CHAPTER SIX

PILOT STUDIES: RESULTS AND METHODOLOGICAL DEVELOPMENT

6.1 Introduction

This chapter presents the results of several small, pilot studies, the conclusions drawn and the subsequent methodological developments arising from those conclusions. It falls into two halves: the first, an investigation of modern teeth that have never been buried (section 6.2) and secondly, archaeological studies from the Neolithic, Late-Roman and Late Mediaeval periods (section 6.3 to 6.5). The archaeological sites are predominantly in the southwest of England and located on sedimentary formations (Figure 1.4). They were all undertaken to address a specific archaeological question of mobility or migration appertaining to the site and period under investigation. There is, however, limited space to develop them as individual archaeological case studies. Consequently, emphasis has been placed on how the results informed future work and the archaeological background and period are dealt with only briefly. The two main case studies presented in Chapters Seven and Eight are developed extensively within their archaeological contexts. The archaeological pilot studies are presented in reverse chronological order from the most recent (late mediaeval) to the earliest (Neolithic). The results are then discussed together (section 6.6) to present the emerging picture of changes through time and on different lithologies. Fortuitously, this was broadly the order in which they were undertaken.

6.1.1 The aims of the pilot studies

1. To investigate the physiological variation in Pb and Sr isotope ratios and concentrations between enamel from the same tooth, enamel and dentine from the same tooth, teeth from the same individual and teeth of siblings.

2. To investigate whether Pb and Sr isotope ratios can reveal mobility in the dental tissues of modern migrants using the sampling technique developed in this project, as demonstrated for Pb by Gulson *et al.* (1997a).
3. To investigate the relationship between Sr and Pb in the locality and period the individual inhabited and whether changes over time were apparent.
4. To use the modern study to inform interpretations of the archaeological data and the investigation of diagenetic change of enamel and dentine.
5. To identify individuals from archaeological case studies who are unlikely to have obtained their Sr and Pb from a local origin.

6.2 Modern teeth

A small number of modern deciduous and permanent teeth were analysed to investigate the first four aims. Samples are listed in Table A8, Appendix II. Unless otherwise stated, enamel samples are core enamel with both the surface and the EDJ removed and dentine is primary crown dentine cleaned of all circumpulpal tissue. Results are presented of teeth from four individuals, two indigenous and two immigrants:

1. **CM (an exfoliated dc_1L) and AM (extracted dm^2R and dm_2L):** siblings born within three years to the same parents and raised in the same household on the Carboniferous Millstone Grit of Halifax, West Yorkshire (British Geological Survey 1979b).

2. **MN (an exfoliated di_2L):** the mother of MN was born in the same locality as CM and AM but emigrated to Johannesburg, South Africa in 1982. After 9 years she moved to Durban and 18 months later returned home to Halifax, West Yorkshire when MN was 5 months old. At 5 months, the deciduous second incisor crowns are usually fully mineralised although the root apex does not normally close until ~18months (Table 3.2). MN, CM and AM have attended the same school since the age of four. The tooth was analysed to test the hypothesis that teeth of inhabitants of ancient geological regions (Johannesburg: Precambrian, Durban: Precambrian/Early

Palaeozoic) should be isotopically distinct from teeth of individuals living in geologically younger areas (West Yorkshire: Late Palaeozoic).

3. **BAB (extracted M^3R and M_3R):** born and raised in New Hampshire, North America (Precambrian/Early Palaeozoic) and moved to England as an adult several years after the third molar crowns would have mineralised. She lived in Nottingham, England (Carboniferous/Triassic) for ~20 years (British Geological Survey 1979b).

Of particular interest with these samples was whether contemporary atmospheric pollution and the worldwide export and import of goods and raw materials would result in widespread homogenisation of Pb and Sr isotope signatures and sever the link between an individual and their place of origin. A shift from radiogenic skeletal Sr isotope ratios characteristic of place of origin to unradiogenic skeletal Sr isotope ratios in modern Norwegian populations has been documented by Åberg *et al.* (1998). Although the geology of Norway is ancient and this is reflected in the skeletal isotope ratios of archaeological inhabitants, modern Norwegians have a much less radiogenic Sr isotope ratio than would be predicted (i.e. <0.709). Norwegians have a high-dairy diet and Åberg *et al.* attribute this low Sr isotope ratio to country-wide distribution of mass-produced cattle feed.

Core enamel and crown dentine samples were taken from every tooth along with a sample of surface enamel from BAB1 following the procedure described in section 5.3.1.2. The trabecular, alveolar bone adhering to the roots of BAB1 was also sampled. Additionally, enamel taken from AM1, AM2 and BAB2 was split into several samples to assess intra-enamel homogeneity (see sections 5.3.3.1 and 5.3.3.2).

6.2.1 Results from modern teeth

Data are presented in Table A3 (Appendix I) and graphically in Figures 6.3 - 6.7 at the end of the chapter. The multiple ratio and concentration determinations made on enamel samples from BAB2, AM1 and AM2 are also listed in Table A3 with the mean measurements in italics. The mean values were not plotted in the figures.

6.2.1.1 Sr results

The enamel samples from the two co-genetic teeth from both AM and BAB replicated extremely well and were within error for both Sr isotope ratio and Sr concentration (Figure 6.3). Despite a very similar upbringing the tissues of CM (no data for enamel-Sr) and AM exhibit a ratio difference of 0.0002, which is an order of magnitude greater than analytical error and observed intra-tooth variation (section 5.3.3.1). The enamel and dentine Sr isotope ratios for the two immigrants to England (BAB and MN) are significantly different from the ratios obtained for the two indigenous children. The Sr isotope ratios of the enamel and dentine of BAB are within analytical error but the bone ratio is different and identical to that of AM. There is a considerable difference between the enamel and dentine Sr isotope ratios for MN. The dentine is intermediate between the enamel ratio and those of CM/AM. All samples of enamel, dentine and bone have < 80ppm Sr. In the three individuals with data for both tissues, the Sr concentration in the dentine (and bone) is *less* than that of the enamel. There is a significant negative correlation between Sr concentration and Sr isotope ratio in enamel and dentine ($r_2 = -0.72$, $p = 0.05$).

6.2.1.2 Pb results

Pb isotopes are presented as ^{206}Pb, ^{207}Pb and ^{208}Pb ratios against the invariant ^{204}Pb in Figure 6.4. They are also presented in Figure 6.5 and 6.6 as ^{207}Pb/^{206}Pb ratios both to facilitate comparison with the environmental and clinical literature as this ratio is commonly used and often the only one published, and to remove the correlated errors associated with the minor ^{204}Pb isotope.

The two co-genetic teeth from both AM and BAB generally replicated well and within error for both Pb isotope ratios and concentration. AM1 has produced one apparently anomalous ratio (Figure 6.4). In Figures 6.5 and 6.6, however, this sample plots with the others from the same tooth indicating the separation visible in Figure 6.4 is perhaps associated with the ^{204}Pb error. Thus, within-tooth tissues from CM, AM and BAB have Pb isotope ratios within analytical error. The enamel and dentine for MN are just outside analytical error for ^{206}Pb/^{204}Pb ratio but close to the intra-enamel variation observed for this ratio in the teeth of AM (section 5.3.3.2). The results for BAB are significantly different from the three children, and BAB's location on Figure 6.4a below the growth curve suggests a source somewhat depleted in thorium relative to uranium, compared to CM, AM and MN. Conversely, the Pb source of CM, AM and MN appears enriched in both ^{208}Pb (i.e. thorium) and ^{207}Pb relative to ^{206}Pb. Although CM and AM have a similar ^{206}Pb/^{204}Pb ratio, they have different ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb ratios. Enamel Pb concentrations for all the children are < 2ppm and for the two indigenous children < 1ppm. Figure 6.5 shows that dentine tends to have *more* Pb than enamel from the same tooth. There is no significant correlation between Pb concentration and ^{207}Pb/^{206}Pb ratio at the $p = 0.05$ level.

6.2.1.3 Sr and Pb combined

For both isotope ratios, the co-genetic teeth of AM and BAB replicated well and within analytical error. The anomalous Pb result from AM1 is not associated with an anomalous Sr isotope ratio, adding weight to the conclusion that it is an analytical error. The difference between Sr and Pb trends is striking. Enamel has *less* Pb than dentine from the same tooth, which was expected, but, unexpectedly, *more* Sr than is present in the dentine. A shift towards a less radiogenic Sr isotope ratio source appears to be present between the tooth and bone of BAB but no change has occurred in the Pb isotope ratios (Figure 6.6). The magnitude of the shift in Sr isotope ratios between the enamel and dentine of MN is not reflected in the Pb isotope ratios, where they are within intra-tooth variation. This may, however, be a function of the magnitude of difference present in the Pb isotope ratios compared to the Sr isotope ratio. CM and AM have the same ^{206}Pb/^{204}Pb ratio and a very similar ^{87}Sr/^{86}Sr ratio but different ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb ratios. There is a significant negative correlation between Pb and Sr concentrations ($r_2 = -0.81$, $p = 0.05$) in enamel and dentine (Figure 6.7) but none between ^{87}Sr/^{86}Sr ratios and ^{207}Pb/^{206}Pb ratios (Figure 6.6).

6.2.2 Discussion of results

6.2.2.1 Subjects CM and AM

The ^{207}Pb/^{206}Pb ratios of the two children of English origin (CM and AM) (Figure 6.5) fall within the range identified by Delves and Campbell (1993, 78) for modern British children (^{207}Pb/^{206}Pb = 0.869 - 0.905). Moreover, according to these authors, such ratios are diagnostic of exposure to mainly low Pb sources but with a greater contribution from geologically younger, British-Pb water pipes (^{207}Pb/^{206}Pb ~ 0.851) than from geologically older U.K. petrol-Pb (^{207}Pb/^{206}Pb ~ 0.939). The majority of U.K. petrol-Pb is imported from the Precambrian, Broken Hill Pb-Zn-Ag mines in Australia (^{207}Pb/^{206}Pb = 0.961) (Gulson *et al.* 1994b, 895) rather than being of domestic origin. As CM and AM live in a semi-rural, soft water area where Victorian Pb pipes (i.e. Pb of British origin) are still in use, this is entirely plausible. However, the Pb isotope ratios of CM and AM do not fall within the range of English ore Pb (Table 2.1) and their enamel-Pb concentrations (< 1ppm) are indicative of low *in vivo* Pb exposure (Gulson 1996, 310; Gulson & Wilson 1994, 281). Such an outcome may have arisen through all their drinking (but not cooking) water being supplied via a tabletop water filter since birth as soft water is known to increase Pb burdens (section 2.3.4). However, in the absence of comparative data from children raised on unfiltered water, this cannot be currently verified.

The amount of variation observed in Pb and Sr isotope ratios between the two teeth of AM and between CM and AM, is clearly pertinent to studies of migration. Variation beyond that explicable by analytical error or intra-tissue heterogeneity, has arisen even between two co-habiting siblings. The differences observed between CM and AM are ~0.0002 (0.03%) in the ^{87}Sr/^{86}Sr ratio, and as much as 0.6 (1.6%) in the ^{208}Pb/^{204}Pb ratio and 0.2 (1.3%) in the ^{207}Pb/^{204}Pb ratio. These variations are an order of magnitude greater than analytical error. The observed Sr isotope ratio difference between the two siblings is, nevertheless, not as great as the spread in Sr isotope ratios reported by Sealy *et al.* (1995, 297) of 0.00032 from a single archaeological bone (see section 5.3.3.1 for discussion). As would be predicted, the difference between siblings is greater than that between enamel samples from the same tooth or between co-genetic teeth from the same individual, the reproducibility of which was better than the standard for Sr and most Pb isotope ratios. The plot of ^{207}Pb/^{206}Pb ratios against ^{87}Sr/^{86}Sr ratios in Figure 6.6 highlights their “relatedness” to each other and their difference from MN and BAB more clearly than the other Pb figures. **Nevertheless, sound conclusions about migration must be based on differences greater than these; neither of these two children are immigrants to the area and their observed variation is likely to represent only the minimum variation within their community.**

The tooth of CM was a deciduous canine, the crown of which is ~30% complete at birth whereas that of the deciduous m2 is present as cusp tips only. Both complete their crown mineralisation at ~9months and ~10months respectively (Table 3.2). Accordingly, the skeletal Pb stores of the mother may have contributed more to the *in utero* Pb signature of CM. Moreover, differences in the mother's diet during each pregnancy (e.g. amount of Ca) could increase or decrease re-mobilisation of Pb from the skeleton (section 2.4). Pb is transferred to the baby both in the womb and during lactation far easier than Sr (section 2.4) and this may account for the greater difference in the observed Pb isotope ratios than Sr isotope ratios between the two siblings.

6.2.2.2 Subject MN

Subject MN has the most similar Pb isotope ratios to those of CM and AM. However, the difference cannot be explained by either analytical error or intra-tooth variation making it possible to propose a shift in the dentine ratio of MN towards that of CM. Moreover, MN is clearly distinguishable from CM and AM on the Sr isotope ratio plot (Figure 6.3). The South African Sr source is more radiogenic than that of CM and AM. This would be expected from the predominantly Precambrian region and is reflected in both the Sr isotope ratio but not so clearly in the Pb isotope ratios. However, MN lived in an urban environment and the Pb may derive from anthropogenic pollutant sources (e.g. petrol, industry) rather than any specific link to the geology.

The Sr and Pb isotope ratios of the crown dentine of BAB's permanent tooth does not differ from the enamel. The observed differences in the dentine and enamel of MN are likely to result from the complete resorption of the root of MN leaving only crown dentine tissue. It is argued that deciduous teeth appear to actively accumulate Pb in the shrinking root during the process of root dentine resorption and that the deciduous enamel is slightly more permeable than permanent enamel (section 3.5 and Whittaker & Stack 1984, 40/1). Based on the data presented here, this process appears to enable both Sr and Pb from the current diet to enter the resorbing dentine, thus rendering resorbing deciduous dentine considerably more dynamic than the crown dentine of a permanent tooth. However, the enamel of MN has not been affected to anything approaching the same degree and still retains an *in utero* signature.

Transfer of old skeletal Pb from mother to child may also explain why the Pb isotope ratios of MN are more similar to CM than her Sr isotope ratio. If Sr was being obtained from the contemporary African diet of both the mother and the baby, rather than from the mother's skeletal stores (which after 10 years may still contain considerable amounts of English Pb and, possibly, Sr) it would be expected to be more radiogenic. Conversely, if Pb was being re-mobilised from the mother's skeleton and transferred to the baby *in utero*, this would be reflected in resulting Pb isotope ratios that were a mix of South African Pb and English Pb. An alternative explanation is that Sr is much more mobile, turnover is far quicker and, thus, residence time in bone is shorter than Pb. As a result, the skeletal signature of MN's mother at the time of conception may have already been heavily influenced by 10 years of exposure to South African Sr but still remained dominated by anthropogenic English Pb.

6.2.2.3 Subject BAB

Subject BAB has very different Pb isotope ratios to the three children. The Precambrian geology of New Hampshire is of similar date to that inhabited by MN but the Pb isotope ratios of BAB appears to be from a more radiogenic source. Such Pb isotope ratios are entirely consistent with average North American anthropogenic Pb (^{207}Pb/^{206}Pb = ~0.847) which derives from a mix of geologically old (^{207}Pb/^{206}Pb = ~0.943) and the anomalous, radiogenic Mississippi Valley type ores (^{207}Pb/^{206}Pb = ~0.763) (Delves & Campbell 1993, 78; Derry 1980, 96; Hurst *et al.* 1996, 307; Veron *et al.* 1992, 342; Yaffe *et al.* 1983, 241). Co-incidentally, BAB's ratios are within the range of native English ore Pb (Table 2.1).

The Pb and Sr isotope ratios of the crown dentine of BAB remain indistinguishable from the co-genetic enamel ratios. This result supports the hypothesis formed in section 3.4 that these two tissues mineralise at the same time and should have the same isotope ratios. Thus, despite the long-term change of residence, crown dentine has remained indicative of origin. Moreover, the surface enamel sample is identical to the core enamel samples in all but Pb concentration. It is often claimed that the surface of tooth enamel absorbs Pb and Sr post-eruption (section 3.5.1 and 3.5.2). LA-ICP-MS profiling of tooth sections indicates that no surface enrichment is present for Sr but that a variable and, frequently very large Pb-peak is present on the surface of both modern and archaeological teeth (Budd *et al.* 1998; Lee *et al.* 1999; Montgomery *et al.* 1999). The results of BAB support these observations as there is no difference in Sr concentration between surface and core enamel samples but considerably more Pb in the surface enamel than in the core enamel (Figure 6.5). However, core and surface enamel have the same isotope ratios, demonstrating that no Pb or Sr has been absorbed by the tooth surface at least since the move to England. As third molars are the last teeth to mineralise and erupt (section 3.3.2), it suggests that surface enrichment of Pb occurs prior to, or very shortly after, eruption and may result from the maturation process itself.

BAB's bone Sr isotope ratio has, however, shifted noticeably towards CM/AM (Figures 6.3 and 6.6). Such a change in trabecular bone only was also observed by Cox and Sealy (1997) (section 4.5). Trabecular, alveolar bone surrounding the tooth root is considerably more dynamic than cortical bone or dentine and substantial turnover would be predicted during 20 years residency (see section 2.4 for discussion). It was, therefore, expected that it would evince the move from North America to England. Curiously, however, no concomitant shift has occurred in the bone-Pb isotope ratios (Figure 6.6), although current British anthropogenic Pb isotope ratios are very different to those in North America. As concluded in the discussion of subject MN, this suggests that the accumulation and residence time of Pb and Sr in bone is very different and the turnover of Sr is much quicker than that of Pb. Consequently, Sr appears much more sensitive to changes of origin than Pb which can be buffered by old Pb stored in the skeleton (Gulson *et al.* 1995, 709).

BAB's bone sample also has significantly lower Pb concentration (1.5ppm) than either the enamel or dentine. This is below the range of typical adult Pb concentrations (~3-60ppm), and neither the low concentration nor the lack of isotope change supports life-long skeletal accumulation; in fact it indicates the exact opposite: loss of Pb with no addition of new Pb. It could be argued that the reduction of Pb in the bone is because of decreased Pb exposure in later life, e.g. the removal of Pb from petrol but there is no concomitant difference in the Pb isotope ratio to support this. The results would be consistent with the conclusions of Gross et al. (1975, 650) that Pb accumulation in bone (and, it is proposed, crown dentine) only occurs in high-exposure individuals (i.e. >15ppm) who continually ingest more than they excrete (see section 2.3.4). Moreover, these results support the conclusions of Jaworowski (1990) that the greater Pb concentration observed in teeth of older subjects, when compared to younger subjects, is due to a recent reduction in anthropogenic Pb rather than accumulation with age. There is no change in Sr concentration between the dentine and the bone of BAB, suggesting equilibrium between loss of old skeletal Sr and incorporation of new Sr. This supports the conclusion that bone and dentine have very similar Sr concentrations rather than that bone Sr increases with age (section 2.3.3) but it also suggests that Sr is considerably more mobile in skeletal tissue than Pb.

6.2.2.4 Combined observations

The obvious discord between Sr and Pb in these samples is striking. It serves to confirm the view put forward in earlier chapters that, apart from divalency of the ion and accumulation in the skeleton, there is no reason and no evidence to suggest they are deposited by the same mechanism or subsequently behave in a similar manner. Sr concentration is reputedly similar across all skeletal tissues but slightly higher in bone and dentine than in enamel (section 2.3.3). Whilst no huge differences were observed between tissues, it appears that, unexpectedly, there is *more* Sr in enamel than in either bone or the co-genetic crown dentine. For Pb, however, the opposite situation exists: where crown dentine concentration is not the same as the enamel, it is greater, as was expected (section 3.5.2). Bone concentration, however, is still less than enamel, which argues against lifelong Pb accumulation. Although based on a few tooth and only one bone sample, these observations are consistent with the findings of Gulson et al. (Gulson & Gillings 1997; Gulson *et al.* 1997a) in their study of Pb in modern human teeth. Moreover, the significant negative correlation between Pb and Sr concentrations suggests that one is excluded in conditions favourable to the incorporation of the other, rather than that they both passively substitute for Ca. High-Sr sources may be low in Pb (e.g. seawater) and vice versa, or their bioavailability or incorporation into skeletal tissues is antagonistic or driven by entirely different factors.

Enamel-Sr concentrations are at the low end (i.e. 50-300ppm, section 2.3.3) of typical modern human concentrations and considered indicative of a high dairy and meat diet rather than one based on plants. None of the four individuals analysed nor the mothers of the three children were vegetarians. The significant correlation observed between Sr isotope ratio and concentration (i.e. the closer to the rain/seawater ratio the greater the Sr concentration) is interesting. CM and AM do not live near the coast and, although they do inhabit a maritime island and live in a rather wet region of the Pennines, rainwater is naturally low in Sr and the underlying bedrock consists of sandstone and not carbonate rocks (section 2.2.1). Carbonate, plagioclase and calcite rocks are highly soluble and a relatively rich source of Sr and may thus contribute large amounts of unradiogenic Sr (e.g. 0.709) to groundwaters, whilst radiogenic Rb-rich rocks such as micas and feldspars are less soluble and Sr-poor. The observed correlation may also arise, as documented by Åberg et al. (1998) from the widespread distribution of foodstuffs with unradiogenic Sr. Neither is the Sr isotope ratio of BAB nor MN quite as radiogenic as would be anticipated from Precambrian geology. Whatever the reason, it appears that there is some buffering of the skeletal Sr isotope ratios toward the rain/seawater signature although it is not clear if this, or a soluble rock phase, is the source.

The lack of correlation found between Sr and Pb isotope ratios in these modern subjects is of note but probably arises because the Pb isotope ratios, and possibly Sr isotope ratios, are not derived directly from the bedrock at the place of origin. The lack of correlation between Pb concentration and Pb isotope ratios would be expected given that there is no reason *per se* to expect Pb concentration to increase or decrease with increasing Pb isotope ratio. Anthropogenic high-Pb sources are taken from a variety of ores ranging from the geologically ancient Pb ores of Broken Hill, Australia and the Sullivan Mine, Canada ($^{206}Pb/^{204}Pb = 16.1$) to the Mississippi Valley type ores of North America which have $^{206}Pb/^{204}Pb$ ratios >20 and anomalous "future" dates that plot beyond the end of the growth curve (Hurst *et al.* 1996, 306).

6.3 Archaeological pilot study I: Site 1 Late Mediaeval burials from Blackfriars, Gloucester

6.3.1 Introduction

The excavation at Blackfriars, Gloucester (Figure 1.4) is part of the burial ground of a Dominican Friary, founded 1239 AD and dissolved in 1538 AD. The site, in the city centre, was excavated in 1991 by Gloucester Archaeological Unit. It was carried out as part of the Blackfriars Assessment Project in a previously uninvestigated part of the city. Approximately 2000 burials have been identified by ground probing radar of which 140 closely packed burials have been recovered from a 20m trench (Figure 6.1). Many females and juveniles were present and a high degree of skeletal pathology was observed, leading to the suggestion that the site was operating as a hospital or hospice (P. Greatorex, pers. comm.). One skeleton, a young adult female (77), displays lesions consistent with tertiary stage venereal syphilis (Roberts 1995). Evidence of a wall demolished in the mid 15th century AD suggests the graveyard was extended westwards beyond the original boundary wall at this time. Burial 77 lies to the east of the wall within the earlier part of the graveyard and is stratigraphically dated to the early-mid 15th century (Figure 6.1). Radiocarbon dating has not helped to confirm the archaeological dating (C. Roberts pers. comm.). Nonetheless, Blackfriars 77 is a rare, but not by any means unique, European case that contributes to the Old World/New World debate on the origin of the treponemal diseases (Pàlfi *et al.* 1992; Roberts 1995; Roberts & Manchester 1995, 158; Stirland 1991).

