Human tooth enamel as a record of the comparative lead exposure of prehistoric and modern people

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Abstract
There is a considerable body of evidence to support the contention that the atmospheric Pb burden is now considerably greater than it was in the remote past. However, as there are a diversity of potential environmental pathways leading to Pb ingestion, it is not clear how atmospheric Pb levels relate to human exposure. It is necessary to establish a baseline for human exposure to Pb from natural sources in the pre-metallurgical past, with which contemporary exposure can be compared. This paper addresses this issue by comparing the Pb content of human dental enamel - an established proxy for Pb exposure - from modern and archaeological, pre-metallurgical individuals using thermal and plasma ionisation mass spectrometry. It is shown that mean Neolithic enamel Pb contents are ~0.31±0.04ppm. These values are only one order of magnitude lower than previously reported data for the same tissues for modern juveniles, despite an established 400 fold increase in the atmospheric Pb burden. The results suggest that 'natural' exposure to Pb in food and water may have been higher than previously thought and that the link between atmospheric Pb and human exposure warrants further investigation.

Keywords: Human dental enamel, teeth, lead, archaeological, human exposure, ID-TIMS, PIMMS.

1. Introduction

Atmospheric lead pollution is a global phenomenon. It is widely believed that the contamination of the atmosphere by anthropogenic lead has led to far greater human exposure today than that which prevailed in the distant past, but this has proved difficult to quantify. The analysis of Arctic ice for time series data on lead deposition has revealed a four hundred fold increase in the atmospheric lead burden in the Northern Hemisphere between the prehistoric period and the mid 1960s (Hong et al. 1994). These conclusions are supported by similar data derived from independent archives of atmospheric lead preserved within European lake sediments (Renberg et al., 1994) and peat bogs (Martínez-Cortizas et al., 1997; Shotyk et al., 1998). These, and previous similar observations, led to widespread concern regarding the potential health effects of elevated atmospheric lead (Patterson et al. 1987). However, as there are a diversity of potential environmental pathways leading to lead ingestion, human exposure may not necessarily be directly related to the atmospheric burden. The need arises to establish a baseline for human exposure to lead from natural sources in the pre-metallurgical past with which contemporary exposure can be compared.

2. Lead exposure estimates from archaeological bone

A number of estimates of early human lead exposure have been made from the analysis of skeletal material recovered by archaeological excavation and thought to preserve lead acquired in vivo. Early measurements produced a wide range of values, some considerably higher than those of modern individuals, which led to speculation regarding the effect of lead on morbidity and mortality (Mackie et al., 1975; Waldron et al., 1976; 1979; Waldron & Wells, 1979; Whittaker & Stack, 1984). Later studies confirmed this pattern, reflecting the diversity of sources of exposure in societies with differing access to metals and little understanding of their potential health effects. We have elsewhere reported tooth analyses for Romano-British and medieval individuals from England which suggest highly variable exposures (Budd et al., 1998). Similarly, Åberg et al. (1998) analysed medieval samples from Norway and found their lead concentrations mostly similar to those of modern people, but with the exception of one tooth which was about fifty times higher than the mean.

Although, Vuorinen et al. (1996) analysed a 16th-17th century population in Finland and concluded that early modern lead exposure there was minimal, other studies of skeletal lead in later historic individuals have been taken to suggest exposures greater than those of today. Two studies of 17th-18th century colonial settlements produced skeletal lead levels taken to indicate exposures up to an order of magnitude higher than modern Americans (Aufderheide et al., 1981; Corruccini et al., 1987). High lead levels have also been recorded in 18th-19th century native American skeletons, where they are thought to be a result of the use of lead-based pigments (Reinhard & Ghazi, 1992; Ghazi et al. 1994), and in the bones of 19th century British sailors (Kowal et al. 1989; Keenleyside et al., 1996).

Whereas skeletal evidence for more recent, historic patterns of lead exposure suggest considerable variability, a number of analyses of samples for earlier, pre-metallurgical populations from various parts of the world have been undertaken which appear more consistent (see Table 1). Pre-metallurgical skeletal lead levels have generally (although not exclusively) been found to be lower than classical, medieval and later historical populations. However, the reported bone lead levels range over more than two orders of magnitude, up to values which would be considered normal for modern people.

