CHAPTER NINE

CONCLUSIONS

9.1 Introduction

This study employed enamel Pb and Sr isotopes and concentrations to identify rural to urban mobility in Mediaeval Gloucester and suggest cyclical mobility during the Neolithic at Monkton-up-Wimbourne. The Viking period case study at Cnip, Lewis, found two migrants to the island (male and female) who originated on Tertiary volcanics, e.g. most probably Iceland, Faeroe Isles or Antrim in Ireland, but not Norway. The study also found evidence for dietary or status differences amongst the Iron Age population at nearby Galson and a male/female pair of possible migrants, but in the absence of more extensive environmental analysis (water, soils, flora, fauna), the results are somewhat tentative. Moreover, there are indications of subsistence change at the Iron Age/Norse boundary, consistent with other stable isotope, archaeological and osteological lines of evidence. Certain skeletal traits appear to be diagnostic amongst the Iron Age and Norse populations but numbers are too small to draw firm conclusions.

The investigation of Anglian settlement at West Heslerton was somewhat inconclusive. The range of Pb and Sr isotope ratios obtained could equally be explained by an English origin and was also present amongst the Iron Age and Neolithic/Early Bronze Age individuals. It is, unfortunately, possible that there is insufficient difference between the geology of eastern England and that of northern Europe, Denmark or coastal Norway to distinguish immigrants by this method. Nevertheless, there is evidence for the presence of two groups in the cemetery, whose differences are not explicable in terms of status or changes over time. Moreover, some promising correlations between origin and certain grave goods were obtained: all burials with wristclasps and cruciform brooches fell into the non-local group whilst only burials in the local group contained girdle-hangers and walnut amulets. Annular brooches were ubiquitous and undiagnostic. Most importantly, the demographic make-up of the "non-local" segment of the West Heslerton population, wherever they originated, did not consist solely of males or weapon burials whilst the majority of juveniles appeared to be of local origin.

In addition to these case-specific conclusions which are discussed in more detail in their relevant chapters, a number of conclusions can be made common to the general application of Pb and Sr isotope ratio analysis to human remains. These are outlined in the following sections, along with suggestions for further investigation of both the fundamentals of the technique and future applications of the method to studies of prehistoric mobility.

9.2 Teeth

Enamel formation should not be confused with mineralisation. Whilst the dentine matrix forms and mineralises within a very short period of time (i.e. a few days) making the two processes to all intents and purposes simultaneous, enamel does not, and *formation* occurs before *mineralisation*. Immature enamel forms on dentine that is already fully mineralised, thus making the two tissues co-genetic. This process is clearly seen in archaeological juvenile dentitions because fully formed, but incompletely mineralised, enamel tends to discolour in a similar manner to bone, whereas fully mineralised enamel does not. It follows, therefore, that enamel crowns are formed morphologically before they are completely mineralised, and the visible incremental structures (neonatal line, lines of Retzius etc.) relate to the formation of the organic enamel protein matrix *not* to crystal growth during the mineralisation period. Moreover, this enamel protein scaffold which guides the nucleation and elongation of the enamel crystallites, is eventually resorbed (contra dentine where it mineralises *in situ*) to permit the enamel crystallites to expand widthways.

There is, therefore, no obvious relationship between Pb and Sr in enamel and such incremental structures. Most of the enamel-Pb and Sr will be incorporated during the massive influx of mineral ions that characterises the maturation phase. As the oldest enamel is at the very centre of the mature enamel crystals with the most recent on the outside (in a manner analogous to the growth of tree rings), any time-dependent analysis should be done at the individual crystal level. It is currently not possible to analyse Pb and Sr isotope ratios *in situ* in such small concentration samples at this resolution. Neither does the enamel mineralise in a simple, progressive wave of advance from the cusp tip down to the cervical region, e.g. enamel nearest the EDJ mineralises first and thin enamel is likely to mineralise before thicker enamel. Moreover, the length of time enamel maturation takes (months to years), the timing of its onset, and how maturation relates to crown formation times as a whole, are not known. Such parameters probably vary between teeth and maturation is likely to occur at the end of the crown formation period rather than at the beginning.

The pilot studies of modern teeth indicate that the Pb and Sr isotope ratios of enamel and the co-genetic crown dentine remain indicative of childhood origin. There is no evidence for post-eruption uptake of Pb or Sr into enamel. However, the resorbing roots of deciduous teeth are more sensitive to changes in origin than permanent teeth and it appears that the dynamic environment of the dentine during this process enables Pb and Sr to be incorporated from the contemporary diet. However, deciduous enamel appears to retain evidence of origin (or the skeletal signatures of the mother) even in exfoliated teeth. Consequently, the dentine of resorbing deciduous teeth reflects recent exposure rather than *in utero/in vivo* signatures. Overall, the data imply that deciduous teeth are considerably more dynamic and Pb and Sr incorporation during mineralisation *in utero* is more complex than for permanent teeth that mineralise after weaning.

Modern co-habiting siblings possessed inexplicable differences in their Sr and Pb isotope signatures that exceeded analytical error and intra-tooth variation. This probably represents only the minimum possible variation within a population and is an important consideration for the identification of migrants in archaeological populations. However, enamel from the same tooth, enamel from antimeres (e.g. upper left canine/upper right canine), and enamel from co-genetic teeth replicated extremely well for both modern and archaeological teeth. In most of the enamel samples, Sr reproducibility in particular was better than the standard, leading to the conclusion that enamel is an exceptionally homogeneous tissue.