Armelagos and Baker, proponents of a New World origin for the disease, dismiss the few isolated cases of Old World pre-Columbian (i.e. pre-1493) syphilis because their distribution does not follow that expected of a newly introduced, highly contagious, disease, e.g. resembling the epidemic in Europe and Japan at the beginning of the 16th century (Armelagos & Baker 1997; Baker & Armelagos 1988). Admittedly, when compared to the abundance of New World cases, there are a far fewer the Pre-Columbian Old World but new cases are still coming to light (C. Roberts pers. comm.). It is possible, however, that the sufferers died too quickly for skeletal involvement to be apparent. However, a misdiagnosis of leprosy either at the time, or by subsequent researchers, could explain the apparent rarity of venereal syphilis prior to the 16th century, or the decline of leprosy may have allowed syphilis to proliferate (Roberts 1995, 106/7). However, if Armelagos and Baker's hypothesis is right and, equally, the diagnoses and dates of the growing number of Old World cases are correct, were these individuals, therefore, isolated immigrants bearing the disease into a treponemal-free area?

Blackfriars 77 was a well-preserved young adult female displaying bone changes characteristic of treponemal disease. The anterior view of the cranium shows destructive gummatous and healed lesions (caries sicca) in the right frontal bone pathognomonic of treponematosis (Hackett 1981; Hackett 1976). There is also extensive destruction and remodelling of the nasal area, palate and alveolar process of the maxilla with subsequent loss of teeth. Healed stellate lesions are also present on the occipital and parietal bones. Extensive osteo-proliferative lesions were observed postcranially on the ribs, clavicles, scapulae, sternum, humeri, right forearm, right ilium, both femora, tibiae and fibulae.

A diagnosis of venereal syphilis is generally accepted on the basis that there is no evidence for congenital syphilis and skull involvement is more likely to occur in venereal syphilis rather than yaws or endemic syphilis. It has been suggested that the long-standing nature of the disease in such a young adult would make venereal and congenital syphilis unlikely; after an active phase during infancy and early childhood, the spirochete lies dormant for some years and may then be reactivated during adolescence (Wiggins *et al.* 1993, 80). This is, however, disputed (C. Roberts per. comm.). Osteological evidence supports an African ethnic origin for Blackfriars 77 (C. Knüsel pers. comm.). A prehistoric, Central African origin is generally accepted for treponemal disease (Roberts & Manchester 1995, 155). Although cranial involvement is rarer in yaws than in venereal syphilis and when present, is somewhat different to that displayed by Blackfriars 77 (C. Roberts pers. comm.), a diagnosis of yaws (restricted today to hot, humid equatorial climates) has nevertheless been proposed (D. Ortner pers. comm.) As several lines of enquiry pointed to an African origin, Pb and Sr isotope analysis was used to investigate the hypothesis that Burial 77 would show different isotopic signatures from indigenous, Blackfriars burials if she had originated from outside Britain.

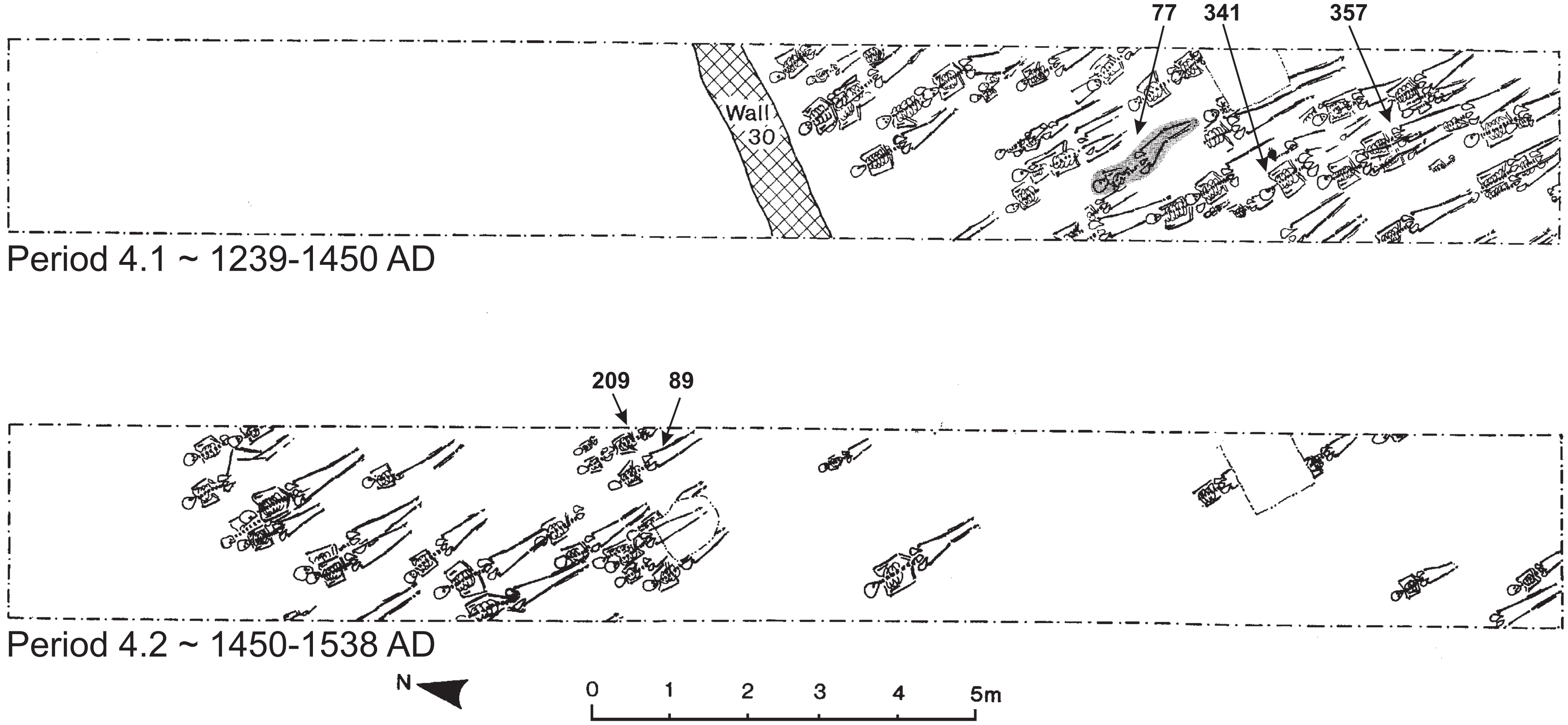


Figure 6.1 Plan of the excavated 20m. trench at Blackfriars, Gloucester. The five individuals analysed in this study are marked. The wall was demolished during the mid 15th century AD and the graveyard extended to the west. Adapted from P. Greatorex, Gloucester Archaeological Unit with additions.

6.3.2 Site geology

The outcropping geology of Gloucester and its immediate environs is relatively straightforward. The city sits squarely at the centre of a band of the Lower Lias, formed during the lower Jurassic, which cuts diagonally across the country from NE-SW (British Geological Survey 1979b). Lower Lias is composed largely of dark clays of sedimentary, marine origin that form broad, clay vales such as the Vale of Gloucester and, to the north, the Vale of Evesham (Ager 1961, 154). However, Gloucester sits very close to the boundary between the sedimentary sandstones, mudstones and limestones dating from the Carboniferous through to the upper Jurassic to the east, and to the west the much older Palaeozoic lithology of Wales. To the south, and separated by the Oxford Clay, are the marine formations of the Greater and Inferior Oolite of the Cotswolds and the Cretaceous chalk of the South Downs (British Geological Survey 1979b).

6.3.3 Samples

The skeletal remains from Blackfriars were, in general, well preserved macromorphologically with little erosion to the bone surface (Wiggins *et al.* 1993). A sampling strategy to provide a selection of British juvenile controls was devised by the author and samples chosen with reference to the skeletal report (Wiggins *et al.* 1993). Unfortunately, there were few juveniles over ~7 years of age. Teeth samples (Table A6) reflected the good preservation of the bone, being in excellent condition (enamel preservation score = 2, dentine = 3) with only slight enamel attrition. Three individuals (77, 357 and 341) were buried close together in the earlier part of the graveyard. The remaining two (89 and 209) were buried in the later, western end of the cemetery (Figure 6.1). Burials 89 and 341 were adolescents and the other three were young or young-middle aged adults. All teeth samples were first premolars (crown formation ~2-7 years, Table 3.3). Three tissue samples: surface enamel, core enamel and crown dentine were analysed (section 5.3.1.2). A site soil sample (Table A9) was obtained from context 371, a large post-Roman pit immediately below the burial layer. It therefore pre-dates the founding of the Friary in 1239. As excavation of the burial trench had been completed several years previously and the car park re-surfaced, this was all that was available from the archives. It had never been sieved, nor handled since the excavation (P. Greatorex pers. comm.).

6.3.4 Results for Blackfriars, Gloucester

6.3.4.1 Sr results

Figures 6.8 and 6.11 show that the five individuals from Blackfriars (black squares) exhibit a wide range of enamel Sr isotope ratios which, without exception, are outside analytical and intra-enamel error of each other. Enamel-Sr concentrations are ≤70ppm, within error of each other and in-line with modern concentrations. As predicted from the modern study, the two co-genetic teeth from burial 341 replicated extremely well for both ratios and concentration. Burial 77, the putative immigrant, has enamel Sr isotope ratio and concentration within the observed range at the site. However, contrary to the observations made in the modern study where dentine contained less Sr than enamel (section 6.2.4), dentine samples from these archaeological teeth (unfilled squares) have an order of magnitude more Sr (185 – 387ppm) and very different Sr isotope ratios to the associated enamel. Moreover, the dentine samples form a cluster of isotope ratios that bear a marked resemblance to the leaches obtained from the burial soil.

Figures 6.12 and 6.15 show enamel and dentine pairs and the burial soil leaches. Core enamel samples (black squares) are hugely variable but dentine samples (unfilled diamonds) are, in all cases, intermediate between the enamel and the soil leaches and very similar to the soil leaches. This is the case whether the enamel ratios are more radiogenic or less radiogenic than the soil leaches. Enamel samples most different from the soil leaches (341 and 77) have dentine that is most different from the soil leaches. As was found with the modern samples, surface enamel is very similar to, or indistinguishable from, core enamel in both Sr isotope ratios and Sr concentration.

6.3.4.2 Pb results

Pb isotope ratios are plotted as $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios in Figure 6.16. For ease of presentation and because dentine and surface enamel results were virtually indistinguishable from the core enamel (Figure 6.20), only core enamel samples are plotted on these two graphs. With the exception of Blackfriars 89, all individuals from Blackfriars plot within the tight cluster of points that is located within the English Pb ore field. Enamel-Pb concentrations for the mediaeval Blackfriars burials, again with the exception of 89, are considerably higher (~7-10ppm) than those obtained for modern individuals in this study (Figure 6.24) and are indicative of high *in vivo* Pb exposure (i.e. >2ppm) (Gulson 1996, 310; Gulson & Wilson 1994, 281). Burial 89 has an extremely low Pb concentration (0.09ppm) which appears to derive from a somewhat different, non-ore Pb source (Figure 6.16). Burial 77 falls within the range of Pb variation observed at the site.

Figure 6.20 shows that, with one exception, surface enamel and dentine samples are indistinguishable from the associated core enamel sample. Accordingly, only burial 209 provides sure evidence for a change in the dentine-Pb ratio to one more like that of the burial soil. As was found with the modern sample BAB1, there is more Pb in the surface enamel than in the core enamel samples but, as this is not associated with a shift towards more soil like ratios, it appears this is *in vivo* rather than post-mortem accumulation. In accord with the modern samples, there is more Pb in the dentine than in the core or surface enamel (0.63 – 74ppm). Considering these dentine samples were primary crown dentine with all circumpulpal dentine removed, this is both a very large range and, at the high end, about three time greater than Gulson *et al.* (Gulson & Gillings 1997; Gulson *et al.* 1997a) found in root dentine samples.

6.3.4.3 Pb and Sr combined

There is considerable variation in the Sr isotope ratios but not in the Pb isotope ratios in the Blackfriars samples (Figure 6.25). Burial 77 is within the variation observed at the site for all parameters. Burial 89 appears to be different on all counts: it has by far the lowest enamel-Pb concentration (Figure 6.24) but the greatest enamel-Sr concentration (Figure 6.8) and it has different Pb isotope ratios (Figure 6.16) and the only Sr isotope ratio less radiogenic than the burial soils (Figure 6.8). The significant negative correlation observed in the modern samples between Pb and Sr concentrations in enamel and dentine is not found with the Blackfriars samples; in fact an insignificant positive correlation is obtained. As only four of each of the three tissue types have concentration data for both elements, numbers are too small to assess each tissue individually. Nevertheless, it is clear that the positive correlation arises because of the dentine data and not the enamel data which still gives a negative correlation, although it falls just short of being significant at the $p = 0.05$ level ($r_2 = -0.64$, critical value = 0.74). This finding suggests post-mortem alteration of the dentine but not the enamel.

6.3.5 Archaeological outcomes of the Blackfriars case study

There is no evidence to suggest Blackfriars 77 did not spend her childhood in Britain or immigrated to England from Africa. All measured parameters fall within the variation exhibited at the site. Her Sr isotope ratio is insufficiently radiogenic to rule out a British origin as more radiogenic ratios were obtained from burial 341. The Sr isotope ratio is however, very different from the local soil, and given the pull effect exerted by rainwater towards Sr isotope ratio of ~0.7092, it is possible to suggest an origin for 77 and 341 on the older lithology of Wales, Scotland or the southwest and northwest of England. However, this in itself cannot *disprove* an African origin as she is also within the wide range of ratios obtained for indigenous Africans (Cox & Sealy 1997; Sealy *et al.* 1995). Her enamel-Pb clusters tightly within the group of post-metallurgical individuals inhabiting the British ore-Pb field. Perhaps most tellingly however, is the Pb concentration of her enamel which, at ~7ppm, is indicative of high anthropogenic Pb exposure (Gulson 1996, 310; Gulson & Wilson 1994, 281). This is certainly not in line with the very low Pb exposure levels deduced from either pre-metallurgical individuals (Budd *et al.* 2000b; Ericson *et al.* 1979; Montgomery *et al.* 2000) or slaves of rural African origin (Aufderheide *et al.* 1981; Corruccini *et al.* 1987, 289). Oxygen isotope analysis of the samples from Blackfriars also supports the conclusion that Blackfriars 77 is within the range of variation observed at the site and within the range of variation expected for western Britain during this period (C. Chenery pers. comm.).

Of particular interest are the unexpected differences displayed by Blackfriars 89. Although there is nothing archaeologically to distinguish this young adolescent, it appears that his/her Pb exposure was much less and from a different (i.e. non-ore Pb) source. Moreover, the Sr isotope ratio is unradiogenic and more indicative of an origin on younger, Rb-poor lithology, perhaps of marine origin, such as chalk or limestone. Blackfriars 89 also has the greatest Sr concentration, which may also support such an origin as may the very low Pb concentration which suggests a rural hard water area rather than a soft water area which can result in high-Pb burdens even in remote areas.

That such differences in Pb exposure are still present in the late Mediaeval period is of great interest as it offers the possibility of distinguishing between rural and urban origins and status, as indicated by access to metal artefacts and products.

Consequently, an urban childhood in western Britain is postulated for burial 77. It has been argued that she was a minimum of 15 years old at the time she contracted the disease (Roberts 1995, 106). Archaeologically her burial is dated prior to the mid-15th century AD. This implies, therefore, that sometime after the mineralisation of the lower first premolar (~8 years of age), burial 77 contracted venereal syphilis which, barring her journeying abroad after this time, must have been present in Britain prior to 1493.

6.4 Archaeological pilot study II: Sites 4, 5 and 6, Late-Roman burials from Mangotsfield, Winchester and Spitalfields

6.4.1 Introduction

Seven individuals were analysed from three sites (Figure 1.4) of Late-Roman date:

1. Mangotsfield, Bristol: two individuals from a limestone sarcophagus.
2. Eagle Hotel, Winchester: three chalk burials and one individual from a Pb-lined coffin.
3. Spitalfields, London: a single individual from a Pb coffin.

6.4.1.1 Mangotsfield, Bristol

The two Mangotsfield individuals (SK1 and SK2) were excavated from a large, soil-filled, limestone sarcophagus, discovered by archaeologists from Avon Archaeological Unit investigating a former school playing field prior to the building of a housing estate. The sarcophagus, aligned NE/SW and dated ~200-400AD, was an isolated burial in the middle of a Romano-British field. The only artefacts found in the sarcophagus were hobnails (Richards 1999, 73). Archaeologists also uncovered remains of stone buildings and enclosures, postholes, pits, coal and lead slag (for more details see Richards 1999, 64-83). The magnificence and isolation of the sarcophagus led to the suggestion that the inhabitant was of considerable status. Pb and Sr isotope analysis was undertaken to look for evidence of a non-British (e.g. Italian) origin and to assess the Pb burden of the occupant. However, the male burial (SK1) turned out to be merely the final burial in the sarcophagus. As the soil fill was removed, a second skeleton of an elderly female (SK2) was discovered. Some years after interment, she had been disturbed from the knees up and her long bones rearranged over this subequent male occupant. Moreover, an additional clavicle and vertebra that belonged to neither of the two complete skeletons were also found, suggesting the presence of an even earlier occupant (Gerry Barber pers. comm.).

6.4.1.2 Eagle Hotel, Winchester

This 4th century Roman cemetery on the site of the Eagle Hotel, Winchester was excavated by Winchester Archaeological Unit. The details reproduced below and more information about the site can be found in Richards (1999, 84-107). Burials at the site were made into the chalk bedrock and many of them had been in wooden coffins. The graves were aligned E-W in what appears to be an early Christian cemetery but many burials had coins in their hand or mouth in accordance with traditional Roman burial practices. A single Pb-lined oak coffin was found at a depth of ~3m. Unlike the other burials at the site, it was aligned N-S and contained a coin of Constantine which was minted in 316/317 AD, i.e. the burial could not have been made prior to this date. Directly overlying the Pb-lined coffin was a child burial aligned E-W and containing a coin of Valentinian dating to between 364 - 375AD. Sr isotope analysis was undertaken to investigate the origins of the occupant of the Pb coffin, i.e. British or non-British, together with comparative data from three adult E-W, chalk burials (G318, G319 and G326) with associated coins minted in 370-380AD. Pb isotope analysis was carried out to investigate enamel diagenesis of the individual in the Pb coffin and also to provide comparative data for other late Roman individuals.

6.4.1.3 Spitalfields, London

A single individual was analysed from this extensive multi-period site excavated by the Museum of London Archaeology Service. The details reproduced below and more information about the site can be found on the Museum's web site (MOLAS 1999). The remains of the young adult female were sealed in a highly decorated Pb coffin contained within a limestone sarcophagus. The coffin was found in a Roman burial ground just south of the mediaeval cemetery of St. Mary's Spital. The burials of adults and children, many containing grave goods, appear to be laid out in rows and are mostly aligned E-W. The coffin was excavated at a depth of 6m and the macromorphologically well preserved skeleton was found to be lying in a layer of silt about 3cm deep. Fine glass and jet grave goods were recovered with the coffin, and remains of silk woven with gold thread and bay leaves were found inside. A date in the early 4th century has been established for the burial.

6.4.2 Site Geology

6.4.2.1 Mangotsfield, Bristol

A mixture of convoluted sedimentary formations of Carboniferous, Triassic and Jurassic age outcrop in the Mangotsfield region (British Geological Survey 1979b), their complexity resulting from the east-west folding of the Hercynian Orogeny (Ager 1961, 116). Mangotsfield itself, sits on a small outcropping of the Upper Westphalian Carboniferous Coal Measure, Triassic mudstones, including the Keuper Marl – a thick, red, fine-grained, blown dust and the same dark clay Lower Lias deposits that form the Vale of Gloucester (Figure 1.4) (Ager 1961, 154). The productive Somerset coalfield extends under the city of Bristol but coal is actually a small part of the formation, the greater part being sandstone, millstone grit and shale (Ager 1961, 113). Triassic rocks are a thick layer of unfossiliferous, sediments that become progressively finer grained and lie between the Coal measures and the marine deposits of the Jurassic (Ager 1961, 121). They outcrop over a larger area in Britain than do any other system and practically all of them are red in colour and of desert origin, being frequently termed the “New Red Sandstone”. To the north, west and south lies the Carboniferous limestone of the Mendips which, because of its solubility, is home to many cave systems such as Cheddar and Wookey Hole. It also hosts the main British Pb ore deposits, in this region and elsewhere in Derbyshire, Yorkshire and the Scottish lowlands. To the east lie the Middle Jurassic Great and Inferior Oolite that form the Cotswolds. The soils on the site itself lay over a mixture of clay and coal measures (Richards 1999, 66) but the soil sample from inside the coffin was noted by the author as being distinctly red in colour.

6.4.2.2 Eagle Hotel, Winchester

Winchester is located on the band of Cretaceous chalk that outcrops extensively over eastern and southern England (Figure 1.4) (British Geological Survey 1979b). Cretaceous chalk is an extremely pure, calcium carbonate deposit of marine origin and almost entirely without any trace of land-derived sediment (Ager 1961, 174), which renders it naturally very low in Pb. The few inclusions that do occur tend to be a sand of possible desert origin found at the base and bands of flints and glauconite (Ager 1961, 175). It was originally deposited over a much greater area than it is found today but has been subjected to extensive erosion so that none of the youngest chalk outcrops in Britain today. Elsewhere, most notably in Denmark, much younger beds of Chalk are preserved (Ager 1961, 178). The Sr isotope ratio of seawater varied from ~0.7072 at the beginning of the Cretaceous to ~0.7077 at its close (Veizer 1989, 157).

6.4.2.3 Spitalfields, London

As the skeleton was sealed inside a Pb coffin no soil samples from the surrounding burial site were taken.

6.4.3 Samples

6.4.3.1 Mangotsfield, Bristol

Two teeth from each skeleton (Table A6) were selected by Gerry Barber, Bristol University who also prepared the skeletal report on the individuals. The skeletons were well preserved macromorphologically but within-dentition tooth preservation and attrition was extremely variable. SK2a was particularly poorly preserved (enamel preservation = 5) whilst SK2b was excellent (enamel preservation = 2) and better than both the teeth from SK1 (enamel preservation = 3). A soil sample from within the cranium of SK1 was removed by the author during the excavation (Table A9).

6.4.3.2 Eagle Hotel, Winchester

The skeletal remains were assessed by the Winchester Archaeological Unit (Winchester Museums Service, Historic Resources Centre) who provided the data in Table A6. The chalk burials produced skeletal material that was well preserved macromorphologically. The adult male buried in the Pb coffin had a well preserved skull but post-cranial preservation was poor (Richards 1999, 101). Teeth samples were selected and removed by the author. The good cranial preservation was reflected in that of the teeth: all teeth produced enamel, and in some cases dentine, that was graded as excellently preserved (preservation score = 2). Two teeth from the Pb coffin burial (G339), a third molar and a premolar were analysed. A grey surface deposit, which was not calculus but possibly precipitated Pb salts, was present on both teeth. A first premolar from each of three chalk burials (G318, G319 and G326) was also analysed. Great care was taken over the cleaning of tools and the mechanical preparation of the enamel samples to ensure no cross contamination from the teeth of G339, which were prepared last in the batch. The teeth were sectioned following the procedure used in previous case studies (section 5.3.1). A sample of chalk bedrock from the site was provided by Paul McCulloch of Winchester Archaeological Unit (Table A9).

6.4.3.3 Spitalfields, London

Skeletal analysis was carried out by Bill White, MOLAS, who selected the premolar tooth requested by the author for analysis. As would perhaps be expected from such an extreme preservation environment, tooth preservation was very unusual. Although outwardly very well preserved with no evidence for the surface precipitation of lead salts which was found in G339, the premolar tooth enamel had an unusual "glassy" appearance and there was a very dark, almost black, band circling the root immediately below the cervical enamel. It is not known if this unusual discoloration in this sample originated *in vivo* or from conditions in the anomalous post-mortem environment. Curiously, a very similar phenomenon has been observed by the author on the teeth of adult seals, where such a band of black stain accumulates with age round the cervical enamel, and is attributed to eating cephalopod prey (i.e. ink) (Montgomery 1996, 25). In this case also, it proved to be a surface feature only and was easily removed with a dental burr. Attrition was slight but the enamel had many cracks and fissures. This tooth sample was the last to be prepared in the sample batch to prevent any cross contamination between samples.