3. Diagenetic accumulation of lead in archaeological bone

This large variation in bone lead between the archaeological individuals and groups investigated is almost certain to be due, at least in part, to diagenesis in the burial environment (Hedges and van Klinken 1995). The biogenic apatite which forms the mineral component of bone is known to sequester various metals from aqueous phases within the soil during burial including strontium and barium (Nelson et al., 1986; Pate et al., 1991), uranium (Millard and Hedges, 1996) and lead (Waldron, 1983; Lower et al. 1998a; 1998b). Diagenesis has also proved a major obstacle for researchers attempting to measure the isotopic composition of strontium from buried material so that sample pre-treatments have had to be devised to remove diagenetic apatite prior to analysis (Sealy et al., 1991; Price et al., 1992; Sillen and Sealy, 1995; cf. Budd et al., 2000).

Some researchers have attempted to monitor and correct for the post-mortem diagenetic accumulation of lead by measuring the Ba/Ca ratio of bone samples (Ericson 1993; Ericson et al., 1979; Patterson et al. 1987; 1991). This approach led Patterson et al. (1991) to reject data from one of their case studies as "obscured by excessive diagenetic additions of Pb" and to adjust their estimate of biogenic lead in the other to one third of the measured value. The Ba/Ca ratio approach is based on the proposition that both Pb^{2+} and Ba^{2+} migrate from aqueous phases in the soil and accumulate in buried bone in a similar way. However recent studies of aqueous lead sorption by hydroxylapatite (Lower et al. 1998b) cast doubt on this.

Analytical studies of archaeological bone in which diagenesis has not been considered are highly unlikely to yield accurate data for biogenic lead. Results from such studies should be disregarded. Even where attempts have been made to account for diagenesis, bone analysis remains problematic (Budd et al., 2000).

4. Lead exposure estimates from tooth enamel

The problem of diagenetic lead accumulation is best minimised by careful sample selection. Dental enamel is the sample material of choice in this respect as its highly mineralised and dense structure make it particularly resistant to diagenesis (Hillson 1996, 181). Dental tissues are considered a reliable reservoir of biogenic lead and have been used widely to reconstruct exposure histories among modern populations, most recently with the addition of source information by lead isotope ratio measurement (Gulson, 1996; Gulson and Wilson, 1994; Gulson et al., 1994; 1997).

Fairly plentiful data are available for contemporary human tooth lead in the UK, although some studies have involved dentine or whole tooth samples rather than exclusively enamel. As dentine is at risk of similar diagenesis to that of bone in archaeological burial, detailed consideration is restricted here to specific measurements of enamel after the extraction of whole crowns or other forms of tissue separation.

A large scale study of the lead content of modern human enamel in UK children was undertaken by Delves et al. (1982) and three subsequent studies have been published (Alexander et al. 1993; Farmer et al., 1994; Lane and Duffy, 1996). The data are summarised in Table 2. The data are relatively consistent and comparable with recent ID-TIMS data which give a mean of 3.8 ppm for lead in the enamel of modern Australian immigrants from sixteen countries world wide (Gulson and Gillings, 1997; Gulson et al. 1997). UK enamel lead burdens measured by Lane and Duffy (1996) appear to be five to ten times higher than other populations without a specific history of high exposure. This is probably a result of the use of a surface sampling technique (PIXE) by these workers. Surface enamel is known to be considerably elevated in lead compared with the bulk tissue (Brudevold & Steadman 1956; Budd et al. 1998), probably as a result of chemical exchange in the oral environment.

Comparatively, very few tooth enamel samples have been analysed for pre-metallurgical individuals world wide, but the available data are summarised in Table 3. As expected, in view of the relative stability of the tissue, there is considerably less variation between the three groups represented than indicated by corresponding bone data. Here we present the results of a new study of tooth enamel lead from a group of four prehistoric burials recently excavated in southern England with the aim of providing accurate base line exposure information for a pre-metallurgical, early agricultural population subsisting in an area with a low-lead geology.

5. Methods

5.1 Samples

Teeth from four prehistoric individuals were selected for analysis. The burials were excavated by Martin Greene in 1997 on farmland near the village of Monkton-up-Wimbourne, Dorset, England. The site is dug into the cretaceous chalk of Cranborne Chase and the skeletons have been radiocarnon dated within the Neolithic period (5500-5100 cal BP). The four individuals, were buried in a single event at the time of initial construction of the site. Macroscopic skeletal preservation was excellent (J. McKinley pers.comm.). Permanent and where present, deciduous tooth samples were taken from all four individuals: a young-middle aged adult female and three juveniles aged 5, 8-9 and 9-10 years.

