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Biologically Available Pb: A Method for Ancient Human Sourcing Using Pb Isotopes from Prehistoric Animal Tooth Enamel

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Abstract: This study analyzes Pb isotopes combining biological (ancient human and prehistoric animal teeth) and geological (soil leachate, whole rock, and rock leachate) samples to determine the origins of prehistoric skeletal elements. It exemplifies how the biologically available Pb method assesses the early lifetime locations of ancient human populations using prehistoric animal teeth and the multivariate/linear nature of Pb isotope data. Lead isotopes provide a valuable technique, in part due to the correlation between their six stable isotope ratios. Other studies have used Pb isotopes for similar purposes, but no clear method for determining a local range has yet been formally defined and tested. The biologically available Pb method uses many prehistoric animal tooth enamel samples to establish a baseline for local ratios in the region, then compares their ratios' linear patterning to human remains to test if they are non-local. The case study compares Pb isotopes from prehistoric animal teeth, human teeth, and whole rocks from southwest Arkansas. These results are compared to animal samples from Louisiana and Mississippi and human data from Illinois and New Mexico. Soil leachates, Pb concentrations of tooth enamel, and trace element analysis are used to assess contamination. Comparisons to southwest Arkansas whole rock Pb isotope ratios suggest they are too variable to be used for direct comparison to ancient human remains, illustrating that prehistoric animal teeth are more appropriate for direct comparison to prehistoric human teeth. The biologically available Pb method provides a key analysis tool needed for studies of ancient human sourcing.

Keywords: Pb; isotopes; sourcing; Sr; Caddo; warfare; contamination

1. Introduction

Many studies use strontium (Sr) isotope ratios to assess ancient human geographic origins by establishing a range of expected local ratios based on the content of archaeologically recovered animal tooth enamel. While this technique is effective in many places, it is clear that in some regions Sr isotope ratios are uniform across large areas. This is particularly true in the midcontinental US (Hedman et al. 2018). Having wide ranges of commonly represented ratios at the locality where human remains are found makes ancient human sourcing difficult, if not impossible, with this isotopic technique alone. Researchers have suggested that using multiple isotope ratios in combination, including Sr and lead (Pb), will lead to clearer results, allowing for better interpretations about human origins (Kamenov and Curtis 2017). Pb isotope analysis is commonly used in geologic studies (e.g. Crocetti et al. 1988; Goldhaber et al. 1995; Potra et al. 2018a, 2018b) and provides a multivariate isotopic dataset which (when combined with other elements like Sr) has the potential to be more sensitive to regional differences than Sr alone.

Pb isotope analysis has been relatively underutilized in studies of ancient human geographic origins in part due to the difficulty in getting sufficient concentrations of Pb from tooth enamel, complications associated with modern Pb pollution, and difficulty with some instrumentation (e.g. Gulson et al. 2018). Studies employing the technique for migration and sourcing studies have greatly increased in the last few years as methods and instrumentation for extraction and analysis improve (Dudás et al. 2016; Giovas et al. 2016; Grupe et al. 2018; Jones et al. 2017; Price et al. 2017; Sharpe et al. 2016; Turner et al. 2009; Valentine et al. 2008, 2015). Nonetheless, unlike the biologically available Sr method (Bentley 2006; Price et al. 2002), a method to use Pb isotopes for assessing ancient geographic origins has yet to be formally defined and demonstrated (Grupe et al. 2017). Studies often add this technique as part of a suite of elements included in multi-isotope migration and sourcing studies where little time or space is given to explicitly outlining and evaluating the effectiveness of methods used to construct Pb isotope backgrounds. This has led to the basic underpinnings of the methods generally being overlooked or assumed, in part due to its inferred similarity to the much better developed Sr isotope technique. However, some researchers have put great effort into developing the technique further (e.g. Dudás et al. 2016; Jones et al. 2017; Kamenov et al. 2018; Sharpe et al. 2016). This study aims to develop and evaluate the effectiveness of one method to establish a local background with Pb isotopes for the purpose of assessing if ancient human populations are local or non-local to a region.

First, this study outlines a clear biologically available Pb method of analysis for ancient human sourcing utilizing the multivariate and linear nature of Pb isotope data from prehistoric animal teeth. This method is demonstrated and evaluated for effectiveness through a case study of ancient human and non-migratory prehistoric animal remains from the southcentral US. Key elements of this analysis include contrasting linear differences between Pb isotope ratios from different regions and using animal samples from multiple sites as a baseline for comparison to incorporate isotopic variation within the regions being compared. Second, the Pb isotope ratios of human and animal teeth are compared to those of whole rocks, rock leachates, and soil leachates to assess the utility of prehistoric animal teeth and geologic samples, particularly whole rocks, for constructing backgrounds. Finally, the potential for evaluating contamination of tooth enamel using soil leachates, Pb concentrations of tooth enamel, and trace element analysis (Dudás et al. 2016; Kamenov et al. 2018) is discussed alongside an assessment of anthropogenic Pb contamination of the burial environment.

1.1 Pb/Sr Isotope Ratios and Geographic Origins

1.1.1 Isotopic Techniques

Sr isotope analysis is a technique commonly utilized in archaeology to determine whether people are local or non-local to a geographic area (Bentley 2006; Bentley and Knipper 2005; Bentley et al. 2004; Buzon and Simonetti 2013; Chenery et al. 2010; Eerkens et al. 2016; Hedman et al. 2009, 2018; Price et al. 1994; Price et al. 2002; Slater et al. 2014; Slovak and Paytan 2011; Thornton 2011). Pb isotope analysis is relatively underutilized but has great potential (Kamenov and Gulson 2014). Pb and Sr are trace elements found in soil, bedrock, water, plants, and animals. Plants absorb these elements from the local geology where they grow. People and animals, in turn, absorb Pb and Sr from the plants and animals they eat. Due to the mechanism of Sr absorption in the stomach, plants are the dominant source of Sr for herbivores and omnivores (Price et al. 1985). Lemons and Kennington (1983) have shown that Pb is even more severely discriminated against than Sr.

There are four naturally occurring Pb isotopes (^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb). The lightest of these, ^{204}Pb, is non-radiogenic while ^{206}Pb, ^{207}Pb, and ^{208}Pb are radiogenic and represent decay products of ^{238}U, ^{235}U, and ^{232}Th, respectively (Dickin 2005; Faure and Mensing 2004; Malainey 2011). Therefore, the radiogenic Pb isotopes are affected by the geologic age of bedrock. These isotopes increase in abundance relative to ^{204}Pb as the deposits age. Depending on the original concentrations of U, Th, and Pb in the local geology, they will have different abundances of radiogenic Pb isotopes, potentially leading to highly sensitive differences in Pb isotope ratios in different areas.

1.1.2 Pb Isotopes and Human Sourcing: Constructing a Local Range

Recent studies have shown that Pb isotopes are revolutionizing biological sourcing research since they result in a multi-dimensional dataset that allows for distinguishing different geographic regions (Kamenov and Curtis 2017; Kamenov and Gulson 2014). Despite the potential for environmental contaminants, Kamenov and Curtis (2017) show that Pb is extremely useful at delineating human remains from different regions, even among modern European populations, which is a strong endorsement of the technique.

Most studies combine Pb isotopes in multi-isotope evaluations of ancient human remains (e.g. Jones et al. 2017; Price et al. 2017; Turner et al 2009; Valentine et al. 2015). Some of these do not attempt to establish if the human remains are local or non-local to a region and instead focus on other questions. Those that do attempt to find non-locals each use a different method to construct an isotopic background and generally evaluate the remains by comparing to known local humans, a few animal samples, or geologic data (e.g. Jones et al. 2017; Sharpe et al. 2016; Valentine et al. 2015).

Direct measurements of soils, bedrock, and water provide an approximation of the isotopic ratios of the geologic source material. These can be highly variable. However, humans and animals amalgamate the Sr isotopes consumed as part of their diet, decreasing the range of local isotopic ratios compared to that of the source materials. Since human and animal isotopic ratios can be expected to be much more similar to each other than to rocks, it increases the value of human to animal comparisons. This has been well established for Sr and forms the basis of the biologically available Sr method (Bentley 2006; Price et al. 2002; Sillen et al. 1998; Slovak and Paytan 2011). There is some disagreement on what samples are most appropriate for defining a local range (Grimstead et al. 2017). While some researchers concentrate on animal samples for defining Sr local ranges (Bentley 2006; Hedman et al. 2009, 2018; Price et al. 2002; Samuelsen 2016), others focus on geologic data or groundwater (e.g. Evans et al. 2010; Hodell et al. 2004).

It is unclear if the Pb isotopes behave similarly. There is a need for Pb isotope studies that combine ancient human tooth enamel, many samples of prehistoric animal tooth enamel, and geologic data from the same region to better understand how these different classes of data can be compared. Grupe et al. (2018) sampled many animal bones (not tooth enamel) and human remains but did not include a comparison to geologic data. Dudás et al. (2016:Figure 9a) showed that the geologic data in surrounding areas was much more variable than the ancient human tooth enamel, but did not include any prehistoric animals for comparison. Giovas et al. (2016) sampled geologic data and both modern and prehistoric animals. They noted great differences between geologic and animal data and suggested that the animal teeth may have been contaminated by anthropogenic Pb. However, they did not include any human remains and most problematic animals were modern samples, which would be more likely to be impacted by anthropogenic Pb. Sharpe et al. (2016), similarly to Hodell et al. (2004) with Sr, constructed a Pb baseline using mostly whole rocks with a few soil, plant, and rock leachate samples, but did not test the baseline with ancient human remains to evaluate its effectiveness. This study attempts to build on this research by combining Pb isotope data from ancient human teeth, prehistoric animal teeth, soil leachates, whole rocks, and rock leachates from a single region to determine the origins of prehistoric skeletal elements. Sampling for constructing a background focuses on prehistoric animal teeth and whole rocks, with limited comparisons to ancient human teeth, soil leachates, and rock leachates.

This study examines whether the biologically available Sr method similarly applies to Pb isotope studies. This is accomplished by obtaining many prehistoric (non-migratory) animal tooth enamel samples from a region and comparing them to ancient human remains to determine if (1) the Pb signatures of animals match those of expected local human remains, (2) the resulting Pb isotope ratios of animals are capable of differentiating humans from other regions, and (3) the use of animals for establishing a local background is more effective than using only expected local human remains. Rather than defining a local range with a formula like +/- two times the standard deviation of the mean, the biologically available Pb method (as defined here) relies heavily on the differentially linear (correlative) and multivariate nature of Pb isotopes to differentiate regions. It is recommended that all six unique ratios be analyzed for linear patterning ($^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$, $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$, and $^{208}Pb/^{207}Pb$), which generates 15 unique bivariate comparisons (Figure 1). Data with linear patterns yielding different slopes may be generated by regional variability, multiple end-members, or "pseudo-isochron" dynamics (Jones et al. 2017). There is also a possibility that linear patterning could be created through in-vivo signatures getting mixed with contaminants, such as anthropogenic Pb. Regardless of the individual causes for these linear patterns defined by the Pb isotope data (assuming it is not contamination), the linear regression lines of various groups reflect the Pb signatures of the region and can therefore be used as a fingerprint to assess origins using bivariate comparisons (or multivariate comparisons, such as a principal component analysis). Thus, even if the isotopic signature of some individuals within a specific group overlap in more than one region, the slope of the regression line combined with all the isotopic values in that specific group can aid in distinguishing it from the isotopic signature of another region. It is suggested that multiple sites within a study area be sampled with multiple animal samples from each site to account for potential intra-site and inter-site variation in Pb isotopes. Once the isotopic signature of an area is defined, the human remains can be confidently compared with the background value of that area. The humans whose isotopic ratios do not match the background value should be considered non-local.

In order for human and animal teeth to be compared, differences in how they incorporate Pb need to be considered. Studies have shown that modern animals can ingest significant amounts of soil (and soil Pb), but these studies generally concentrate on modern, anthropogenic Pb contaminated environments (e.g. Johnsen and Aaneby 2019; Johnsen et al. 2019; Thornton and Abrahams 1983). In environments with heavily contaminated soils, as much as 97% of the absorbed Pb can be due to soil ingestion (Abrahams and Steigmajer 2003); however, as much as 60% can still be attributed to plant consumption in lesser contaminated environments (Thornton and Abrahams 1983). These studies also tend to investigate pasture animals that are restricted in location and consume in areas with short grass, potentially increasing root and soil ingestion. Durkalec et al. (2015) found that wild deer and bore absorbed significantly less Pb in a modern, uncontaminated environment. This is particularly important when estimating the relative contributions of plant and soil Pb as Yan et al. (2012) showed that plants may absorb less Pb relative to soil Pb as soil Pb concentrations increase. Their findings are consistent with plant Pb absorption from nutrient solutions which show that plant Pb concentrations can scale well below a 1:1 rate with increasing soil Pb concentrations (Kabata-Pendias 2011:Figure 19.4). This suggests that plant Pb could contribute significantly more Pb to animals relative to soil Pb in uncontaminated environments.

Two factors, (1) modern anthropogenic Pb contamination of soils and (2) restricted grazing areas, do not apply to prehistoric animal populations in the US. Since these factors did not affect prehistoric animals, it is likely that plants provided more Pb to their diet. Even if animals were consuming significant amounts of soil with their plants, the soil and plant isotopic ratios would have been similar (soil and dust are the major sources for plant Pb [Chenery et al. 2012]). Therefore, soil ingestion should not greatly impact the comparisons between Pb isotopes in prehistoric human and animal tooth enamel but could influence trace element concentrations. Inhalation of dust (not affected by anthropogenic Pb prehistorically) would have affected both humans and animals and therefore should not be a significant factor in comparisons.

1.1.3 Contamination

There is typically very little Pb in tooth enamel compared to Sr and therefore contamination of Pb in tooth enamel is of greater concern. Care has to be taken to sample only those materials which are least affected by this contamination (Dudás et al. 2016; Giovas et al. 2016; Kamenov 2008; Kamenov et al. 2018). Tooth enamel is generally used for Pb and Sr isotope analyses and has been shown to be more resistant to contamination compared to bone (at least with Sr). The isotopic signatures absorbed during tooth formation are locked into the enamel, making it particularly useful in sourcing studies (Bentley 2006; Turner et al. 2009).

There are two major considerations related to contamination that are particularly important for Pb isotope studies. First, the burial environment may be contaminated with modern, anthropogenic Pb. Tooth enamel has the potential to be contaminated by this anthropogenic Pb, but for this to be the case, the burial environment (i.e. soil) itself must be contaminated. If the burial environment can be shown to be mostly free of such anthropogenic Pb contamination, then the effect of this contamination on the teeth can be ruled out as a significant factor. Second, the teeth may be contaminated by soil whether anthropogenic Pb is a factor or not.

This study uses soil leachates (water and weak-acid leaching) and trace element analysis of soils to assess if the burial environment has been contaminated by anthropogenic Pb. This includes a trace element analysis of Pb, Cu, and Zn concentrations and comparisons to contaminated and uncontaminated locations in northern Arkansas. Soil contamination of tooth enamel is assessed using three methods. (1) Pb isotope ratios of soil leachates and teeth are compared to each other. Differences in their isotopic signature would suggest the teeth were not overwhelmed with soil Pb. (2) Pb concentration thresholds outlined by Dudás et al. (2016) are used to assess contamination of human teeth. (3) Duplicate tooth samples are assessed for contamination using trace elements based on Kamenov et al.'s (2018) study. The results of these three methods are compared to assess the effectiveness of the methods.