Variations were, however, found *within* the dentition of a single individual, which indicate a change in food or water sources between teeth mineralising at different ages, and between the two mother and child pairs identified by aDNA analysis (Monkton-up-

Wimbourne and West Heslerton). In both cases, the mother appears to have been a non-local and a move towards the place of burial can be observed in the permanent teeth of the offspring. However, in the case of Monkton A, the deciduous tooth is intermediate between that of the mother and the child's permanent tooth, suggesting the mother had moved prior to pregnancy but A received some Pb and/or Sr contribution from the mother's skeletal stores. Results from several studies demonstrate that individuals may have the same Sr but different Pb and *vice versa* and, whilst highlighting the differential behaviour of Pb and Sr generally, it also suggests that the transfer and source of Pb and Sr from mother to child is not the same. This may have implications for the interpretation of enamel formed before weaning if the mother is an immigrant. Whilst teeth formed pre and post-weaning may provide complementary information it is vital to consider their different origins when choosing samples. However, the difficulty with assessing weaning age in archaeological populations is acknowledged. The decreased likelihood of juveniles being immigrants has been shown to provide a useful additional parameter for assessing the local range of Pb and Sr at a site where the majority of the population is likely to have been sedentary. Contrary to this, the juveniles at Monkton displayed a range of movement that was non-random and demonstrates the potential of within-dentition analyses to identify cyclical mobility practices such as transhumance amongst mobile populations.

9.3 Diagenesis

The results indicate that enamel is highly resistant to diagenetic alteration by Pb or Sr from the burial environment under all the burial conditions investigated in this thesis and, thus, a reliable reservoir of biogenic Pb and Sr. This is concluded from the following trends:

1. Consistently small enamel concentrations in the face of large increases in co-genetic crown dentine concentrations which are not observed in modern correlates
2. Consistent movement of archaeological crown dentine ratios towards burial soil-leach ratios
3. Variability amongst enamel ratios from different individuals in contrast to the trend towards homogeneity in the dentine ratios

4. Where enamel homogeneity is observed in modern samples (i.e. between samples from a single tooth, antimeres and co-genetic teeth from the same dentition) this is present in archaeological enamel samples also, but not in the associated crown dentine

Whilst it cannot be determined with certainty what the *in vivo* enamel ratios were at the point of burial, even in the case of the two Pb coffin burials isotope ratios were obtained that did not have solely coffin Pb as their source. This retention of even the smallest vestige of biogenic Pb in such a Pb-contaminated burial environment is reassuring. It is also of interest that the molar enamel was considerably less contaminated than the premolar from the same individual (G339) implying that Pb was unable to penetrate the thicker enamel core to the same extent. However, whether this was simply because the molar enamel was thicker or because more hairline cracks and fissures were present in the premolar facilitating diffusion, is unknown. It should be considered, therefore, whether molar teeth are a better choice of sample than the premolars originally preferred in this study.

Dentine is considerably less resistant to diagenetic Sr and Pb incorporation, to the extent that it can be proposed as a useful proxy for mobile soil Sr and Pb over the period of time the remains have been buried. This would apply to bone also by implication, but because bone is continually remodelling *in vivo*, there is no guarantee that bone and enamel ratios would have been the same at the time of burial. However, those of primary crown dentine and enamel should, and the results of this study made it possible to suggest that enamel would provide a good control for testing Sr decontamination techniques developed for bone, on the associated dentine. If the technique was successful and only Sr incorporation rather than turnover had taken place, it should be possible to recover the enamel Sr isotope ratios from the co-genetic crown dentine.

Enamel and dentine preservation scores indicate that even macromorphologically poorly-preserved teeth retain enamel isotope ratios that are very different from the burial soil leaches. The evidence suggests that only incompletely mineralised enamel is an unreliable archive of *in vivo* signatures and should be avoided or treated with the greatest caution. The opposite is true with dentine. Even dentine that appeared very well preserved had equilibrated to some extent with the burial soil; the final dentine ratio appears to be a function of how different the enamel ratio is (and hence, the dentine ratio was) from the soil, rather than the tooth preservation score. The Monkton study, for example, where exceptionally well-preserved burials were excavated from chalk, produced dentine Sr and Pb isotope ratios intermediate between the enamel and the chalk ratios despite the teeth being almost indistinguishable from modern examples. It is concluded that gross morphological preservation is no guide to the integrity of the Pb and Sr isotope ratio in either enamel or dentine but for different reasons. Mature enamel retains biogenic ratios irrespective of macromorphological preservation whilst dentine does not, irrespective of macromorphological preservation. Radosevich's (1993) assertion that good survival of skeletal material occurs precisely *because* of incipient fossilisation and hence, incorporation of Sr from the burial soil, corroborates these observation. This strongly suggests that it is the level of *in vivo* mineralisation that is most relevant for samples submitted for Sr and Pb isotope analysis not post-mortem preservation. Mature enamel, already a virtual fossil, is thus stable and resistant to change. Dentine is not. The isotopic integrity of incompletely mineralised unerupted permanent teeth is also, therefore, questionable. Furthermore, these conclusions have important implications when choosing the sampling location from which to remove enamel, as the histologically complex enamel beneath the cusps and fissures is less well mineralised than that of the flatter, tooth walls.

Under certain burial conditions, e.g. chalk, Sr diagenesis of the dentine is much more likely to occur than Pb diagenesis. This may simply be a factor of greater Pb solubility and mobility in acidic environments whereas Sr is more mobile under alkaline conditions (section 3.6.3.2). However, *in vivo* Pb concentration will determine the extent to which soil Pb is able to alter the biogenic Pb isotope ratios. The similarity observed between enamel and dentine Pb isotope ratios in the Roman and Mediaeval samples is present because they were the same prior to burial, already at a high concentration and hence, not so easily swamped by burial soil Pb. Susceptibility to diagenesis will, therefore, depend on how great the enamel-Pb concentration was before the tooth was buried and the subsequent availability as well as the solubility and mobility of Pb in the burial environment. For example, although Winchester chalk-Pb was very different to that in the Roman teeth, it had no discernible effect on the enamel or dentine ratios, whereas amongst the low-Pb, Neolithic Monkton samples the dentine was altered towards chalk ratios even in the low-Pb environment of the chalk. Interestingly, this was also the case with the Bronze Age soil burial at Monkton where surface Pb of anthropogenic origin had no effect on the resulting dentine Pb-ratio, suggesting particulate Pb is insoluble, and thus immobile, in chalk soils.