The individuals lived about a thousand years before the advent of metallurgy in the British Isles. As early agriculturalists, it is also considered unlikely that they imported significant quantities of foodstuffs over large distances. We believe that they are likely to have had lead exposures in vivo representative of the 'natural' background for southern England. Lead available to the skeleton post-mortem would also be restricted due to the exceptional purity and virtual absence of sedimentary inclusions of the cretaceous chalk. Anthropogenic and diagentically derived lead should therefore both be minimal.

The teeth (Table 4) were cleaned ultrasonically for 5 minutes in Millipore Alpha Q H_2O (< 1ppb total heavy metal content) and then high-purity acetone (< 5ppb Pb and Sr). Each tooth was then divided longitudinally using a flexible diamond edged stainless steel rotary dental saw to produce half-tooth samples for mass spectrometry. Samples were then cleaned with a tungsten carbide dental bur to remove all traces of adhering dentine. The enamel

was also abraded from the surface to a depth of >100μm using the bur and the surface material discarded to produce a sample of core enamel tissue. Although the enamel surface lead enrichment may be physiological (Budd et al. 1998), it was removed in order to maximise the probability that the sample analysed contained only biogenic lead and to give the lowest estimate of exposure compared with those reported for modern individuals. Clean core enamel samples were then sealed in acid-leached Teflon (Savillex) beakers and samples were transferred to a clean (class 100, laminar flow) working area at NIGL for further preparation.

Samples of both the underlying chalk and soil excavated from the shaft on site were collected for lead isotope analysis by PIMMS to assess the isotopic composition of lead available to aqueous phases in the burial environment. Aqueous leaches failed to produce sufficient lead for analysis. Approximately 2g of chalk was isolated from the centre of a large block taken from the site in the low-lead laboratory, ground in an agate mortar and pestle and placed in an acid-leached Teflon (Savillex) beaker. A finely divided soil sample of similar size was placed in a second beaker. Both samples were then leached overnight at room temperature in 5ml of 0.1M Teflon-distilled acetic acid and the leachates removed for analysis.

5.2 Plasma Ionisation Multi-collector Mass Spectrometry (PIMMS)

Samples were first cleaned ultrasonically in Millipore Alpha Q H_2O for 5 minutes to remove dust, rinsed twice, dried down in high-purity acetone and then weighed into pre-cleaned Teflon (Savillex) beakers. Solid samples were dissolved in 1ml 16M Teflon-distilled HNO_3 and an aliquot of 100μl of this solution was transferred to a second pre-cleaned Teflon (Savillex) beaker for ID-TIMS analysis. Lead separation was performed on the remaining solution by conventional anion exchange column chemistry. Lead isotope analyses were carried out on the VG Elemental P54 instrument at NIGL, Keyworth. Data were corrected for mass discrimination using the Tl-spiking technique (Ketterer et al., 1991; Walder et al., 1993). Lead isotope ratio errors were calculated from replicate analyses of the NBS981 standard as 0.011% for $^{207}Pb/^{206}Pb$ and 0.014% for $^{208}Pb/^{206}Pb$ (2σ, n=4).

5.3 Isotope Dilution - Thermal Ionisation Mass Spectrometry (ID-TIMS)

Aliquots of the sample solution were spiked with a known volume of ^{208}Pb-enriched solution and lead separation performed by anion exchange column chemistry. The overall mean procedural lead blank was 122 ± 30pg (1σ, n=3). Lead concentrations were determined by ID-TIMS using a silica gel-phosphoric acid emittor on Re filaments. Analyses were carried out on the Finnigan MAT262 instrument at NIGL, Keyworth. Data were corrected for mass fractionation which was monitored by repeated analyses of the NBS 981 standard from which the ratio measurement error was estimated as 0.07% (2σ) for $^{208}Pb/^{204}Pb$.

