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# SAYING SO DOESN'T MAKE IT SO

## PAPERS IN HONOUR OF B. FOSS LEACH

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# Diet Reconstruction From Human Bone Trace Element Analysis

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## INTRODUCTION

The reconstuction of prehistoric subsistence is fundamental to an understanding of past human cultural ecology. Traditional methods used by archaeologists to reconstruct past diets include the analysis of economic debris, coprolite analysis and environmental reconstruction. It has become increasingly clear, however, that these methods do not provide a full quantitative picture of prehistoric diet. This is because of factors such as sampling discrepancies, different rates of preservation of dietary components, biases due to the seasonal occupation of sites and the almost total lack of direct archaeological evidence of plant foods in some regions. The main problem in reconstructing diets is the lack of methods to determine accurately the relative contribution of plant and animal foods.

In the Pacific region it has been difficult to establish the significance of horticulture. Indeed, it has often been impossible to identify the presence of cultigens, even in areas where they are known to have been important. In New Zealand the importance of vegetable foods and the significance of horticulture in prehistoric subsistence have not been established satisfactorily despite efforts by a number of researchers to develop research strategies, analytical methods and models of subsistence economics to assist with the accurate reconstruction of past diets (e.g., Nichol 1978; B. F. Leach 1976; H. M. Leach 1976; Shawcross 1967). Although Helen Leach's work, particularly in the southern Wairarapa, clearly showed the importance of horticulture among the activities of the earliest inhabitants, such interpretations are usually not achieved. The development of new methods to overcome the deficiencies in traditional methods of dietary reconstruction is, therefore, essential.

In recent years, the investigation of the chemical composition of human bone has begun to provide new information about past subsistence. Two techniques for extracting information from prehistoric skeletal material are stable isotope and trace element analysis. To date, the application of stable isotope analysis in archaeological research has centred on the examination of the isotopic ratios of carbon and nitrogen in human and animal bone collagen to measure the importance of certain plants or animals in the diet (De Niro 1987; Sealy and Van der Merwe 1986; Schoeninger and De Niro 1984; Van der Merwe and Vogel 1977). Both nitrogen and carbon isotope ratios in human bone collagen reflect the isotopic

composition of the foods consumed. However, a number of different dietary components exhibit similar isotopic ratios. This is a problem in situations, such as the Pacific islands, where marine foods are available. In these areas an overlap in values is evident between the isotopic ratios of some marine foods and some terrestrial plants. Carbon isotope ratios for marine foods and sugar cane are similar. Coral reef food resources and terrestrial plants exhibit similar nitrogen isotope ratios. This makes it difficult to identify the precise food resources represented by the nitrogen and carbon isotope ratios. A more recent application of isotope analysis to archaeological dietary research has been through the examination of the stable isotopes of sulphur in combination with carbon and nitrogen (Leach *et al.* 1989). A clear distinction is evident between the sulphur isotope ratios of marine plants and animals and terrestrial plants and animals. This shows the potential of multiple isotope analysis in resolving the difficulties outlined above.

For more than a decade researchers have been exploring the potential of trace element analysis of human and animal bone as a means of providing information on past diet. The analysis of strontium in archaeological human bone was first undertaken in the United States in 1973. Since then, the method has been modified considerably. Other elements have been incorporated, although strontium is still the focus of most studies to date. Specialist collaboration has helped to identify and resolve problems inherent in material, methodology and interpretations. Applications have expanded to incorporate many aspects of diet related human behaviour such as status, health and interpopulation variability. However, in the Pacific region in general and in New Zealand in particular, archaeologists have not taken advantage of the developments in this field. This is surprising in view of the many problems encountered in traditional methods of dietary analysis.

The purpose of this paper is to describe trace element analysis of archaeological human bone as this has been applied to dietary reconstruction. One method by which human bone can be examined for its trace element content is reported.