9.4 Sample selection and preparation

This study has highlighted the absolute necessity of ensuring that it is clearly understood which tissue is being extracted during sample preparation. The analysis of whole teeth, modern or archaeological, is a meaningless exercise, as the sample would consist of two very different types of tissue. The accidental incorporation of just a small amount of dentine in archaeological enamel samples may drastically alter both the concentration and the isotope ratio of the final result. This is especially so with Sr, where the concentration of the archaeological dentine is frequently several times that of the enamel, and the isotope ratios are almost always intermediate between the enamel and the burial soil leaches. Moreover, if the sample is not constrained to a specific tissue it is impossible to compare results between studies and different archaeological sites where burial conditions may be very different. Interpretation also becomes problematic, because formation, mineralisation and subsequent behaviour ante- and post-mortem are very different between dentine and enamel. Even within the tissues of enamel and dentine there are age and exposure-dependent changes that will invalidate comparisons between adults and juveniles, if such tissues are not identified and removed.

It is, therefore, strongly recommended that core enamel (enamel removed of all surfaces and the EDJ) is the best way of standardising archaeological samples, as the analyst can then be assured the tissue is homogeneous, resistant to diagenetic alteration and indicative of a specific time during childhood when the enamel was mineralising. During sampling it is vital to be conversant with the 3-dimensional internal tooth structure and to remove the enamel sample from the thicker part of the flat tooth wall; enamel beneath the cusps and in the occlusal fissure region is less well mineralised and hence, susceptible to diagenesis, and more difficult to clean and prepare. Moreover, results from this study suggest that molar enamel may be preferable to that of bicuspids or cuspids, as it is thicker and possibly less prone to hairline cracks and fissures. It is further recommended that incompletely mineralised enamel is not used for Pb and Sr isotope analysis as it appears, like the less well-mineralised dentine, to be highly susceptible to incorporation of elements from the burial environment. Such immature teeth are easy to identify in archaeological juvenile dentitions because they are opaque and discolour during burial in a similar manner to the surrounding bone, whilst fully mature teeth remain white and translucent. Finally, it is important to realise that teeth formed from the diet of the mother *in utero*, or during a mixed diet prior to weaning may be extremely complex to interpret. The transfer of Pb and Sr from mother to child *in utero* and during breastfeeding cannot be assumed to be the same for both elements, in either quantity or source (section 2.4).

9.5 Pb versus Sr

No evidence was found that Pb and Sr behave in the same manner in the geosphere or biosphere. Neither do they exhibit many similarities in biological organisms specifically, either *in vivo* or post-mortem; in fact, the reverse appears to be true. This observation appears contrary to the vast majority of statements made in the associated literature that assume that both Pb and Sr substitute in an identical manner at Ca sites in the carbonate hydroxyapatite lattice. This assumption appears to be based on two circumstantial facts: that Pb and Sr, like Ca, form divalent ions; and that the vast majority of the Pb and Sr body burden in animals is found in the skeleton. Divalency of itself is no guide – there are many elements that form divalent cations and some have very different functions in the body. Zn, for example, does so and also concentrates at areas of on-going bone mineralisation but its role in bone formation is to mediate and enable rather than as a structural building block. Moreover, all evidence suggests that whilst elemental Sr is a harmless Ca substitute, this is far from the case with Pb and there are complex antagonisms at play between Ca and Pb. As discussed in section 3.5.3, other researchers such as Neuman and Neuman (1958, 94/95) have expressed such doubts for many years but have been little heeded.

Empirical observations from modern teeth, whilst admittedly few in number, suggest that modern enamel has slightly *more* Sr than dentine and *less* Pb than dentine. In archaeological samples the situation with Pb does not change but for Sr it is reversed and dentine almost always has more (usually an order of magnitude more) than enamel. A negative correlation was found between Sr isotope ratios and Sr concentration but this may be site-specific and dependent on the underlying geological substrate. There is no correlation between Pb concentration and Pb isotope ratios, except that ore Pb isotope ratios are associated with the greatest enamel-Pb concentrations. Interestingly, a significant *negative* correlation also exists between Sr and Pb concentrations but no correlation between $^{87}Sr/^{86}Sr$ and $^{207}Pb/^{206}Pb$ ratios, adding further weight to the observation that the same mechanisms are not controlling their uptake or incorporation in skeletal tissue. Sr is more sensitive to changes in origin than Pb and it is suggested that this is due to greater mobility, faster turnover and hence, shorter skeletal residence time. Pb appears to be tenaciously retained in dental tissues and bone but no evidence for age related accumulation was found. Sr does not appear to increase but did turnover in the single bone sample analysed. Accordingly, all evidence suggests that there are very different factors controlling the uptake of Pb and Sr into skeletal tissue.