Isotope dilution accuracy can exceed 1% (2δ) for homogenous samples in favourable circumstances (Dickin 1995), but dental tissues have not been well characterised with respect to compositional heterogeneity. Previous LA-ICP-MS studies suggest a degree of Pb and Sr variation within tooth enamel (Budd et al. 1998, Montgomery et al. 1999). Estimates of reproducibility for the current study are based on three replicate tooth enamel measurements undertaken for both a permanent and deciduous tooth. These suggest errors of ±14% (2δ, n=3).

6. Results and Discussion

Lead concentration and isotope ratio data for core tooth enamel from the four Neolithic individuals are presented in Table 4. The lead isotope ratio data are also plotted in Figure 1 to illustrate differences between the enamel values and those of the chalk and soil leachates. Lead isotope ratios are presented as $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$ as these ratios can be measured with the greatest accuracy. Tooth enamel lead concentrations determined by ID-TIMS range from 0.15 to 0.68ppm (dry weight) giving a mean concentration of 0.31ppm.

Small differences in the isotopic composition of the chalk and soil leachates suggest that some variability may be expected for soil derived lead depending on those soil fractions contributing to aqueous phases which might be expected to contribute to diagenesis. Nevertheless, only one of the tooth samples (D_{perm}) plots in this area of the graph and even this has an isotopic composition significantly different to either of the leachates. All of the remaining tooth samples gave lead isotope ratios significantly more radiogenic that those of either soil or chalk leachates and clearly separable in $^{207}Pb/^{206}Pb$ vs. $^{208}Pb/^{206}Pb$ space. The adult (C) has an enamel lead isotope composition very different from that of the Monkton geology. Although for one of the juveniles (B), deciduous and permanent tooth enamel lead has a similar isotopic composition, for the other two it is significantly different. This suggests the preservation of lead ingested in vivo at different stages of life from sources other than the local Monkton geology. The wide range of tooth enamel lead isotope compositions and clear differences between them and soil derived lead from the site demonstrate the preservation of biogenic lead and make it highly unlikely that the tissue was subject to diagenesis on a scale which would cast doubt on the concentration data.

We believe that our estimate of the mean tooth enamel lead concentration for these four Neolithic individuals of ~0.3ppm is a reasonably accurate estimate of biogenic lead ingested in vivo which can be compared with previous studies of pre-metallurgical populations and with data for modern individuals. Given that surface enamel is known to be elevated in Pb in vivo (Budd et al. 1998) and that this was removed by us, but was not in most studies of modern people, our results are likely to underestimate rather than overestimate the exposures of the archaeological relative to the modern population.

Our measurements of lead concentration in prehistoric enamel for individuals in southern England are about twice those reported by Patterson et al. (1991) for pre-contact Arizona and almost three times those reported by Ericson et al. (1979) for pre-Columbian Peru (Table 3). They are an order of magnitude higher than those reported by Patterson et al. (1991) for pre-contact Malibu. This degree of variation may reflect true differences in lead exposure depending on the local environment. Whereas the Monkton individuals were buried in, and presumably

living on, a very low-lead geology, it appears likely that they were not sedentary and had spent part of their lives elsewhere exposed to more radiogenic lead of different geological origin.

The four Neolithic individuals from southern England that we have examined had tooth enamel lead concentrations about one order of magnitude lower than those reported by Delves et al. (1982) for the contemporary UK population. They are similarly different to the mean value for modern Australian immigrants recently surveyed by ID-TIMS (Gulson and Gillings, 1997; Gulson et al. 1997). We conclude that prehistoric human lead exposure in southern England was no more than about one order of magnitude lower than that of modern people in the same area and not two to three orders of magnitude as previously proposed for pre-metallurgical individuals elsewhere (Ericson et al. 1979; Patterson et al. 1987; 1991). These data, suggesting human lead exposure was about one order of magnitude lower than it is today, is at variance with the proxy records for the atmospheric lead burden which is thought to have increased 400 fold (Hong et al. 1994; Renberg et al., 1994;Martínez-Cortizas et al., 1997; Shotyk et al., 1998). The discrepancy suggests that the dominant source of lead to which prehistoric people were exposed was geologically derived and ingested via the food chain. Records of past atmospheric lead pollution may thus not be used directly to estimate human exposure.

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Tables

Table 1. Summary of data from studies of the lead concentration of bone from pre-metallurgical individuals from various locations world wide. Techniques used are: AAS, Atomic Absorption Spectroscopy; ICP-AES, Inductively Coupled Plasma - Atomic Emission Spectroscopy; ID-TIMS, Isotope Dilution - Thermal Ionisation Mass Spectrometry.
Notes: *median values; †estimated sensitivity; ‡these data are for femur, rib data are also reported.