Although the primary reason for analysis was to establish if the Sr isotope ratio provided evidence for a non-British origin, this tooth was prepared with great care with the aim of producing a sample of core enamel that may not have been completely swamped with coffin Pb. The enamel broke into many small pieces along the cracks and fissures during sampling and was very difficult to handle. However, for every piece that was used in the analysis, the outer and inner (i.e. EDJ) enamel surfaces and all edges were diligently and consistently abraded to a much greater depth than usual (i.e. >100µm). A sample from the Pb coffin from Spitalfields was kindly provided by Chris Thomas, MoLAS and analysed by Dr. R.A.R. McGill at the NERC Isotope Geosciences Laboratory, BGS, Keyworth.

6.4.4 Results for Late-Roman burials

6.4.4.1 Sr results

The male and female burials from the Mangotsfield sarcophagus (black diamonds) and the female in the Spitalfields Pb coffin, have the same Sr isotope ratios but widely varying concentrations (i.e. 37 – 189ppm) (Figures 6.9 and 6.11). SK1 and SK2 data points are for surface enamel, as concentrations were not obtained for any of the core enamel samples. SK1 has considerably more Sr than any of the modern or archaeological samples in the pilot studies but is still within the range of typical Sr concentrations reported in modern people (i.e. 50-300ppm, section 2.3.3). The four individuals from Winchester, including the Pb coffin burial, have more variable and less radiogenic Sr isotope ratios, which fall mostly between the ratios obtained for the chalk bedrock and rain/seawater. All their enamel-Sr concentrations are <100ppm.

Enamel samples from the three individuals where two teeth were analysed (SK1, SK2 and G339) have replicated well and within analytical error, although only those of SK1 (upper canines) were co-genetic teeth (Figures 6.13 and 6.15). The two teeth of SK2 were a lower first premolar and a lower canine which have broadly similar crown formation times. However, those from G339 (third molar and lower second premolar) do not (section 3.3.2 and Table 3.3), which may account for the small observed Sr isotope ratio difference between the two teeth. Both SK2 and G339 had an intra-dentition difference of 0.00016 in the Sr isotope ratio of enamel samples which is slightly less than that observed in the modern siblings (0.00022) but an order of magnitude more than that observed in the co-genetic teeth of AM, BAB, SK1 and 341.

As with the Blackfriars samples, dentine samples contain more Sr than their associated enamel sample and the ratios are less variable (Figure 6.9). The Winchester dentine samples are intermediate between the co-genetic enamel and the chalk leaches and enamel samples most different from the soil leaches have dentine most different from the soil leaches (Figure 6.13). The teeth from SK1 and SK2, Mangotsfield appear to be behaving differently, with the dentine of SK1a and b moving towards less radiogenic ratios whilst that of SK2a and b are moving to more radiogenic ratios. Despite the very different preservation of SK2a and SK2b, the two teeth have very similar Sr isotope ratios in all three tissues. There is clearly a considerable spread between the soil leaches from within the stone sarcophagus and it encompasses the total variation observed in the tooth samples. This may result from inputs from two sources of Sr: the soil and another less radiogenic source in the limestone sarcophagus.

6.4.4.2 Pb results

The majority of the Late Roman enamel samples cluster very tightly and within analytical error at the centre of the English Pb ore field (Figure 6.17). Curiously, the two samples that do not, are teeth from the two Pb coffin burials. Of the two teeth analysed from G339, it is the later forming third molar (G339a), rather than the very heavily contaminated premolar (G339b), that is different. The Spitalfields tooth also shows a difference, inexplicable solely by analytical error, from the Pb in the coffin (^{206}Pb/^{204}Pb = 18.40, ^{207}Pb/^{204}Pb = 15.62, ^{208}Pb/^{204}Pb = 38.36, R. McGill pers.comm.). Both these two enamel ratios are on the edge of the English ore Pb field. Nevertheless, both teeth have what would be considered an extremely high *in vivo* enamel-Pb concentration (30ppm and 42ppm) that is highly unlikely to originate purely from non-anthropogenic, country rock sources.

For the four Mangotsfield teeth, core enamel and surface enamel are indistinguishable from each other, and the associated dentine is within the intra-tooth variation observed in the modern samples (i.e. 0.35% on ^{207}Pb/^{206}Pb ratios). The same is true for the Winchester samples (Figure 6.21). The water and acetic soil leaches from the Winchester chalk are, however, hugely variable but the samples show much more similarity to the water leach than the acetic acid leach. No data from the two Pb coffin burials are plotted on Figure 6.24 as the enamel-Pb concentrations were excessive (i.e. > 10ppm) when compared to all other samples. Enamel-Pb concentrations for individuals not buried in Pb coffins range from 0.59 – 8.6ppm and the dentine samples from 2.2 – 45ppm. As was found in the modern study, dentine Pb is greater than enamel Pb from the same tooth. Unexpectedly, dentine from G339a and G339b has a Pb concentration within the range observed for non-Pb coffin burials (22ppm and 43ppm) and *less* than the associated enamel (42ppm and 1540ppm).

6.4.4.3 Sr and Pb combined

All samples from this period have very similar Pb isotope ratios but more variable Sr isotope ratios, although the majority of the Sr isotope ratio variation is present in chalk burials from Winchester (Figure 6.25). Too few samples in this study had concentration data for both Pb and Sr so the relationship between these two variables could not be investigated.

6.4.5 Outcomes of the Late-Roman studies

6.4.5.1 Mangotsfield, Bristol

SK1 and SK2 have Pb isotope ratios within the English Pb ore field. The soil sample Pb ratio is very similar to that of the Mendips ore field close by, but their enamel-Pb concentrations (i.e. ≥1ppm, see sections 7.4.2 and 9.5) suggest a considerable input from anthropogenic (i.e. ore-Pb) sources. Pb was being mined in the nearby Mendips by 49AD (Tylecote 1992, 71). Pb was used to make drinking vessels, cooking pans, Pb pipes etc., so it is possible such Pb entered the diet. Crops grown on local soils would be very likely to reflect the underlying mineralisation and contain Pb of similar isotope composition (Gulson 1986, 89). Their Sr isotope ratios are also similar to each other and to the soil leach and within the range of other results from this region. The data are consistent with both individuals having a local origin but do not, by themselves, rule out an origin elsewhere. However, oxygen isotope analysis indicates SK1 and SK2 fall within the range exhibited by other individuals from southwest Britain and that expected of the Bristol area today but would be inconsistent with Mediterranean Italy (C. Chenery pers. comm.).

6.4.5.2 Eagle Hotel, Winchester

The Sr isotope ratios obtained from all individuals are consistent with a British origin in a region such as Winchester where relatively unradiogenic sources of Sr occur widely, i.e. between chalk and rainwater. This, on its own, does not rule out an origin elsewhere but oxygen isotope analysis indicates an origin in a climate very similar to southern England and, therefore, inconsistent with Mediterranean Europe (C. Chenery per. comm.). For the three individuals buried in chalk, the Pb isotope ratio and concentrations are consistent with exposure to anthropogenic British ore Pb during childhood. Such British ore Pb isotope ratios were also obtained from the Pb-coffin burial (G339) but, clearly, it cannot be assumed that this represents *in vivo* exposure. Despite the aggressive sample preparation, one tooth had a Pb content, which, if it derived purely from *in vivo* sources, is indicative of extremely high exposure (42ppm) and the other contained so much Pb (1570ppm) to be physiologically implausible. As would be predicted from what is, perhaps, the ultimate opportunity for Pb diagenesis, the dentine and enamel Pb isotope ratios for the Pb coffin burial are within error, but this is the case with the chalk burials also. This implies that the Pb isotope ratios of the enamel and dentine of G339 are liable to have been the same prior to burial and that the Pb in the post-mortem burial environment is the same as that encountered ante-mortem, during childhood. Obviously, clear proof that tooth samples have not suffered post-mortem contamination is problematic in this case.

6.4.5.3 Spitalfields, London

The Sr isotope ratio and Sr concentration data obtained from SPR-1 are consistent with a British origin. The Sr isotope ratio is identical to that obtained from Late Roman Mangotsfield and within the range of those obtained from mediaeval Blackfriars. Notably, however, the Pb isotope ratios are not the same as those of the coffin (Figure 6.17). Not only does this result validate the sampling method devised in this project and repay the care and skill brought to bear during the sample preparation but it is also evidence that the integrity of the *in vivo* Pb isotope signature has not been completely swamped by coffin Pb. Nevertheless, at 30ppm the enamel-Pb is still suspiciously high, although perhaps not implausibly so given the oft-quoted ubiquitous Pb use during the Roman period (Gilfillan 1965; Nriagu 1983). The enamel-Pb isotope ratios are on the very edge of the British ore Pb field, which does not appear to be a plotting location for indigenous high-Pb exposure (Figure 9.2). It seems extremely unlikely that a Pb concentration of this magnitude was obtained solely through natural exposure to the sedimentary, country rocks found in southeastern England.

It is possible to consider two scenarios: either the tooth enamel is not contaminated at all which means SPR-1 had a very high childhood Pb exposure (quite possible given the period) to a source of Pb ore that was unlikely to be entirely of British origin. This does not rule out imported food and drink. Or, the enamel is *partly* contaminated by coffin-Pb, which would make her *in vivo* signature even more anomalous and *very* unusual indeed for Britain; in fact it would then be possible to exclude all known British ore Pb sources and suggest an origin outside the British Isles. This unknown and anomalous Pb source appears enriched in both ^{208}Pb (i.e. Th) and ^{207}Pb (i.e. ^{235}U) relative to common Pb ore.

Figure 6.26 shows, however, that similar ratios have previously been obtained from a Pb coffin (970) from the Roman-British cemetery at Poundbury and, moreover, an individual from this site (1405) was found to have a Pb signature even more enriched in ^{208}Pb and ^{207}Pb (Molleson *et al.* 1986, 251). The authors were unable to identify possible sources for the Pb found in this individual but did not believe that imported food and drink were the cause (Molleson *et al.* 1986, 252). Clearly, in the absence of data from contemporary inhabitants of Rome, it is impossible to know if such a combination of Sr and Pb would be compatible with an origin there, or elsewhere in the Roman Empire, but SPR-1 is evidently not the only individual in Roman England that had an unusual Pb isotope signature.

6.5 Archaeological pilot study III: Site 8 Neolithic burials from Monkton-up-Wimbourne

The Monkton-up-Wimbourne case study has been published (Budd *et al.* 2000b; Montgomery *et al.* 2000) and the reader is referred to these papers for further details and the analytical procedure.

6.5.1 Introduction

The site was discovered through aerial photography in a field of peas and subsequently excavated by Martin Green in 1997 (for more details see Green 2000, 77-84). It lies on farmland near the village of Monkton-up-Wimbourne, Dorset and is dug into the Cretaceous chalk downs of the Cranborne Chase (Figure 1.4), a region rich in archaeological sites and megalithic monuments. The site is ~70m^2 and consists of a central pit 1.2m deep and 12m in diameter that had almost vertical sides and a flat-bottomed base that was described as having a "polished" appearance through use (Figure 6.2). It was surrounded by an oval of 14 smaller, soil filled pits and a raised chalk rubble bank resembling a Neolithic henge monument (M. Green pers. comm.). In the centre of the large pit just beneath the soil, was a flint cairn, which covered the complete but somewhat poorly preserved flexed skeleton of an adult male (Figure 6.2a). Pottery fragments and flint arrowheads nearby suggested an early Bronze Age date (~3700 BP) for the burial and radiocarbon analysis indicated a slightly later date in the middle Bronze Age (Richards 1999, 20/36).

Excavation of the central pit then revealed further features including a 7m deep shaft filled with chalk rubble and soil that had been hacked out of the bedrock with antler picks and stone tools (Figure 6.2a). The pit contained antler fragments, cattle bones, auroch bones, a carved chalk block and Neolithic Peterborough ware pottery, suggesting a date in the Early Neolithic (Richards 1999, 23). Partly surrounding the shaft rim was the low rubble bank and as this was removed another pit was discovered. Chalk rubble and large chalk blocks were removed to reveal a further four skeletons in context F23 (Figure 6.2). The construction of the pit and articulated, undisturbed nature of the bones was interpreted as one burial event at the time of initial construction of the site, i.e. they were all buried at the same time (Richards 1999, 26).

The skeletal analysis was performed by Jackie McKinley at Wessex Archaeology (Table A9). Three were juveniles (A, B and D) aged ~5yrs, 8-9yrs and 9-10yrs respectively, one an adult female (C) and a Bronze Age adult male (E). All three juveniles exhibited cribra orbitalia, and the youngest had a tooth abscess and small tumours in the cranial vault, but none were likely to have caused or contributed to death (J. McKinley pers. comm.). Radiocarbon dating of the skeletons confirmed a calibrated date range in the Early Neolithic of 5500-5100BP for the four chalk burials and a date in the mid 3rd millennium BC for the Bronze Age male (M. Green pers. comm.). aDNA analysis was performed by Christine Flaherty (University of Columbia) to obtain sexing and familial information. Her results indicated that:

1. A and D were female
2. B was male
3. Only A was the offspring of C
4. B and D were possibly siblings

The four Neolithic burials pre-date the advent of metallurgy in the British Isles and their Pb burden should derive from natural, rather than anthropogenic, sources. It should, therefore, be closely tied to the geology in the region they sourced their food and drink. Additionally, the Cretaceous chalk is remarkably pure with few sedimentary inclusions, creating a burial environment very low in Pb (section 6.4.2.2). Consequently, these burials represented an opportunity to obtain prehistoric baseline enamel concentrations from natural Pb exposure. Furthermore, a settled farming community not importing large quantities of foodstuffs, as is often suggested to be the case in this period, would provide evidence for the natural range of isotopic variation that could be expected from a sedentary community. The identification of a mother/daughter pair also provided the opportunity to investigate the transfer of Pb and Sr from mother to offspring.

6.5.2 Site geology

Like Winchester, Monkton-up-Wimbourne is located on the same band of Cretaceous chalk that outcrops extensively over eastern and southern England (Figure 1.4). See section 6.4.2.2 above for discussion.

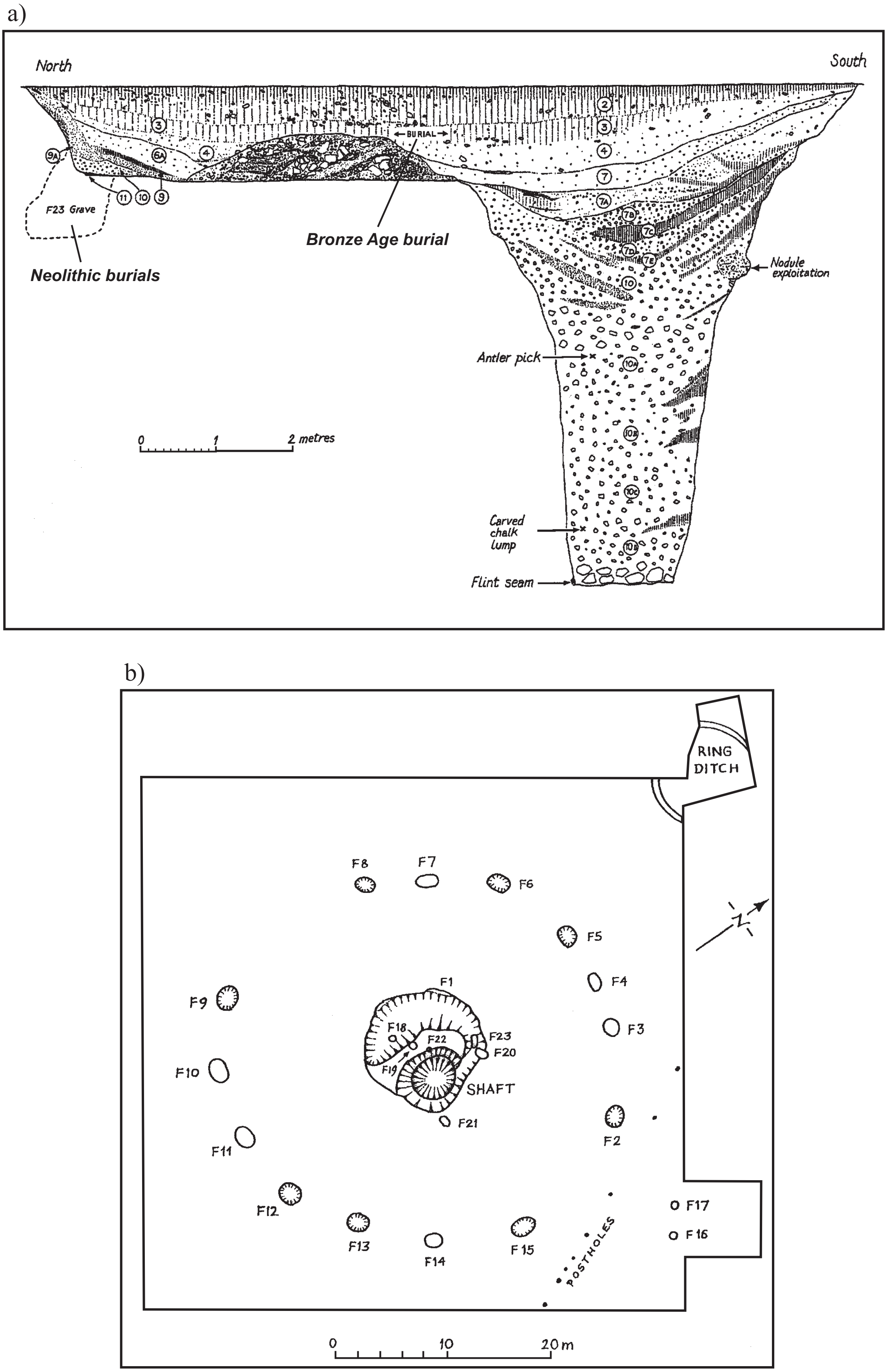


Figure 6.2 Section(a) and plan(b) of the Neolithic site at Monkton-up-Wimbourne
The four Neolithic burials were in context F23.
Adapted from: Green 2000 with additions.

6.5.3 Samples

All teeth samples were selected by the author. Macromorphological skeletal preservation of the Neolithic individuals (A, B, C and D) buried directly into the chalk bedrock, was observed by the author to be excellent and this is clearly reflected in the tooth preservation scores (enamel preservation = 2, dentine preservation = 2/3). The skeletal preservation of the Bronze Age soil burial (E) was not as good, despite being buried for 2000 years less. Many of his bones were broken and the cortex eroded but the tooth preservation was still comparatively good (enamel preservation = 3, dentine preservation = 3). The case study provided the rare opportunity to analyse deciduous and permanent teeth from three Neolithic juveniles of different ages. Although there was no certain evidence that the group had been moving together over the last ten years of their lives, they had clearly been buried together. The sampling strategy was devised by the author to provide Pb and Sr isotope information spanning the last ~10 years of the group's existence both *in utero* and during childhood. Two teeth (a deciduous and a permanent) were taken from each of the juveniles and one permanent tooth from each of the adults. Three soil samples were obtained from this site (Table A9): a sample from inside a chalk block, a sample of soil excavated from the bottom of the 20m shaft (Figure 6.2) and a sample of the surface soil from within the skull of the Bronze Age male (E).

6.5.4 Results for Monkton-up-Wimbourne

6.5.4.1 Sr results

The Monkton enamel samples from deciduous and permanent teeth (black triangles) have variable Sr isotope ratios, and Sr concentrations ≤ 103ppm (Figures 6.10 and 6.11). By contrast, the associated dentine samples have similar Sr isotope ratios to each other and that of the burial chalk and much greater Sr concentrations (162 – 282ppm). Burial C has the most radiogenic Sr isotope ratio of both the enamel and dentine samples. Figures 6.14 and 6.15 show that, as with the other pilot studies, dentine samples are, without exception, intermediate between the Sr isotope ratio of the associated enamel samples and that of the chalk in which they were buried, despite the excellent macromorphological preservation of the skeletal remains. Of the three juveniles, the deciduous tooth of A has a Sr isotope ratio most like that of the adult female C. There is considerable variation between the deciduous and permanent enamel of A and D but not of B.

6.5.4.2 Pb results

It is clear from the Pb plots (Figure 6.18 and 6.19) that the individuals from Monkton have different isotope ratios and distribution pattern to those from the later periods. The majority of points fall on an apparent mixing line between the Winchester and Monkton chalk leaches and plot below and to the right of the ore Pb growth curve (Figure 6.19a). Ocean rocks are known to plot in the same region as the Monkton and Winchester acetic leaches (Faure 1986, 328; Gulson 1986, 150; von Blanckenburg *et al.* 1996, 4960), consistent with the marine origins of the chalk. Only burial C plots away from this line and has a Pb isotope ratio that appears to be similar to the ore Pb ratio, although no Pb ore (or any metals) was being mined or used in this period. Contrary to more recent individuals, the enamel Pb isotope ratios are widely variable but dentine ratios still plot at a point intermediate between the enamel ratio and that of the burial chalk (Figure 6.22). Figure 6.24 shows that the enamel Pb isotope ratios from Monkton are both low in Pb and have extremely variable ^{207}Pb/^{206}Pb compared to the high-Pb and invariant samples from later periods. Dentine has a greater Pb concentration than the associated enamel. Of the juveniles, the deciduous enamel of A has Pb isotope ratios most like that of the adult female C.

6.5.4.3 Sr and Pb combined

Figure 6.25 illustrates that very variable ^{207}Pb/^{206}Pb ratios are present amongst the Monkton individuals who, compared to other sites, have relatively invariant Sr isotope ratios. The opposite situation is clearly the case for the post-metallurgical individuals who have invariant ^{207}Pb/^{206}Pb ratios but very variable Sr isotope ratios. The dentine-Sr appears to have equilibrated with the chalk burial soil to a greater degree than the dentine-Pb (Figure 6.14 and 6.22). Burial C has the most radiogenic Sr isotope ratio and Pb isotope ratios that are most like Pb ore but the very low-Pb concentration supports a natural country rock origin. C is on both counts less like the chalk burial rock than any of the other Monkton individuals analysed. The deciduous enamel (i.e. *in utero*) of A is most like that of C for both Pb and Sr isotope ratios.

6.5.5 Outcomes of the Monkton study

The isotope data show that there are differences between the four Neolithic individuals in both the Pb and Sr isotope ratios that exceeds analytical error, intra-tooth variation and the variation observed between modern siblings. There is also a considerable difference in both isotope systems between C and A, the mother/child pair. Although the deciduous tooth of A, which mineralised *in utero*, is the tooth most similar to C's childhood signature, A's permanent tooth (first molar) is not, and this suggests considerable difference in dietary Sr and Pb. Such a magnitude of variation is unlikely to have arisen from inter-individual variation in a community of sedentary, self-sufficient farmers. It suggests, therefore, that there was still considerable mobility during the Neolithic, either in search of food sources or following wild or domestic herds. Moreover, the evidence from the Sr isotope ratios suggests the movement was systematic and ended with the move back to the Cranborne Chase a few year prior to death (Montgomery *et al.* 2000, 379). Of course, this patterning may be an artefact produced by too coarse an investigation tool and better resolution would undoubtedly be obtained by using all the teeth in the dentition of all three juveniles. However, access is impractical when the analytical technique is destructive and requires at least half a tooth crown to provide sufficient enamel to analyse low-Pb samples such as these. Nevertheless, recent results from surface-only sampling by LA-MC-ICP-MS are promising for Sr isotope ratios of tooth enamel (J. Evans pers. comm.) and should greatly increase the number of samples that can be analysed for such studies.

The enamel-Pb concentrations of these individuals (0.15 – 0.65ppm) are somewhat higher than expected, being of the same magnitude as those found in the modern children, CM and AM. **This is consistent with the observations that Pb pollution and exposure is currently far lower than it has been for several centuries** (Jaworowski *et al.* 1985, 105). Nonetheless, Pb burdens an order of magnitude lower than the Monkton individuals have been found in both Blackfriars 89 and Iron Age, Bronze Age and Neolithic individuals from Cnip and West Heslerton. Post-mortem contamination cannot provide an overarching explanation for the Monkton samples, however, as the Pb isotope ratios are widely variant and not the same as the burial soil leaches. Permanent tooth dentine from Monkton has very similar Pb concentrations to the enamel, but neither A, B or D were fully formed teeth. The deciduous teeth exhibit more variation in Pb concentration between enamel and dentine but this should, perhaps, be expected given that the roots of B and D were actively resorbing.