Several broad trends were observed in Sr and Pb isotope ratios during the course of this study. Sr isotope ratios remain indicative of geological origins in all periods so far investigated. However, Pb enamel ratios are only linked to the local geology until the advent of widespread metal usage at which point the link is severed (Figure 9.1). Accordingly, there is a clear trend of Pb isotopes in geologically related communities to move away from the natural country rock signature and converge on anthropogenic ore Pb along with a concomitant rise (i.e. >1ppm) in enamel-Pb concentration (Figure 9.2). This phenomenon has been termed cultural focussing (J. Evans pers. comm.). It is consistent with the findings of Carlson (1996) (section 4.4). Data from this study indicate that this has occurred by the Roman period (Figure 9.3). The timing is, however, clearly geographically and culturally variable and the link may be re-established if lifestyle and status changes. On Lewis, which was little affected by the Roman occupation of southern Scotland, individuals (i.e. Gals IV, Gals 93) with an anthropogenic Pb ratio and elevated concentration were present by the 1st – 3rd centuries AD, although it is possible that the individuals concerned may not be indigenous to the islands. If further work establishes this Pb isotope focussing as a distinctive characteristic of British populations during certain periods, it would provide a means for identifying immigrants who are focussed on a different ore source. This

"British Pb" ratio changes once importation of Pb commences in the mid 19th century and becomes progressively less radiogenic. Contemporary English subjects have skeletal Pb that is shifted from native English Pb isotope ratios to ratios dominated by Australian Precambrian Pb ores (i.e. $^{207}Pb/^{206}Pb$ >0.872). A modern British signature can still be identified, but it is different to that prior to the 20th century.

After the Roman period, individuals may still retain a Pb signature indicative of geological origin if it is not swamped by anthropogenic Pb. Pb may, therefore, be used as a status indicator in certain periods and localities, for example between urban and rural origins, although it must be remembered, for example, that a hard water supply can significantly reduce Pb uptake. Apart from the Pb coffin burials, enamel-Pb concentrations >1ppm were only obtained at a restricted range of isotope ratios that converge on the lower centre of the English Pb ore field (Figure 9.2). Pb ores from around Dublin also plot here. Low enamel-Pb concentrations (<1ppm) have variable ratios and frequently plot outside the Pb ore field.

At the sites investigated, variable Pb isotope ratios (i.e. prehistoric) tend to go hand in hand with invariant Sr isotope ratios, whereas variable Sr isotope ratios all have fairly invariant (i.e. ore) Pb (Figure 9.1). The second observation can be explained by the swamping of the natural low-Pb country rock ratios by the ubiquitous high-Pb anthropogenic ratios but no certain explanation for a reduction in Sr variability in the face of increased Pb variation can be currently proposed. The results may have arisen solely because of the specific case studies investigated in this project. For example, in the Lewis study, which produced some of the most variable enamel-Pb isotope ratios, the machair environment was heavily dominated by marine Sr, which is extremely invariant and, therefore, renders Sr somewhat undiagnostic in coastal habitats. Moreover, it is not known if the prehistoric individuals genuinely had very variable Pb; it may just appear so because it is more variable than the relatively invariant ore Pb.

Enamel Sr isotope ratios obtained in this study ranged from 0.7078 – 0.7143, which in geological terms and in comparison with other studies, is quite a restricted range but highly typical for the predominantly Cretaceous/Jurassic bedrock at the sites investigated. Populations at all the sites investigated were found to have a range of Sr isotope ratios that were a mixture of local geological Sr and rainwater Sr, i.e. if the underlying geology is *less* radiogenic than rainwater the indigenous populations will have a ratio *more* radiogenic than the geology; if the geology is *more* radiogenic than rainwater the indigenous population will have a ratio *less* radiogenic than the geology. It is concluded, therefore, that inputs from rain and seawater bias the enamel Sr of humans inhabiting a maritime environment towards the marine ratio of ~0.7092 thus resulting in a tendency for Sr isotope ratios to cluster around 0.709 ±0.001 in the studies investigated. Moreover, such an observation implies that when searching for possible origins of a British burial exhibiting a Sr isotope ratio of 0.7078 or 0.7143, a source <0.7078 or >0.7143 should be sought in order to balance the marine-derived contribution.

For this reason, there is a clear need to define the "local" environmental Sr isotope signature before conclusions can be drawn between immigrant and local populations beyond the immediate burial ground. Juveniles are potentially the group that will contain the highest proportion of "locals", although, it is acknowledged that very few are present at some burial sites and in certain periods. Moreover, the transfer of Pb and Sr from the mother to the child both *in utero* and whilst breastfeeding would benefit from closer study. Results presented here suggest that it is not at all straightforward nor the same for both elements. Although confirmed mother and child pairs are rare, aDNA analysis may enable more to be identified, and if so, it would be very useful to compare the mother's enamel with enamel formed at various stages during the child's development.

Analysis of contemporary plant remains and animal teeth from associated settlement sites, along with soil leaches and water samples is recommended, although such dietary inputs are obviously only valid if children were eating them. A community's range of Pb and Sr isotope ratios is an averaging over time of many different inputs which ultimately characterises their culturally-mediated dietary sphere and it may not be possible to recreate them or the social systems controlling access to food and water sources. Neither will every such input have equal loading. For example, animal meat and milk are very low in Sr, and thus may not have a great impact on the resulting Sr signature of humans, whereas plants, a much richer bioavailable source, will. Analysis of animal teeth may therefore, be of limited value unless the community were, for example, pastoral and almost entirely carnivorous. Moreover, it is concluded that for both Sr and Pb, water source carries a considerable weighting for the resulting enamel signatures. All Sr signature ranges are markedly biased towards 0.7092 and the amount of soluble Pb ingested is very dependent upon whether hard (high Ca, low Pb) or soft (low Ca, high Pb) water sources are present. The analysis of modern water sources and local plants is clearly problematic and require careful interpretation when projecting the results back onto archaeological populations. Direct evidence from the people themselves is still obligatory because no matter how many environmental samples are analysed, many assumptions will have to be made to reconstruct, in its entirety, the diet of the children in a community.