## TRACE ELEMENT ANALYSIS

Trace element analysis of prehistoric human bone is potentially capable of evaluating the relative importance of animal and plant foods in the diet of past populations. Strontium has been the main focus of most studies to date, because the behaviour of strontium in its movement through the food chain is reasonably well understood. However, other useful elements include zinc, copper, molybdenum and selenium, which are usually associated with animal protein; and strontium, magnesium, manganese, cobalt and nickel, which are generally found in greater amounts in vegetable matter (Gilbert 1985: 347). Strontium and zinc, the most frequently utilised and reliable elements so far investigated, are described in this paper.

### STRONTIUM AND ZINC

Environmental strontium levels vary geographically. The amount of strontium present in soil dictates the uptake by plants and therefore determines the amount present throughout the food chain. As plants do not discriminate between strontium and calcium, they have strontium levels similar to those present in the soil and adjacent water catchments. Strontium ingested by animals is discriminated against in favour of calcium during incorporation

into bone lattice (Szpunar 1977: 38). Strontium absorption in the female is further affected by pregnancy and lactation (Price *et al.* 1986: 369).

Discrimination against strontium in favour of calcium during incorporation into bone tissue has been estimated as occurring about five times for every step of the food chain. For this reason the amount of strontium absorbed by herbivores is about 20% of that present in the plant food ingested. This has been described as a ratio of strontium to calcium, which, in a herbivore, will be about five times greater than that in plant matter (Price *et al.* 1985b); or as a bone to diet ratio which has been determined experimentally in laboratory rats as about 0.26 (Price *et al.* 1986).

A herbivore, which obtains strontium directly from plants, ingests a relatively large amount of strontium, most of which is excreted by the kidneys into the urine. Virtually all that remains moves into the skeleton, because strontium is a bone seeker (Schoeninger 1979b). The ingestion of animal flesh will, therefore, provide little strontium to organisms higher up the food chain. For this reason, the bone strontium levels of carnivores are comparatively low. Conversely, because of the higher concentrations of strontium in plant matter than animal flesh, herbivores will have comparatively high bone strontium levels. Omnivores will exhibit intermediate levels. This principle enables the relative proportions of plant and meat foods consumed by past populations to be estimated by a comparison of human with herbivore and carnivore bone from the same locality. This is very difficult in Pacific Islands where carnivores are essentially absent.

The major dietary sources of zinc are protein-rich foods, particularly red meats and fish (Prasad 1978: 298). Certain nuts and legumes contain zinc in levels similar to if not higher than red meats (Rheingold *et al.* 1983: 233). Zinc is essential for proper bone mineralisation, some endocrine functions, wound healing, disease resistance, and growth (Szpunar 1977: 63).

Absorption and retention of zinc can be influenced by a number of factors. Phytate (inositol hexaphosphate), which is present in cereal grains, greatly impairs zinc absorption, as it binds zinc to the fibre of grains so that it is not degraded by the digestive system. This limits its availability for metabolism (Prasad 1978).

The interaction between zinc and copper has been defined as mutually antagonistic (Kirchgessner *et al.* 1982: 486); that is, zinc affects copper metabolism and vice versa. The proportions of these elements present in bone are not necessarily a reflection of dietary intake. A high level of copper intake will act to reduce zinc, while the impairment of zinc absorption due to the presence of phytate, for example, will probably permit an elevation in copper levels (Blakely and Beck 1981: 421).

## PROBLEMS

There are a number of problems associated with this type of research which warrant consideration. These problems relate to the analytical method and the specific material examined and produce variation in trace element concentrations between individuals and populations resulting from a number of factors other than diet. Environmental, anatomical, age and postdepositional factors have been shown to affect trace element levels within prehistoric human bone.

The environmental variation of strontium prevents comparison of populations from different geographical areas unless some means of normalising human bone strontium values is found. One way of achieving this is by determining bone strontium concentrations in

faunal samples from each area and using these as a baseline for comparing the human bone values.

There appears to be some variation in trace element levels between different bones of the same individual, although this is disputed (see Price *et al.* 1985a; Schoeninger 1979a). Therefore, there might be discrepancies in results if different anatomical components were used in the same study. Variation with age also appears to be significant, although once again there have been inconsistencies in results from various researchers (e.g., Gilbert 1975; Price *et al.* 1986; Schoeninger 1979a). To reduce interpretative error, adult bone only should be utilised in studies of dietary differences. Variation between sexes appears to be insignificant (see Price *et al.* 1985a, 1986).

