations left behind after their colonising friends and relatives moved east, or as representing a return west after most of the eastern Pacific islands had been settled. Recent studies suggest rather more complex histories, including cultural replacement, and highlight the need to consider each island individually rather than as a group (Bayard 1976, Kirch1984, Davidson 1992). No matter what the case is for individual "Outliers", evidence is emerging from the middle of the second millennium AD for a major rise in Polynesian influence west of the "triangle" (Spriggs 1993). Some of the strongest evidence for this comes from the islands of Vanuatu, where burials (e.g. Garanger 1982) and social systems (Spriggs 1986) are recognised as having Polynesian traits.

For archaeologists the frustration has been in the inability to identify whether this influence was due to actual migration of Polynesian communities into the islands, or through cultural transmission requiring contact rather than relocation of people. A third, but less likely possibility is independent development of traits similar to Polynesian due to a shared ancestry (Spriggs 1993). Archaeologists have recovered a number of human burials in Vanuatu and the Solomons that have been said to represent the original Polynesian migrants to these islands. The proposed method of isotope analysis would provide the a rigorous test for these claims. If burials such as could be shown to belong to first generation immigrants, as claimed, or conversely if it can be shown categorically that they do not, then our understanding of the movement of people within the Pacific in prehistory would require a re-evaluation and challenge current theories regarding the constitution of Pacific societies and their interaction over time.

Archaeological study of human skeletal material

Considerable science-based archaeology research has been devoted to the study of skeletal material derived from buried deposits. Numerous studies have been undertaken to study trace elemental and isotopic data in order to reconstruct dietary and palaeo-environmental factors which affect skeletal remains. In part, this research effort is a reflection of the ubiquitous nature of bone and other skeletal material on most archaeological sites. It is also, however, a reflection of a desire to increase the sophistication of the types of material that can be addressed in the archaeological material record. Amongst this research are some significant studies of heavy metal pollutants and their influence on the archaeological populations (Patterson *et al.* 1991 and refs. therein). As with modern day environmental counterparts, however, these studies have generally been conducted on total accumulated lead. They can therefore only provide circumstantial evidence for past exposure. Results and general conclusions of the source of lead-exposure, based upon total element analysis, are equivocal. In order to determine geographical information as to sources of exposures another class of data is required. With current concern over the long term effects of lead pollution there is considerable interest in quantifying "pre-industrial" human exposure and in evaluating changes in the sources and levels of lead pollution over time. In recent studies various materials have been suggested as potential archives of ancient or historical lead pollution, including soils, lake sediments (Renberg *et al.* 1993), tree bark, and fossil bivalves. A more direct approach has been to quantify lead levels in human bones from historical or archaeological populations (Mackie *et al.* 1975, Waldron *et al.* 1976, 1979, Waldron and Wells 1979, Aufderheide *et al.* 1981, Rogers and Waldron 1985, Corruccini *et al.* 1987, Kowal *et al.* 1989, Vuorinen *et al.* 1990).

It has become clear from this work that modern bones have considerably elevated lead compared with the "natural", pre-industrial levels in their archaeological correlates. However, accurate quantification of the differences between modern and ancient bone has proved problematic in view of the difficulty of measuring the ultra-trace levels of lead typically found in tissue from pre-industrial individuals. In recent years there has been a continuing programme of development of stringent sampling and analytical techniques making use of ultra-clean laboratories a high-purity reagents. As a result, estimates of biogenetic lead levels in uncontaminated archaeological tissue have continuously decreased. Typical estimates in the studies cited above range from 0-200 ppm, to give more specific estimates of 0.1-12ppm (Grandjean 1988, Hisanaga *et al.* 1988). Patterson and co-workers (Patterson *et al.* 1991), currently provide the lowest specific estimate at 13ppb. Interest in contemporary levels of lead exposure has been complemented in recent years by a desire to explore the sources of the pollution, particularly amongst 'high-risk' groups such as sub-adult urban or mining community populations (Yaffe *et al.* 1983, Rabinowitz 1987, Gulson and Wilson 1994). In this work lead isotope analysis has been used to distinguish between various pollutants which were then compared with the isotopic composition of blood lead. The possibility now arises to extend this approach to a consideration of the sources of lead to which ancient or historic peoples were exposed. Lead ingestion from plumbing was once suggested as a serious problem for the Roman urban population (Gilfillan1965). More recently it has been proposed that widespread air pollution from early metallurgy is represented by elevated lead levels in soil profiles or lake sediments (Renberg *et al.* 1993); although this seems implausible given the small scale of ancient metallurgy and the extremely low vapour pressure of lead at smelting temperatures. Considering the more recent past, Kowal and co-workers (Kowal *et al.* 1991) used lead isotope analysis in an attempt to assign the cause of the lead poisoning that befell the members of the ill-fated Franklin