In machair and coastal environments, marine Sr (0.7092) swamps the Sr from terrestrial sources. Sr is, thus, an inappropriate element when attempting to differentiate between different geological coastal areas. It is currently unclear how far inland this marine effect on the soils will extend but clearly this may be a problem for small, maritime island studies. Enamel Sr concentrations may help to differentiate between marine fish and mammal meat or milk (low Sr) and cereal crops (high Sr) and may provide supporting evidence for C and N-isotope analysis. For prehistoric maritime environments, before access to metal artefacts becomes widespread, Pb may prove a better discriminant. Pb is only present at very low concentrations in seawater and, as a consequence, terrestrial Pb will predominate over marine Pb.

9.6 Utility of archaeological indicators

Only the study at West Heslerton had sufficient numbers of individuals to make it possible to consider skeletal variation amongst the population. Unfortunately, the very poor bone preservation made skeletal variation impossible to assess in most individuals. A preliminary assessment of skeletal variation and isotope analysis of the Iron Age and Norse populations on Lewis suggested some differences between and within the two populations which may indicate immigrants (e.g. see Gals 93 and Cnip D Table 8.2) but the small number of individuals precluded any firm conclusions being made. Unfortunately, the cost and sample preparation time needed to carry out Pb and Sr analysis by TIMS means that it is not likely that enough samples can be analysed to make population-level studies feasible, and it is at this level that biological variation needs to be assessed. However, the availability of the new generation of LA-MC-ICP-mass spectrometers may provide the necessary means to increase the sample throughput sufficiently to enable population size studies to be carried out, at least for Sr. However, the very low concentrations of enamel-Pb in most prehistoric populations may still constitute an analytical obstacle.

Burial practices and grave goods at both West Heslerton and Lewis were investigated and some promising lines of enquiry suggested. Such an endeavour clearly needs embedding within archaeological theory and the context of the site and period. It was interesting that Anglian burials with wristclasps, an artefact that Hines (1984) considers the clearest indication of the presence of migrants from Norway, all fell into the "non-local" group at West Heslerton. Although their enamel Sr isotope ratios are within the English range, they do not rule out a coastal Norwegian origin. On Lewis, Gals 93 was the only individual not buried in a stone long cist and buried with grave goods (pot, bone pin, iron brooch). He had the most radiogenic Sr of all burials analysed. Again, such a ratio cannot currently be ruled out as indigenous, but it was not matched by any other samples, animal or human, so far analysed on the island, although the female Gals IV showed similar trends in both Sr and Pb.

9.7 Soil analysis

The soil leaches performed during this study used deionised water and weak acetic acid reagents. They were performed principally to provide a comparison between the burial environment and the tooth samples. Whilst many were from sealed contexts or from within the cranium and should, therefore, be contemporary, it is difficult to be certain that soils have not been redeposited or altered in some way since burial. However, where enamel and dentine samples were analysed it is clear that, almost without exception, the crown dentine sample is intermediate between the soil and the enamel suggesting that the co-genetic dentine has incorporated Pb and Sr from the burial environment post-mortem. At some sites, e.g. Blackfriars, it would seem that dentine constitutes a good proxy for the time averaged mobile soil Sr and to a lesser extent, Pb.

Water leaches of alkaline chalk and surface chalk soils, that produced enough Pb to analyse, appeared to be removing predominantly particulate Pb of an entirely different origin to the chalk. It is, therefore, recommended that a deionised water leach be performed prior to the acetic acid leach to remove historical anthropogenic Pb. Acetic acid leaches characterise the diagenetic Pb signature of the chalk well which would, perhaps, be anticipated given the increasing mobility of Pb as soil pH decreases. For Sr, there is little appreciable difference between the water and acetic leaches of the chalk but for the soils at Blackfriars, the acetic leaches appear to better characterise the mobile Sr that has contaminated the dentine. However, acetic acid soil leaches do not appear to extract the mobile Sr from the alkaline coastal soils of Lewis quite as well as the water leaches and this may be due to reduced Sr mobility as pH decreases.

At Lewis and West Heslerton, soil leaches were analysed from contemporary cultivation areas if these were different from the soils in which the burials were made. It is clear from both studies, and particularly from the Pb results from Lewis, that soil leaches do not, on their own, explain the range of isotope signatures obtained from humans or herbivores. Evidently, either some humans and animals were immigrants to the area/island or a broader environmental sampling programme must be embarked upon if the local environment is to be properly characterised. Nevertheless, as a method for assessing and identifying diagenetic change, soil leaches were very useful and enabled a site-specific diagenetic vector to be established. At Cnip and Blackfriars, this provided a means to identify individuals who showed a different diagenetic vector to other individuals in the cemetery.

9.8 Suggestions for future work

It is evident that there is much that can be done on the fundamentals of this technique that will greatly aid future applications. The problem is the multi-disciplinary nature of the work, which unfortunately, or perhaps fortunately, can create obstacles that are unlikely to be overcome without inter-disciplinary collaboration. The following future directions are suggested:

1. Technical developments, such as the new generation multi-collector ICP-MS, that enable smaller sample sizes to be used would open up access to rarer and more important collections such as hominid, Neanderthal and Mesolithic remains. Currently, analysis by TIMS and solution ICP-MS requires a substantial portion of the tooth enamel to be destroyed. The necessary innovation requires Sr and Pb isotope ratio data to be collected at trace and ultra-trace levels using laser ablation sampling in high (~37%) calcium-matrix enamel samples. This would enable the sample to be ablated directly from the enamel surface with only microscopic change to the intact tooth.