1.2 The Case Study

1.2.1 The Crenshaw Skull and Mandible Cemetery

The Crenshaw site (3MI6) is centrally located in the Southern Caddo Area (Figure 2). It is a multiple-mound, Caddo ceremonial center (Samuelsen 2014). Caddo archaeologists recognize its importance in the region due its clear ritual importance to the prehistoric Caddo (Hoffman 1970, 1971; Schambach and Early 1982). Crenshaw is located on Quaternary deposits in the West Gulf Coastal Plain and is surrounded by Tertiary and Cretaceous deposits in the uplands (Figure 3). Further to the north, the Ouachita Mountains consist of bedrock of varying ages, from Mississippian to Pre-Cambrian. The streams and rivers that lead to Crenshaw are sourced from these mountains and therefore their weathered and redeposited sediments help make up the Quaternary landscape around Crenshaw and other sites in the Red River and Little River drainages.

Crenshaw was used as a cemetery between at least A.D. 900 and 1400 based on burial artifacts and radiocarbon dates (Durham and Davis 1975; Moore 1912; Samuelsen 2014; Weinstein et al. 2003; Wood 1963). Geophysical investigations have uncovered evidence of occupation areas, although the timing is unclear (Samuelsen 2010). Clarence B. Moore's (1912) survey of the site revealed there were at least six mounds (A through F). Mound C was destroyed by collectors in 1961, but a salvage excavation of the mound was executed by the University of Arkansas Museum which resulted in the preservation of some material and information from the mound (Durham and Davis 1975; Wood 1963). Burials were excavated from Mound F by landowners and Arkansas Archaeological Survey (ARAS) staff in 1968 (Samuelsen 2009; Schambach 1982).

In 1983, Frank Schambach and volunteers salvaged human skulls and mandibles deposited in clusters on the southern portion of the site representing 344 individuals (Zabecki 2011). These areas are referred to as the West Skull Area (WSA) and North Skull Area (NSA). Other clusters of skulls were occasionally uncovered southwest of Mound C by relic hunters. The Rayburn Skull Cluster, consisting of eight skulls, was excavated by Schambach from this area in 1968. Clusters had various numbers of individuals represented. Some consisted of a single person, others had a few skulls and a mandible included, while others consisted of as many as a hundred mandibles (Zabecki 2011). Accelerator mass spectrometry dating shows they were deposited over time between A.D. 1253 and 1399, indicating that this was not a single event, such as a large local massacre (Samuelsen 2014). They are unlikely to represent victims of local warfare. The prevalence of local warfare suggested by a deposit of such size would be expected to have major impacts in the rest of the cultural system (e.g. fortification/nucleation). This does not occur in Late Prehistoric southwest Arkansas and neither does any strong evidence of violent trauma (Samuelsen 2016). One potential exception is evident in the Ouachita region of southwest Arkansas, also culturally affiliated with the Caddo. Three burials were excavated at the Hardman site (3CL418) that consisted of articulated skeletons without skulls (Early 1993).

Upside-down bowls were placed over their missing heads. Most studies on the topic suggest the skulls and mandibles at Crenshaw are victims of warfare from other regions (Akridge 2014; Brookes 1999; Burnett 2010; Powell 1977; Schambach 2014; Schambach et al. 2011; Zabecki 2011).

1.2.2 Limitations of Sr and the Need for Pb Isotope Studies

Only one study (Samuelsen 2016) has suggested that the human skulls and mandibles found at Crenshaw were locals buried in accordance with local cultural traditions. Samuelsen (2016) reanalyzed the Sr isotope ratios of samples taken from the skulls and mandibles and concluded they were most likely local since they matched the local isotopic signature defined by the animal remains. However, key weaknesses highlight the need for additional data. The success of Sr studies relies on a small range of Sr isotope ratios at the locality where the remains were buried and different ratios in other areas (e.g. Eerkens et al. 2016). The skulls and mandibles match the local Sr range, but the ancient human Sr data from the rest of the eastern US is too similar to distinguish people from other regions based solely on Sr isotopes (Figure 4). Recently published Sr isotope data on animal teeth from the midcontinent confirm the similarity (Hedman et al. 2018). This greatly weakens the ability to draw conclusions based on Samuelsen's (2016) analysis since it would suggest that even if the remains came from elsewhere, they would also likely match the local Sr isotope signature.

Kamenov and Curtis (2017) note the same concern with using Sr isotopes alone, namely that they can be similar in many different regions. They state that combining them with Pb isotopes can solve this problem since different regions yield different Pb isotope ratios that can be used to differentiate between groups of people. In order to achieve this, a clear method for analyzing Pb isotopes from human remains to evaluate if they are local or non-local must be defined and demonstrated. The current study accomplishes this by comparing Pb isotope ratios from 22 distinct human teeth from Crenshaw and Hardman to 80 animal teeth from southwest Arkansas, Louisiana, and Mississippi. Published data from the Elizabeth site in west Illinois and the vicinity of Pueblo Bonito in northwest New Mexico also allowed for human to human comparisons (Dudás et al. 2016; Jones et al. 2017; Price et al. 2017). These published results are compared to illustrate the utility of the method, although are not considered a possible area of origin for the skulls and mandibles. Hedman et al. (2018), however, studied migration in the midcontinent and noted similarities in Sr isotope ratios in west Illinois (American Bottom) humans and southwest Arkansas animals, potentially indicating migration between these areas. Comparing Pb isotopes of individuals from west Illinois and Mounds C and F at Crenshaw will evaluate the possibility of migration to southwest Arkansas from west Illinois. The geologic setting in west Illinois (mostly Paleozoic) and northwest New Mexico (mixture of Cretaceous and Lower Tertiary) are quite different from southwest Arkansas. The current teeth data are also compared to current Pb isotope data from 18 whole rock and 26 soil leachate samples, and to published whole rock (n=46) and rock leachate (n=9) data from southwest Arkansas (Cains 2019; Duke et al. 2014; Simbo et al. 2019).

2. Materials and Methods

2.1 Sample Selection

Human teeth were selected from articulated and bundle burials from Mounds C (n=7) and F (n=6) at Crenshaw to serve as a local comparison (Table 1). The Rayburn Skull Cluster (n=5) was selected as a potential non-local group. These represent 5 skulls from an 8-skull cluster which are part of 352 excavated skulls and mandibles from the site. Second molars were used for most samples to assess their location of geographic origin during early childhood. Two samples required the use of first molars, more closely assessing their location during infancy, and one sample required the use of a third molar, assessing their location during late adolescence (Buikstra and Ubelaker 1994:47, Figure 24). Animal teeth from Mound F and near an ash bed structure on the south edge of the site (n=12) were sampled to provide a set of Pb isotope ratios from the site (Table 2). Other local animal samples (n=44) were selected from six sites in southwest Arkansas, north of Crenshaw: Martin Farm, Tom Jones, Bell, Millwood Site #35, Old Martin, and Graves Chapel. Human teeth (n=4) were selected from Hardman in the Ouachita region to test if the skulls could relate to headless burials from this adjacent region (would still be considered Caddo) while comparative animal teeth (n=8) were selected from both Hardman and Hedges (3HS60). In addition to published data from other regions (Jones et al. 2017; Price et al. 2017), animal teeth from the Fish Hatchery 2 site in northwest Louisiana (n=8) and the Austin site in northwest Mississippi (n=8) were selected (Figure 5). Both sites are situated in Quaternary alluvial deposits near major rivers. Austin was sampled because it has evidence of prehistoric violence, and headless burials recovered from the nearby Bonds site have been interpreted to be possible victims of raiding parties from Crenshaw (Brookes 1999). All animal samples were taken from prehistoric contexts and were non-migratory animal specimens. Soil leachate samples (n=26) were taken from soil cores and from previous excavations at Crenshaw (Table 3). Igneous whole rock samples of syenites and carbonatites (n=18) were collected from two sources in central-southwest Arkansas (Table 4), Granite Mountain and Magnet Cove (in the immediate vicinity of Hedges). The published literature on whole rock and rock leachates from southwest Arkansas includes a variety of other locations, including Prairie Creek, which is nearest to the sites where most of the local animals were sampled (see Figure 3).

2.2 Lab Methods

2.2.1 Tooth Drilling and Pre-treatment

Methods for processing tooth samples generally followed El Mugammar (2014), Slater et al. (2014), and Turner et al. (2009) for strontium and lead on enamel. Each tooth was cleaned through sonication in ultra-pure water for 30 minutes and dried overnight. A microscope was used to allow for high accuracy drilling of the teeth and removal of dentin from enamel. The surface of the enamel was abraded with a drill bit to clean and remove any potential contaminants. A diamond wheel bit was used to cut approximately 50mg of enamel from each tooth. Small animal teeth often did not have 50mg of enamel present, so amounts closer to 20mg were used for these samples. Given the potential for dentin to be contaminated, it was clearly removed from all human and deer samples. While every effort was made to remove dentin from all samples, some samples of small animal teeth may have included very small portions of dentin with the enamel due to the need to maximize enamel recovery. To remove any additional contamination, the enamel was then sonicated for 60 minutes in ultra-pure water, sonicated for 30 minutes in 0.1M (all humans and animals AN57 B-AN156) or 1M (animals AN1-AN56) high-purity acetic acid, sonicated in fresh acetic acid a second time for 5 minutes, and rinsed to a neutral pH with ultra-pure water.

2.2.2 Soil Leaching

Soil leachates were processed following Potra et al. (2018b) and Church et al. (1994). The soil samples were placed in acid-leached polypropylene cups. Some samples had dried out and formed large chunks. About 5g of each sample was placed in an agate mortar. The soil samples were broken up with an agate pestle until they no longer formed large (>2mm) chunks. About 4g of each sample was placed in an acid-leached Teflon beaker along with 20ml of ultra-pure water, shaken, and left to settle for 24 hours. Any organic material or clear portions of water were removed from the surface with a pipette and the remaining portion was dried down on a hot plate at 80°C. The samples were then leached in 15ml of 2N HCL in a Dubnoff metabolic shaking incubator at 55°C for 2 hours. The leachate was then pipetted into acid-leached 15ml centrifuge tubes and centrifuged for 30 minutes. A 0.25ml portion of the leachate was removed for trace element analysis. A 5ml portion of the leachate was taken from each sample for isotope analysis. This was dried down and digested in 1N HBr three times. The final digestion consisted of 4ml of 1N HBr. It was then centrifuged for 10 minutes at 3900 rpm to consolidate any undigested organic material. The top 3ml of 1N HBr was removed for isotope analysis. Processing water soil leachates followed a similar procedure: 2g of sample was used, no water rinse and dry down was executed, 10ml of ultra-pure water was added instead of 15ml of 2N HCl, and a 7ml portion of the leachate was used for Pb isotope analysis. Methods for processing the whole rocks are as presented in Simbo et al. (2019).

2.2.3 Column Chemistry for Teeth and Soil Leachates

Column chemistry (ion chromatography) was executed in a class 100 clean room at the University of Arkansas Radiogenic Isotope Laboratory. The samples were digested in 1M HBr in acid-cleaned Teflon beakers. The columns, containing 0.1ml of Dowex 1X-8 Pb resin, were cleaned with 2ml of 0.5N HNO_3, followed by 2ml of ultra-pure water. The columns were then conditioned with 2ml of 6N HCl. Each enamel sample was loaded and then the columns were washed three times with 1ml of 1N HBr. The Pb fraction from the sample was then eluted into a Teflon beaker using 1-2ml of 20% HNO_3 and subsequently dried down on a hot plate inside a class 10 laminar flow hood. The loaded sample and wash from the Pb column processing were collected in a separate Teflon beaker. Column separation methods for whole rocks are as presented in Simbo et al. (2019) and followed Pin et al. (2014).

2.2.4 Pb Isotope Ratios and Concentration Analyses

The Pb fraction was analyzed on a Nu Plasma multi-collector inductively-coupled plasma mass spectrometer (MC-ICP-MS) using a desolvating system at the University of Arkansas' Trace Element and Radiogenic Isotope Laboratory (TRAIL). The dried down Pb samples were redissolved in 2% HNO_3 spiked with a thallium (Tl) standard created just before analysis, following the procedures outlined by Kamenov et al. (2004). The Pb isotopes were corrected to NBS 981 Pb standard values ($^{208}Pb/^{204}Pb = 36.7006$, $^{207}Pb/^{204}Pb = 15.4891$, $^{206}Pb/^{204}Pb = 16.9356$) based on Todt et al. (1996) using a time-based bracketing method. A standard was run after every fourth sample. All standard and sample Pb data were normalized to $^{205}Tl/^{203}Tl = 2.38750$ (Kamenov et al. 2004). The standards (190) were run from August 2016 to November 2019 ($^{208}Pb/^{204}Pb = 36.675 \pm 0.006$ 2σ, $^{207}Pb/^{204}Pb = 15.484 \pm 0.002$ 2σ, $^{206}Pb/^{204}Pb = 16.931 \pm 0.002$ 2σ). Average 2σ standard error for standards was low ($^{208}Pb/^{204}Pb = 0.002$, $^{207}Pb/^{204}Pb = 0.001$, $^{206}Pb/^{204}Pb = 0.001$). Given the small amount of Pb in many teeth, aiming to a consistent concentration in solution for all samples is generally not possible. Sensitivity on ^{204}Pb for the 80ppb Pb standard and higher concentration samples was about 0.24v. Lower Pb standard concentration (35ppb) was used for some teeth samples with lower Pb concentrations. Samples with the lowest concentrations were analyzed using the time-resolved analysis method which followed the procedure outlined by Valentine et al. (2008) and Kamenov et al. (2006). Blank Pb concentration levels were less than 1‰ of all human, soil, and whole rock samples. Blank levels exceeded 1‰ when compared to some animal samples with low concentrations (see Table 2). Pb concentrations were similarly measured on the MC-ICP-MS and normalized to the

Pb standard concentrations, which were independently verified on a Thermo Scientific iCAP Q ICP-MS. Pb concentrations for teeth are based on the post-column Pb fraction. However, it should be noted that several procedures can affect these concentrations, including drilling technique, acid pre-treatment, and column yield if using the post-column fraction.

Trace element analysis of teeth (taken from the digested portion prior to column chemistry), following Kamenov et al. (2018), and soil leachates (Pb, Cu, and Zn) were carried out on the iCAP Q ICP-MS and corrected to multiple concentrations of elemental standard ICP-MS-68A. Duplicates of soil leachates were run from SO1 and SO20 (noted as "Dup"). SO1 and SO1 Dup were not homogenized before sampling (taken from two soil chunks from the same depth) to provide a measure of differences between samples at the same depth. SO20 and SO20 Dup were homogenized as soil dust prior to sampling to provide a measure of analytical procedure accuracy. Differences between SO20 and SO20 Dup were within 2SD of the standard error in all Pb isotopes. In terms of trace element concentrations, SO20 recorded 12%, 13%, and 15% higher Pb, Cu, and Zn values compared to SO20 Dup. Small differences in Pb isotope ratios between SO1 and SO1 Dup can be explained by the heterogeneity of the samples. Differences between SO1 and SO1 Dup in Pb, Cu, and Zn concentrations (13%, 18%, and 26%, respectively, higher in SO1) were similar to SO20 and SO20 Dup. This reflects strong consistency in analytical procedures for isotopes, but some variability in concentrations from leachates.