The similarity of C to individuals from Gloucester, Winchester and Mangotsfield suggests an origin to the northwest, in the region of the Mendips orefield. It is known that soil Pb levels are raised in areas of mineralisation (Gulson 1986, 114) although the Pb is not necessarily bioavailable (Gulson *et al.* 1994b, 904). Clearly, even in periods when metallurgy was unknown, Pb could be ingested from the fingers and via the inclusion of soil and grit in food (e.g. from grinding/milling stones). Ingestion of dust and soil Pb, rather than inhalation or exposure to insoluble metal ore, is still regarded today as the major pathway for Pb uptake in children (Jaworowski *et al.* 1985, 121; Mielke & Reagan 1998, 224; Yaffe *et al.* 1983, 244).

6.6 Discussion and methodological developments

6.6.1 Diagenesis

6.6.1.1 Inter-tissue comparisons

There was no significant difference between the Sr isotope ratios of core and surface enamel (paired *t*-test: $P = 0.90$). However, in all modern and archaeological samples which had data for both tissues, Sr concentration was *greater* in the core enamel than the surface enamel and this difference is significant (n = 5, paired *t*-test: two-tailed $P = 0.02$, one-tailed $P = 0.01$). The actual concentration differences were not great, however, particularly when viewed in the context of the inter-enamel range of concentrations obtained for BAB and AM (section 5.3.3.1). Nonetheless, this strongly suggests Sr enrichment of the enamel surface is absent both *in vivo* and post-mortem. A difference was also suspected in the Pb concentration between these two tissues; in six out of eight teeth the surface enamel samples had more Pb than the corresponding core enamel in both modern and archaeological teeth but this difference proved not significant statistically (n = 8, paired *t*-test: $P = 0.10$). However, the observed surface enrichment was clearly not associated with any measurable change in Pb isotope ratio even for known immigrants, as no significant difference was found between the Pb isotope ratios of the core and surface enamel (n = 7, paired *t*-test: $P = 0.14$, 0.16 and

0.81 for $^{208}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ respectively). This indicates that the origins of the surface Pb enrichment lie in the early days of tooth mineralisation rather than later life or post-mortem. Alternatively, the surface enamel layer may be just too thin to have any appreciable affect on the Pb isotope ratios of the bulk of the core enamel beneath. Consequently, it is recommended for Pb and Sr analysis that the surface enamel layer is mechanically removed from samples in order to standardise the sampling protocol, clean the samples of surface accretions and facilitate comparisons between studies.

For all archaeological teeth analysed, dentine contains *more* Sr than the associated core enamel and this is highly significant (n = 17, paired one-tailed *t*-test: $P = 6.1^{-7}$). For non-Pb-coffin burials, the Pb concentration increase between enamel and dentine is also significant (n = 18, paired one-tailed *t*-test $P = 0.01$). Likewise, the differences in the Pb isotope ratios of dentine and enamel are all significant (n = 22, paired *t*-test: P = 0.02, 0.02 and 0.04 for $^{208}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ respectively), whilst the difference between enamel and dentine Sr isotope ratios is highly significant (P = 0.001). Moreover, where an isotope ratio change is apparent, the dentine ratios are intermediate between those of the co-genetic enamel and the burial soil (Figures 6.15 and 6.23). Again, however, Pb and Sr appear to be behaving in a different manner. Generally, the modern samples have slightly *less* Sr in the dentine than in the associated enamel and the *same* Sr isotope ratio. This change in the archaeological samples is quite startling and is strong evidence for post-mortem Sr contamination from the burial environment. There appears to be no systematic process to ascertain whether this concentration increase arises from simple addition or addition coupled with turnover of the carbonate hydroxyapatite itself (Budd *et al.* 2000a), but evidence from the modern samples would suggest that Sr is more mobile in skeletal tissue than Pb. This evidence suggests that primary crown dentine is extremely slow to change, particularly in permanent teeth. If the change in concentration and Sr isotope ratios is only from addition, it should be possible to remove the extraneous Sr and recover the Sr isotope ratios of the co-genetic enamel. Clearly, from the indications of subject BAB, permanent teeth of juveniles should have had insufficient time to register any change of residence ante-mortem. If post-mortem turnover has occurred to any great extent, techniques such as solubility profiling, which remove the more soluble diagenetic phases, would not be able to recover the *in vivo* Sr isotope signature.

The author’s hypothesis was subsequently tested by Trickett (1999; Trickett *et al.* in press) using premolars co-genetic with those previously analysed by the author from the two adolescents from Blackfriars: one which had more radiogenic Sr (341) and the other less radiogenic Sr (89) than the soil leaches. Bone and dentine Sr isotope ratios moved progressively further away from the soil Sr isotope ratio toward those of the core enamel with each leaching stage, i.e. in different directions but never quite reaching the enamel ratios. However, he found only small changes (< 0.0006) in the Sr isotope ratio of core enamel leaches or the final residue and the original, untreated (but physically abraded) tissue. Clearly, archaeological dentine, and by implication bone, is heavily and possibly irrevocably contaminated with Sr from the burial environment and cannot be used to represent *in vivo* signatures. It may, however, be a very useful proxy for the time-averaged, mobile soil Sr, something which it may not be possible to obtain from contemporary soils.

The evidence is clearer for Sr diagenesis of the dentine than it is for Pb. With few exceptions, both modern and archaeological teeth have the same or more Pb in the dentine than in the core enamel. It is, therefore, not clear from the Pb concentrations that Pb has been incorporated from the burial soil (or indeed the Pb coffins) into the dentine. It would appear that dentine has accumulated Sr from the burial environment to a greater degree than Pb. This would, perhaps, be expected given the greater sensitivity of Sr to *in vivo* change illustrated in the modern samples. Figure 6.23 shows a clear difference between the results from Winchester and Monkton although both were chalk burials. Enamel Pb isotope ratios are much more variable in the Monkton samples and the dentine is intermediate (with the exception of the permanent tooth of D, where enamel and dentine samples are within analytical error of each other) between the enamel and the chalk. Although chalk is depleted in Pb relative to typical crustal rock abundances (Faure 1986, 283), Pb concentrations were also small in the enamel and dentine samples, perhaps making them prone to swamping by burial soil Pb. The samples from Winchester have dentine and enamel ratios indistinguishable from each other and very different from the chalk. G339 was buried in a Pb coffin so it would not be expected to be diagenetically affected by the chalk. However, the three other Winchester chalk burials have the same Pb ratio as G339. This observation implies that in these cases, large *in vivo* Pb burdens were not swamped by chalk-Pb. Very similar ratios for the two individuals from Mangotsfield, buried in a limestone sarcophagus, are also evident. The mediaeval individuals from Blackfriars have more variable Pb isotope ratios than the Late Roman individuals, and in most cases the dentine Pb remains indistinguishable from the enamel Pb. The only exception is 209 where the dentine is intermediate between the enamel and the burial soil leach but there were no criteria by which this individual could be differentiated from the other Blackfriars samples; tooth types and preservation scores etc. were very similar for all Blackfriars individuals.

The inter-dentition Sr and Pb isotope ratios from the Monkton juveniles varied considerably. The deciduous teeth from B and D had roots that were partially resorbed whilst the permanent teeth from A, B and D had incomplete root formation (Table A6). The modern samples suggested that completely resorbed roots may produce a considerable change in crown dentine Sr isotope ratio, and to a lesser extent Pb isotope ratios, if those of the contemporary environment are different from those obtained when the enamel was mineralising. However, only primary, crown dentine was analysed in these samples and none had completely resorbed roots. Moreover, similar dentine isotope changes are also seen in the two adults (C and E) and the full-rooted deciduous tooth of A. There appears, therefore, no reason to assign the observed inter-tissue differences to mobility rather than post-mortem incorporation. All the Monkton teeth had exceptionally well-preserved enamel and dentine; indeed, they were almost indistinguishable from modern examples and the comments of Radosevich (1993) that excellent preservation results precisely *because* of Sr accumulation are worth considering here. Despite the excellent preservation, there appears to be a diagenetic trend towards homogenisation with the burial environment in both Pb and Sr isotope ratios in the dentine. The Winchester teeth were also very well preserved but there has been little change in the dentine Pb isotope ratios towards those of the burial environment. Two factors are suggested: the much shorter burial period of the Winchester group or, as discussed above, the greater ante-mortem Pb concentration in the teeth which makes the *in vivo* Pb signature much more robust in such a low-Pb environment.

6.6.1.2 The Pb coffin burials

Results from the three teeth from the two Pb coffin burials suggest that enamel and dentine behave very differently in this unusual burial environment although the reasons are not immediately apparent. Curiously, dentine samples from the Pb coffins have not absorbed large amounts of Pb and contain less than dentine samples from individuals (e.g. G326, 341) not buried in Pb coffins. Conversely, at least one enamel sample (G339b) has incorporated a lot (1540ppm). Surface accretions were present on the enamel of G339 but not on SPR-1, suggesting that water could percolate freely through the coffin of G339. It was hypothesised that small fissures in the enamel may have enabled Pb to penetrate from the surface and that these were left uncleaned during sample preparation. As described in the subsequent Spitalfields sample preparation (section 6.4.3.3), this possibility was addressed very carefully in the Spitalfields study. Two teeth (G339a and SPR-1) have Pb concentrations which, today, would be considered indicative of extremely high childhood exposure, and the first reaction is to invoke post-mortem contamination. However, SPR-1 enamel-Pb is different from the coffin-Pb, and G339a is different from the heavily contaminated G339b, implying that the sample preparation has succeeded in extracting enamel that has not been entirely swamped by Pb from the burial environment. As G339a was a third molar and G339b a premolar it is possible that the thicker enamel of G339a resulted, either through the core being better protected or more resistant to hairline cracks and fractures, in a less contaminated sample. Second or third molar enamel may, therefore, provide a more robust post-weaning sample in situations where substantial Pb diagenesis is suspected.

Although it cannot be proposed that Pb isotopes are an ideal system for identifying migration in individuals buried in Pb coffins, these results are nevertheless positive, and validate the sample preparation technique developed in this project and the skill and experience brought to bear when carrying it out. There is no reason *per se* to believe that *in vivo* Pb is not still present in the teeth as, in the absence of groundwater, it is difficult to suggest a mechanism for its removal. Although hydroxyapatite is used to "mop-up" Pb on contaminated land, the mechanism appears to be dissolution of the carbonate hydroxyapatite and re-precipitation as hydroxypyromorphite rather than substitution into the carbonate hydroxyapatite lattice (section 2.3.4). Moreover, results from the modern studies suggest Pb signatures are actually quite robust and reluctant to change even in trabecular bone. The study by Molleson et al. (1986) demonstrates that the *in vivo* Pb signature persists even in rib bones. This positive outcome could be because individuals with a high-Pb exposure and buried in chalk where Pb is immobile and present at a very low concentration (as was also found at Winchester) were analysed. The same result may not have been obtained from, for example, low exposure individuals in an acidic Pb-rich soil.

The Sr results for G339 merit discussion as there is still evidence for Sr diagenesis from the chalk in both teeth. The Sr isotope ratio of the dentine is intermediate between those of the enamel and the chalk. Dentine Sr concentration is slightly elevated compared to that of the enamel, but not to the same degree as observed at other sites such as Monkton or Blackfriars and G339a does not appear to have been affected to the same extent of G339b.

6.6.1.3 Soil leaches

The soil leaches from Winchester and Monkton raise several questions. At rural Monkton-up-Wimbourne, there was too little Pb to analyse in the water leach from either the chalk or the soil sealed at the bottom of the Neolithic shaft. However, the surface soil in which the Bronze Age male (E) was buried did yield enough Pb from the water leach to analyse. It produced a very different Pb isotope signature to any of the acetic leaches of the soil or the chalk bedrock. At urban Winchester, sufficient Pb was obtained from the chalk water leach but it had a very different Pb isotope signature to the acetic leaches from either Winchester or Monkton but bore marked similarities to the surface soil at Monkton (Figure 6.23). This suggests that the water leach is removing solid Pb particulate pollution from the surface soils that has been deposited in the intervening centuries, rather than Pb intrinsic to the chalk. As these Pb isotope ratios are very similar to anthropogenic British ore Pb, and there is evidence from a chalkland study that *"non-petrol sources of lead, in particular historical inputs, dominate the store of lead in sediments and rural soils"* (Bacon *et al.* 1996, 2516), this would support this conclusion. Clearly, the ancient soil at the bottom of the Neolithic shaft has been protected from subsequent surface pollution.

Accordingly, it is suggested that an initial wash with de-ionised water to remove recent Pb particulate should precede the acetic acid leach to extract the mobile soil Pb. The results from the Bronze Age burial from Monkton indicates that this surface Pb was either immobile or insoluble and patently did not supply the diagenetic Pb incorporated into the dentine; Figure 6.23 clearly shows it to be of chalk origin. For Sr isotope ratios, the data suggest that the acetic acid leach was slightly better than the water leach for obtaining the mobile Sr at the Blackfriars site although, for leaches of the alkaline chalk and associated soils, there was very little appreciable difference. Further work on soil leaches using a standard sequential leaching protocol such as that of Tessier *et al.* (1979), one developed from it (e.g. Li *et al.* 1995) or alternatively, that of the Community Bureau of Reference (Quevauviller *et al.* 1997), would be very useful and would facilitate inter-study and inter-laboratory comparisons. Used in conjunction with bone or dentine data from the same burial contexts this would enable refinement of the soil leaching method to ensure a good representation of what the mobile Pb and Sr signatures have been over the period of burial. The most appropriate leach reagent may vary with different soil types and the element of interest. It is suggested that, in the case of archaeological studies where changes in land use or alteration of the soils are suspected, bone or dentine may offer a more pertinent indication of the ancient time-averaged signature since burial than the soil.

6.6.2 Implications for archaeological studies of migration and exposure

6.6.2.1 Sr

The variation in Sr isotope ratio from these samples (0.708 – 0.714) is not particularly large on the geological scale and there is a clear tendency for the vast majority of individuals to cluster around 0.709 ± 0.001. A very similar range and distribution of enamel ratios has been reported by Grupe et al. (1997, 523) from sites on Tertiary and Jurassic deposits in Bavaria. This reflects the age and type of the geological sites studied and is probably typical for central, southern and eastern England. It may be very different for areas of much older or younger geology.

Sr enamel concentrations are very consistent and rarely outside the range of 40-100ppm, whereas those of Pb vary by four orders of magnitude. The striking difference between archaeological dentine, i.e. increased Sr concentration coupled with more soil like ratios against the unchanged dentine of a modern immigrant, appears to be clear evidence for post-mortem Sr diagenesis. The enamel Sr isotope ratios still appear to be linked to the local region: at Winchester and Monkton a similar range of enamel Sr isotope ratios were found but this was not the case for Pb. The difference in time periods between these two sites accounts for the Pb differences but has made little difference to the Sr isotope ratio. The negative correlation found between Sr concentration and Sr isotope ratio is interesting but may only have arisen as a result of the two types of case study analysed, i.e. Cretaceous chalk and slightly more radiogenic Triassic/Jurassic sediments. However, it does suggest that there is some relationship between the amount of Sr in the underlying geology and the amount incorporated into the enamel irrespective of other nutritional factors.

6.6.2.2 Pb

The enamel-Pb isotope variation found in these studies is quite large but primarily restricted to the Neolithic samples. The pre-metallurgical individuals are the only ones that do not plot within the Pb ore isotope field. Based on data in this study and other data in the literature (Kowal *et al.* 1991; Molleson *et al.* 1986), the Pb isotope ratios of English people appear very consistent from the Roman period through to the 19th century, i.e. they reflect domestic Pb sources and suggest anthropogenic sources contribute the most to the Pb burden at these times. Moreover, results show that anthropogenic enamel-Pb isotope ratios cluster even tighter in the lower centre of the English ore field than the spread of ore Pb isotope ratios would suggest they might. Despite the large variations seen in Sr isotope ratios there is little concomitant variation in Pb during the Roman or Mediaeval periods when access to metals and possibly urban pollution were high. Obviously there is a big time gap between the Neolithic samples and those from the late-Roman period so it is impossible to identify, based on this sample set, when the link between an individual and their place of origin is severed by widespread anthropogenic Pb. The evidence from Blackfriars illustrates it is still possible to obtain country rock (i.e. non-ore) Pb isotope ratios after the advent of metallurgy, and these are likely to go hand in hand with Pb concentrations < 1ppm, as was also found at Monkton. Similarly, it is suggested that the purported "Greek" child of Molleson et al. (1986), for whom the authors could find no corresponding ore Pb other than those of Laurion, may also be an individual whose bone-Pb burden is indicative of natural, geological Pb, either *in vivo* or from the chalk in which it was interred, rather than one dominated by Pb artefacts (Figure 6.26).

6.6.2.3 Sr and Pb

The negative correlation found between Pb and Sr concentrations in enamel in modern and archaeological teeth is interesting. It is not known why this should occur but it indicates that Pb and Sr incorporation in enamel is not controlled by the same factors. Direct antagonisms such as Sr substituting passively for Ca whilst Pb is suppressed in high Ca diets and Pb-rich geology/soils/plants being Sr-poor can all be suggested but the underlying reason is unlikely to be simple. Generally, there is little statistical evidence for any correlation between Sr and Pb isotope ratios, however, in the few individuals with country rock Pb (89 and the Monkton burials) there does appear to be some geological relationship between the two isotope systems. This connection between the geology and the individual survives until the advent of anthropogenic Pb. This is demonstrated by the lack of variation of Pb isotope ratios, but variation of Sr isotope ratios in Roman and Mediaeval individuals. The modern study indicates that Sr is much more sensitive to changes of origin than Pb in resorbing deciduous roots and alveolar bone. This may be evidence that skeletal Pb is immobile or re-mobilised and then re-incorporated *in situ*, whereas Sr is far more easily exchanged. These characteristics would also seem to affect the respective susceptibilities to diagenesis also.

6.6.2.4 Inter and intra-tooth variation

As was found in modern samples, archaeological permanent tooth enamel replicates well both within a single tooth and between co-genetic antimeres from the same dentition. Different teeth from the same dentition also gave very similar results. However, considerable differences in Pb and Sr isotope ratios were found between deciduous and permanent enamel from the same individual at Monkton. Interestingly, the small Sr isotope ratio difference, coupled with a much greater Pb isotope ratio difference found between the modern siblings AM and CM, was also found between Monkton B and D, also purported siblings. The source of these differences is not known but it is suggested that it is more likely to arise from *in utero* dietary differences during transfer from mother to offspring than mobility in both cases. There is clearly no simple explanation for the differences between Sr and Pb isotope ratios in the teeth of the Monkton juveniles, many of which were either actively resorbing or still in the process of root formation. In accord with the aDNA results, it would be possible to suggest from the Pb and Sr isotope ratios of the deciduous teeth that B and D were not the offspring of C and that, of the three juveniles, A was most like C. Overall, the data imply that deciduous teeth are considerably more dynamic and their origin *in utero* more complex than permanent teeth. As a result, they will be more difficult to interpret.

6.7 Conclusions

6.7.1 Conclusions drawn from modern teeth

1. Enamel has *more* Sr than dentine and *less* Pb than dentine.
2. Sr isotope ratios are negatively correlated with Sr concentration but there is no correlation between Pb concentration and Pb isotope ratios. A significant negative correlation also exists between Sr and Pb concentrations but not between ^{87}Sr/^{86}Sr and ^{207}Pb/^{206}Pb ratios.
3. Enamel and the co-genetic crown dentine retain Pb and Sr isotope ratios indicative of childhood origin. There is no evidence for post-eruption uptake of Pb or Sr into enamel.
4. Sr is more sensitive to changes in origin than Pb, suggesting 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 does turnover in bone.
5. All evidence suggests that there are very different factors controlling the uptake of Pb and Sr into skeletal tissue.
6. The resorbing roots of deciduous teeth are more sensitive to changes in origin than permanent teeth, although deciduous enamel does appear to retain evidence of origin or the skeletal signatures of the mother. Deciduous teeth with resorbing roots must be treated with caution.
7. Co-habiting siblings were found to possess different 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 studies of migration.
8. Contemporary English subjects have skeletal Pb that is shifted from native British Pb isotope ratios to older ratios dominated by Australian Precambrian Pb ores.

6.7.2 Conclusions drawn from archaeological teeth

1. Core enamel from the same tooth is a relatively homogeneous tissue with regards to Pb and Sr and results replicate well both within a tooth and between two co-genetic teeth.
2. During burial, enamel-Pb and Sr appears to retain biogenic concentrations and isotope ratios. Even Pb coffin burials suggest that *in vivo* signatures have not been entirely swamped by the Pb dominated environment. Dentine however does change even in apparently well-preserved samples. It has more Sr than enamel and very different Sr isotope ratios to enamel, but is similar to other dentine samples from the same cemetery, making it possible to propose it as a time-averaged proxy for mobile soil-Sr.
3. British Pb isotope ratios change once importation of Pb commences in the mid 19th century and become progressively less radiogenic. A modern British signature can still be identified but it is different to that prior to the 20th century.
4. The range of Sr isotope ratios obtained was 0.7080 – 0.7143. In geological terms this is quite a restricted range but typical for the Cretaceous/Jurassic bedrock at the sites investigated. Individuals tend to have a Sr isotope ratio that is a mixture of 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 geology. This results in a tendency for Sr isotope ratios to cluster around 0.709 ±0.001 in these studies but it will not apply to all areas.
5. Sr and Pb isotope ratios are connected to the geology until some time after the advent of metal use. After this time, individuals may still retain a Pb signature indicative of geological origin if they have a very low Pb exposure that is not swamped by anthropogenic Pb. It may, therefore, be used as a status indicator in certain cases – remember that some rural origins produce high Pb exposure due to soft water.
6. Enamel-Pb concentrations >1ppm are only obtained at a restricted range of isotope ratios, i.e. at the lower centre of the English ore field. Low enamel-Pb concentrations (<1ppm) tend to have variable ratios and plot outside the ore Pb field. Variable Pb isotope ratios tend to go hand in hand with invariant Sr isotope

ratios, whereas variable Sr isotope ratios all have fairly invariant (i.e. ore) Pb. Variable Pb isotope ratios were only obtained for prehistoric individuals. Elevated enamel-Pb was only obtained from Roman and Mediaeval burials.

7. Susceptibility to diagenesis will depend on how great the enamel Pb concentration was before the tooth was buried and the subsequent availability, solubility and mobility of Pb in the burial environment.
8. Soil leaches are complex but vital to site interpretation. Water leaches of alkaline chalk and chalk soils appear to be removing predominantly particulate Pb (if present) of an entirely different origin to the chalk and are recommended prior to obtaining the acetic acid leach. Acetic acid leaches characterise the diagenetic Sr and Pb signature of the chalk well. For Sr there is little difference between the water and acetic leaches of the chalk, but on the less alkaline soils at Blackfriars the acetic leaches appear to better characterise the mobile Sr that has contaminated the dentine.