Expedition (but cf. Farrer 1993). In pre-industrial societies it is highly likely that lead incorporated in skeletal material was derived mainly, or solely, from the diet. The isotopic ratios of such dietary lead reflect those of the soil and therefore of the underlying geology. Lead isotope ratios vary in a systematic manner in the geosphere as a result of radiogenic isotope evolution and therefore as a function of the age, parent isotope abundance, and subsequent remodeling of crustal material. The potential to use these differences to trace mobility and residence patterns by virtue of variations in the isotopic composition of lead between individuals in burial groups and between human remains and their burial environment has been recognised for some time (Ericson 1985). The principle has been demonstrated by recent studies based on strontium isotopes (Price *et al.* 1992, 1994, Sealy *et al.* 1995) and by combined lead-strontium isotope investigations to source modern rhinoceros horn to specific National Parks in southern Africa (Hall-Martin *et al.* 1993).

Skeletal lead incorporation in vivo and the diagenetic alteration of buried tissue

From the proceeding discussion it can be seen that there is considerable potential in examining both the absolute abundance and isotopic composition of lead in preserved archaeological human tissue as a means of determining the archaeological lead-burden and as a means of provenance analysis. In any study of the elemental or isotopic composition of skeletal hard tissue there are, however, a number of potential complications. The most serious of these concern the selection of suitable tissue for analysis (given the differential uptake and turnover of lead in different tissues during life) and the effects of post-depositional diagenesis. Diagenesis was investigated by us through the examination of tissue from adult marine mammals - Cape Fur Seals (*Arctocephalus pusillus*). Skeletal seal tissue is considered sufficiently similar to human material to act as a useful analogue. Marine mammals offer distinct advantages for a study of this nature since lead levels at the top of the marine food chain can be assumed to have been relatively consistent overtime and not subject to the widespread geographical variation which would be expected in terrestrial animals. Lead concentrations in bone and tooth samples from modern seals could therefore be taken as a useful base-line with which to compare samples of similar tissue recovered from archaeological contexts in order to evaluate post-depositionaldiagenesis. Pb^{2+} cations appear to reside, almost exclusively, in the inorganic mineral phase of bone (Spadaro *et al.* 1970, Brätter *et al.* 1977), where they are incorporated into the calcium phosphate lattice at sites normally occupied by Ca^{2+}.