While variability in results can be introduced by the drilling technique and acid pre-treatment of tooth enamel, considerable intra-tooth differences in isotope ratios can also be due to teeth being formed over years and reflecting variable food sources (Buikstra and Ubelaker 1994; Lugli et al. 2017; Müller and Anczkiewicz 2015; Willmes et al. 2016). Teeth analyzed in this study (processed as tooth enamel chunks, not powder) cannot be homogenized until just before column separation. Therefore, intra-tooth differences can make duplicates (separate enamel chunks) more a measure of intra-tooth variability than a measure of analytical procedure accuracy. Since SO20 and SO20 Dup were homogenized and processed exactly the same as teeth during and after column separation, they provide a means of testing that portion of the analytical procedure. Four duplicate tooth samples were analyzed to ensure that intra-tooth differences (or potentially analytical issues) would not affect interpretations (Table 5).

3. Results and Discussion

3.1 Assessment of Contamination

3.1.1 Assessment of Anthropogenic Pb in the Burial Environment (i.e. Soil)

There are several lines of evidence that suggest the burial environment at Crenshaw has not been contaminated with anthropogenic Pb. This should not be surprising as Crenshaw is located in a remote environment, far from any major roads, mines, or industry. First, previous studies have shown that anthropogenic Pb generally remains in the topsoil and is relatively immobile in soil (Clemens 2013; Kede et al. 2014). Kamenov et al. (2009) tested soil at multiple depths and detected evidence of anthropogenic Pb contamination by illustrating different isotopic signatures and higher concentrations closer to the surface. At Crenshaw, soil samples from multiple depths within the same cores were tested at multiple locations (Table 3). The samples 25cm below the surface and deeper did not display evidence of consistently elevated Pb isotope ratios, as shown in Kamenov et al. (2009), suggesting anthropogenic Pb is not a significant factor.

Second, Pb, Cu, and Zn concentrations of soil leachates are far below what would be expected of environments contaminated with anthropogenic Pb (Figure 6). Soil leachates within

Pb contaminated environments in northern Arkansas (Potra et al. 2018b) averaged 92.0ppm Pb, 42.7ppm Cu, and 344.9ppm Zn. By contrast, soil leachates at Crenshaw averaged 2.9ppm Pb, 1.4ppm Cu, and 2.5ppm Zn. Pb, Cu, and Zn concentrations at Crenshaw are 32, 30, and 137 times lower, respectively, than the contaminated environments of northern Arkansas. The average Pb, Cu, and Zn concentrations at Crenshaw are also lower than all uncontaminated sites in northern Arkansas. This indicates that Crenshaw is a relatively uncontaminated environment.

Third, there was a possibility that higher isotope ratios in the soil could be explained by anthropogenic Pb; however, Pb isotope signatures of water and weak-acid soil leachate samples contradict this (Table 3). The results of SO10 (B) and SO20 (B) show that the weak-acid leachates have slightly higher Pb isotope ratios than the water leachates. If anthropogenic Pb with elevated isotope ratios were impacting the soil, the higher ratios should have been reflected in the water leachates as these would reflect the more labile surface contaminant. Instead, the weak-acid leachates recorded higher ratios, indicating that the soil contains the higher ratio Pb and that anthropogenic Pb is not a contaminant source.

Fourth, along similar lines, whole rocks in southwest Arkansas also have both low and high Pb isotope ratios. Whole rock data (Cains 2019; Duke et al. 2014; Simbo et al. 2019) illustrate that both low and high ratios are entirely consistent with naturally occurring ratios in southwest Arkansas. Therefore, there is no need to explain high isotope ratios at Crenshaw or elsewhere in southwest Arkansas by citing foreign materials.

Fifth, it is unlikely that high ratios reflected in the human remains in Mounds C and F are due to anthropogenic Pb because of their burial depth. As mentioned earlier, anthropogenic Pb tends to be constrained close to the surface of the soil (Clemens 2013). Many of the burials tested in Mounds C and F were buried several meters below the surface (in mounds) while the Rayburn

Cluster, with lower Pb isotope ratios, was buried relatively close to the surface (but below the topsoil). This is the opposite of what would be expected if high-ratio anthropogenic Pb were affecting the soil and, therefore, the teeth.

Establishing the lack of significant anthropogenic Pb contamination is extremely important as it indicates that the linear patterning in the current results is not due to a local ratio range being stretched by a contaminant. The evidence overwhelmingly indicates that anthropogenic Pb is not the cause of the linear patterning in the data and validates the linear method of analysis used in this study.

3.1.2 Assessment of Soil Contamination of Human Tooth Enamel

Soil contamination of human tooth enamel is assessed using three methods: (1) comparisons to Pb isotope ratios of soil leachates, (2) tooth enamel Pb concentrations, and (3) trace element analysis. First, Pb isotope data from weak-acid soil leachates and Pb concentrations from human tooth enamel were analyzed to assess contamination (Figures 7, 8). Figures 7b and 8 show that there are differences between the Pb isotope ratios of the soil and those of the human and animal teeth at Crenshaw. The linear pattern defined by the soil is different from that defined by the Crenshaw humans and animals (Figure 8). There are three exceptions to this trend, with samples HU5, HU6, and HU13 better matching the linear pattern of the soil. These samples include the highest Pb concentration in Mound C (HU6) and the second highest concentration in Mound F (HU13). This suggests that if soil contamination of the tooth enamel is present, it does not significantly affect most of the samples as they still display different Pb isotope signatures from the soil.

Four duplicate tooth samples and two duplicate soil leachate samples confirmed the separation between these two groups of data (Figure 9). The four tooth duplicates were consistent with other samples from Mounds C and F. The homogenized samples (SO20 & SO20 Dup) were nearly identical and the heterogenous soil leachate samples (SO1 and SO1 Dup) were very similar. Some tooth duplicates were very similar, and some had more pronounced differences, most likely due to intra-tooth variation in Pb isotopes. Regardless, these differences did not contradict the linear patterning established by the original samples.

The second method, which assesses tooth enamel contamination using Pb concentrations, did not provide a clear understanding of contamination. Dudás et al. (2016:28) used an upper threshold of 0.7ppm and a lower one of 0.15ppm for Pb in prehistoric human tooth enamel, outside of which the likelihood of contamination would increase. The authors based the thresholds on ancient human teeth from New Mexico, so these values may only be applicable to New Mexico and not to other environments. Humans with higher Pb concentrations could also simply reflect a greater in-vivo exposure to Pb in their environment, and not post-burial contamination.

No human samples at Crenshaw were below the lower threshold (0.15ppm) set by Dudás et al. (2016) with the exception of HU15 B. Human teeth from Hardman were all below this threshold, but so were most animal teeth, suggesting that the environment in this area has less bioavailable Pb. Most samples from Mound C were above the 0.7ppm upper threshold set by Dudás et al. (2016). The highest of these (HU6) was also the sample with the highest Pb isotope ratios at the site (human, animal, or soil). A third of the samples from Mound F were above 0.7ppm and none of the Rayburn Cluster samples were above 0.7ppm. In terms of Pb isotope ratios, most of the samples above the 0.7ppm threshold separated themselves from the soil leachates, suggesting this threshold is not appropriate for Crenshaw. The human and animal samples from Crenshaw that had Pb concentrations above 0.7ppm also tended to have higher

$^{208}Pb/^{204}Pb$ ratios (Figure 10). The pattern between elevated Pb isotope ratios and Pb concentration was also somewhat represented in soil concentrations. The two soil samples (SO10 and SO20) with the highest Pb concentration also recorded the highest Pb isotope ratios, but there was no correlation. The linearity of the human, animal, and soil Pb isotope data suggests that the source for the elevated ratios has a higher concentration of Pb compared to the source for the lower ratios.

The third method, using trace element concentration analysis, was key to indicating a lack of contamination. Most teeth processed in this study were analyzed prior to Kamenov et al.'s (2018) study. Therefore, four duplicate tooth enamel samples were analyzed for V, Mn, Fe, La, Ce, Nd, Dy, Yb, Th, and U concentrations to determine if evidence of strong contamination could be detected among some samples with high Pb isotope ratios in Mounds C and F. The results indicated that all elements, except for V, were below the contamination threshold (Figure 11). If the value for V (0.3ppm) obtained on deciduous teeth by Curzon et al. (1975) is used, then HU15 B would be below all thresholds. This is consistent with Kamenov et al.'s (2018:Figures 1, 3) figures showing no alteration to weak alteration and is inconsistent with Kamenov et al.'s (2018:Figure 4) figure illustrating strong diagenetic alteration. This indicates that the high ratios in the teeth are not due to anthropogenic Pb or soil contamination and that the linear pattering is representative of in-vivo values.

The interpretation that is most in line with all these factors is that samples HU5 and HU6 have possibly been contaminated by soil. While HU13 is also close to the soil trend line, HU13 B suggests that HU13 has not been heavily contaminated, if at all. On bivariate diagrams, the remainder of the teeth samples from Crenshaw show distinct isotopic patterns compared to the soil leachates and are likely not contaminated (Figures 8, 9). Instead, the elevated Pb concentrations and elevated Pb isotope ratios in some samples are likely reflecting input from a local geologic end-member that is also contributing to the soil. As previously suggested, this indicates that anthropogenic Pb is not the source of these isotopic ratios.

When these three methods (soil leachate companions, Pb concentrations, and trace element analysis) are compared, the use of a particular Pb concentration threshold to assess contamination at Crenshaw may not be appropriate. According to the soil leachates and trace element data, the samples that are possibly contaminated range from 0.5ppm to 31.5ppm while the relatively uncontaminated samples range from 0.12ppm to 5.3ppm. A threshold of 6ppm could be used, but only one sample (HU6) is above the threshold and this sample has already been identified as potentially contaminated by the soil leachate comparison.

3.2 Demonstrating the Validity of the Biologically Available Pb Method

Analysis of linear patterning of human and animal Pb isotope data from Crenshaw and comparisons to other regions illustrate three significant results (Figures 12, 13). First, animal samples from Crenshaw match the expected local human population, verifying that the animal samples are capable of defining a local range that is directly comparable to human remains (Figure 12a). The articulated burials in Mound C and the bundle burials in Mound F match the Pb isotope data from animals in southwest Arkansas in all 15 bivariate comparisons. Second, the animals define a linear pattern that also identifies the potential non-local group (the skull cluster) as within the local range. If only the expected local human data were used, this group would have been considered non-local (Figure 12b).

Third, the Pb isotope signatures of animals and humans from southwest Arkansas do not match those of human remains from New Mexico or west Illinois (Figure 13a,b). While there is some overlap, isotopic comparisons of each group show that the slope of the regression lines are different enough to distinguish different groups. Even in the case of the Rayburn Cluster, where individual samples are close to Illinois' range, the Rayburn Cluster distinguishes itself from west Illinois when compared as a group (Figure 13c). This clearly illustrates that Pb isotopes can discriminate between human remains from New Mexico and southwest Arkansas where Sr isotopes alone cannot. Similarly, the Illinois human remains would have been considered local at Crenshaw if using Sr isotope ratios alone; however, Pb isotope ratios clearly indicate that they are non-local (Figure 13a). The Pb isotope ratios of samples from Mounds C and F are not consistent with the linear patterning of those from west Illinois, suggesting that migration of these individuals from west Illinois is unlikely. This conclusion is, nevertheless, based only on the one locality, the Elizabeth site, that was tested.

Human and animal teeth from nearby regions were sampled to test if more proximal areas to Crenshaw could be distinguished. This included samples from the Ouachita region of southwest Arkansas and individuals from Hardman where headless bodies were buried. The Pb isotope ratios of humans and animals in this area indicate that the Rayburn Cluster did not originate from these localities in the Ouachita region, since the Pb isotope values of the Ouachita samples are too low (Figure 14a). The Pb isotope ratios from the Ouachita region are generally consistent with the southwest Arkansas linear patterning defined by the other sites in southwest Arkansas, despite their considerable distance and different geology. The Fish Hatchery 2 site down the Red River in northwest Louisiana and the Austin site in northwest Mississippi were also selected for sampling. Given the proximity to Crenshaw, it was not expected that Fish Hatchery 2 would be distinguishable. Also, it was not clear if northwest Mississippi would be different either, given the similarly young geologic setting. However, the data showed that the Pb isotope ratios for each location were restricted to a small range (Figure 13d). Even though they overlap with a small part of southwest Arkansas, the data clearly suggest that the Rayburn Skull Cluster, Mound C, and Mound F remains did not come from these sites in Louisiana and Mississippi. It is important to note that the sampling is not sufficient to define a range for each of these other regions and this analysis is only capable of distinguishing these specific localities. However, the current results have established that the Pb isotope ratios from the Rayburn Skull Cluster are consistent with the Pb isotope ratios of southwest Arkansas and that they are inconsistent with the ratios from localities in the Ouachita region of southwest Arkansas, northwest Louisiana, northwest Mississippi, west Illinois, and New Mexico. This is the same cluster that has been repeatedly referred to as "trophy skulls" in the literature based on Powell's (1977) research.

This study has shown that 1) the Pb isotope ratios of animals match those in expected local human remains, 2) the Pb isotope ratios of animals are different than those of known non-local humans, and 3) the isotopic background created by animals was more effective at defining a local isotopic range than using expected local human remains. This clearly demonstrates that the method presented in the current study is valid and that, when additional clusters, localities, and regions are tested, it will provide answers to the questions surrounding the origins of the remains at Crenshaw that have otherwise been unobtainable.