Country	Mean Pb (ppm)	SD	n	Technique	Reference
Croatia	0.24	0.11	9	ICP-AES	Kniewald et al., 1994
Denmark	<0.2	-	17	AAS	Grandjean, 1988
France	1.5	-	1	AAS	Jaworowski et al., 1985
France	<0.1	-	3	ID-TIMS	Jaworowski et al., 1985
Greenland	0.71*	-	3	AAS	Grandjean & Jørgensen, 1990
Japan	1.2	-	67	ICP-AES	Kosugi et al., 1986
Mexico	<2†	-	16	PIXE	Solis et al., 1996
Egypt (Nubia)	2.0*	-	9	AAS	Grandjean & Jørgensen, 1990
Peru	0.4	0.4	2	ID-TIMS	Ericson et al., 1979
Tenerife	4.12	2.38	15	AAS	ArnayDeLaRosa, 1998
UK	14.3	4.7	11	AAS	Rogers & Waldron, 1985
USA (Arizona)	0.06‡	0.02	10	ID-TIMS	Patterson et al., 1991
USA (California)	0.27‡	0.16	13	ID-TIMS	Patterson et al., 1991

Table 2. Tooth enamel lead concentrations for contemporary UK individuals. The 'high exposure' group are children known to have suffered a high environmental lead exposure. Concentrations are by weight in dry tissue unless otherwise stated.
Notes: * Concentrations in ashed tissue.

Population Group	**n**	**Mean Pb Concentration (ppm)**	**Technique**	**Reference**
Non-industrial	12	4.1	AAS	Delves et al. 1982
High exposure	20	16.4	AAS	Delves et al. 1982
Enfield	1243	4.7*	AAS	Delves et al. 1982
Hounslow	235	5.6*	AAS	Delves et al. 1982
Hounslow	160	4.3*	AAS	Delves et al. 1982
London	130	4.1	AAS	Delves et al. 1982
London	65	4.2	AAS	Delves et al. 1982
Reading	52	4.1*	AAS	Delves et al. 1982
Reading	31	4.8*	AAS	Delves et al. 1982
Birmingham	70	4.8*	AAS	Delves et al. 1982
Hammersmith	22	5.3*	AAS	Delves et al. 1982
Hammersmith	14	5.7*	AAS	Delves et al. 1982
Manchester	11	6.3*	AAS	Delves et al. 1982
Berks./Hants. (rural)	103	3.6	AAS	Delves et al. 1982
Blackpool (Lancs.)	66	3.9	ICP-MS	Alexander et al. 1993
Cleveleys (Lancs.)	10	1.9	ICP-MS	Alexander et al. 1993
Gleetwood (Lancs.)	56	2.9	ICP-MS	Alexander et al. 1993
Garstang (Lancs.)	46	2.1	ICP-MS	Alexander et al. 1993
Other UK	59	2.3	ICP-MS	Alexander et al. 1993
Edinburgh	3	3.8	GF-AAS	Farmer et al. 1994
Oxfordshire	97	27.2	PIXE	Lane and Duffy 1996

Table 3. Tooth enamel lead concentrations for pre-metallurgical individuals. Concentrations are by weight in dry tissue.

Population Group	**n**	**Mean Pb Concentration (ppm)**	**Technique**	**Reference**
US, Malibu	10	0.039	ID-TIMS	Patterson et al. 1991
US, Arizona	12	0.147	ID-TIMS	Patterson et al. 1991
Peru	4	0.11	ID-TIMS	Ericson et al. 1979

Table 4. Lead concentration and isotope ratio data for the adult female (C) and three juveniles (A, B and D) from Monkton with lead isotope ratio data for the chalk bedrock and soil leachates from the site. The suffixes 'perm' and 'decid' refer to permanent and deciduous teeth respectively. Uncertainties for the ID-TIMS concentration data are ±14% (2σ, n=3) and for the ratios, 0.011% for $^{207}Pb/^{206}Pb$ and 0.014% for $^{208}Pb/^{206}Pb$ (2σ, n=4).