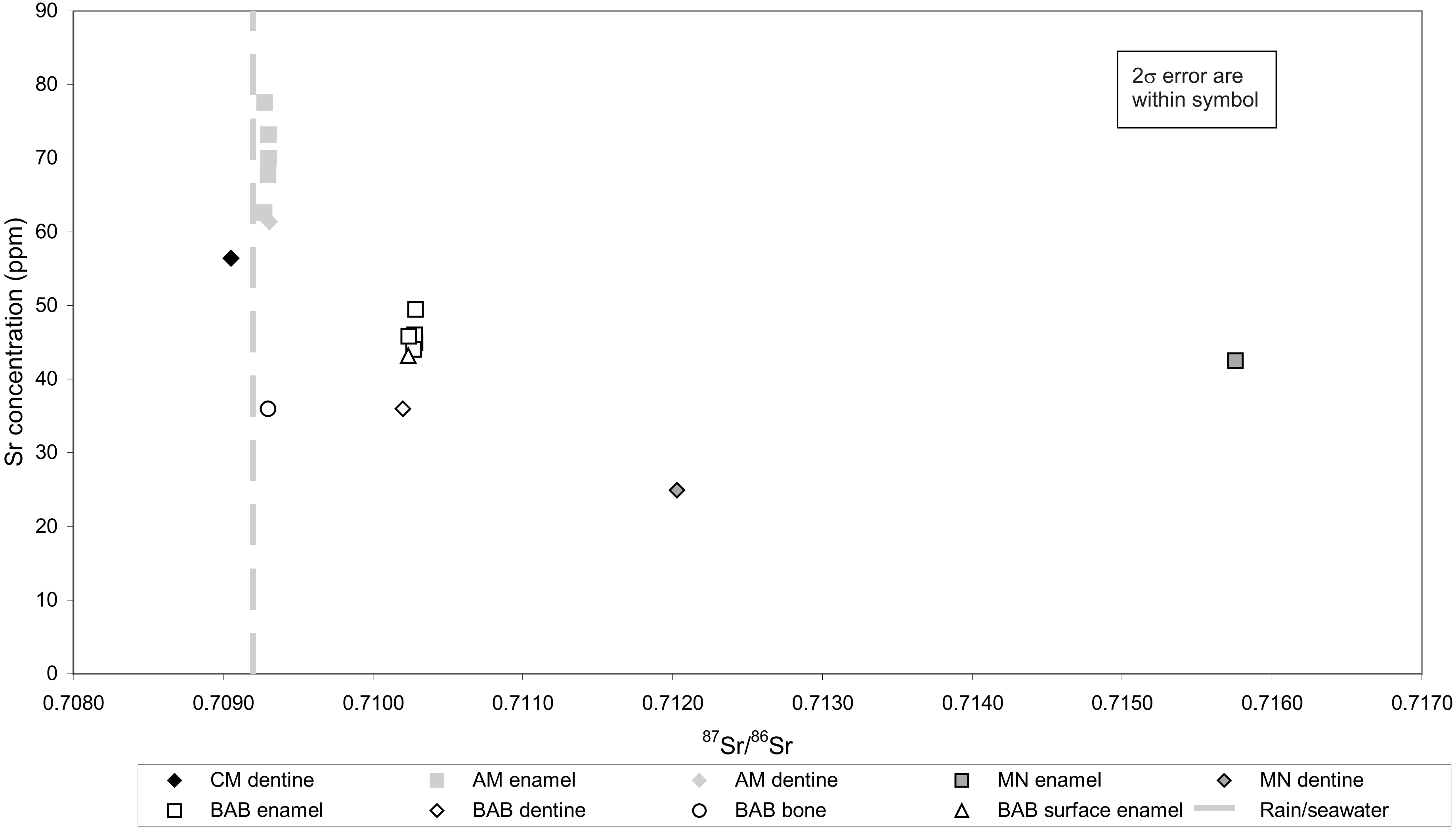


Figure 6.3 Plot of ^{87}Sr/^{86}Sr ratio versus Sr concentration for modern subjects. BAB and MN are modern migrants to England, CM and AM are indigenous siblings. Enamel contains more Sr than dentine. Note that even after 20 years residency only the bone of BAB has shifted and dentine has remained indistinguishable from the co-genetic dentine. The dentine of MN was from an exfoliated deciduous tooth and is clearly a more dynamic tissue. Despite CM and AM being co-habiting siblings they still exhibit a Sr isotope ratio difference that exceeds analytical error.

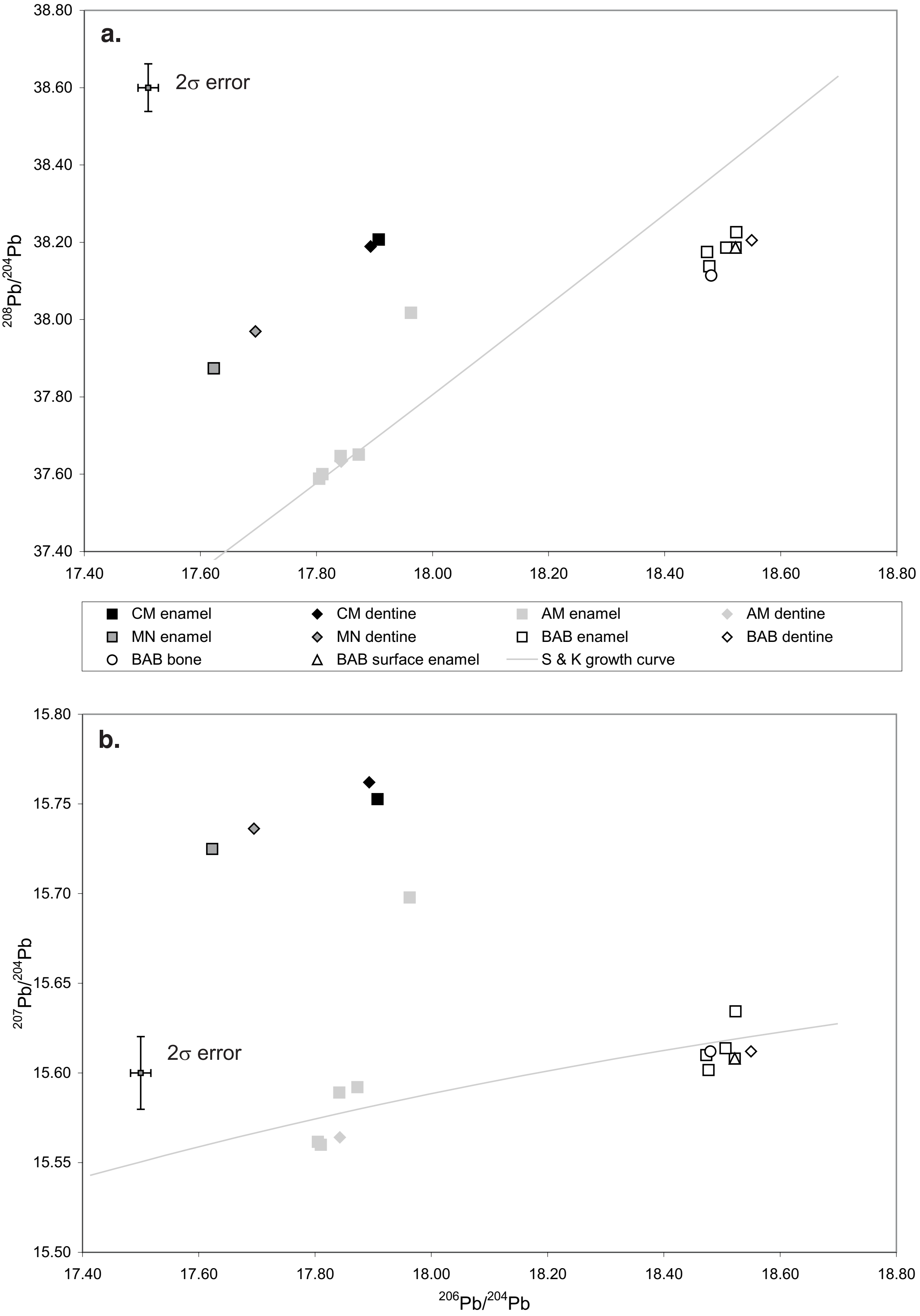


Figure 6.4 Plots of ^{206}Pb/^{204}Pb versus ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb ratios for modern subjects. BAB and MN are modern migrants to England. CM and AM are indigenous siblings. Note that even after 20 years residency all tissues of BAB are still within analytical error. The dentine of MN, an exfoliated deciduous tooth appears to have shifted towards that of CM. Note also the difference between CM and AM.

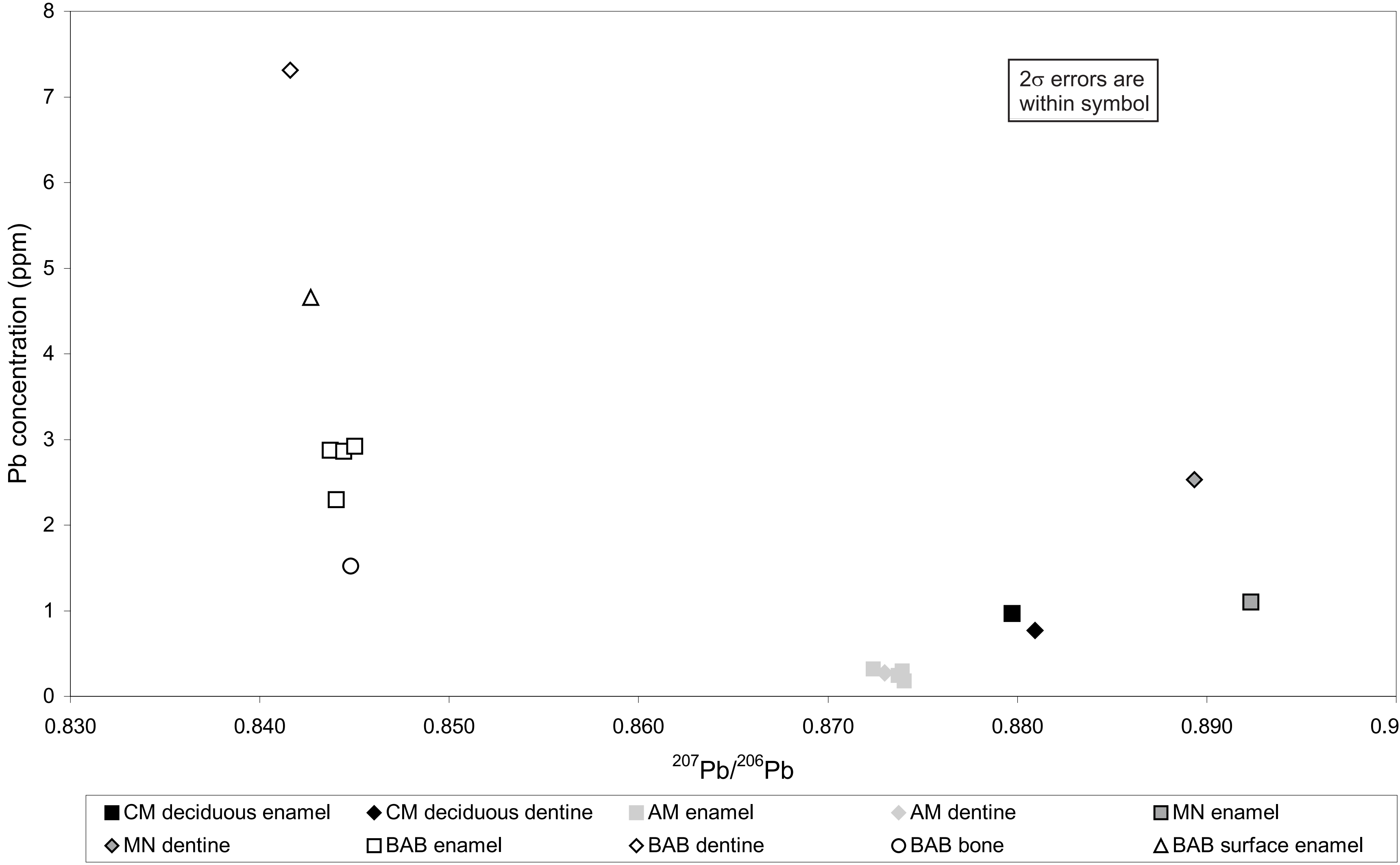


Figure 6.5 Plot of ^{207}Pb/^{206}Pb versus Pb concentration for modern subjects. Note that all samples for AM have the same ^{207}Pb/^{206}Pb ratio including the sample that appeared to be aberrant in Figure 6.4.

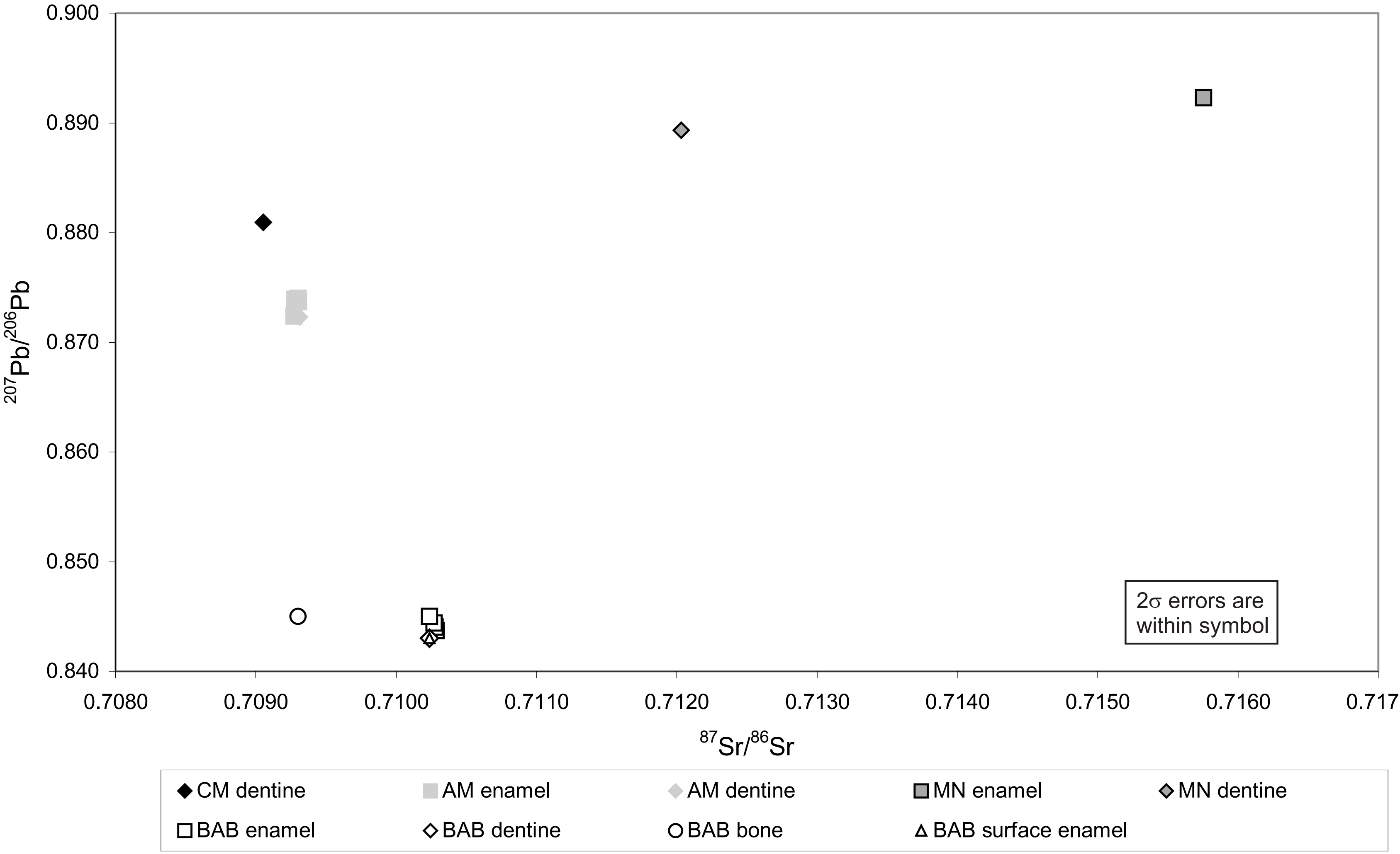


Figure 6.6 Plot of ^{87}Sr/^{86}Sr ratio versus ^{207}Pb/^{206}Pb ratio for modern subjects. Note the excellent reproducibility of the tooth tissues of AM and BAB.

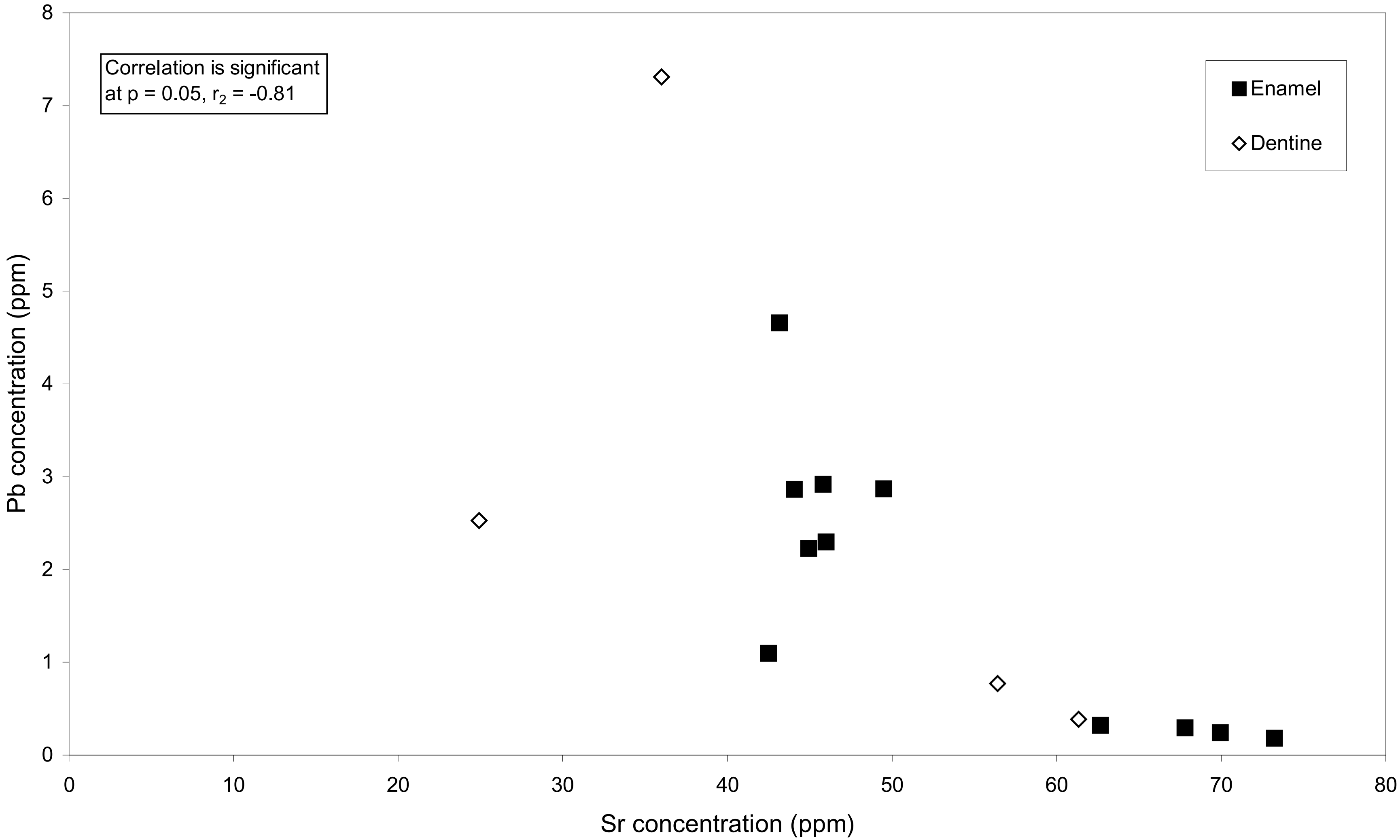

Correlation is significant at $p = 0.05$, $r_2 = -0.81$

Figure 6.7 Plot of Sr versus Pb concentrations in enamel and dentine of modern subjects.

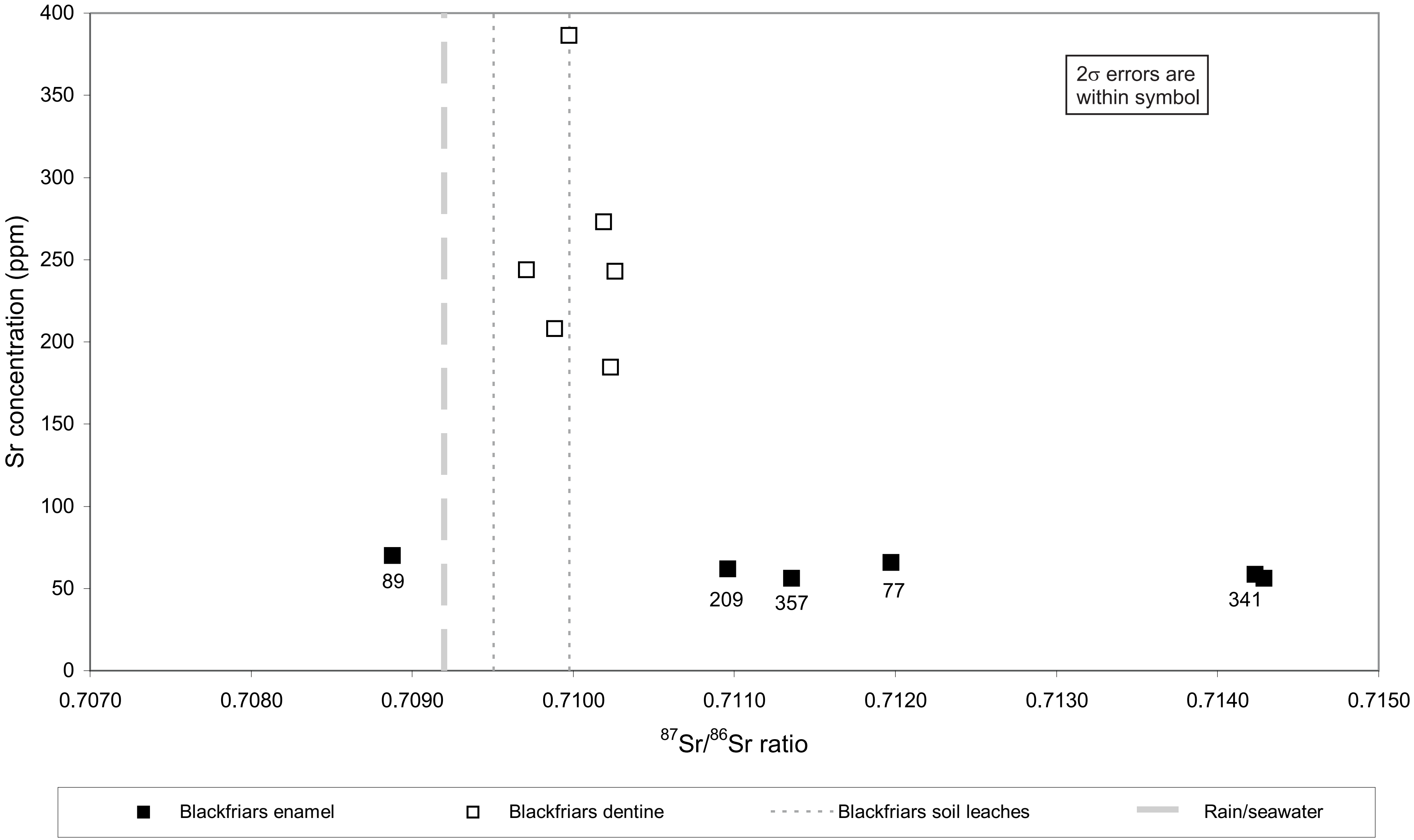


Figure 6.8 Plot of $^{87}Sr/^{86}Sr$ ratio versus Sr concentration for Blackfriars samples. Enamel samples have similar low Sr concentrations but widely varying ratios whereas dentine samples contain much more Sr and exhibit a much more restricted $^{87}Sr/^{86}Sr$ ratio range that is very similar to the burial soil leaches. Excellent reproducibility was obtained from the two antimeres of Burial 341. Burial 77 lies within the variation observed at the site. Note that Burial 89 is less radiogenic than the soils and rain/seawater.