3.3 Comparisons to Geologic Data

While the human and animal data can be directly compared, Pb isotope data from whole rocks in southwest Arkansas indicate that they are far too variable to be useful for sourcing human remains (Figure 15a). One concern is that the whole rock samples and the human and animal samples were collected from different locations. There are two exceptions. First, Prairie Creek (see Figures 3 and 15a) is close to the majority of sites and has extreme whole rock Pb isotope ratios compared to those of humans and animals. Second, many whole rock samples were gathered from Magnet Cove, which is in the immediate vicinity of one of the Ouachita region sites. Comparisons between the Ouachita region sites and the Magnet Cove whole rocks confirm that the whole rock Pb isotope signatures are significantly different from those of the humans and animals (Figure 14b). Therefore, it is suggested that whole rocks not be used for direct comparison to human remains, but they still provide some utility when the geologic regions are different enough (e.g. Kamenov and Gulson 2014). Simbo et al. (2019) showed that the Pb isotope ratios of whole rocks were more variable than those of rock leachates from the same rock samples. Despite a relatively small number of samples (n=9), rock leachates in southwest Arkansas also are much more variable than human or animal data and may not be directly comparable (Figure 15a); however, the limited sampling requires further research to test this. By contrast, the ratios of soil leachates from Crenshaw compare more favorably with those of southwest Arkansas animals (Figure 7a), but there are still differences between the soil and the human and animal remains from Crenshaw (Figures 7b, 8, 10). The reasoning for this pattern in geologic variability in Pb isotope ratios (whole rock > rock leachates > soil) can be inferred from the process of weathering and eventual deposition of Pb at the sites under study. The labile fraction of soils is a major source of bioavailable materials (Anderson and Hillwalker 2008; John and Leventhal 1995). Whole rocks include silicates that may not ever be biologically available due to the difficulty in their dissolution. Rock leachates are made up of the more easily mobile fraction, which is more likely to end up in river sediments and soils. Whatever is redeposited in the form of soil is mixed up and redeposited, likely reducing extreme Pb isotope ratios and creating a more amalgamated signature as is seen in the soil. This is the portion of Pb that is absorbed by plants and ingested by humans and animals. Soil and modern plants could be useful as direct comparisons to ancient humans as well but are more likely to be impacted by anthropogenic Pb. Therefore, it is suggested that the biologically available Pb method is best implemented using prehistoric animal teeth for defining the local isotopic background.

Even though the whole rocks may not be as appropriate for defining backgrounds as prehistoric animal tooth enamel, such data can provide insights into the provenance of Pb in the region. Pb isotope ratios of whole rocks from the Ouachita mountains suggest they could represent the source of the end-member with lower isotope ratios that define the linear pattering in southwest Arkansas (Figure 15). Some whole rock samples also indicate the presence of higher Pb isotope ratios in the region.

4. Conclusions

The biologically available Pb method compares the human remains to a large number of animal tooth enamel samples and utilizes their multivariate and linear nature to detect differences between possible regions of origin. This method successfully identifies non-local individuals and aids in evaluating of the origin of a skull cluster from the Crenshaw site in southwest Arkansas. It clearly demonstrates that Pb isotope ratios from prehistoric animal tooth enamel are most appropriate (particularly compared to whole rocks) for direct comparison to ancient human remains. It is unclear at this stage if the method is capable of uniquely identifying areas of origin and future studies, employing further development of the method will resolve this. The linear patterning section of this method relies on the presence of linear trends in the data, so this section may not be applicable if linear trends do does not exist within a region. Each region may have varying amounts of Pb in the environment, making a particular threshold of Pb concentration in tooth enamel difficult to apply in different regions. Based on the analysis of contamination provided here, it is suggested that trace element analysis, like the one described by Kamenov et al. (2018), be used in combination with soil samples when assessing contamination rather than relying on Pb concentrations alone. Further research in the area of contamination and decontamination of Pb isotopes in ancient teeth is needed.

Two issues prevent broad conclusions in the case study. First, the sample size of a single skull cluster is too small to make conclusions about the origin of the skull and mandible deposits at Crenshaw (8 of 352 individuals). Second, the number of animals from other regions is too small to provide a detailed regional map of Pb isotope ratios. All of the above-mentioned issues are being researched as part of a Doctoral Dissertation Research Improvement Grant from the National Science Foundation (grant number 1830438) and should result in further resolution of these concerns.

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Figure Captions

Figure 1 – The 15 possible unique comparisons of different Pb isotope ratios utilizing animal data from southwest Arkansas and human data from northwest New Mexico (Dudás et al. 2016; Price et al. 2017). The Pb isotope ratios of humans must match those of animals in all 15 comparisons in order to be considered local. If they overlap in 14 but are different in one bivariate diagram, they are considered non-local. The slopes of the regression lines are clearly different in many comparisons, identifying that the humans from New Mexico are non-local to southwest Arkansas.

Figure 2 – Map of Crenshaw (southwest Arkansas) within the Caddo Area on the border between the Southern Plains and the Eastern Woodlands. The Caddo Area is considered to be part of the Eastern Woodlands. Sites sampled also include Hardman, Hedges, Austin (northwest Mississippi) and Fish Hatchery 2 (northwest Louisiana). The geology around both is made up of Quaternary alluvium. Austin is in a very large area defined by this alluvium while there are nearby Tertiary deposits around Fish Hatchery 2.

Figure 3 – Maximum age of geology in southwest Arkansas. Sampled locations and previously published data are identified. Animal teeth (red dots) were selected from sites in several counties in southwest Arkansas. Soil samples were selected from Crenshaw and human samples were selected from Crenshaw and Hardman (purple dots). New whole rock data (black diamonds) are from Magnet Cove and Granite Mountain while previously published whole rock and rock leachate data (black diamonds) are from Cains (2019), Duke et al. (2014), and Simbo et al. (2019).

Figure 4 – Comparison of Sr isotopes of humans from Crenshaw and ancient humans from the Eastern US. All but five out of 670 prehistoric human teeth tested for Sr ratios in the eastern US would be considered "local" to southwest Arkansas. The data from other regions look similar to Crenshaw humans, but they are more skewed towards lower ratios (Beehr 2011; Hedman et al. 2018; Jones et al. 2017; Price et al. 2007; Samuelsen 2016; Slater et al. 2014). The five non-local teeth (representing three individuals) from the American Bottom are also non-local to the American Bottom. Available Sr ratios in the eastern US appear similar to southwest Arkansas (see also, Hedman et al. 2009).

Figure 5 – Map showing counties sampled for Pb isotope analysis in this study and counties sampled for prehistoric human Pb isotope ratios in previous studies (Dudás et al. 2016; Jones et al. 2017; Price et al. 2017).

Figure 6 – Mean Cu, Pb, and Zn concentrations in weak-acid leachates from Crenshaw and northern Arkansas (Potra et al. 2018b). The mean from northern Arkansas uncontaminated sites included all sites upstream of mines that were interpreted to not be impacted by anthropogenic Pb contamination by Potra et al. (2018b) while the mean from northern Arkansas contaminated sites include all locations downstream from mines. The results clearly show that Crenshaw has lower concentrations than both the contaminated and uncontaminated sites in northern Arkansas, supporting the lack of significant anthropogenic Pb contamination.

Figure 7 – Bivariate Pb isotope diagrams comparing weak-acid soil leachate data to human and animal data. a) Comparison of the weak-acid soil leachates to all southwest Arkansas humans/animals shows they are much more consistent with the humans/animals than other geologic data (i.e. whole rocks and rock leachates). b) Comparison of the weak-acid soil leachates to only Crenshaw humans/animals shows that they have similar Pb isotope ratios, but that there is a clear difference between the soil and the human and animal tooth enamel. The teeth with lower ratios have a different linear pattern than the soil. The soil samples do not have ^{208}Pb/^{204}Pb ratios higher than 40 while many teeth do have higher ratios. The tooth samples generally do not match the soil, suggesting they are not being replaced by soil contaminated Pb.

Figure 8 – Linear comparison of Pb isotope ratios from weak-acid soil leachates and human and animal samples at Crenshaw. Extending the fit line on soil samples to higher ratios shows that HU5, HU6, and HU13 better match the linear pattering defined by the soil than by the human and animal samples at Crenshaw. Otherwise, the soil and human/animal samples maintain different linear pattering which suggests the other human/animal samples were not greatly impacted by contamination.

Figure 9 – Linear comparison of Pb isotope ratios from weak-acid soil leachates and human and animal samples at Crenshaw, including duplicate soil leachate and tooth enamel samples. The lone homogenized samples (SO20 and SO20 Dup) were nearly identical (Δ ^{208}Pb/^{204}Pb=0.000, Δ ^{207}Pb/^{204}Pb=0.000, Δ ^{206}Pb/^{204}Pb=0.002). Soil leachate samples SO1 and SO1 Dup were not homogenized but were still very similar (Δ ^{208}Pb/^{204}Pb=0.012, Δ ^{207}Pb/^{204}Pb=0.003, Δ ^{206}Pb/^{204}Pb=0.002). Tooth duplicates showed greater differences (Δ ^{208}Pb/^{204}Pb=0.052, Δ

^{207}Pb/^{204}Pb=0.007, Δ ^{206}Pb/^{204}Pb=0.078), but these are most likely due to intra-tooth differences. Regardless, the duplicate teeth and soil leachate samples verify the Pb isotope trend lines and continue to show differences between the soil and teeth Pb isotope trends.

Figure 10 – Comparisons involving Pb concentrations on Crenshaw humans, animals, and soil. a) Bivariate plot comparing ^{208}Pb/^{204}Pb ratios and Pb concentrations of humans and animals. Samples with higher Pb ratios tend to have higher Pb concentrations. All but one sample above 0.7ppm had a ^{208}Pb/^{204}Pb ratio above 40. HU6 had the highest Pb concentration (31.5ppm) and ^{208}Pb/^{204}Pb ratio (40.96) and is not depicted. b) Comparison of ^{208}Pb/^{204}Pb ratio between weak-acid soil leachates and humans/animals with Pb concentrations above 0.7ppm. All 20 weak-acid soil leachates (duplicates excluded) have ratios below 40 and the highest two samples are statistical outliers. All but one tooth sample above 0.7ppm have ratios above 40.

Figure 11 – Trace element concentrations of elements in duplicate human tooth enamel based on Kamenov et al. (2018). All element concentrations (C), with the exception of V, are below the maximum threshold concentrations (MTC). Lines below 1 C/MTC suggest the lack of diagenetic alteration. The results are consistent with Kamenov et al. (2018:Figures 1,3) showing no alteration to weak alteration and are inconsistent with Kamenov et al. (2018:Figure 4) showing strong alteration. This suggests these tooth enamel samples were not subject to significant diagenetic alteration and supports the conclusion that the high ratios and linear patterning reflect in-vivo values rather than anthropogenic or soil Pb contamination.

Figure 12 – Illustration of the importance of the biologically available Pb method. The study sampled human remains from Mound C (n=7), Mound F (n=6), and the Rayburn Cluster (n=5) from Crenshaw and burials (n=4) from Hardman. It also sampled prehistoric archaeological animals in southwest Arkansas (n=64) from nine sites (3CL418, 3HE40, 3HE92, 3HO11, 3HS60, 3LR49, 3MI6, 3SV15, and 3SV20), Louisiana (Fish Hatchery 2 [16NA70], n=8), and Mississippi (Austin [22TU549], n=8). a) Pb isotopes of human teeth from Crenshaw and Hardman and animal teeth from southwest Arkansas indicate that all the human remains are "local" to southwest Arkansas. Differences between local human groups could be explained by a number of factors (e.g. settlement patterns). Animal samples included raccoon (7), opossum (8), squirrel (8), rabbit (18), deer (33), and other small animals (6). No significant difference was seen between deer and other animals. b) Pb isotope ratios from human teeth at Crenshaw show that the articulated and disarticulated remains from Mounds C and F do not match the skull cluster, suggesting the skulls are non-local. This illustrates that not using the biologically available Pb method would lead to the wrong conclusion.

Figure 13 – Linear comparisons of Pb isotope ratios, each consisting of 15 different combinations of ratios (selected comparisons shown). a) Comparison of southwest Arkansas data to humans from Pueblo Bonito (Price et al. 2017). b) Comparison of southwest Arkansas data to west Illinois humans (Jones et al. 2017). c) Comparison of southwest Arkansas animals, the Rayburn Cluster, and west Illinois. d) Comparison of southwest Arkansas animals, the Rayburn Cluster, Fish Hatchery #2 (northwest Louisiana) animals, and Austin (northwest Mississippi) animals.

Figure 14 – Comparisons of Pb isotope ratios of human and animal samples from the Ouachita region sites (Hardman and Hedges) to other southwest Arkansas samples (Duke et al. 2014; Simbo et al. 2019). a) Ouachita region humans and animals are distinguishable from the skull cluster (and Mounds C and F), suggesting the skulls did not come from this area. b) Despite their close proximity, Ouachita region humans and animals are not consistent with Magnet Cove whole rocks. Magnet Cove whole rocks have extreme ratios, making the Ouachita region humans and animals better match the more distant animals from the Red/Little River sites in southwest Arkansas.

Figure 15 – Comparisons of southwest Arkansas humans, animals, and geologic data (Cains 2019; Duke et al. 2014; Simbo et al. 2019). a) Comparisons of humans/animals and whole rocks show that the whole rocks are far too variable to be directly compared to human remains. The whole rock sample locations closest to the animal sampling sites, Prairie Creek and Magnet Cove, have extreme isotope ratios that are inconsistent with the animals and humans. Humans/animals from other regions (e.g. New Mexico) would also be considered local to southwest Arkansas if this data was used to construct a local regional background. Higher ratios among some whole rocks indicate that the higher ratios in the human and animal data could be defined by local geology. b) Comparisons of humans/animals to rock leachates show the rock leachates are more consistent with the humans and animals than their whole rock counterparts. However, they have linear patterns that are not consistent with the humans/animals and are still more variable. Lighter leaching methods may result in more comparable data. These data suggest that these rocks could form the lower end member that defines the linear patterning in southwest Arkansas.