Sample	**Tooth**	**ID-TIMS Pb concentration (µg g^{-1} in solid tissue)**	**PIMMS**	
			$^{207}Pb/^{206}Pb$	**$^{208}Pb/^{206}Pb$**
A_{decid}	c^1right	0.26 ± 0.04	0.8378	2.0484
A_{perm}	M^1right	0.33 ± 0.05	0.8321	2.0501
B_{decid}	c^1left	0.29 ± 0.04	0.8324	2.0502
B_{perm}	P^1left	0.15 ± 0.02	0.8318	2.0499
C_{perm}	P^2right	0.23 ± 0.03	0.8498	2.0786
D_{decid}	c^1left	0.68 ± 0.10	0.8356	2.0532
D_{perm}	C^1left	0.25 ± 0.04	0.8279	2.0391
Chalk leachate	-	-	0.8275	2.0449
Soil leachate	-	-	0.8245	2.0391
Mean		**0.31 ± 0.04**		

Figures

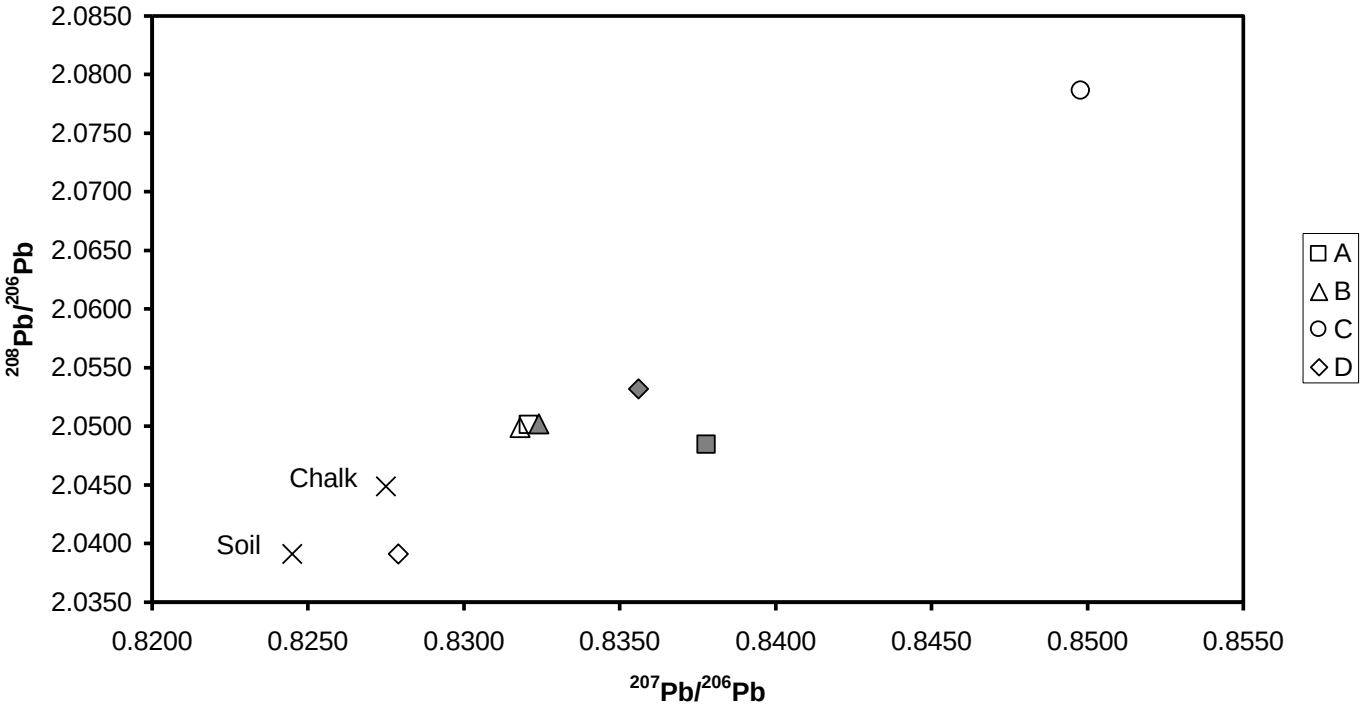

Figure 1
Lead isotope ratio data for the four Neolithic individuals. Deciduous teeth (filled symbols) are plotted with permanent teeth (open symbols) and leachates of soil and chalk from the site. Errors (2σ) are smaller than the symbols on this scale.