The examination of post-depositional alteration of human bone trace element levels has been undertaken by numerous researchers (for a review of the literature on diagenesis see Pate and Brown 1985). It is important to identify the extent to which diagenetic factors have altered bone trace element concentrations. The use of faunal bone from the same site for intrasite and intersite comparisons will compensate for the possibility that diagenetic effects have altered trace element levels. The use of subsurface samples of cortical bone has been recommended as it is less subject to diagenetic effects than trabecular bone (Pate and Brown 1985: 489).

Large sample sizes are desirable for the assessment of trace element variability within a population. Several studies have addressed this issue. Schoeninger (1979a), in an attempt to clarify the amount of variation in strontium between individuals of one species, examined bone strontium levels of a group of 35 mink raised on the same diet. From this she suggested a coefficient of variation of 19.26 (mean $= 270$, $\sigma = 52$) as a conservative estimate of variability. Humans consuming the same diet would probably have a lower coefficient of variation because of their larger body size and different metabolism (p. 19). In another study, Price *et al.* (1985b) examined bone from 53 white-tailed deer. The results of this study produced higher variability than Schoeninger's study with a coefficient of variation of 34.4% (mean $= 125$, $\sigma = 43$). These researchers consider this may be representative of a natural population from a small area; the variability being a result of differences in age, sex, individual metabolism and measuring error (pp. 430, 434).

A further complicating factor for bone trace element studies is the consumption of shellfish. Schoeninger and Peebles (1981) found that the inclusion of freshwater molluscs in the diet of a hunting and gathering population produced increased bone strontium concentrations relative to an agricultural population. This was the opposite of the expected result. The meat of filter feeding molluscs is known to contain large amounts of strontium, because strontium present in water is continually taken up by filter feeding and is then concentrated in these organisms.

Rosenthal *et al.* (1970) undertook a study where strontium/calcium ratios were compared for marine and freshwater shellfish. Their results showed that crustaceans had the highest strontium values in both environments, followed by shellfish, then bony fish; marine shellfish exhibited considerably higher strontium levels than freshwater shellfish (cited by Katzenberg 1984: 13). This is because sea water contains comparatively high concentrations of strontium (about 8 ppm) while freshwater varies between 0.003 to 0.8 ppm depending on the location (Katzenberg 1984: 14).

The opposite appears to be the case for zinc. Hunter and Tyler (1987) examined the distribution of zinc and reactive silicate in a New Zealand harbour. Their results indicated that zinc is present in freshwater in considerably higher concentrations than in sea water

(500 to 1000 ng/kg in fresh water as opposed to 5 to 20 ng/kg in sea water) (Hunter and Tyler 1987: 386, Figure 5).

Strontium is not the only heavy metal accumulated by shellfish, particularly filter feeders such as bivalve molluscs. Nielsen and Nathan (1975) undertook a comprehensive survey of heavy metal levels in 13 species of molluscs from around New Zealand, concentrating on edible bivalves. Of the 12 bivalve species examined, the green mussel, *Perna canaliculus* and the cockle, *Chione stutchburyi*, had a predilection for accumulating lead, while the rock oyster, *Crassostrea glomerata*, appeared to accumulate zinc preferentially. The one gastropod included in their study (*Haliotis iris*) exhibited comparatively low concentrations of the various heavy metals examined. Goldberg *et al.* (1978) have presented similar results. Their research indicated that oysters accumulated more silver, zinc, copper and nickel than mussels, while mussels were better concentrators of lead (pp. 111–12).

This shows that not only strontium but also zinc, lead, copper and nickel accumulate in the meat of some filter feeding molluscs. However, browsing gastropods such as *pāua* (*Haliotis iris*), do not appear to accumulate heavy metals to the same extent as filter feeders.

As filter feeding shellfish appear to concentrate strontium above natural water abundance, it has been suggested that studies involving trace element analysis of human bone must be carried out with some prior knowledge of the range of plants and animals available for consumption by the population (Schoeninger and Peebles 1981: 396). The consumption of certain nuts and berries will produce similar effects because of the extremely high elemental levels in these foods, and this has to be anticipated.