Figure 1

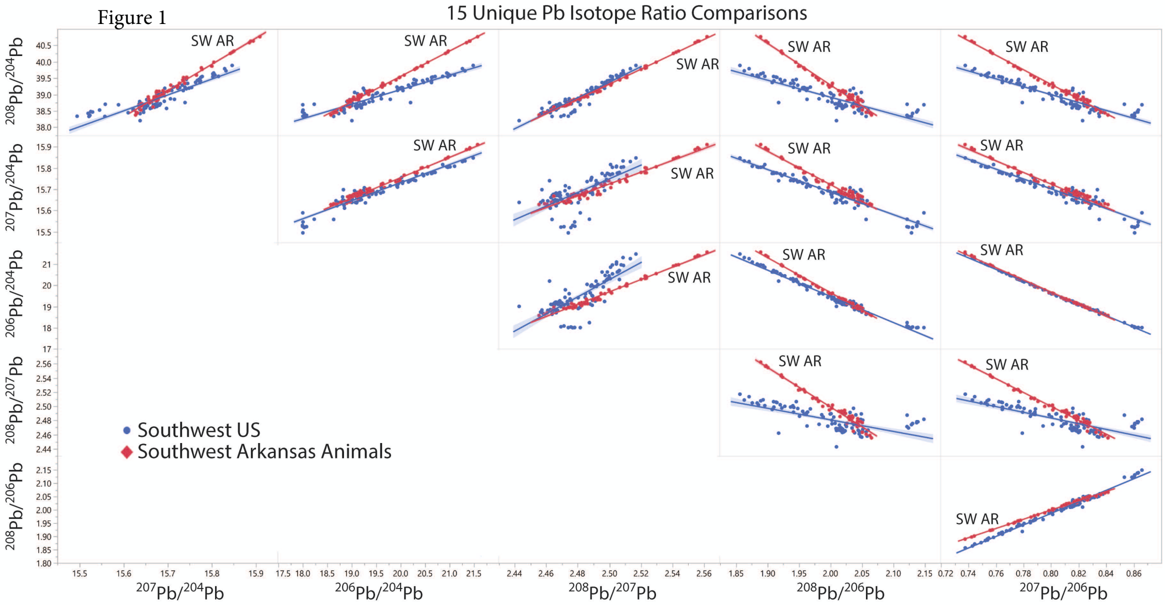

15 Unique Pb Isotope Ratio Comparisons
SW AR
Southwest US
Southwest Arkansas Animals
$^{208}Pb/^{204}Pb$
$^{207}Pb/^{204}Pb$
$^{206}Pb/^{204}Pb$
$^{208}Pb/^{207}Pb$
$^{208}Pb/^{206}Pb$
$^{207}Pb/^{206}Pb$

Figure 2

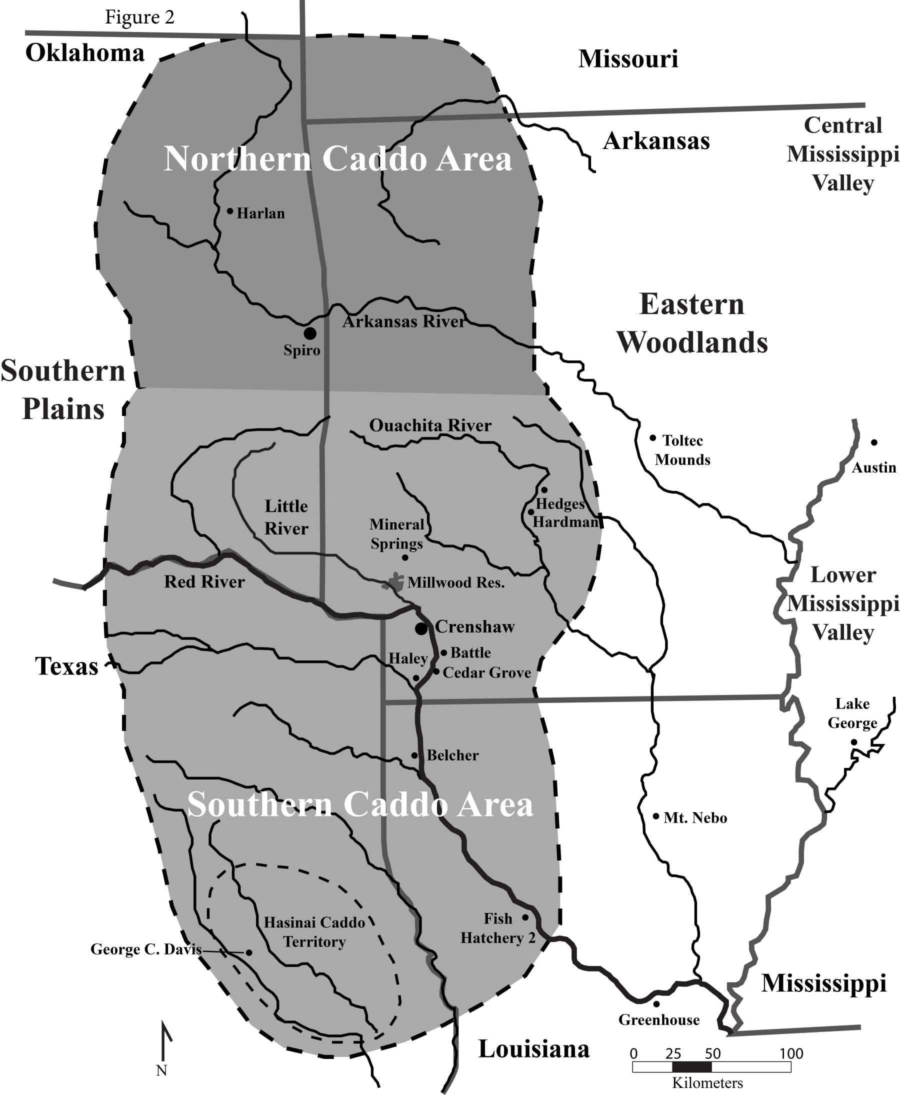

Oklahoma
Missouri
Arkansas
Central Mississippi Valley
Northern Caddo Area
Harlan
Arkansas River
Spiro
Eastern Woodlands
Southern Plains
Ouachita River
Toltec Mounds
Austin
Little River
Mineral Springs
Hedges
Hardman
Red River
Millwood Res.
Lower Mississippi Valley
Crenshaw
Texas
Haley
Battle
Cedar Grove
Lake George
Belcher
Southern Caddo Area
Mt. Nebo
Hasinai Caddo Territory
Fish Hatchery 2
George C. Davis
Mississippi
Greenhouse
Louisiana
N
0 25 50 100
Kilometers

Figure 3

Maximum Age of Geology and Sampled Sites in Southwest Arkansas

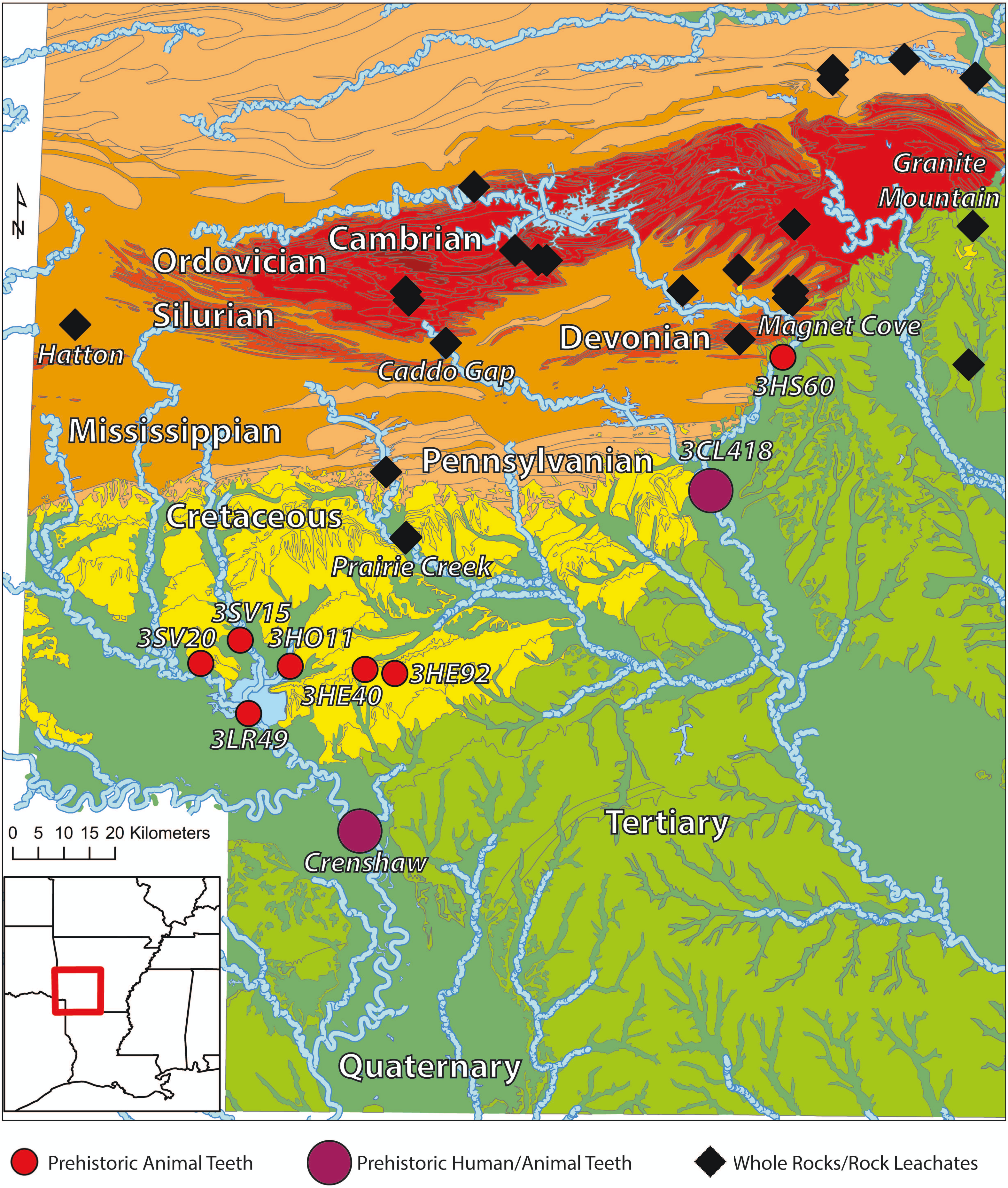


Figure 4 Sr ratios - SW Arkansas vs Eastern US Prehistoric Humans

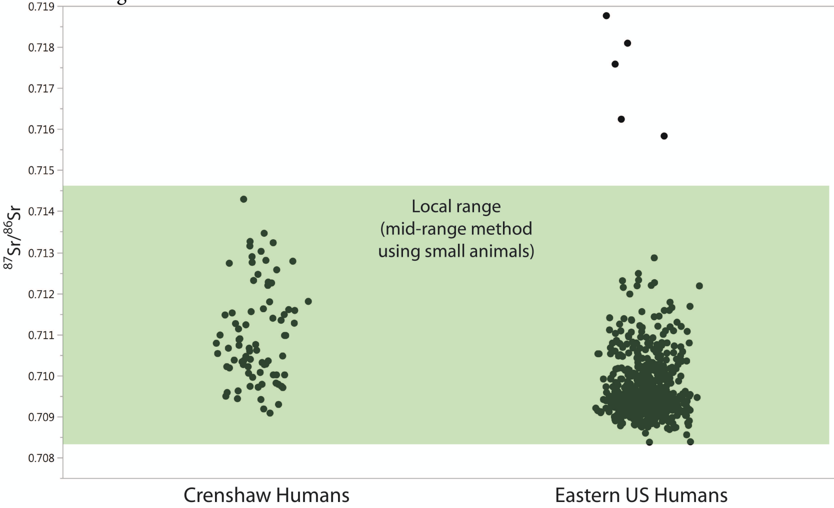


Figure 5

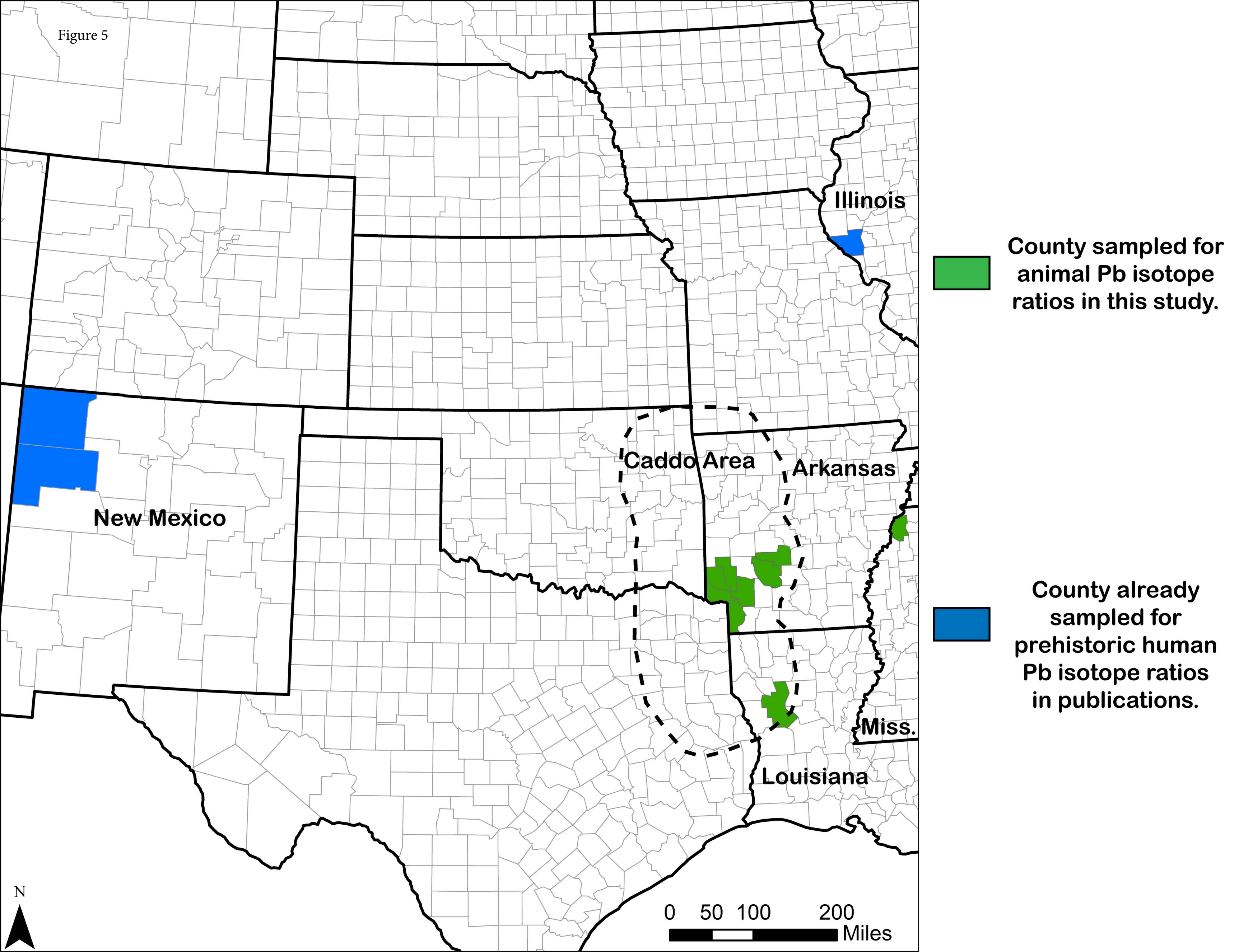
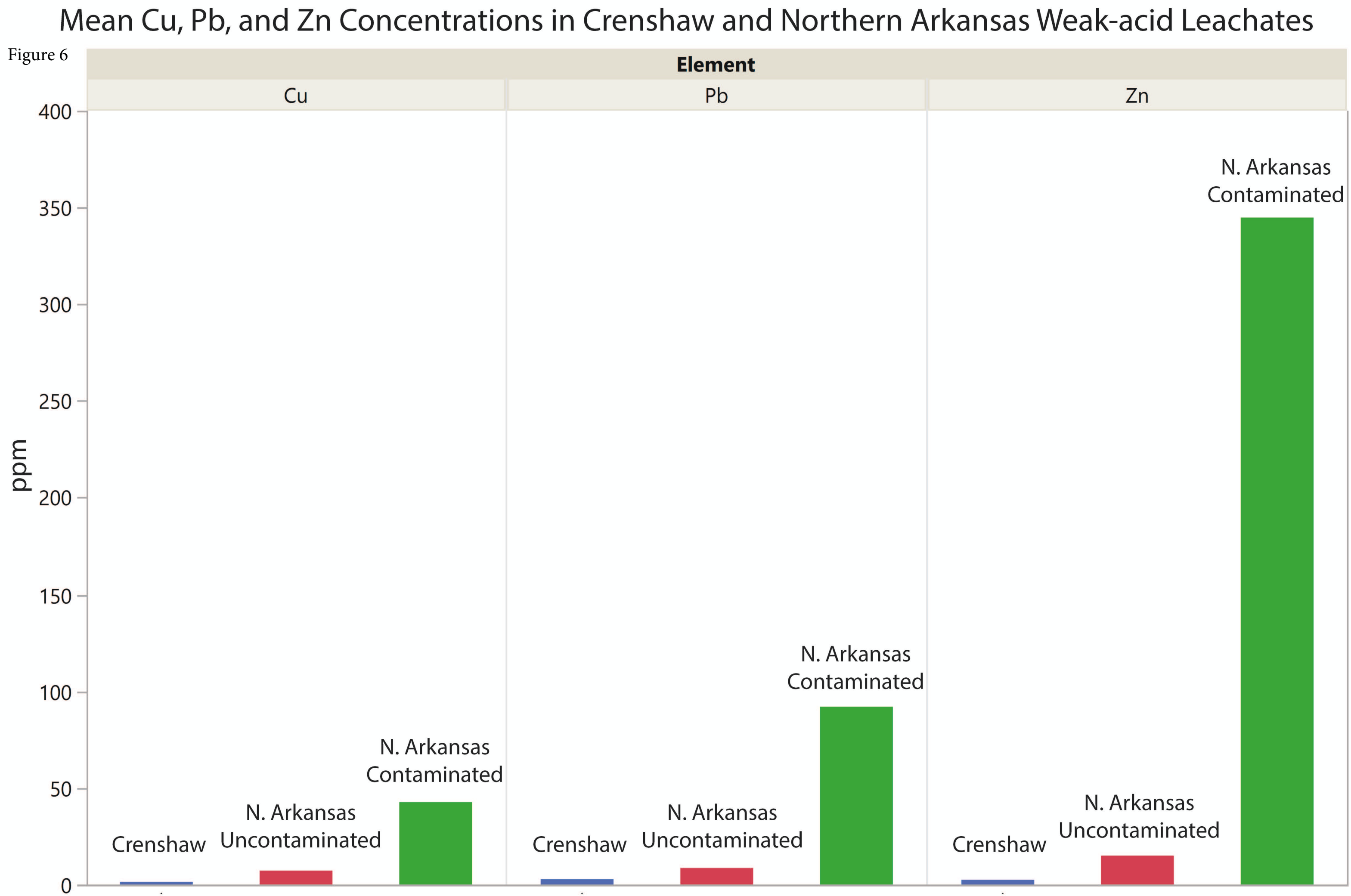