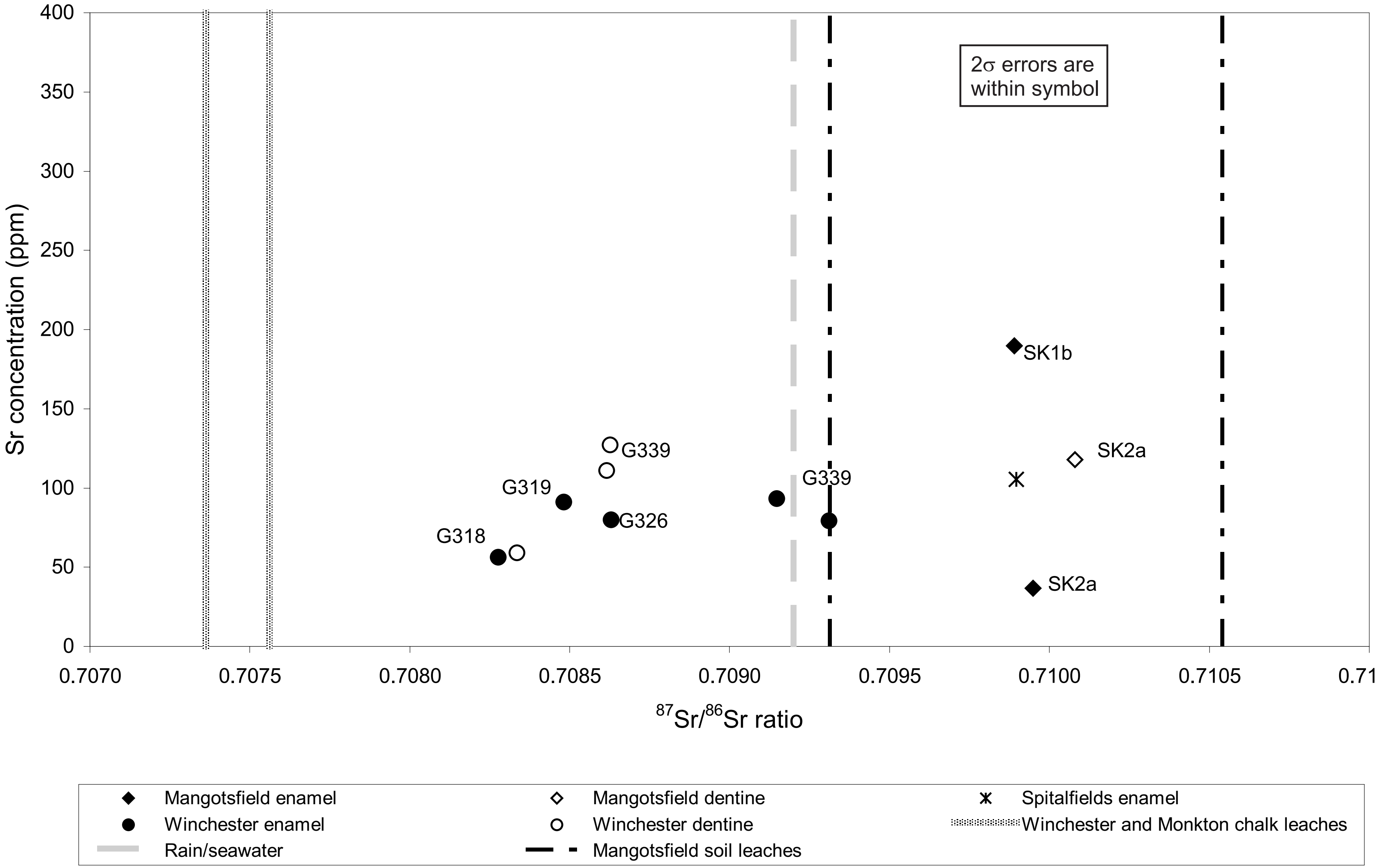


Figure 6.9 Plot of ^{87}Sr/^{86}Sr ratio versus Sr concentration for Late-Roman samples. Dentine Sr concentrations were not obtained for G319 and G326 but in both cases the Sr ratio was less radiogenic than that of the enamel, i.e. between the co-genetic enamel and the chalk ratios (see Figure 6.13).

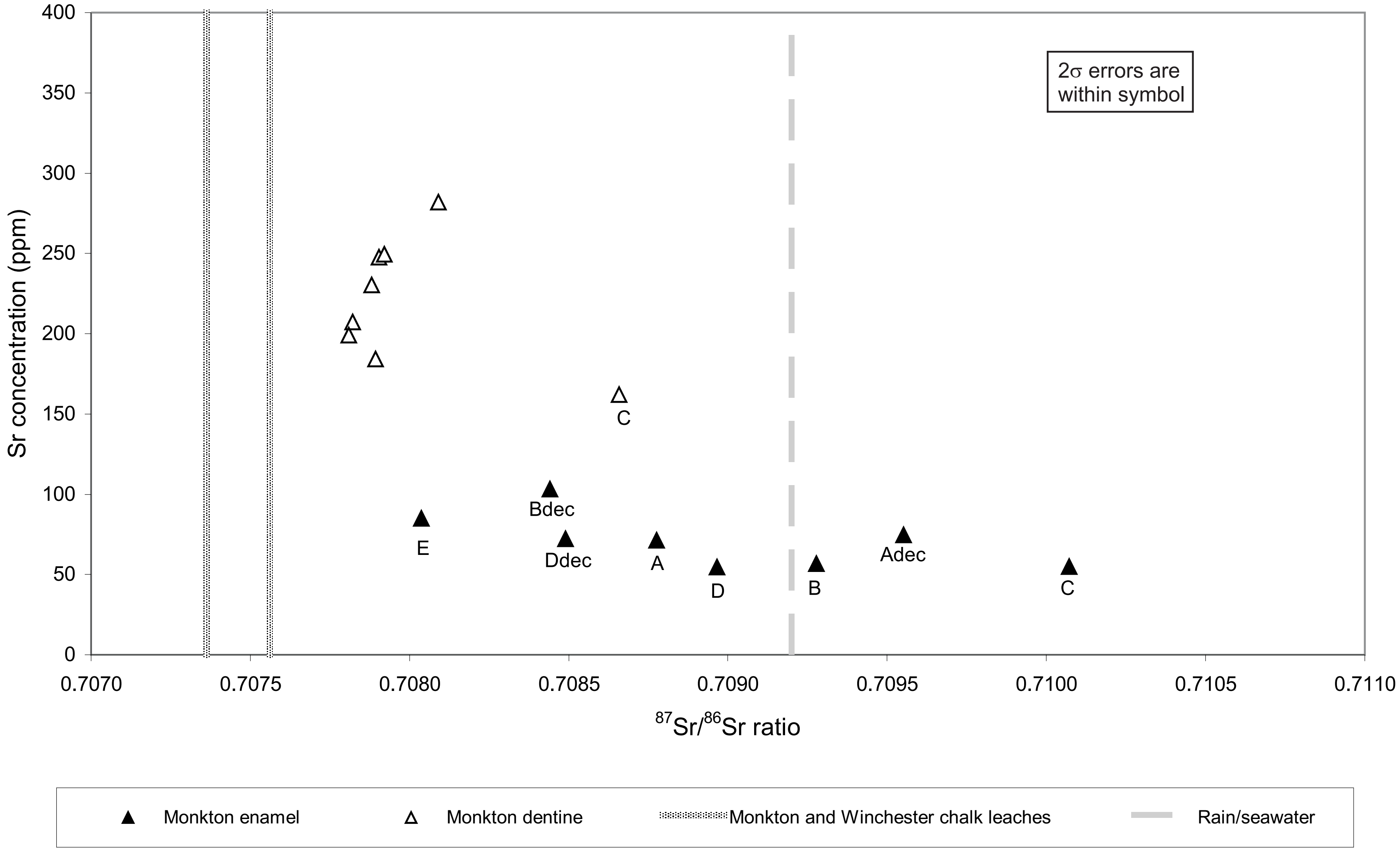


Figure 6.10 Plot of ^{87}Sr/^{86}Sr ratio versus Sr concentration for Monkton samples. Enamel samples have similar low Sr concentrations but varying ratios whereas dentine samples contain much more Sr and exhibit a much more restricted ^{87}Sr/^{86}Sr ratio range that is very similar to the burial soil leaches. Note that for the eldest children, D and B, the permanent enamel has a more radiogenic Sr ratio than the deciduous (i.e. in utero) enamel whilst the opposite is true for A, the youngest child.

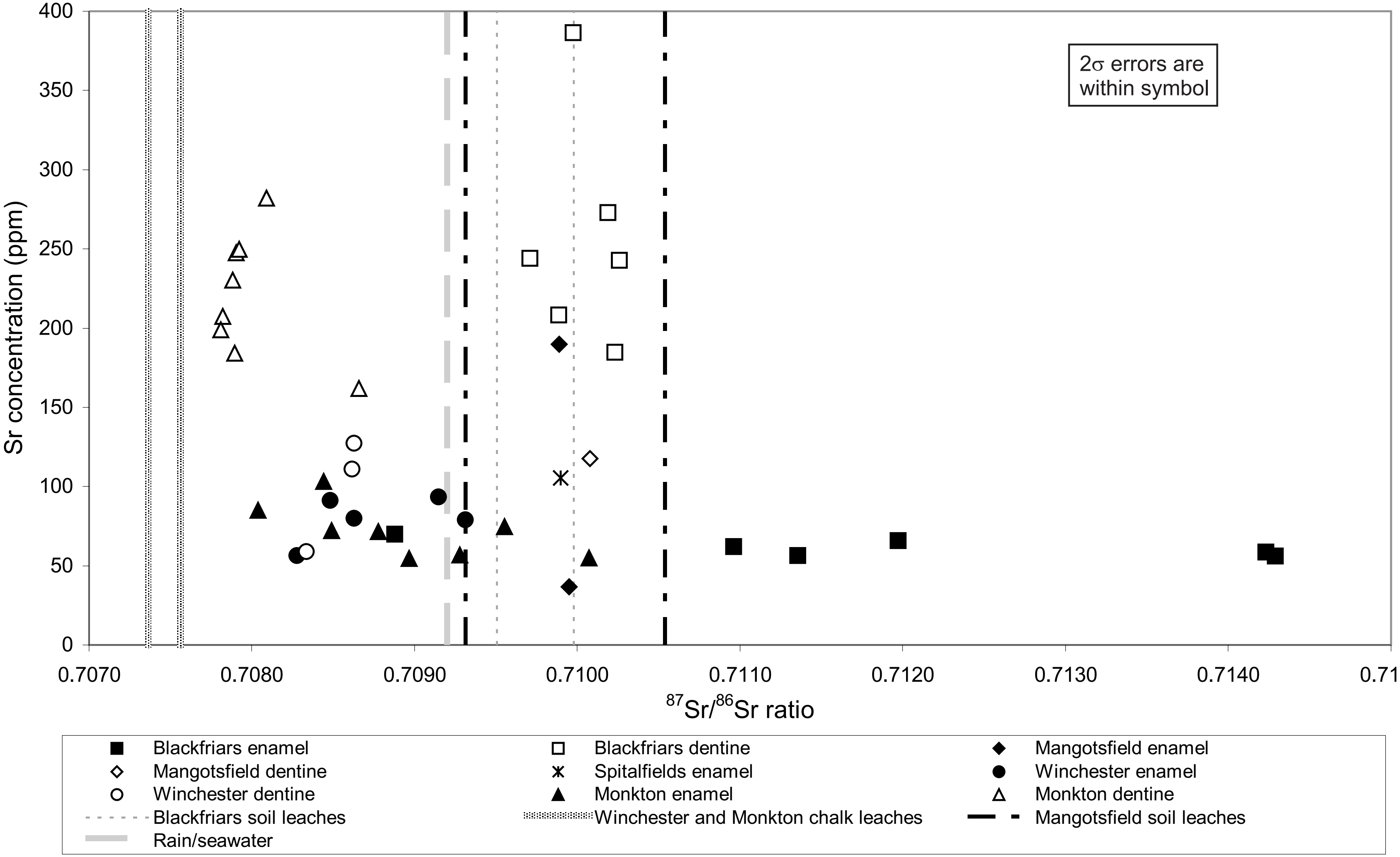


Figure 6.11 Plot of ^{87}Sr/^{86}Sr ratio versus Sr concentration for all archaeological pilot studies. Note that contrary to the modern samples, dentine contains more Sr than enamel and tends to cluster close to the Sr isotope ratio obtained from the burial soil leaches.

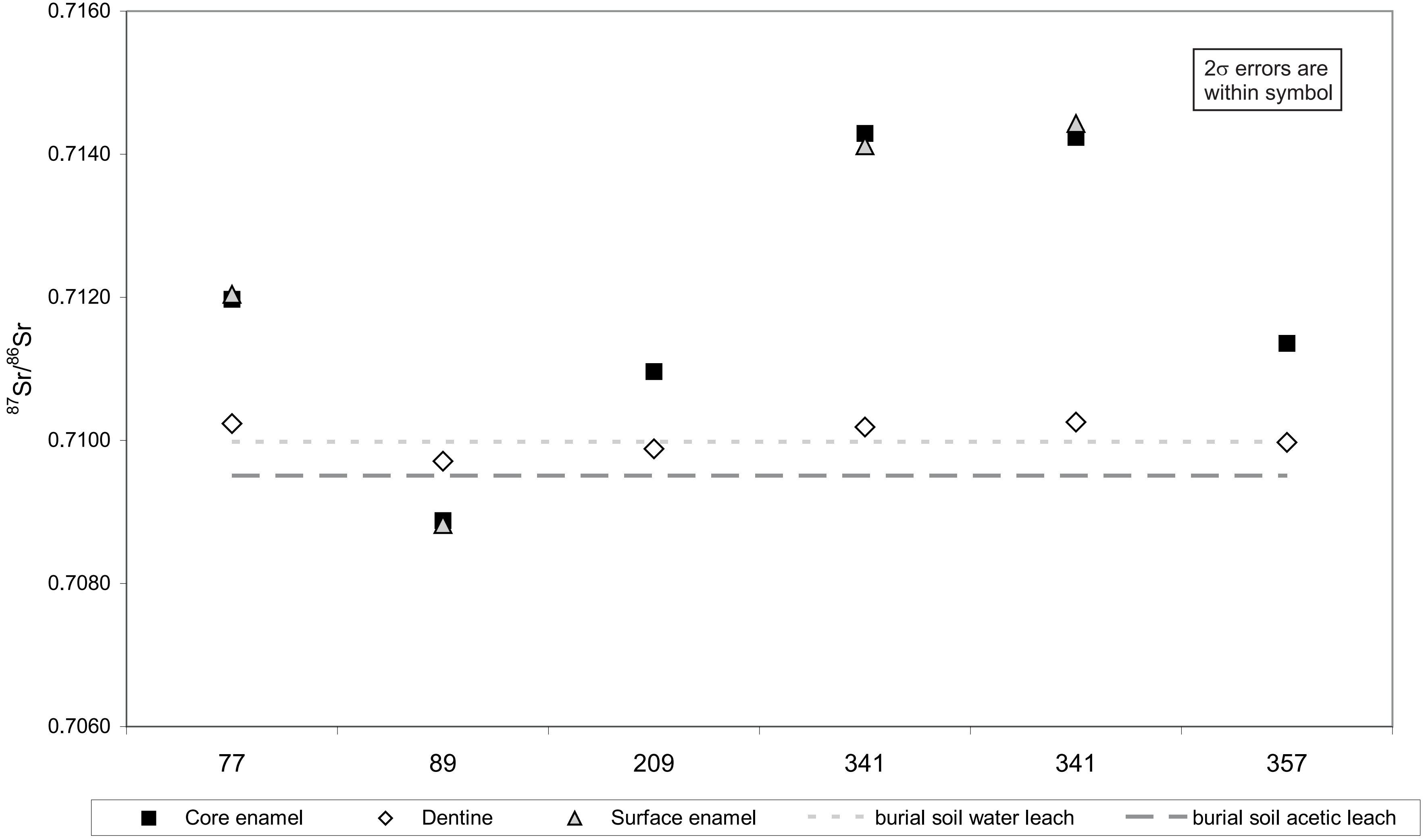


Figure 6.12 Plot of ^{87}Sr/^{86}Sr ratios of intra-tooth tissues for Blackfriars samples. As with modern examples, core enamel and surface enamel samples have the same Sr ratio. For all samples, the dentine Sr ratio is virtually indistinguishable from the burial soil ratios. Note the excellent reproducibility of the antimeres from Burial 341.

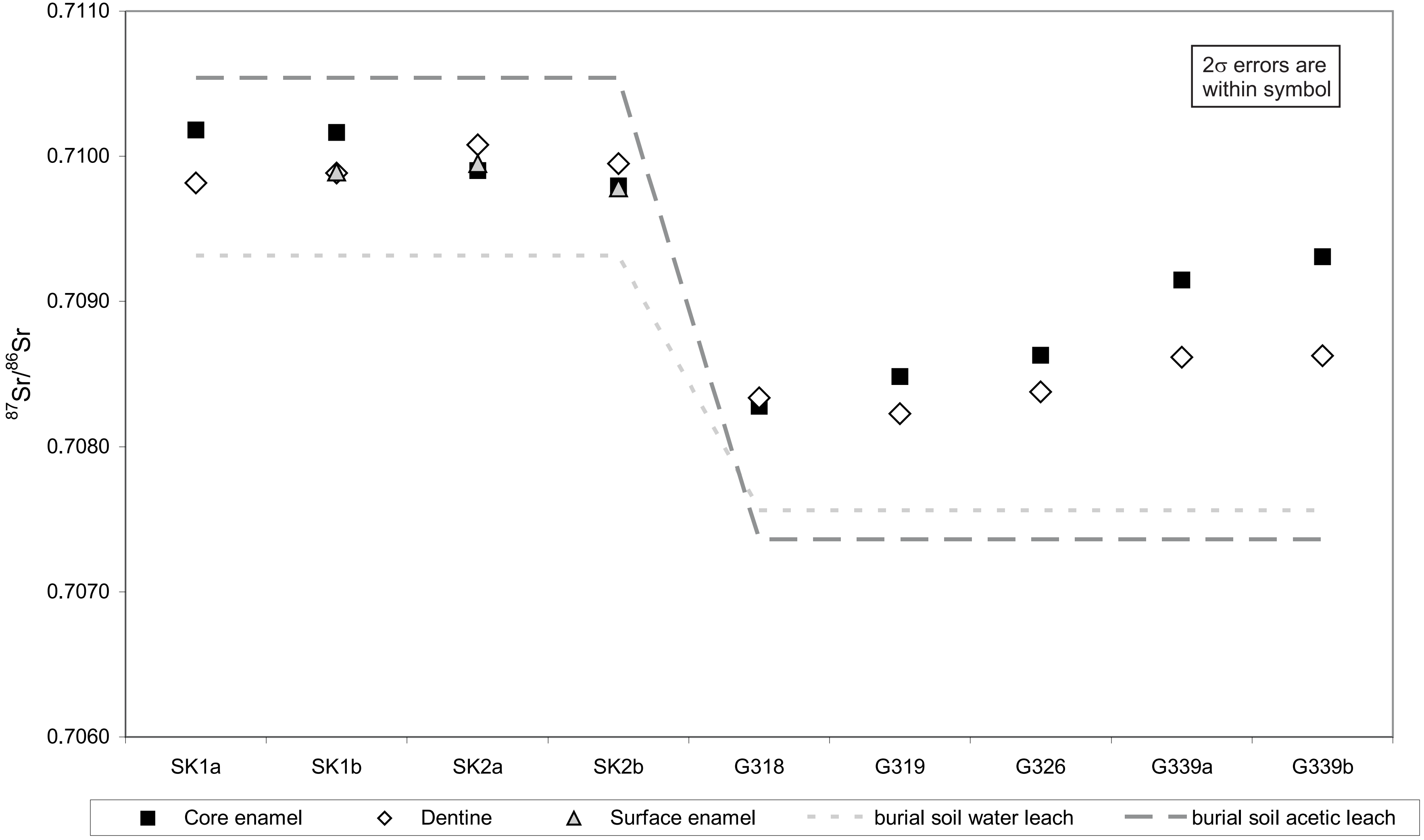


Figure 6.13 Plot of $^{87}Sr/^{86}Sr$ ratios of intra-tooth tissues for Late-Roman samples. For the Winchester samples, dentine ratios are intermediate between those of the enamel and the chalk burial soil. SK1 and SK2 were excavated from a limestone sarcophagus filled with local soil but they were not contemporaneous burials. The burial environment is clearly more complex and the soil leaches variable. The tissues of the two individuals appear to be reacting differently with the enamel-dentine vector moving in opposite directions.

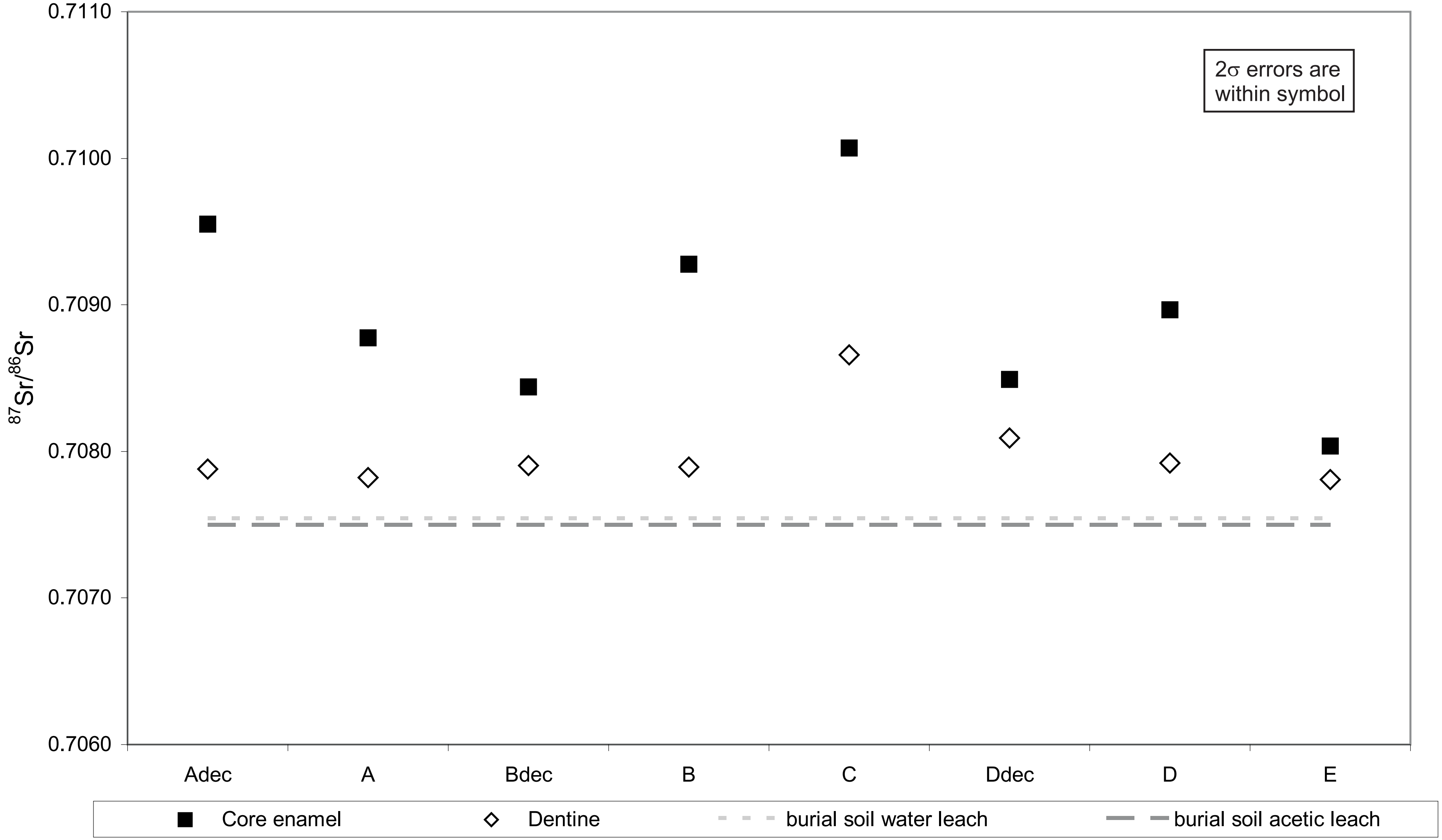


Figure 6.14 Plot of $^{87}Sr/^{86}Sr$ ratios of intra-tooth tissues for Monkton samples. In every case, dentine ratios are intermediate between those of the enamel and the chalk burial soil leaches.