2. Instrumental advances, such as the MC-ICP-MS, which result in increased sample throughput, and hence lower cost, would be advantageous to archaeological applications as many more samples can be analysed. Again, using a laser ablation sampling method removes the lengthy sample preparation required for TIMS and solution ICP-MS analysis. This would aid the characterisation of local signatures and considerably expand the extremely limited existing database of geographical human variation. Larger sample numbers will also enable a more useful investigation of the skeletal and craniometric traits to be carried out and compared with the isotope data.

3. Analysis of cremated remains is currently untested. Before archaeological cremations can be analysed with confidence, it would be useful to compare before and after enamel isotope signatures to ascertain whether any fractionation or incorporation of Sr or Pb occurs during the cremation process. The excellent reproducibility seen here between antimeres suggests this could be achieved by removing a tooth from an animal carcass prior to experimental cremation and then analysing the remains of the antimere cremated *in situ*.

4. Further feeding experiments to clarify which dietary component makes the greatest contribution to metabolised Sr and Pb are necessary to investigate

dietary variation. The results of this thesis suggest that enamel Sr derives predominantly from the water source and plants, rather than meat and milk which are very low in Sr. The isotope signatures of plants and water sources are clearly more difficult to establish in archaeological populations, but carbonised plant seeds and pollen do survive in many archaeological contexts.

5. The advent of localised human anthropogenic Pb exposure could be investigated using the simultaneous changes observed in both Pb concentration and isotope signature in the studies presented here.

6. The method is particularly suited to the investigation of mobility amongst island communities and the initial and subsequent population of islands, as they represent a discrete geological environment rather than one continuous with adjoining localities. However, as discussed in Chapter Eight, Pb may prove more useful than Sr in these applications.

7. Archaeological questions other than large-scale migrations may be addressed using this method: exogamy/endogamy, i.e. whether males and/or females were moving for marriage; trading links; transhumance and cyclical mobility, e.g. between coastal and interior areas or different geological substrates; and warfare.

8. Pb and Sr isotope analysis would clearly be highly complementary to stable isotope systems which have also been used to examine human mobility such as oxygen (e.g. Fricke *et al.* 1995), carbon and nitrogen (e.g. Cox & Sealy 1997; Richards *et al.* 1998; Sealy *et al.* 1995) and, most recently, sulphur (Richards *et al.* 2001).

There are many unanswered questions concerning the fundamentals of Pb and Sr uptake in enamel and dentine, how and when they are incorporated and how ensuing concentrations and isotope ratios reflect (if at all) the locality and concentration in the diet. It would be helpful to narrow the exact onset and timing of mineralisation by investigation of juvenile dentitions. Whilst this is a difficult endeavour in living children, archaeological dentitions offer an alternative as there is a clear colour difference between fully mineralised, and fully formed but incompletely mineralised, crowns. Once mineralised, it would be useful to investigate which teeth provide the best sample of post-weaning Pb and Sr signatures. Preliminary conclusions suggest this may be second or third molar enamel from the tooth wall but this warrants further study. Studies of mother and child pairs and siblings would greatly help to illuminate the differential transfer of Pb and Sr from mother to child before and after birth.

This study has concluded that there is little evidence that Pb and Sr behave in the same manner as passive Ca substitutes but it is not known if Pb and Sr uptake is entirely independent of each other or whether complex synergisms or antagonisms are present. Studies of pre-metallurgical individuals where both Pb and Sr exposure is derived from natural sources would clarify whether they do vary together and in accord with the local geology, or whether increased uptake of one affects, directly or indirectly, the other. Much of the interpretation of what Pb and Sr isotope ratios are bioavailable from which geological substrates is obtained by extension from other studies and geological maps, rather than direct investigation. Unfortunately, no geographical atlas of *bioavailable* Pb and Sr isotope variation exists. Neither is it firmly established which reagents are most suitable for each soil type, or indeed each element, to begin compiling one. Such an endeavour is, however, vital if migration studies are to progress beyond the identification of immigrants to tracing the migration stream back to the place of origin.