Illinois
Caddo Area
Arkansas
New Mexico
Miss.
Louisiana
County sampled for animal Pb isotope ratios in this study.
County already sampled for prehistoric human Pb isotope ratios in publications.
N
0 50 100 200
Miles

Figure 6



Mean Cu, Pb, and Zn Concentrations in Crenshaw and Northern Arkansas Weak-acid Leachates
Element
Cu
Pb
Zn
ppm
400
350
300
250
200
150
100
50
0
Crenshaw
N. Arkansas Uncontaminated
N. Arkansas Contaminated
Crenshaw
N. Arkansas Uncontaminated
N. Arkansas Contaminated
Crenshaw
N. Arkansas Uncontaminated
N. Arkansas Contaminated

Figure 7

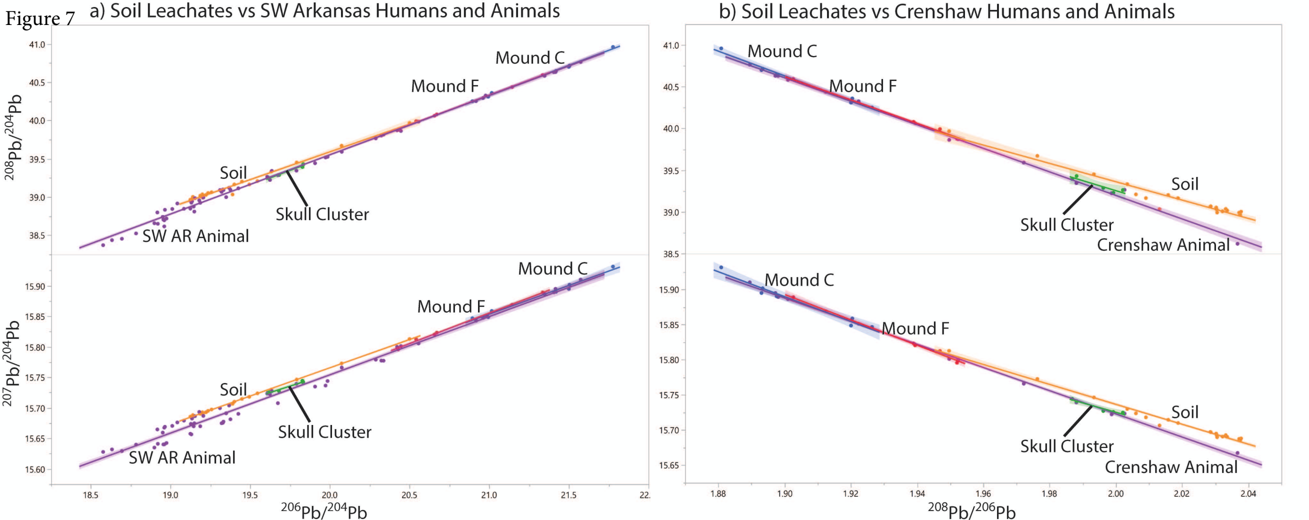
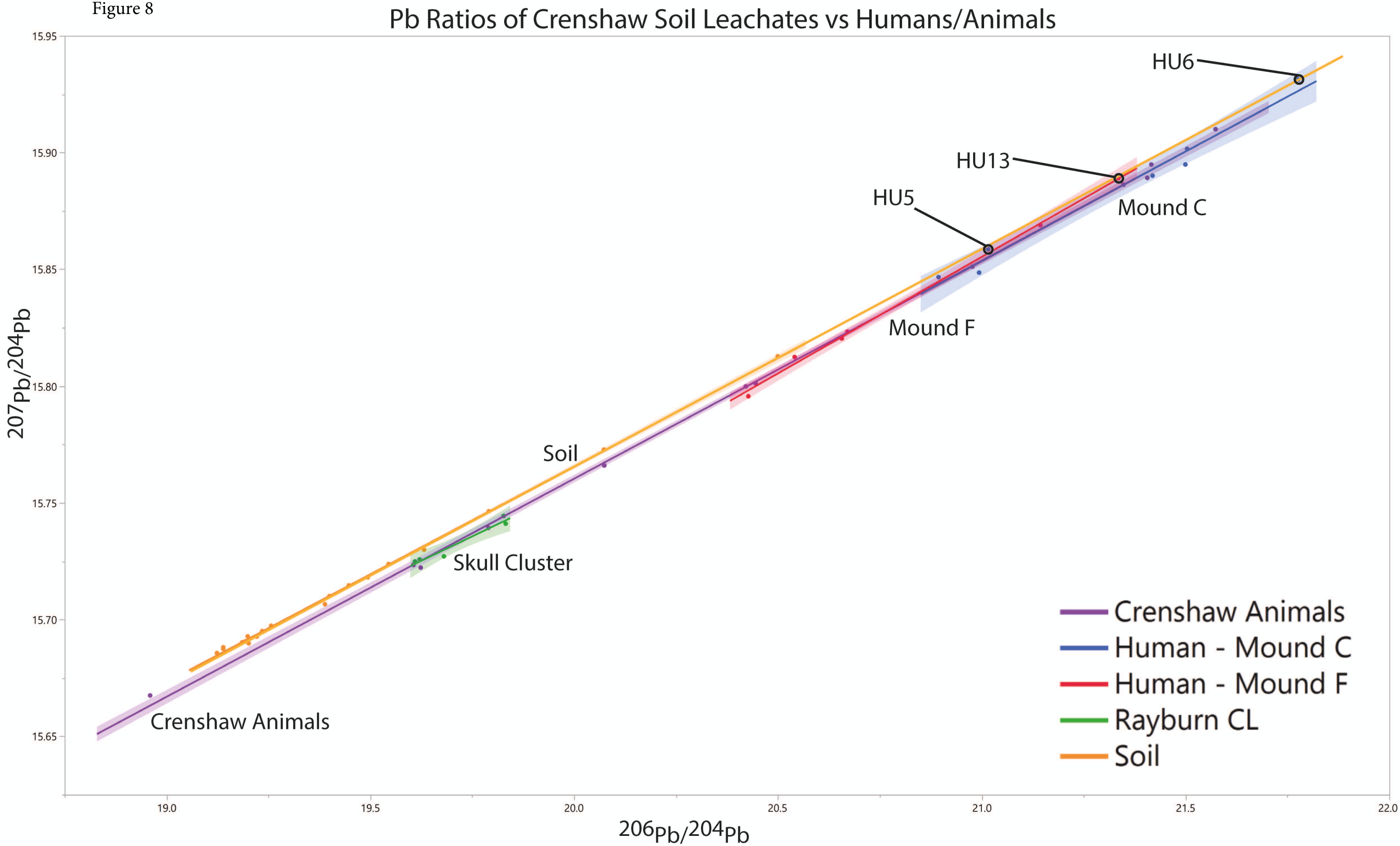

a) Soil Leachates vs SW Arkansas Humans and Animals
b) Soil Leachates vs Crenshaw Humans and Animals
Mound C
Mound F
Soil
Skull Cluster
SW AR Animal
Crenshaw Animal
$^{208}Pb/^{204}Pb$
$^{207}Pb/^{204}Pb$
$^{206}Pb/^{204}Pb$
$^{208}Pb/^{206}Pb$

Figure 8



Pb Ratios of Crenshaw Soil Leachates vs Humans/Animals
15.95
15.90
15.85
15.80
15.75
15.70
15.65
207Pb/204Pb
19.0
19.5
20.0
20.5
21.0
21.5
22.0
206Pb/204Pb
HU6
HU13
HU5
Mound C
Mound F
Soil
Skull Cluster
Crenshaw Animals
Crenshaw Animals
Human - Mound C
Human - Mound F
Rayburn CL
Soil

Figure 9

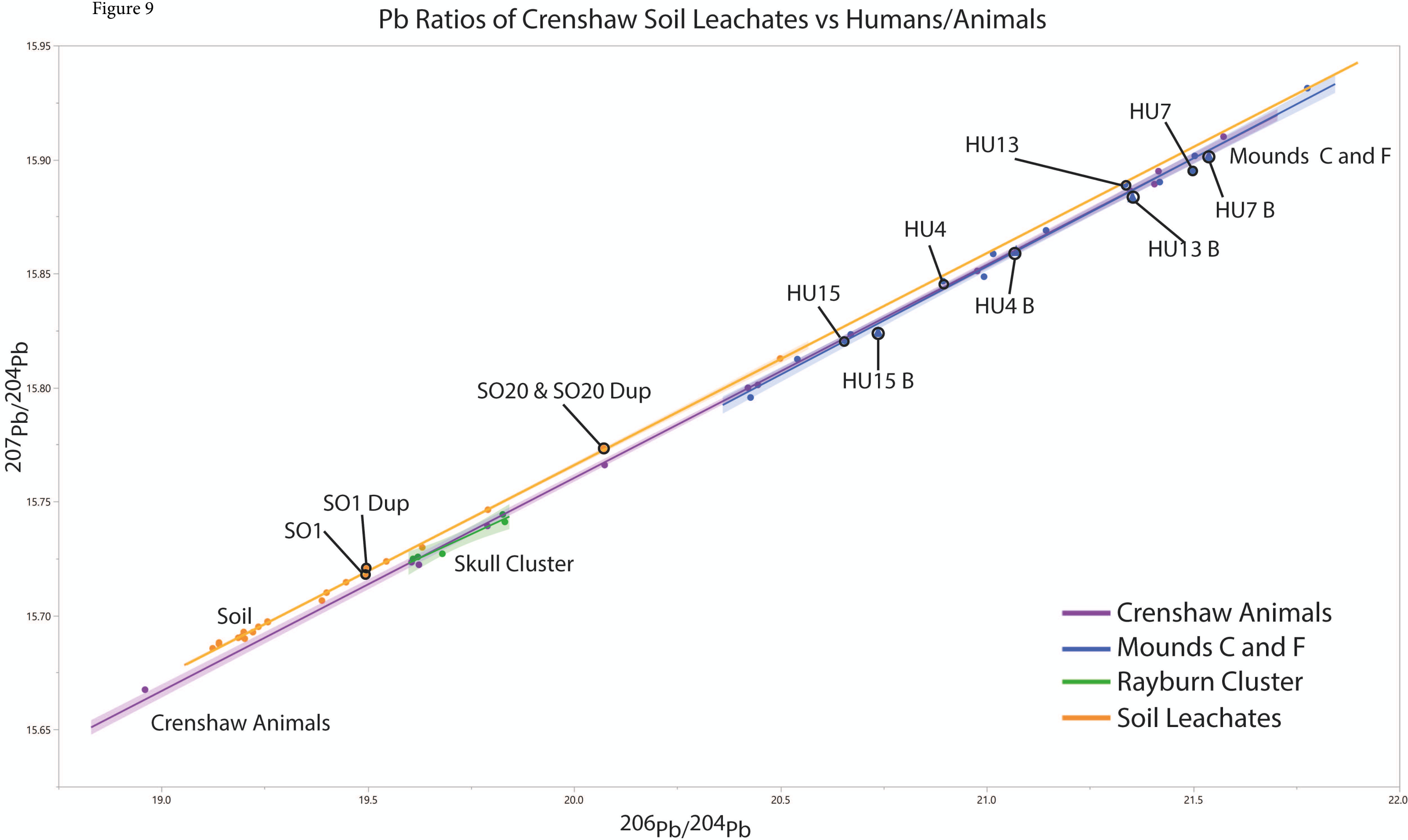
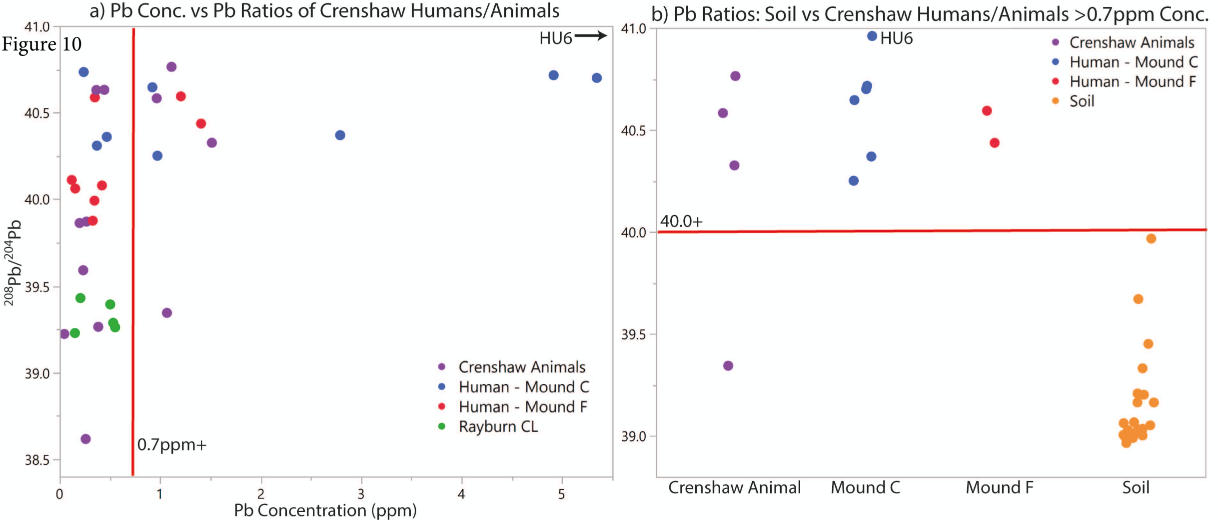