Pb^{2+} is preferentially incorporated into bone tissue, as free energy differences favour Pb^{2+} over Ca^{2+} by about 27 $kJmol^{-1}$ (Patterson *et al.* 1991). Once there, the lead ions are harder to re-metabolise than Calcium ions; which are metabolically active in bone: acting as a dynamic-pool for calcium homeostasis and moving easily from bone to the bloodstream and back as required. The relative affinity of bone for the uptake of lead also suggests that post-mortem, diagenetic incorporation of lead is more likely to occur than leaching of the element (Patterson *et al.* 1987).Skeletal tissues display differential lead uptake, due to varying density, function, time of formation and rate of remodelling. Compact bone has a mainly mechanical and protective function, whereas trabecular bone fulfills the metabolic function of mineral homeostasis. Most bone turnover therefore occurs in the trabecular bone at the interface with bone marrow (Baron1990). There are also far more remodelling sites in trabecular bone due to its high surface to volume ratio (Teitelbaum 1990). However, varying levels of lead exposure during an individual's lifetime may lead to quite different patterns of skeletal lead distribution. A discrete period of high-lead ingestion would be apparent sooner in trabecular bone and on cessation, bone lead levels may be quickly reduced by remodelling. A more gradual accumulation would occur from the slower remodelling rate of cortical bone but removal of the lead source would leave elevated levels for far longer. Bone is continually remodelled in vivo so that lead continues to be incorporated throughout most of adult life. Indeed, the rates of lead accumulation in various skeletal tissues have been estimated by comparison with the prevailing mean isotopic composition of lead pollution in the USA (Manea-Krichten *et al.* 1991). In life history reconstruction it is extremely useful to be able to target lead ingested in childhood or adolescence with which to compare lead from the adult burial environment. Fortunately, dental enamel offers just such a possibility and is also highly resistant to diagenesis. Enamel is a dense, felted structure which, once formed, is not remodeled. Dentine is also of interest as it too is not remodeled in the same manner as bone. Dentine is successively deposited as linings in the pulp chamber cavity in response to tooth attrition, throughout an individuals life (Hillson 1986). Both enamel and dentine have a lower organic and water component than bone but enamel, particularly, is almost wholly composed of hydroxy-apatite crystals which are significantly larger than those of bone or dentine and give enamel considerable stability and resistance to organic decay, mechanical erosion and chemical attack (Parker and Toots 1980). Studies on the incorporation and distribution of lead in tooth enamel indicate that the amount of lead does increase somewhat during an individual's lifetime, despite the static nature of the tissue (Brudevold and Steadman 1956, Manea-Krichten *et al.* 1991). Analysis of lead concentration gradients in tooth cross-sections, show a lead concentration in surface enamel and secondary dentine: being highest in the layer immediately adjacent to the pulp cavity (Brudevold and Steadman 1956, Shapiro *et al.* 1972). Like bone, calcification of secondary dentine is a lifelong process and increasing lead levels here are, perhaps, unsurprising. The abrupt concentration gradient present in enamel may, however, indicate that the lead is adsorbed onto the surface of the tooth both in vitro and in vivo but the dense, impermeable structure prevents lead penetration into deeper enamel layers (Brudevold and Steadman 1956). In order to make an assessment of diagenetic alteration, particular attention has been paid in this study to the extraction and analysis of core enamel and to comparisons between this and other tissues from the same individuals.

Application of isotope analysis to pacific islander population dynamics

The technique to be used here is Thermal Ionisation Mass Spectrometry(TIMS). The high experimental precision (0.1%), obtainable using TIMS since the 1980s make it particularly suitable for this work and enable us to characterise the populations under review. In part, the technique is be applied in an analogous manner to application on ancient metalwork, however, additional precision is available in the study of teeth, through the measurement of strontium isotope ratios as well as lead.

Human and animal bones contain trace amounts of strontium and lead that is ultimately from local rocks and ground-waters and thence enter the food-chain. Study of Pacific islander populations offers some incidental advantages over equivalent studies based on mainland archaeological populations (Carlson 1996). The islands under investigation are of small size, and metal production is unknown in the region. Consequently, the number of possible sources of lead exposure for the archaeological population of any single island is extremely limited and will be unaffected by lead derived from re-melting of metal artefacts derived from long-distance trade. The total lead exposure in any Polynesian Outlier population therefore certainly relates to aspects of the food-chain which are unlikely to vary widely between members of the same population. Provided that the isotopic signature of the islands under investigation are sufficiently different to allow discrimination, it is likely, therefore, that any population studied will form a relatively homogenous group in terms of its isotope signature. Any migrant arriving from a geologically different setting will be relatively easy to spot on a multi-dimensional projection of the data.

There are, however, a certain number of obstacles to the use of radiogenic isotopes for provenance analysis. It is imperative that these difficulties are recognised, from the outset, in order that they can be taken into account before archaeological conclusions are drawn form the data.

If meaningful conclusions are to be derived from the results of the isotope analyses, separation must exist between proposed isotopic source fields, the boundaries of which may be difficult to define (Gulson 1986). The variability of ratios result from rocks having different ages, uranium/thorium/lead ratios and geo-chemical histories. Separation would therefore, be much clearer between two areas of very different geology than those between rocks that are close in both geographical distance and age, as mixing during formation may have occurred (Gulson 1986). It is possible to estimate the age of a geological source from its lead ratios, in this case 208Pb:204Pb / 206Pb:204Pb using the single stage Holmes-Houtermans model. For archaeology, it could be used to estimate the lead ratios that would be concordant with bedrock from a particular location and whether their "fingerprints" may be resolved.