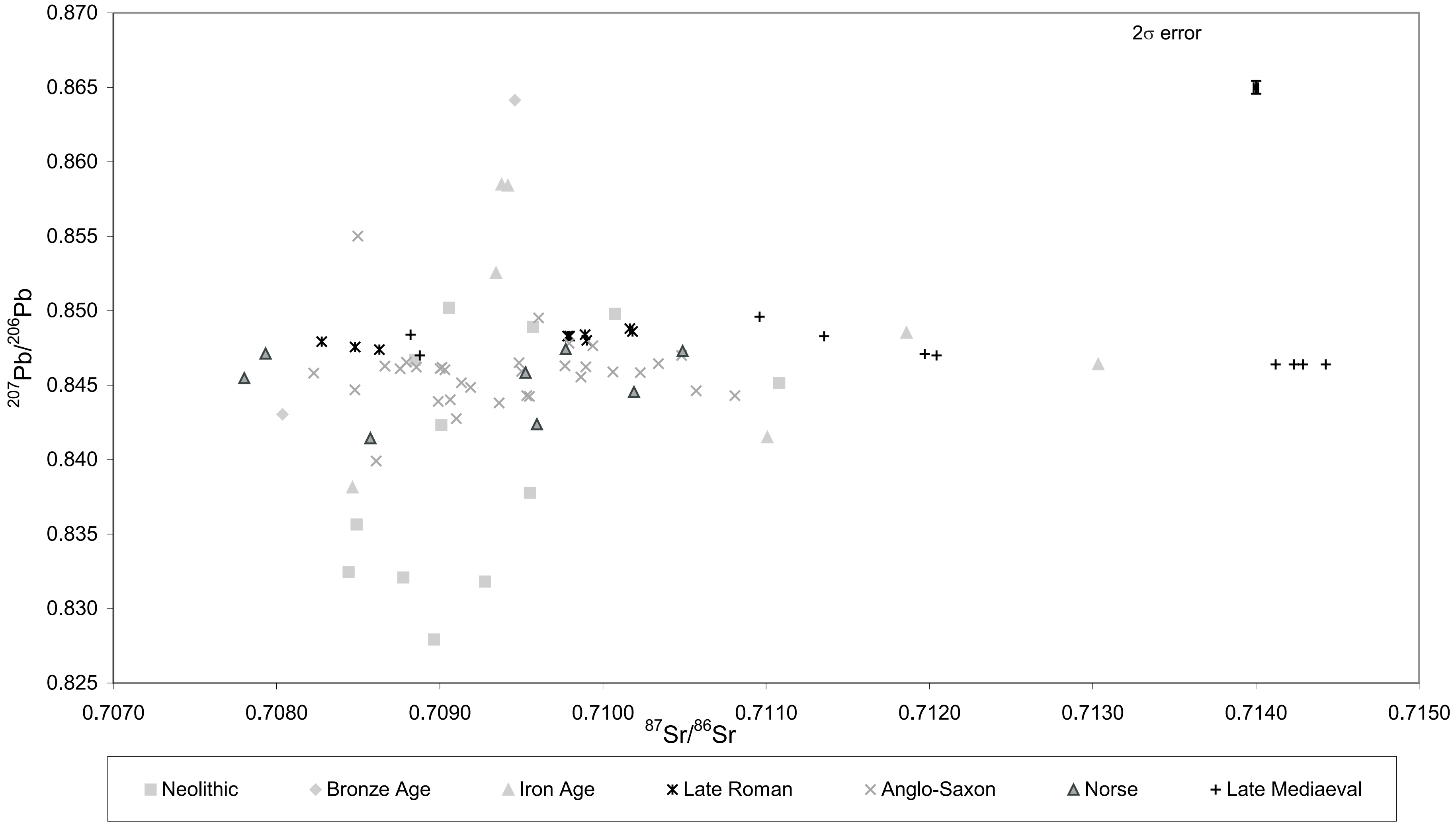

Figure 9.1 ^{87}Sr/^{86}Sr ratio v ^{207}Pb/^{206}Pb plots of combined case study data showing differential variability of enamel-Sr and Pb. Note how the Late Roman and Late Mediaeval individuals have invariable Pb but variable Sr isotope ratios suggesting no geological information is present in the Pb isotope ratios. Conversely, prehistoric samples exhibit very variable Pb isotope ratios but much less Sr isotope variation. This could be site specific, however, and arise from Sr homogeneity in the chalk and machair-dominated environments where most of the prehistoric burials were located.