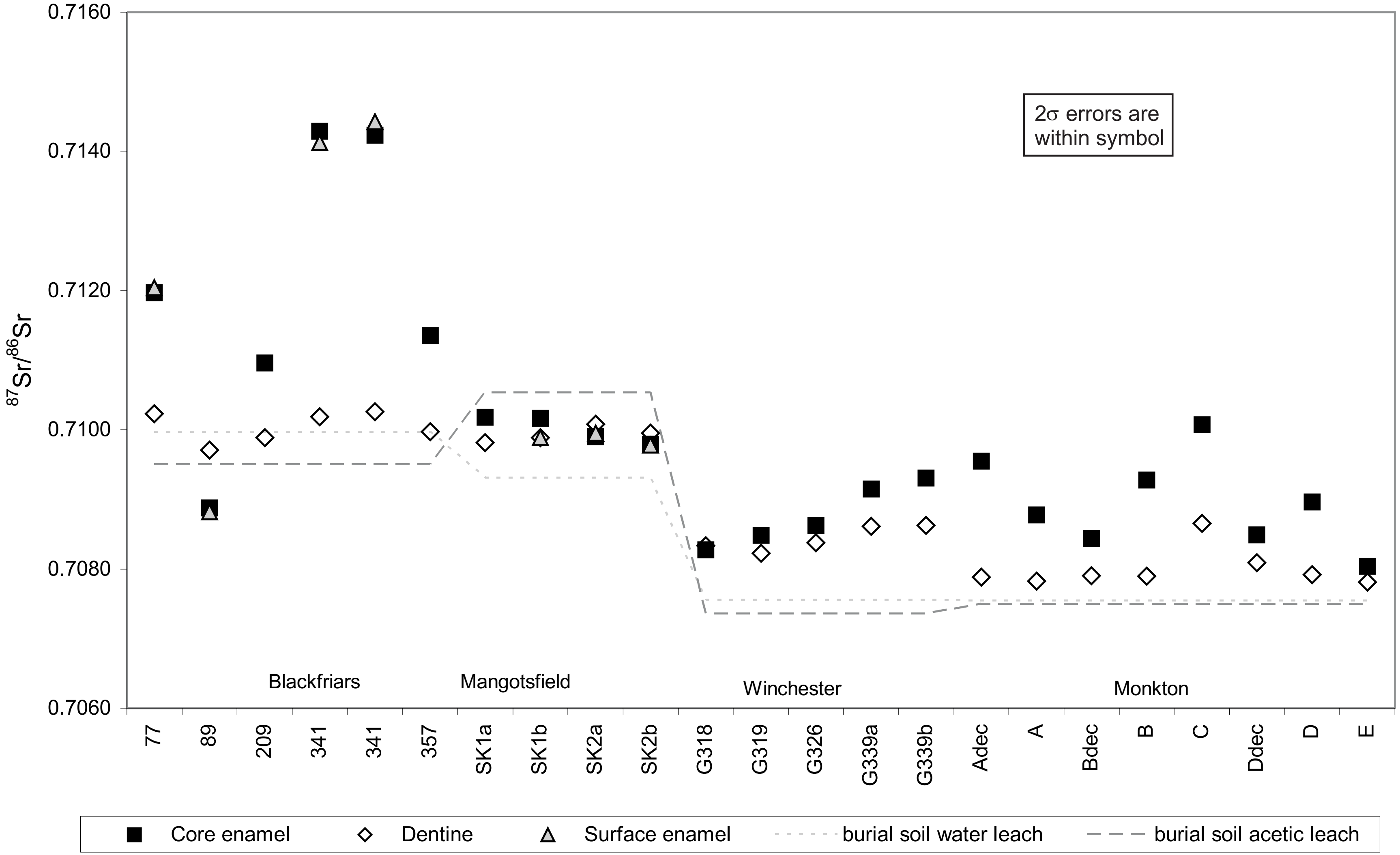


Figure 6.15 Plot of $^{87}Sr/^{86}Sr$ ratios of intra-tooth tissues for all archaeological pilot studies. Note that in virtually every case the dentine ratio is intermediate between that of the co-genetic enamel and the burial soil leach.

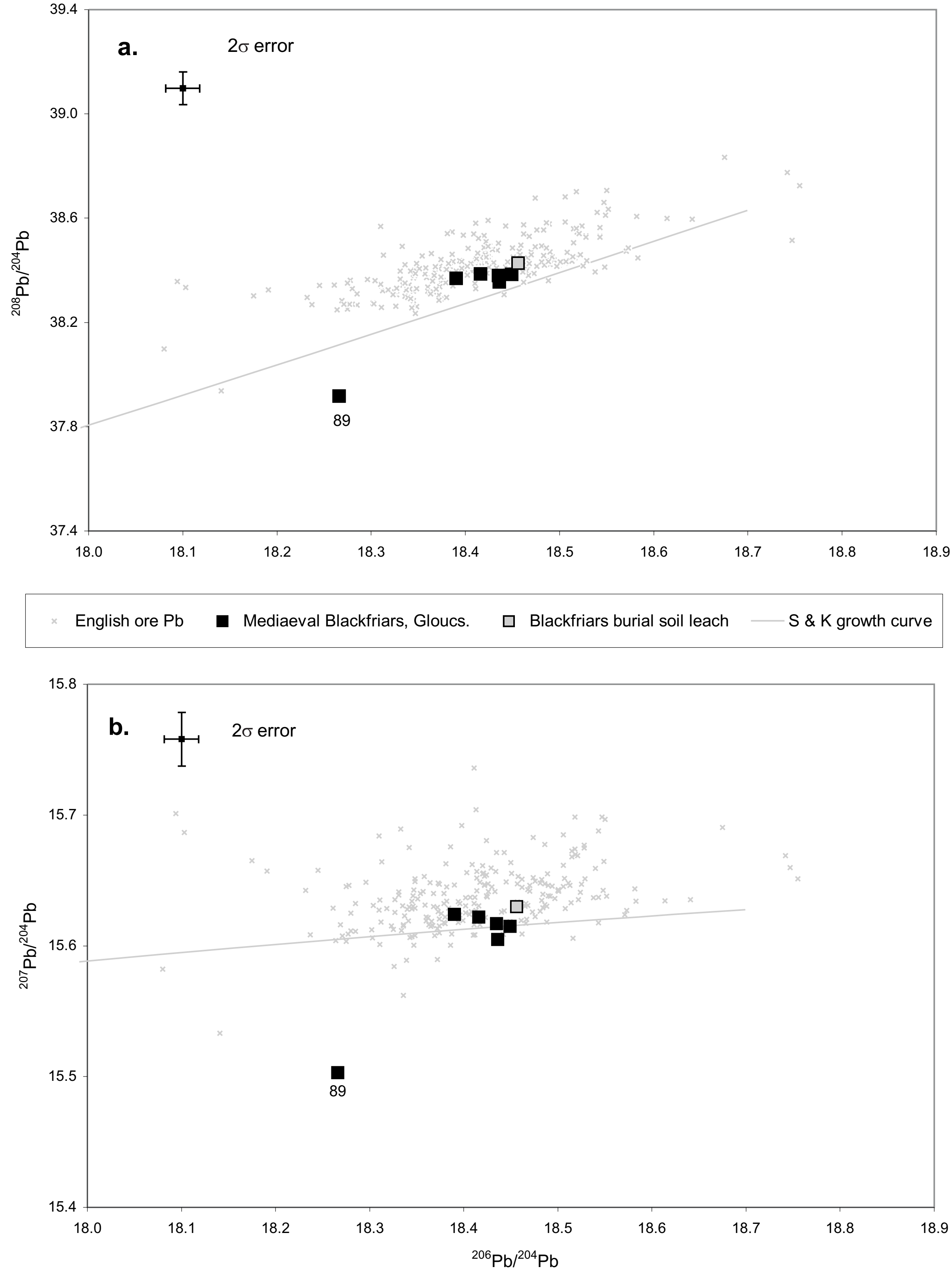


Figure 6.16 Plots of ^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb ratios for Blackfriars enamel samples. With the exception of Burial 89 which also had a very low Pb concentration, note the tight clustering of enamel ratios and the soil leach in the lower centre of the English ore Pb field.

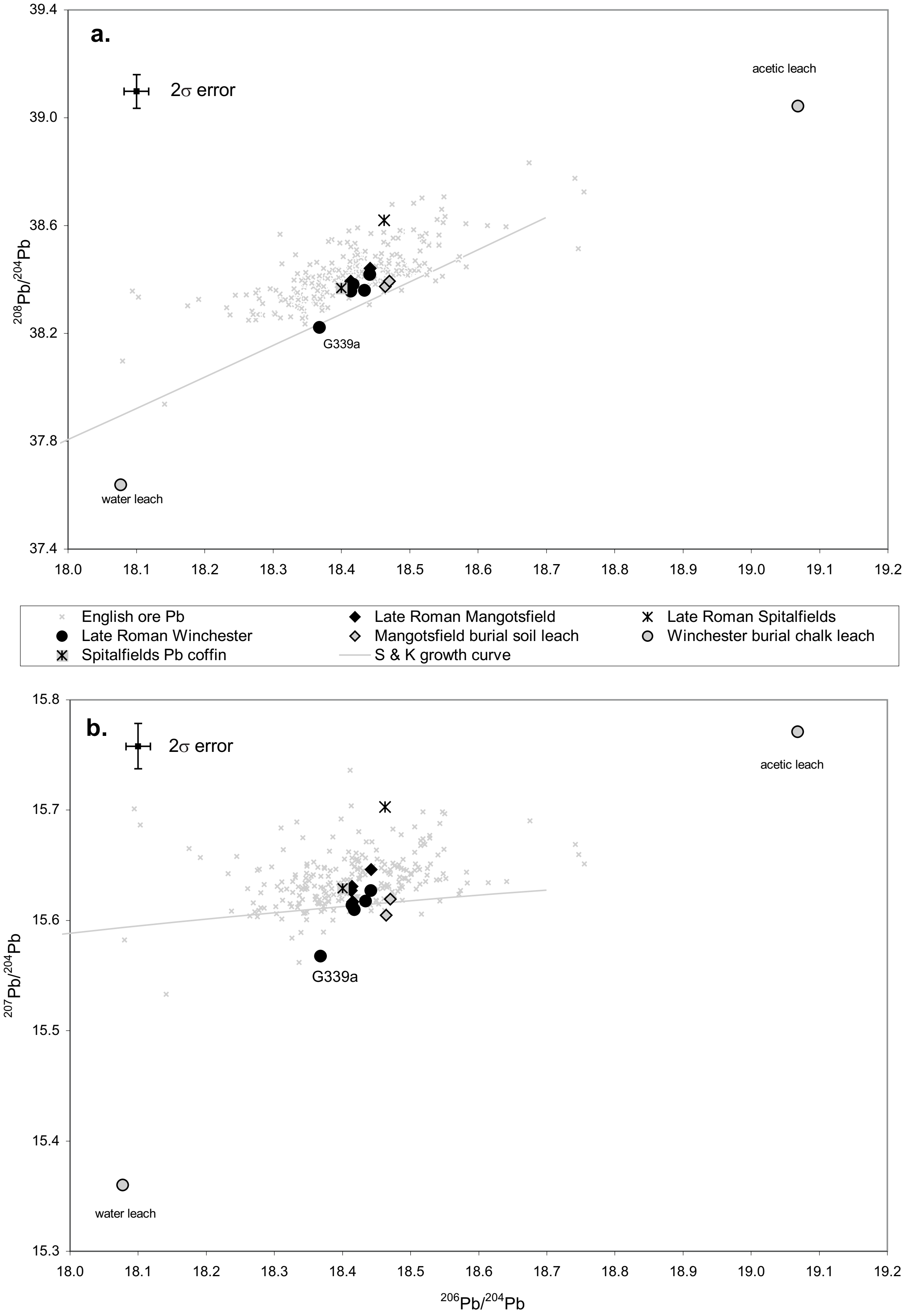


Figure 6.17 Plots of ^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb ratios for Late-Roman enamel samples. Most samples cluster tightly in the lower centre of the English ore Pb field. Both the outliers (Spitalfields and G339) were Pb-coffin burials. Note the difference between the Spitalfields enamel Pb isotope ratio and that of the Pb coffin.

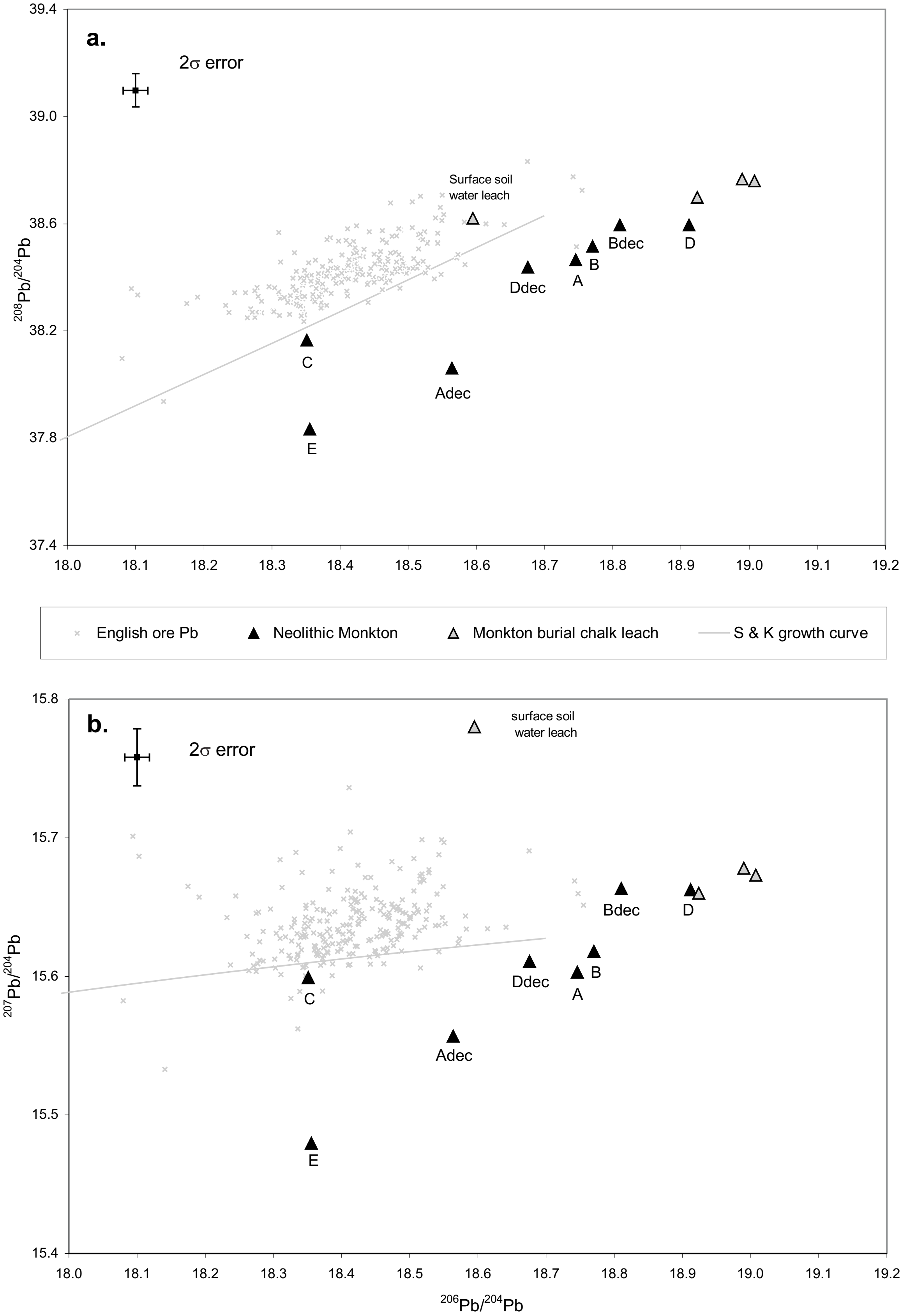


Figure 6.18 Plots of ^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb ratios for Monkton enamel samples. Only C and the surface soil water leach fall within the English Pb ore field, the remaining samples fall on a line below and to the right of the Pb ore growth curve.

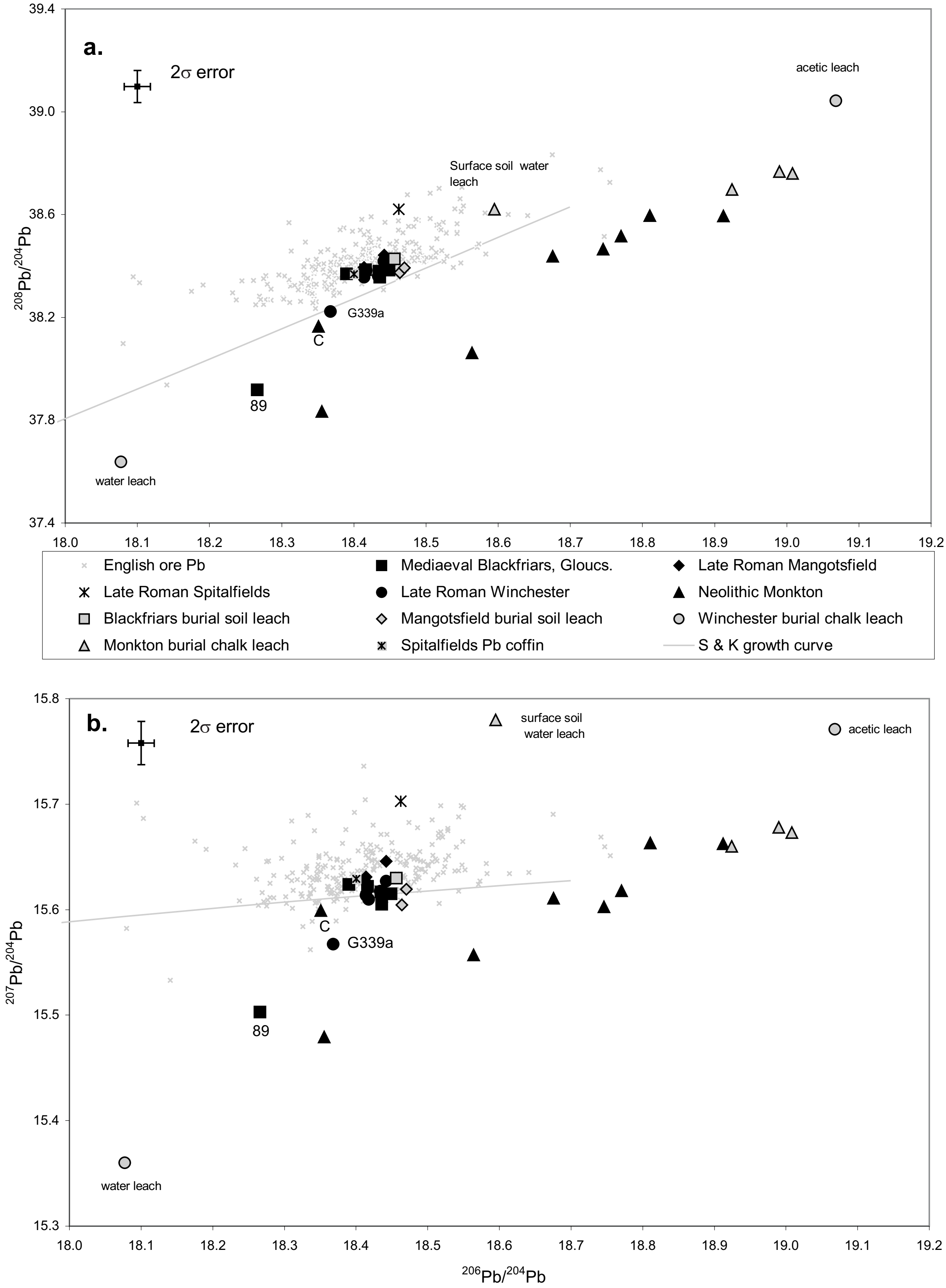
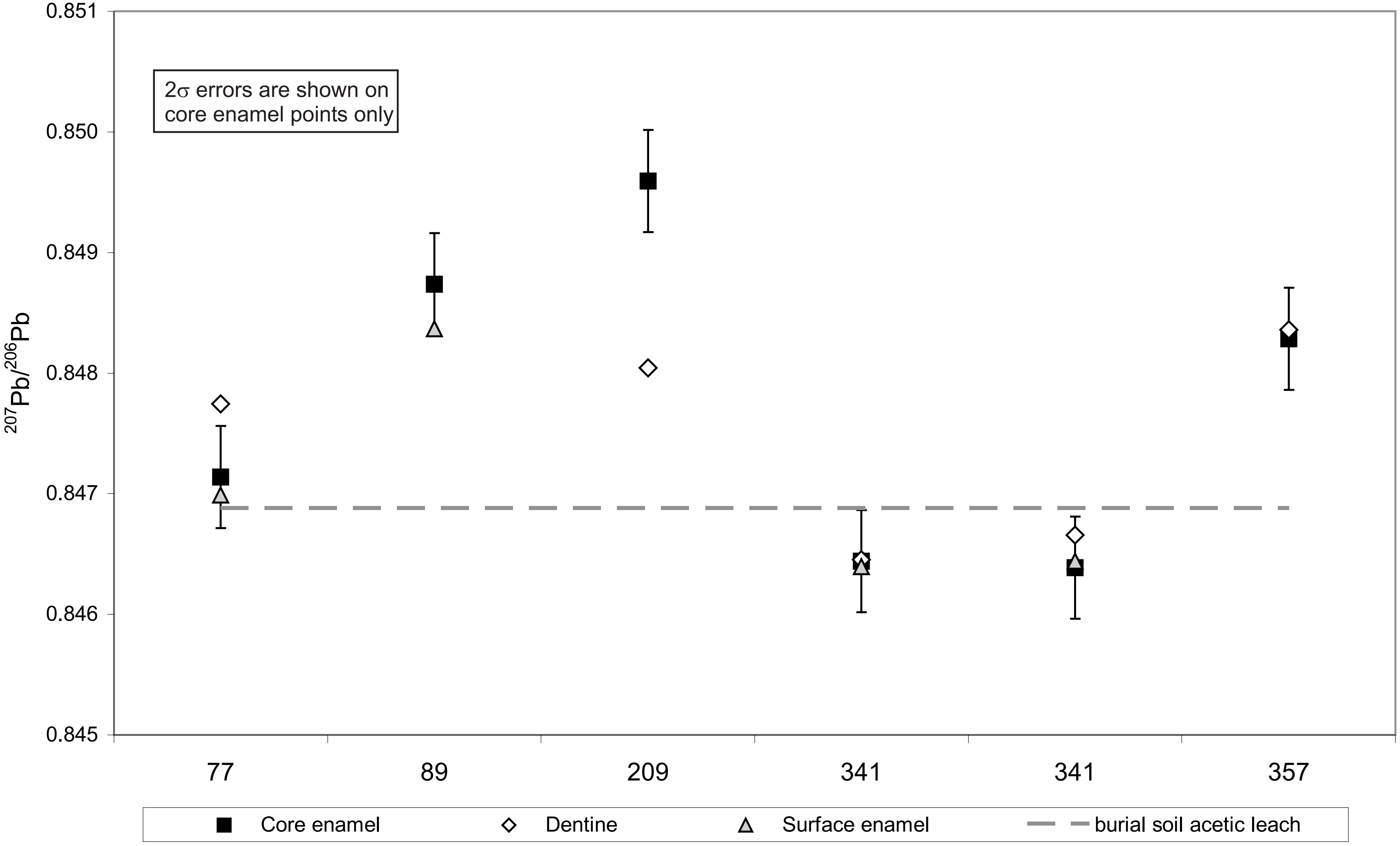


Figure 6.19 Plots of $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios for all archaeological pilot studies enamel samples.



2σ errors are shown on core enamel points only

Figure 6.20 Plot of ^{207}Pb/^{206}Pb ratios of intra-tooth tissues for Blackfriars samples. There is little variability between intra-tooth tissues (note the expanded scale); in most cases core enamel, surface enamel and dentine are within analytical error.