Pb Ratios of Crenshaw Soil Leachates vs Humans/Animals
15.95
15.90
15.85
15.80
15.75
15.70
15.65
207Pb/204Pb
19.0
19.5
20.0
20.5
21.0
21.5
22.0
206Pb/204Pb
HU7
HU13
Mounds C and F
HU7 B
HU4
HU13 B
HU4 B
HU15
HU15 B
SO20 & SO20 Dup
SO1 Dup
SO1
Skull Cluster
Soil
Crenshaw Animals
Crenshaw Animals
Mounds C and F
Rayburn Cluster
Soil Leachates

Figure 10



a) Pb Conc. vs Pb Ratios of Crenshaw Humans/Animals
HU6
0.7ppm+
Crenshaw Animals
Human - Mound C
Human - Mound F
Rayburn CL
^{208}Pb/^{204}Pb
Pb Concentration (ppm)
b) Pb Ratios: Soil vs Crenshaw Humans/Animals >0.7ppm Conc.
HU6
Crenshaw Animals
Human - Mound C
Human - Mound F
Soil
40.0+
Crenshaw Animal
Mound C
Mound F
Soil

Figure 11

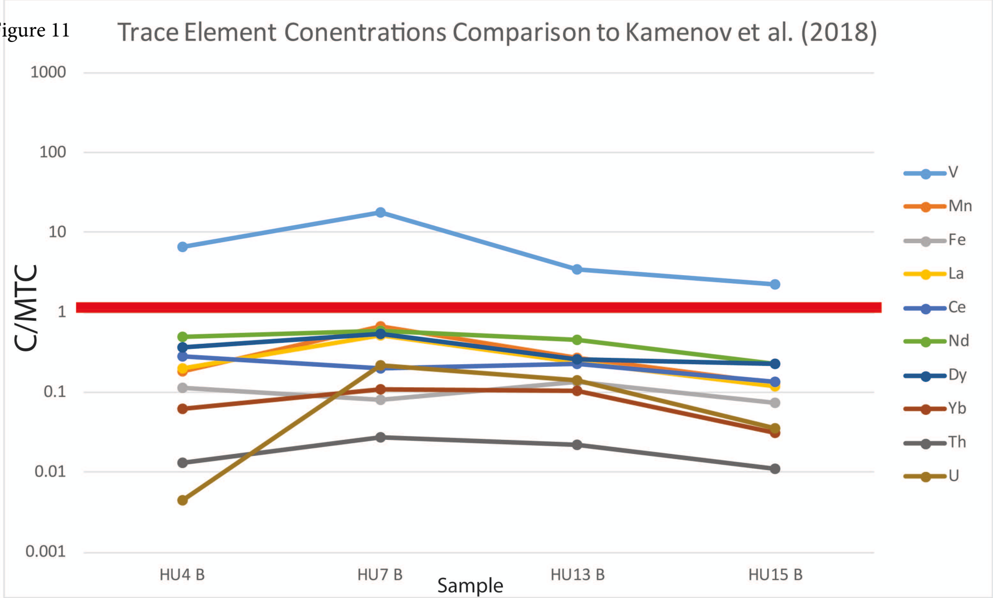
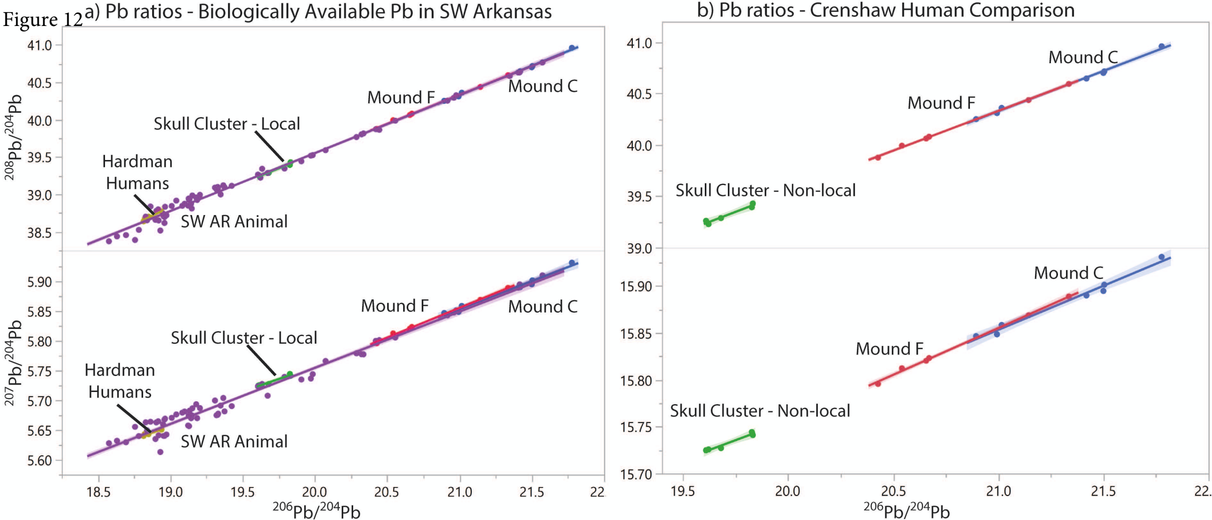

Trace Element Conentrations Comparison to Kamenov et al. (2018)
1000
100
10
1
0.1
0.01
0.001
C/MTC
HU4 B
HU7 B
HU13 B
HU15 B
Sample
V
Mn
Fe
La
Ce
Nd
Dy
Yb
Th
U

Figure 12



a) Pb ratios - Biologically Available Pb in SW Arkansas
b) Pb ratios - Crenshaw Human Comparison
Mound F
Mound C
Skull Cluster - Local
Hardman Humans
SW AR Animal
Skull Cluster - Non-local
$^{208}Pb/^{204}Pb$
$^{207}Pb/^{204}Pb$
$^{206}Pb/^{204}Pb$

Figure 13

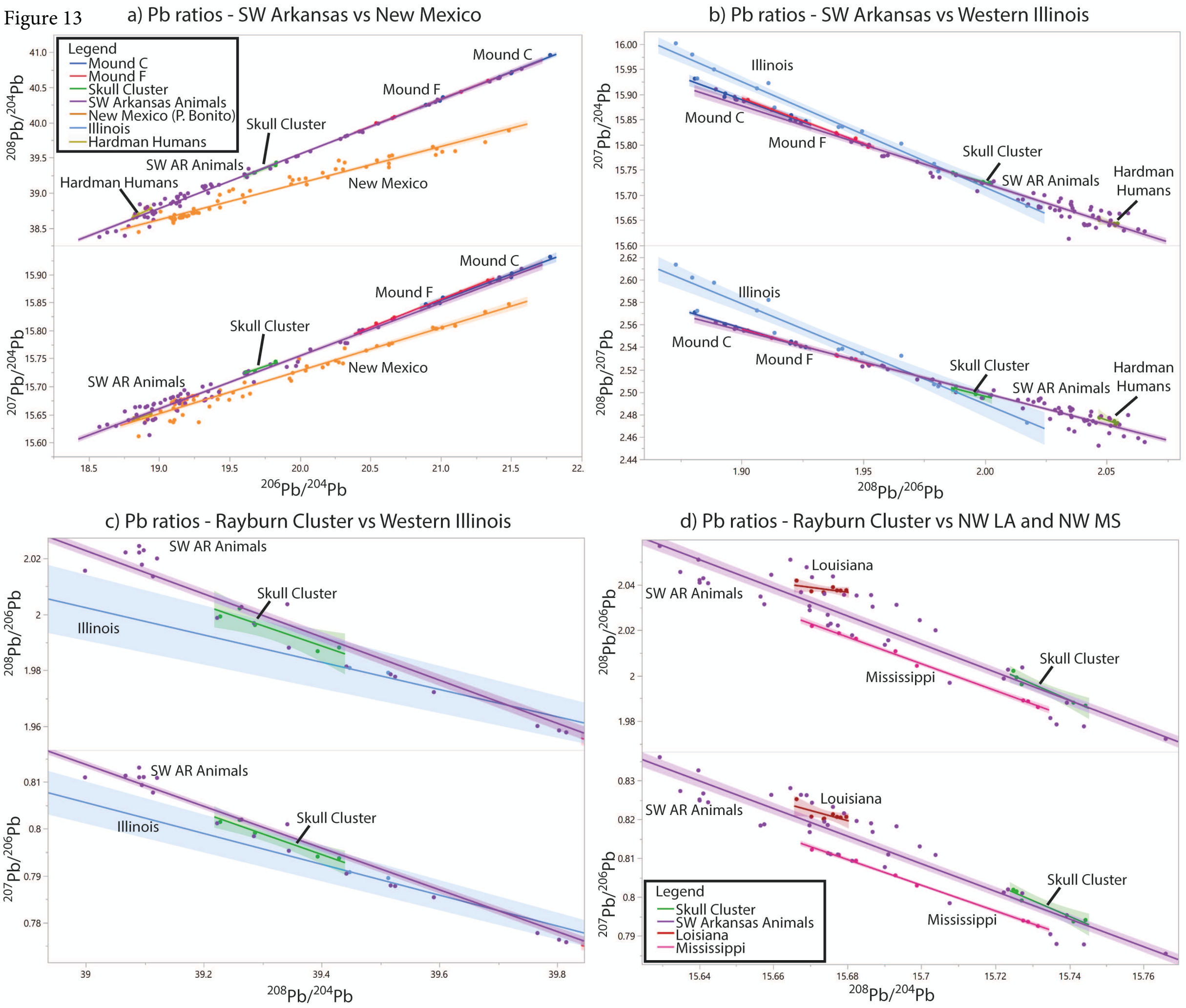

a) Pb ratios - SW Arkansas vs New Mexico
Legend
Mound C
Mound F
Skull Cluster
SW Arkansas Animals
New Mexico (P. Bonito)
Illinois
Hardman Humans
Mound C
Mound F
Skull Cluster
SW AR Animals
Hardman Humans
New Mexico
Mound C
Mound F
Skull Cluster
SW AR Animals
New Mexico
208Pb/204Pb
207Pb/204Pb
206Pb/204Pb
b) Pb ratios - SW Arkansas vs Western Illinois
Illinois
Mound C
Mound F
Skull Cluster
SW AR Animals
Hardman Humans
Illinois
Mound C
Mound F
Skull Cluster
SW AR Animals
Hardman Humans
207Pb/204Pb
208Pb/207Pb
208Pb/206Pb
c) Pb ratios - Rayburn Cluster vs Western Illinois
SW AR Animals
Skull Cluster
Illinois
SW AR Animals
Skull Cluster
Illinois
208Pb/206Pb
207Pb/206Pb
208Pb/204Pb
d) Pb ratios - Rayburn Cluster vs NW LA and NW MS
Louisiana
SW AR Animals
Skull Cluster
Mississippi
Louisiana
SW AR Animals
Skull Cluster
Mississippi
Legend
Skull Cluster
SW Arkansas Animals
Loisiana
Mississippi
208Pb/206Pb
207Pb/206Pb
208Pb/204Pb

Figure 14

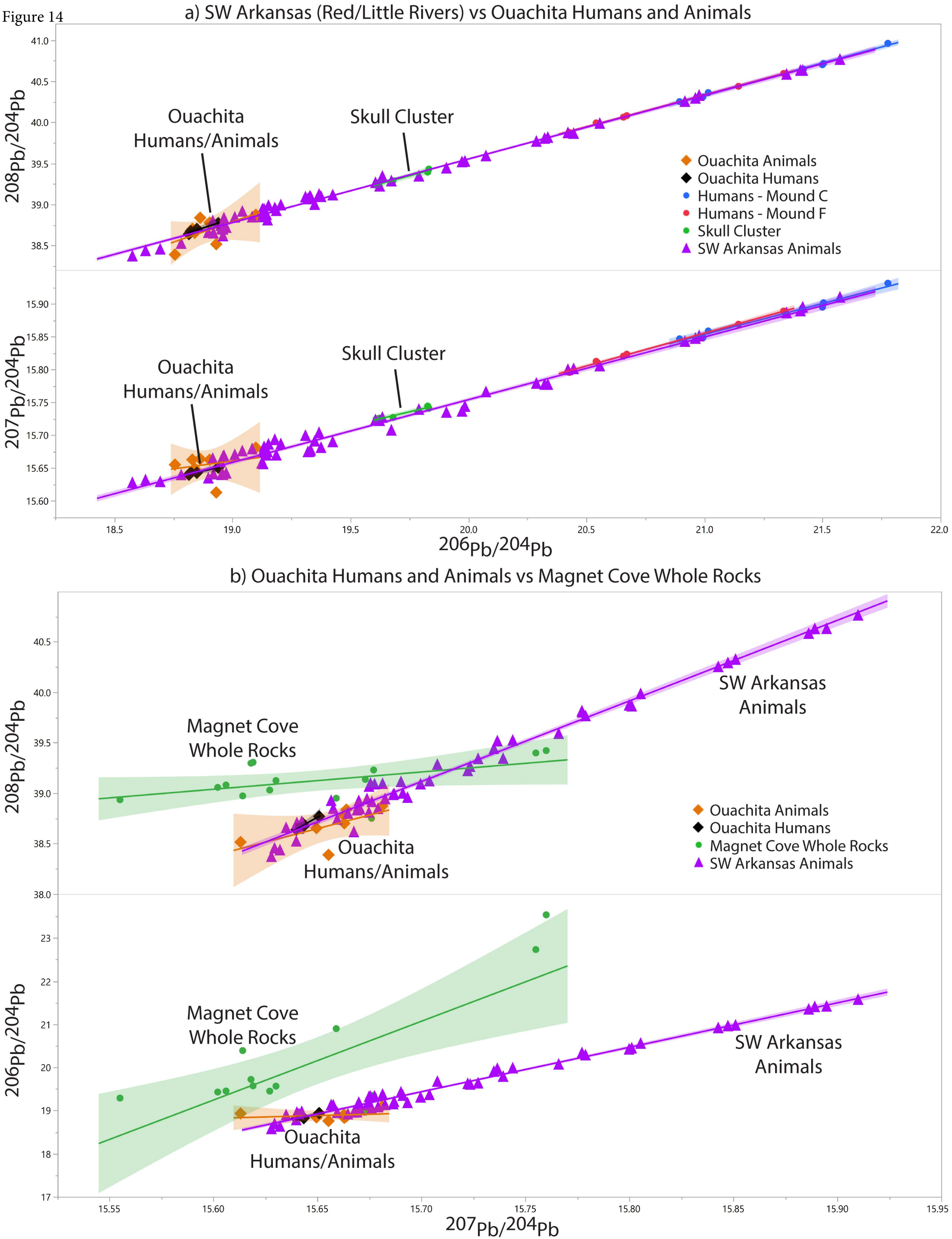

a) SW Arkansas (Red/Little Rivers) vs Ouachita Humans and Animals
208Pb/204Pb
207Pb/204Pb
206Pb/204Pb
Ouachita Humans/Animals
Skull Cluster
Ouachita Animals
Ouachita Humans
Humans - Mound C
Humans - Mound F
Skull Cluster
SW Arkansas Animals
b) Ouachita Humans and Animals vs Magnet Cove Whole Rocks
208Pb/204Pb
206Pb/204Pb
207Pb/204Pb
Magnet Cove Whole Rocks
SW Arkansas Animals
Ouachita Humans/Animals
Ouachita Animals
Ouachita Humans
Magnet Cove Whole Rocks
SW Arkansas Animals