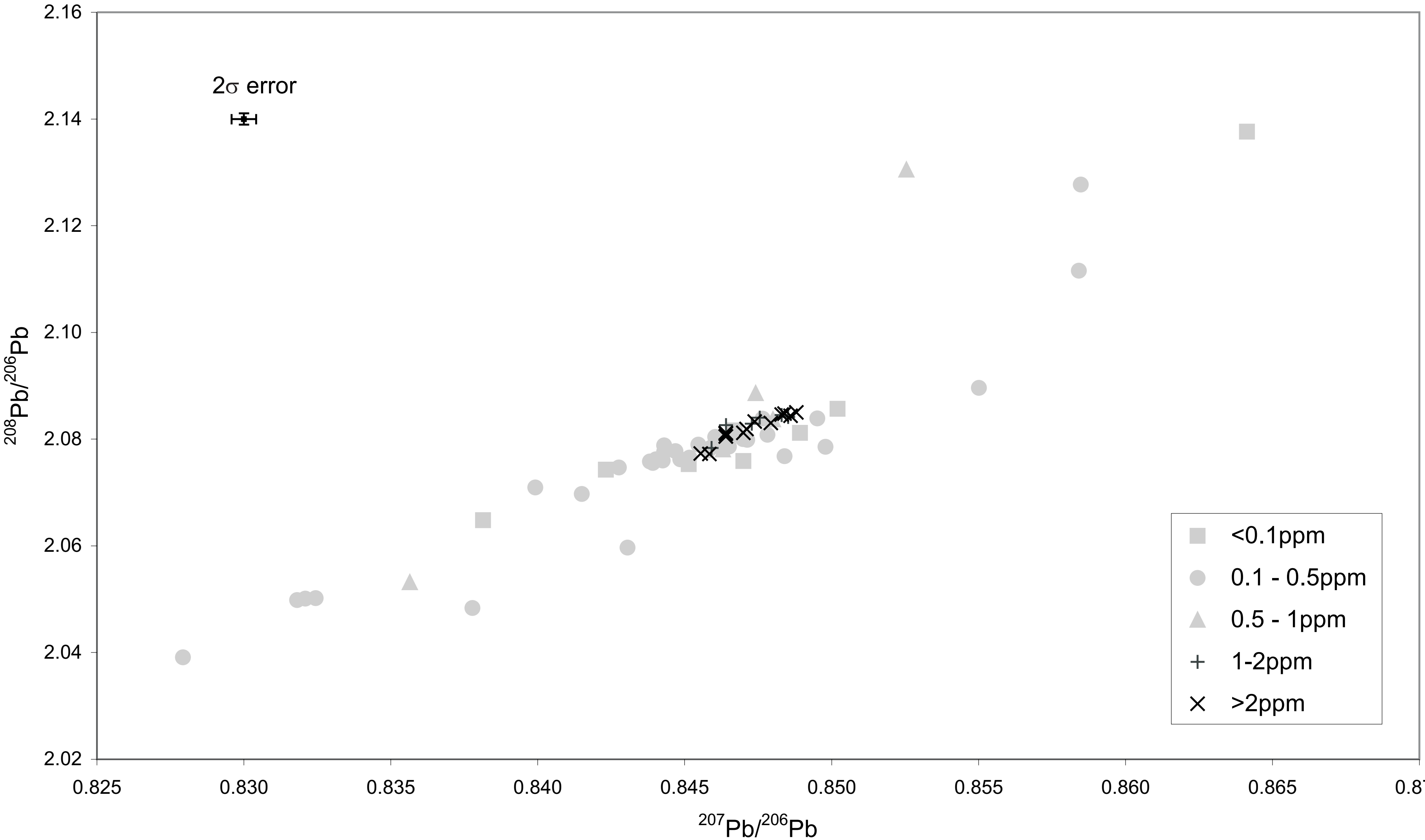

Figure 9.2 ^{207}Pb/^{206}Pb v ^{208}Pb/^{206}Pb plots of combined case study data showing cultural focussing of the enamel-Pb isotope ratios with increasing concentration. Individuals with the greatest Pb concentrations are tightly clustered at the centre of the plot whilst low concentration individuals have a much more widespread distribution.

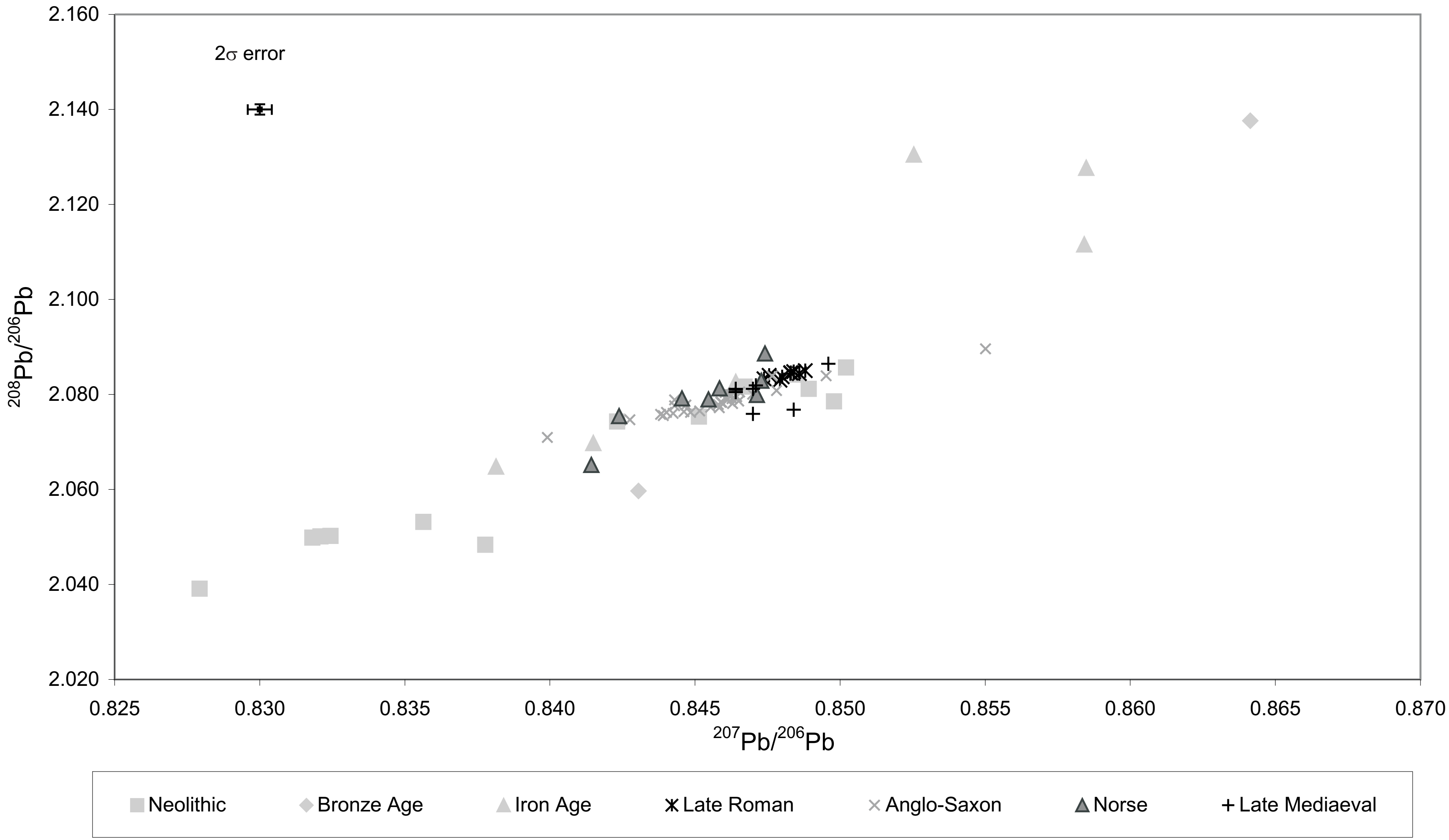

Figure 9.3 $^{207}Pb/^{206}Pb$ v $^{208}Pb/^{206}Pb$ plot of combined case study data showing cultural focussing of the enamel-Pb isotope ratios through time. Note how Late Roman and Late Mediaeval individuals cluster tightly at the centre of the plot whilst Anglo-Saxon and Norse samples are slightly more diffuse. Only prehistoric individuals are widely scattered. The two Late Mediaeval points that are below and out of error of the main cluster are from Blackfriars 89 who had an extremely low core enamel-Pb concentration (0.09ppm) and a less radiogenic Sr isotope ratio.