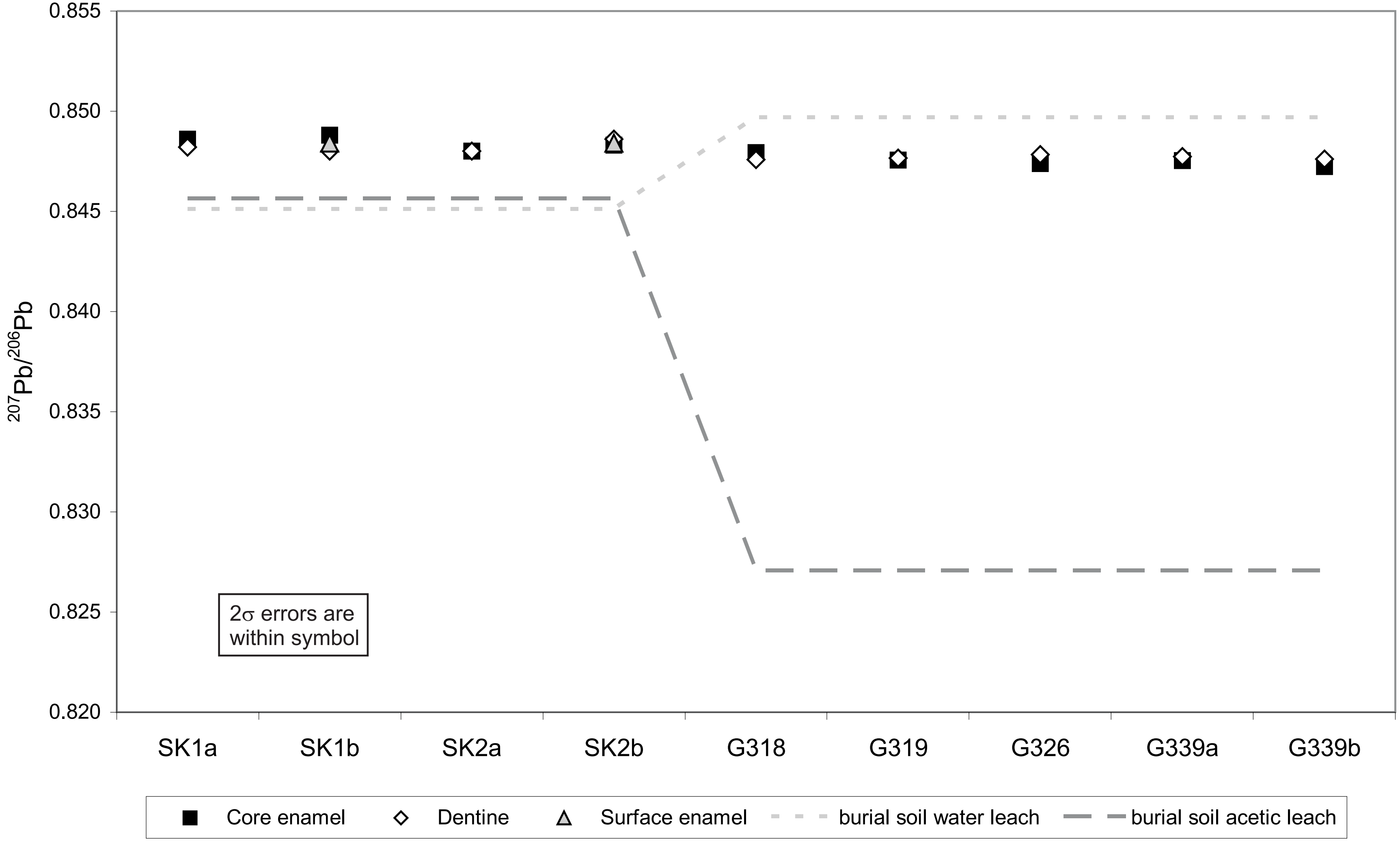


Figure 6.21 Plot of ^{207}Pb/^{206}Pb ratios of intra-tooth tissues for Late-Roman samples. As with the Blackfriars samples (Figure 6.20 and 6.23), there is virtually no difference between any of these tissues. Note however, that the Pb obtained from the acetic chalk leach at Winchester appears to have no effect on the dentine Pb ratio which is not the case with the Neolithic samples from Monkton (Figure 6.22 and 6.23).

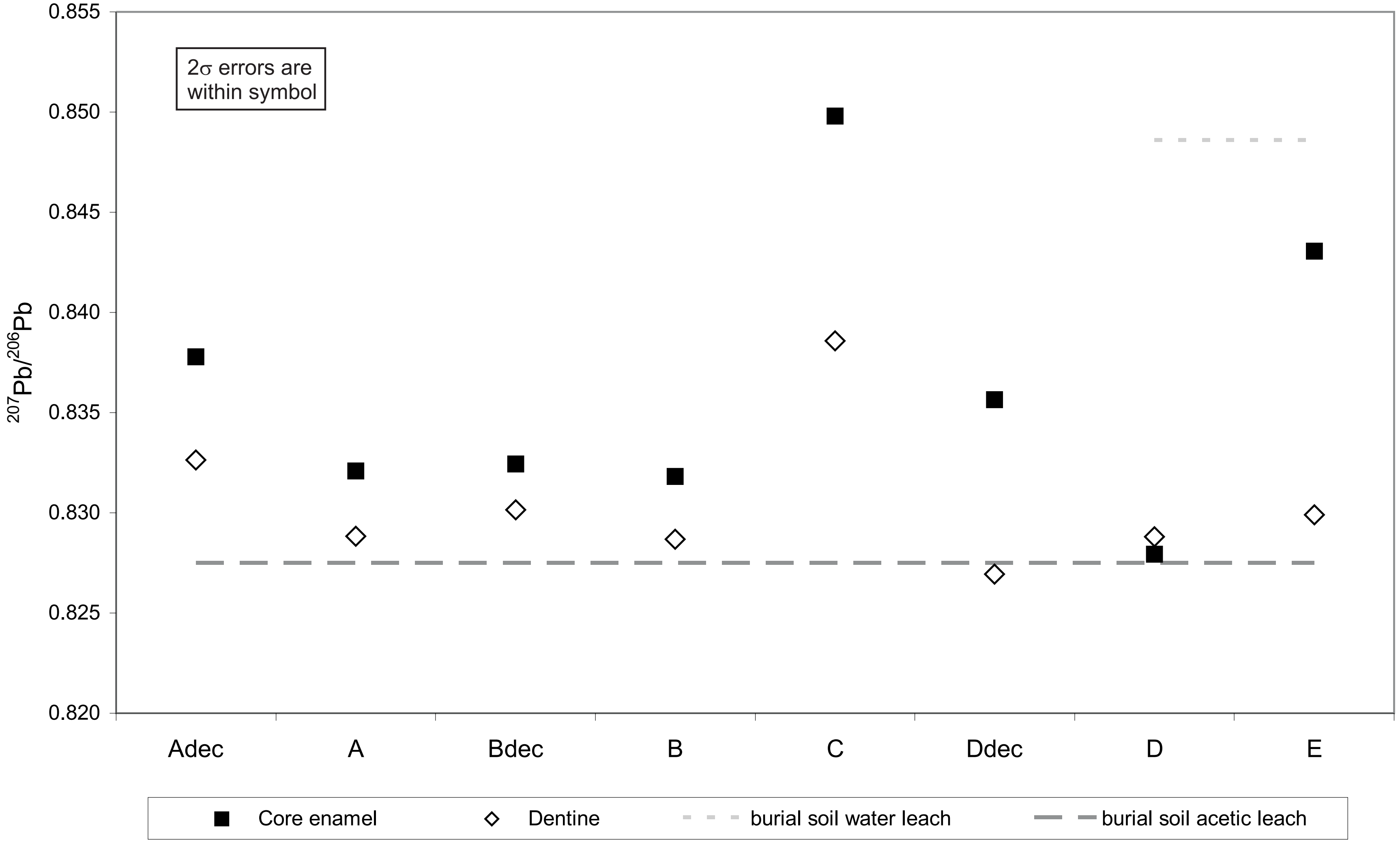


Figure 6.22 Plot of ^{207}Pb/^{206}Pb ratios of intra-tooth tissues for Monkton samples. Dentine Pb ratios are intermediate between those of the co-genetic enamel and that of the acetic chalk leach. Sufficient Pb for analysis could only be obtained from the surface soil water leach, in which only E was buried, but this appears to have no influence on the dentine Pb of these samples.

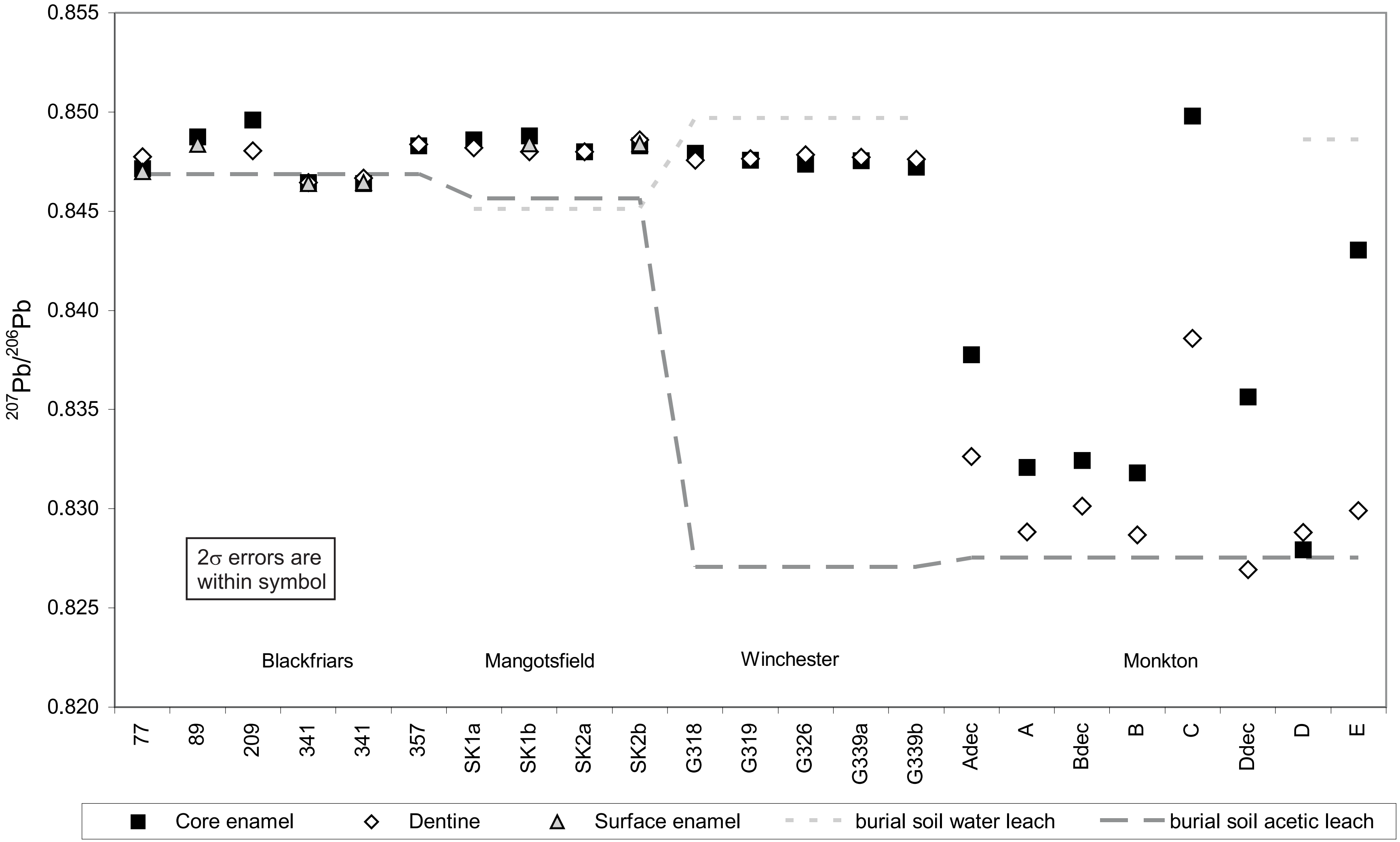


Figure 6.23 Plot of ^{207}Pb/^{206}Pb ratios of intra-tooth tissues for all archaeological pilot studies. Note the restricted variability in the Mediaeval and Late-Roman samples from Blackfriars, Mangotsfield and Winchester when compared with the Neolithic samples from Monkton.

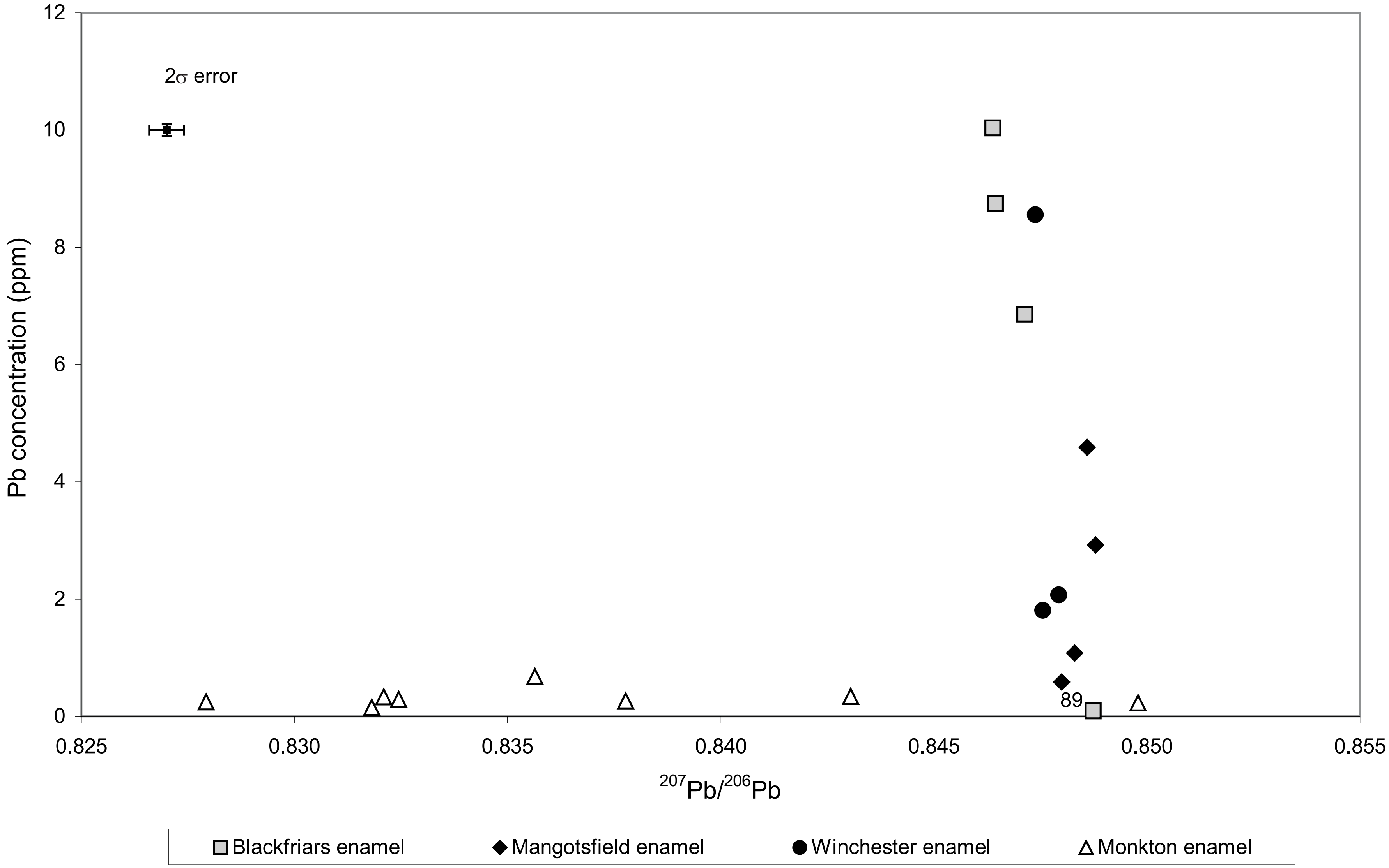


Figure 6.24 Plot of ^{207}Pb/^{206}Pb versus Pb concentration for all archaeological pilot studies. Note that Pb concentrations >1ppm are only present at a restricted range of ^{207}Pb/^{206}Pb ratios. All Neolithic samples (Monkton) have Pb concentrations <1ppm. The Pb-coffin burials from Spitalfields (0.851, 30.1ppm) and Winchester G339 (0.848, 41.8ppm and 0.847, 1540ppm) are not plotted.

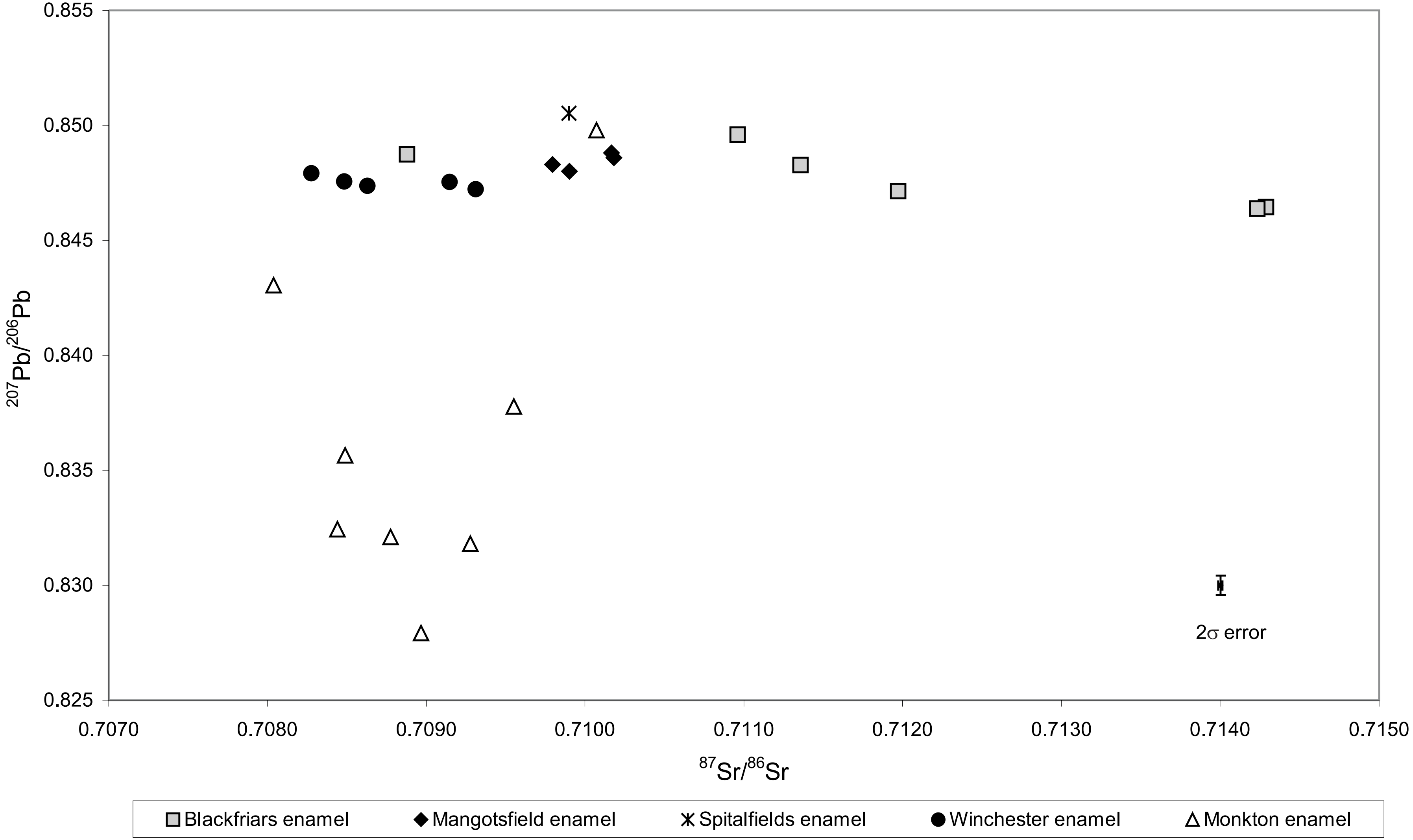


Figure 6.25 Plot of ^{87}Sr/^{86}Sr ratio versus ^{207}Pb/^{206}Pb ratio for all archaeological pilot studies. Note that variable ^{87}Sr/^{86}Sr ratios are seen in samples with a restricted range of ^{207}Pb/^{206}Pb ratios, whereas samples with variable ^{207}Pb/^{206}Pb ratio have a restricted range of ^{87}Sr/^{86}Sr ratios.

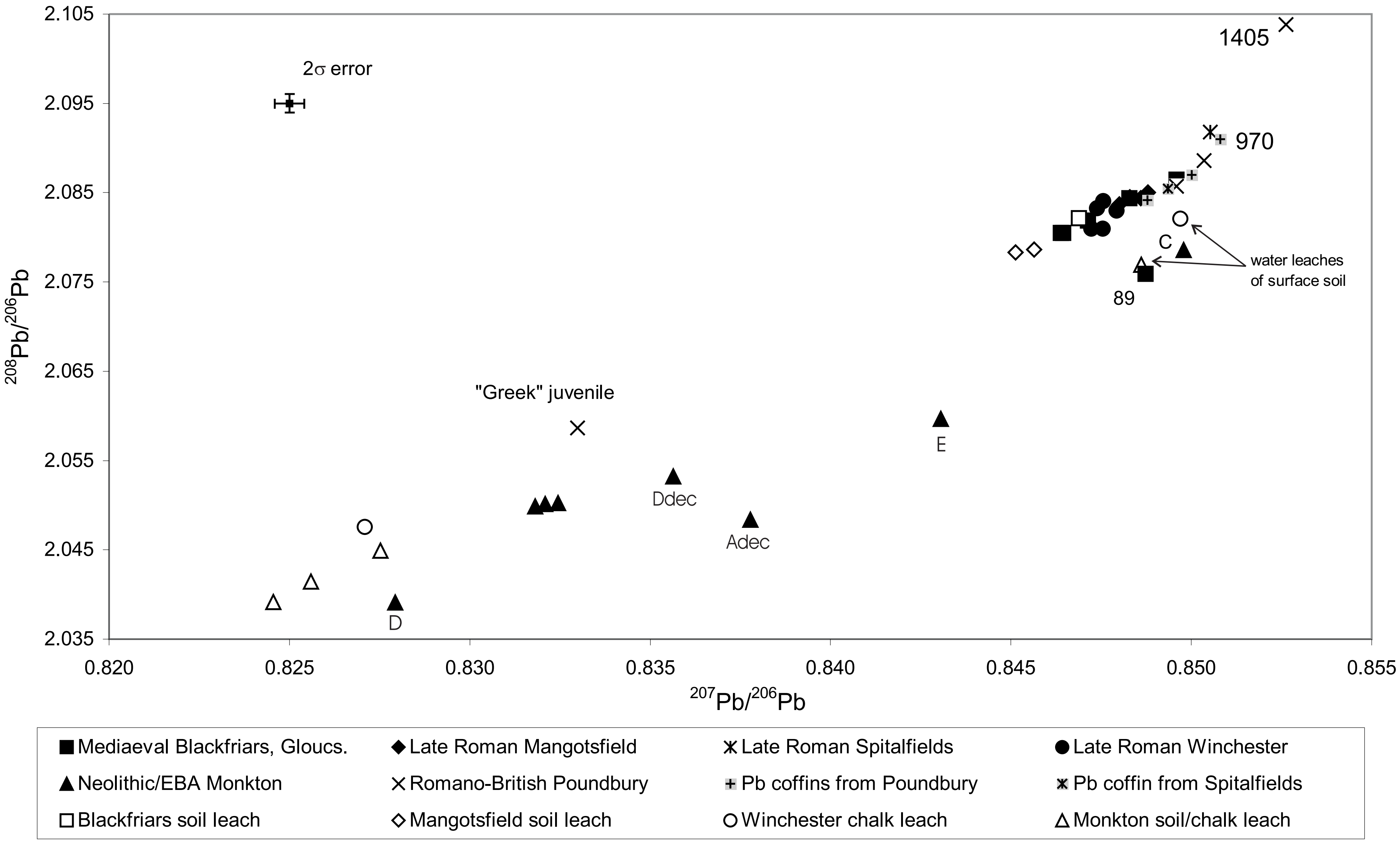


Figure 6.26 Plot of ^{207}Pb/^{206}Pb v ^{208}Pb/^{206}Pb ratios for all archaeological pilot studies.