As strontium and zinc are similarly affected by the consumption of nuts or molluscs it may be advisable to analyse both in any particular study. A high concentration of bone strontium should, in theory, indicate a high intake of vegetable foods, whereas high bone zinc levels should indicate a diet rich in red meat. High concentrations of both strontium and zinc may, however, suggest the consumption of a significant quantity of nuts and/or filter feeding shellfish.

It had been claimed that sea mammals absorb more strontium than terrestrial mammals because some heavy metals are concentrated in sea water (Price *et al.* 1985a: 423; Schoeninger 1979b: 297). Leach *et al.* (1979) have provided some conflicting evidence which indicates that strontium/calcium ratios in sea mammals (with the exception of *Dugon dugong*, a herbivorous sea mammal) are comparable to those in terrestrial fauna. This has been confirmed by similar studies (Connor and Slaughter 1984; Wessen *et al.* 1977). The actual status of strontium in sea mammals, however, requires further research.

## ANALYTICAL METHOD

Various methods have been used to analyse the trace element content in bone. These include atomic absorption spectrophotometry, x-ray fluorescence, electron microprobe and neutron activation analysis. Atomic absorption analysis, the method described here, is particularly suitable for strontium and zinc analysis because the analytical equipment is accurate and sensitive to these elements, and analysis time is short once the spectrophotometer is tuned to specific requirements. This has advantages for multi-element analysis of large samples. One benefit of atomic absorption over other analytical methods is that it more readily yields absolute figures, rather than ratios of trace element concentrations, which are obtained from emission spectroscopy for example. Also, the detection limits are more suited to the analysis of bone material than is the case for any other method except

neutron activation (Gilbert 1985: 353). The main disadvantage of this method, however, is that it is destructive; that is, samples are consumed by analysis. Details of one method of bone sample preparation and analysis by atomic absorption spectrophotometry are given in Appendix 1.

### CHEMICAL INTERFERENCE AND IONIZATION

Elemental determination by atomic absorption is particularly prone to chemical interferences and ionization effects which effectively depress absorbance values. When strontium concentrations are determined by this method the absorption is reduced by interference due to the formation of stable compounds between strontium and calcium, phosphate or sulphate for example. The addition of lanthanum to sample solutions removes the interference of the strontium-phosphate compound as lanthanum will bond more readily than strontium with phosphate. It has been demonstrated that ionization effects which result in depression of the strontium signal (Kirkbright and Sargent 1974: 684) can be alleviated by the addition of a more easily ionized metal such as potassium.

The incorporation of 1.0% lanthanum oxide, plus 0.5% potassium chloride into sample solutions during preparation is therefore necessary.

## APPLICATIONS TO PACIFIC AND NEW ZEALAND SAMPLES

Trace element analysis using atomic absorption spectrophotometry has been applied to human and faunal bone from a number of sites in the Pacific and New Zealand. Material from a range of sites was studied to provide information to answer specific questions about past diet. Some results of this research have been published elsewhere (Horwood 1989).

The following sites were included in the study: Namu, which is a burial mound dated to about A.D. 1250–1600 on Taumako, a Polynesian outlier in the southeast Solomon Islands; Waihora mound (New Zealand Archaeological Association Site Number C240/283) in the Point Durham area of the Chatham Islands, occupied in or about the 16th century A.D.; Site 8 on the northeastern corner of Watom, an island situated off the northeast New Britain coast and dated to about 500 B.C. (Green and Anson 1987); Nebira and Eriama, two sites in the Port Moresby region of Papua New Guinea, the former dating to about A.D. 1000–1300 and the latter to about A.D. 1500–1625. Individuals recovered from a *pā* site on the northern shore of Lake Rotoiti in the central North Island of New Zealand were also examined.