Not all rocks are conformable and they may have gone through several stages of formation when their lead, uranium or thorium contents were changed e.g. metamorphic and igneous rocks, thus producing a blip in the smooth lead growth curve (Faure 1977). Particularly complex geology in a specific area will also lead to a situation which is far from clear cut and various combinations of isotopes ratios may have to be analysed in order to resolve these differences. For example, extremely small variations may not be apparent when 204Pb ratios are used and a comparison between 208Pb:206Pb and 207Pb:206Pb may optimise results in this case (Gulson 1986). It is problems along these lines that have plagued the provenance analysis of metals trade in the Mediterranean (Budd *et al.* 1995, 1996). There is, however, good evidence to suggest that an isotopic study of dental remains within the Pacific will avoid many of the pitfalls encountered in the studies of ancient metallurgy. The variation of abundance in radiogenic isotopes amongst the Polynesian islands are determined as a function of both parent/daughter isotopic ratios in the mantle source of oceanic island basalts and the age of this source. Specific isotopes of lead and strontium are derived exclusively from the decay of specific parent isotopes uranium, thorium and rubidium. Information regarding the rate of these decay processes have important implications for our current understanding of geological processes. In terms of the island sunder consideration, although their formation is thought to a relatively recent event it is believed it involved the remobilisation of ocean basalts of considerably greater age. Although, being basaltic in origin, the islands are extremely lead-poor (2-3ppm), considerable success has been achieved in constructing isotopic databases of lead and strontium isotopes. Characteristic signatures of island groups can therefore be identified isotopically. Considerable success has been achieved in using these isotope fields for the sourcing of prehistoric basalt artefacts in the region (Weisler and Woodhead 1995, Woodhead and Weisler in press). The ratio of 87Sr/86Sr has for some time been used archaeologically to determine the relative importance of foods with different isotopic environments (Ericson 1985, 1993; Yesner 1988 Sealy *et al.*1991). 87Sr is derived uniquely from the decay of 87Rb in a manner analogous to lead and is plotted as a ratio 87Sr/86Sr. When the additional data provided by 87Sr has been applied to archaeological provenance analysis of stone implements in Polynesia it has been possible to comment on inter-island trade interactions within archipelagoes as well as between island (Woodhead and Weisler in press). The same level of analysis using archaeologically-derived dental enamel offers exciting possibilities for the study of human movement between Polynesian islands.

Choice of material

Due to the ethical considerations incumbent on any research conducted using human remains some care has been exercised in the selection of material for this research project. The team is aware of the sensitivities attached to the study of the ancestral remains of some cultures of the Pacific region and have consequently arranged to study only material that is already excavated and for which permission has been obtained to carry out primary research. The samples requirements for the research are very small (50mg) and although the sample is destroyed will not result in any visible change to the integrity of the remains. The material under investigation in this project will offer unique opportunities to test theories regarding migration in the Polynesian Outlier populations and to test the robustness of the method. The research team recognise, however, that selectivity in gaining to the material for this study may artificially skew the results in favour of certain archaeological conclusions. For this reason every effort will be made to obtain study material that is both chemically and archaeologically representative of the area under consideration.

Conclusions

Lead isotope analysis of dental enamel could offer the opportunity to characterise different archaeological individuals as members of distinct island populations. The study offers unique possibilities as a means by which skeletal material of unknown origin can be ascribed a provenance. A method of determining geographical origin would be of considerable importance to the study of archaeology in the Pacific region. The results will be used to determine whether a number of burials, found on islands in the Pacific truly belong to migrant Polynesian individuals as they have previously been described or whether they were born on the island in which they were excavated. As well as determining where migration has occurred, it may be further possible to determine the direction of the migration. This information would be of importance in the chosen study of the Polynesian Outlier populations and has important possibilities for testing the veracity of the local oral tradition and previous archaeological theories. The research may contribute to our understanding of patterns of lead exposure in island communities. In the absence of contribution to the community's lead burden from leaded petrol or from metallurgical activities, the lead signature will reflect some aspect of the food chain. This signature may be the background signature of the island's geology ingested via agricultural activities, that of the local marine fauna or indeed a mixture of the two. Due to the limited size of individual islands and the absence of any additional lead burden derived from metallurgical activity, the populations of the Pacific Islands are particularly suitable for this application of lead and strontium isotope analysis. Isotopic analysis will therefore provide a discrete characterisation free of the mixing problems that constrain similar studies based on mainland populations. The project will have a dual function to both establish systematic distinctions in the provenance of human populations of the Pacific region and to answer a number of archaeologically important questions regarding the movement of individual island populations.

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