## CONCLUSIONS

Trace element analysis offers a new way of using skeletal material to further our knowledge about past human culture in general and diet in particular. This method approaches human subsistence from a different perspective than that of traditional methods of economic analysis. It enables information to be recovered directly from the consumers, for surely the focus of research should be on the human populations themselves, as the prime rationale for archaeological investigation and interpretation. This approach to the study of past diets should prove more successful than the analysis of prehistoric refuse dumps, for example, and certainly warrants more consideration than it has received in the Pacific to date.

There are still problems inherent in this form of analysis which will, with time and more information, be overcome. These problems must be taken into consideration in the interpretation of trace element results.

The use of trace element analysis as a method of dietary reconstruction requires multi-element analysis as well as some understanding of the food resources available to the prehistoric populations being investigated. Strontium, reflecting the vegetable component in diet, and zinc as an indication of meat consumption, are the most frequently used and reliable elements so far investigated. Their reliability for accurately reflecting the composition of diet is dependent upon analytical method and accuracy, and on the ability to overcome problems of interpretation resulting from environmental, anatomical, age and postdepositional factors.

## APPENDIX 1
## BONE SAMPLE PREPARATION AND ANALYSIS BY ATOMIC ABSORPTION SPECTROPHOTOMETRY

A suitable method of cleaning bone samples for analysis is as follows. Remove all adhering soil by gentle brushing, followed by immersion in an ultrasonic bath containing deionized water. Dry the samples (48 hours at 35°C is adequate) then grind to powder. The ground samples should then be dried once again to remove as much moisture as possible, using a vacuum oven with a liquid nitrogen vacuum trap. Following this, samples should remain in a drying room or equivalent so that reabsorption of moisture is at a minimum.

Digest one gram of bone powder from each sample in 3 ml concentrated nitric acid on a hot plate. Dilute the solution to 100 ml with deionized water containing 1.0% lanthanum oxide and 0.5% potassium chloride. Following this, filter the solution into polyethelene bottles using weighed Toyo glass filter papers (equivalent to Watman GF/C) to remove all remaining soil/silica residue. Storage of samples in polyethelene containers is recommended. Storage in glass may result in contamination of samples caused by the movement of ions from the glass surface into the solution. Dry and reweigh the filters to determine the residue weights which need to be accounted for when converting trace element concentrations into ppm by the method of standard additions. This method was found to be necessary to avoid chemical interference caused by the high level of phosphate in bone, which is not completely offset by the addition of lanthanum. The method of standard additions also involves the bone from one sample acting as the standard for all samples from the same area/site. This enables the value of the standard to fall in the same range as that for the samples, thereby increasing accuracy during conversion to final trace element concentrations. To provide calibration curves, prepare standards with the addition of 3 ml nitric acid, 1.0% lanthanum oxide, and 0.5% potassium chloride as for the bone samples. To this add the following concentrations of Atomic Absorption Standards of strontium nitrate and zinc nitrate, for five strontium and four zinc standards:

| Element | Concentration (ppm) | | | | |
|---|---|---|---|---|---|
| | 1 | 2 | 3 | 4 | 5 |
| Strontium | 2.0 | 4.0 | 6.0 | 8.0 | 10.0 |
| Zinc | 0.5 | 1.0 | 1.5 | 2.0 | |

To produce a stock lanthanum solution, dissolve 58.65 g lanthanum oxide in 250 ml concentrated hydrochloric acid. Dilute this to one litre to provide a 5.0% lanthanum solution. Then add 4.66 g of potassium chloride to provide a 2.5% potassium concentration. Dilute the stock solution five times when adding to bone samples.

I have undertaken atomic absorption analysis using a PYE UNICAM PU900 atomic absorption spectrophotometer. Trace element concentrations of strontium and zinc were determined in an air acetylene flame with the burner head centered horizontally. The values for the machine parameters were as follows:

| AAS Parameters | Strontium | Zinc |
|---|---|---|
| Maximum lamp current (mA) | 10.0 | 9.0 |
| Fuel flow rate (l/m) | 0.8 | 0.8 |
| Wavelength (nm) | 460.7 | 213.9 |
| Band pass (slit) (nm) | 0.5 | 0.5 |
| Lamp current (mA) | 7.5 | 6.6 |

The machine was recalibrated after every ten samples were analysed.

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