Figure 15

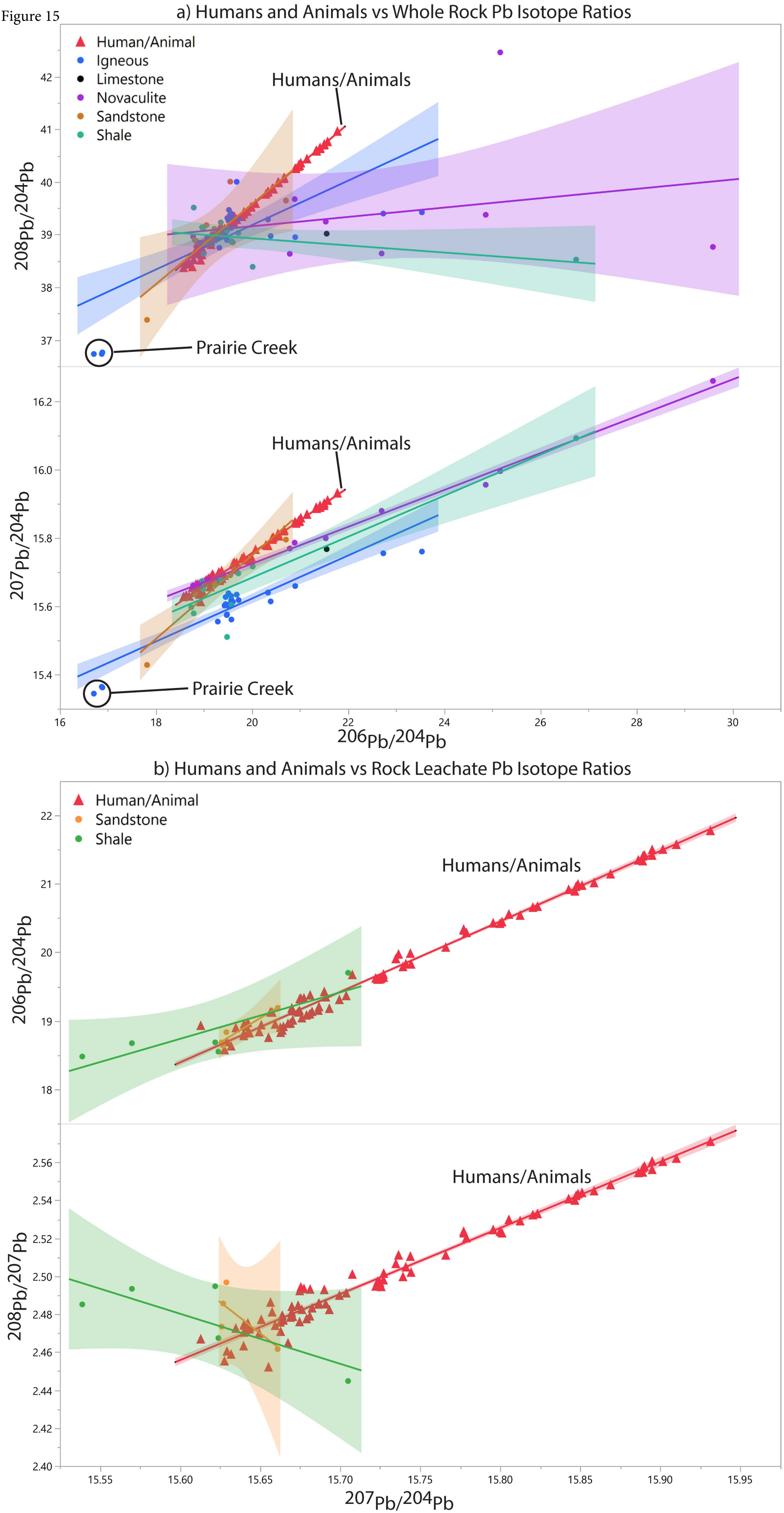

a) Humans and Animals vs Whole Rock Pb Isotope Ratios
Human/Animal
Igneous
Limestone
Novaculite
Sandstone
Shale
Humans/Animals
Prairie Creek
208Pb/204Pb
207Pb/204Pb
206Pb/204Pb
b) Humans and Animals vs Rock Leachate Pb Isotope Ratios
Human/Animal
Sandstone
Shale
Humans/Animals
206Pb/204Pb
208Pb/207Pb
207Pb/204Pb

Table 1. Lead Isotope Ratios of Human Teeth from Crenshaw (3MI6) and Hardman (3CL418).

Lab Number	Site	Catalog Number	Context	Tooth	Sex	Age	$^{208}Pb/^{204}Pb$	2*Std. Err.	$^{207}Pb/^{204}Pb$	2*Std. Err.	$^{206}Pb/^{204}Pb$	2*Std. Err.	Pb ppm
HU1	Crenshaw	62-40-34	Mound C	1st L Mand Molar	Not Possible	3-4	40.715	0.001	15.902	0.001	21.504	0.001	4.915
HU2	Crenshaw	62-40-47	Mound C	3rd L Mand Molar	Ambiguous	20-35	40.646	0.002	15.890	0.001	21.419	0.001	0.923
HU3	Crenshaw	62-40-48	Mound C	2nd L Mand Molar	Probable Male	35-50	40.309	0.002	15.849	0.001	20.994	0.001	0.371
HU4	Crenshaw	62-40-49	Mound C	2nd R Mand Molar	Not Possible	20-35	40.251	0.002	15.847	0.001	20.894	0.001	0.973
HU5	Crenshaw	62-40-50	Mound C	2nd R Mand Molar	Not Possible	20-35	40.360	0.002	15.859	0.001	21.016	0.001	0.470
HU6	Crenshaw	62-40-58	Mound C	2nd R Max Molar	Not Possible	5	40.960	0.001	15.931	0.000	21.777	0.001	31.482
HU7	Crenshaw	62-40-121	Mound C	1st R Mand Molar	Probable Male	50+	40.700	0.001	15.895	0.000	21.500	0.001	5.345
HU13	Crenshaw	83-376-1	Mound F	2nd L Mand Molar	Probable Female	20-35	40.594	0.001	15.889	0.001	21.336	0.001	1.207
HU14	Crenshaw	83-376-2	Mound F	2nd R Mand Molar	Not Possible	12	40.437	0.001	15.869	0.001	21.144	0.001	1.408
HU15	Crenshaw	83-376-3	Mound F	2nd L Mand Molar	Female	20-35	40.062	0.002	15.820	0.001	20.657	0.001	0.156
HU16	Crenshaw	83-376-6	Mound F	2nd R Max Molar	Not Possible	18	39.876	0.003	15.796	0.001	20.428	0.001	0.330
HU17	Crenshaw	83-376-6-2	Mound F	2nd R Max Molar	Not Possible	15	39.992	0.002	15.812	0.000	20.541	0.001	0.347
HU18	Crenshaw	83-376-7	Mound F	2nd L Max Molar	Probable Female	20-35	40.079	0.002	15.823	0.001	20.671	0.001	0.422
HU30	Crenshaw	69-66-589-1	Rayburn	2nd L Mand Molar	Probable Male	35-50	39.430	0.002	15.741	0.001	19.832	0.001	0.208
HU31	Crenshaw	69-66-589-2	Rayburn	2nd R Mand Molar	Probable Female	20-35	39.287	0.002	15.727	0.001	19.681	0.001	0.533
HU32	Crenshaw	69-66-589-3	Rayburn	2nd L Mand Molar	Probable Female	20-35	39.393	0.001	15.744	0.001	19.827	0.001	0.504
HU33	Crenshaw	69-66-589-4	Rayburn	2nd R Max Molar	Probable Male	Older adult	39.261	0.003	15.725	0.001	19.610	0.001	0.553
HU36	Crenshaw	69-66-589-7	Rayburn	2nd R Mand Molar	Possible Male	20-35	39.228	0.002	15.726	0.001	19.621	0.001	0.153
HU105	Hardman	87-710-274	Burial 2	2nd L Mand Molar	Probable Female	35-45	38.770	0.017	15.651	0.008	18.940	0.013	0.085
HU106	Hardman	87-710-326	Burial 7	2nd L Mand Molar	Probable Female	35-40	38.701	0.008	15.643	0.003	18.849	0.004	0.133
HU107	Hardman	87-710-708	Burial 12	2nd R Max Molar	Probable Male	39-44	38.674	0.006	15.643	0.003	18.824	0.003	0.090
HU108	Hardman	87-710-912	Burial 13	2nd R Max Molar	Probable Male	50+	38.638	0.008	15.640	0.004	18.816	0.004	0.082

Note : Age/sex classifications are from Burnett (1993) and Harvey et al. (2014).

Table 2. Lead Isotope Ratios of Prehistoric Animal Teeth from southwest Arkansas, northwest Louisiana, and northwest Mississippi.

Lab Number	Location	Catalog Number	Site Number	Animal	$^{208}Pb/^{204}Pb$	2*Std. Err.	$^{207}Pb/^{204}Pb$	2*Std. Err.	$^{206}Pb/^{204}Pb$	2*Std. Err.	Pb ppm
AN1	SW Arkansas	69-66-591	3MI6	Deer	40.306	0.003	15.847	0.001	20.974	0.001	1.515
AN2	SW Arkansas	69-66-587	3MI6	Rabbit	39.244	0.002	15.720	0.001	19.603	0.001	0.385
AN3	SW Arkansas	69-66-261	3MI6	Opossum	39.323	0.003	15.735	0.001	19.787	0.001	1.068
AN4	SW Arkansas	69-66-317	3MI6	Cottontail	39.569	0.005	15.762	0.001	20.071	0.001	0.235
AN5	SW Arkansas	69-66-230	3MI6	Wood Rat	38.595	0.007	15.663	0.003	18.956	0.003	0.260
AN6	SW Arkansas	69-66-389	3MI6	Swamp Rabbit	39.851	0.002	15.796	0.001	20.418	0.001	0.268
AN7	SW Arkansas	69-66-469	3MI6	Deer	39.201	0.005	15.718	0.002	19.620	0.003	0.047
AN8	SW Arkansas	69-66-389	3MI6	Deer	39.842	0.003	15.797	0.001	20.442	0.001	0.199
AN9	SW Arkansas	90-634	3MI6	Deer	40.609	0.004	15.885	0.001	21.403	0.003	0.366
AN10	SW Arkansas	90-634	3MI6	Deer	40.560	0.003	15.882	0.001	21.344	0.001	0.967
AN11	SW Arkansas	95-449	3MI6	Cottontail	40.741	0.002	15.905	0.001	21.570	0.001	1.114
AN12	SW Arkansas	90-634	3MI6	Swamp Rabbit	40.611	0.002	15.891	0.001	21.413	0.001	0.445
AN13	SW Arkansas	83-379-114	3HE92	Squirrel	40.269	0.003	15.843	0.001	20.957	0.001	1.433
AN14	SW Arkansas	82-450-20	3HE92	Pocket Gopher	38.788	0.003	15.670	0.001	19.145	0.001	3.154
AN15	SW Arkansas	83-379-264	3HE92	Rabbit	39.963	0.003	15.801	0.001	20.552	0.001	0.533
AN16	SW Arkansas	83-379-101	3HE92	Rabbit	39.505	0.004	15.740	0.002	19.982	0.002	0.203
AN17	SW Arkansas	83-379-141	3HE92	Rabbit	40.231	0.002	15.839	0.001	20.913	0.001	0.319
AN18	SW Arkansas	83-379-281	3HE92	Rabbit	39.745	0.004	15.775	0.001	20.284	0.002	0.172
AN19	SW Arkansas	84-380-158	3HE92	Pocket Gopher	38.978	0.003	15.687	0.001	19.345	0.001	1.295
AN20	SW Arkansas	2002-700-76	3HE40	Squirrel	38.638	0.004	15.631	0.001	18.895	0.001	0.321
AN21	SW Arkansas	2002-700-345	3HE40	Squirrel	38.698	0.003	15.638	0.001	18.970	0.001	0.274
AN22+	SW Arkansas	2003-685-84	3HE40	Squirrel	38.686	0.006	15.637	0.002	18.951	0.003	0.870
AN23	SW Arkansas	2002-700-34-5	3HE40	Squirrel	38.675	0.004	15.636	0.001	18.956	0.002	0.226
AN24	SW Arkansas	2002-700-34-5	3HE40	Squirrel	38.631	0.006	15.638	0.002	18.917	0.003	0.134
AN25	SW Arkansas	61-114-4686	3HO11	Raccoon	39.075	0.003	15.671	0.001	19.323	0.001	0.299
AN26	SW Arkansas	61-114-4686	3HO11	Opossum	39.065	0.003	15.672	0.001	19.327	0.001	0.443
AN27	SW Arkansas	61-114-607	3HO11	Small Rodent	38.902	0.003	15.651	0.001	19.126	0.001	0.270
AN28	SW Arkansas	61-114-685	3HO11	Rabbit	38.507	0.004	15.637	0.002	18.780	0.002	0.154
AN29	SW Arkansas	61-114-473	3HO11	Opossum	39.091	0.003	15.686	0.001	19.421	0.001	0.948
AN30+	SW Arkansas	61-114-676	3HO11	Deer	38.831	0.009	15.655	0.004	19.121	0.006	0.044
AN31	SW Arkansas	61-114-694	3HO11	Deer	39.076	0.006	15.678	0.002	19.372	0.004	0.077
AN32	SW Arkansas	61-114-638	3HO11	Deer	38.904	0.003	15.666	0.001	19.182	0.001	0.098
AN33	SW Arkansas	61-114-468a	3HO11	Deer	39.048	0.003	15.671	0.001	19.317	0.001	0.269
AN34	SW Arkansas	61-114-554	3HO11	Deer	38.856	0.003	15.666	0.002	19.141	0.002	0.096
AN35+	SW Arkansas	64-51-1	3SV20	Deer	39.422	0.008	15.731	0.003	19.903	0.004	0.040
AN36	SW Arkansas	64-51-1	3SV20	Rabbit	39.795	0.004	15.774	0.001	20.333	0.001	0.660
AN37	SW Arkansas	64-51-1	3SV20	Opossum	39.266	0.003	15.704	0.001	19.670	0.001	0.515
AN38	SW Arkansas	64-51-1	3SV20	Small Rodent	39.784	0.003	15.774	0.001	20.320	0.001	1.367
AN39	SW Arkansas	64-51-1	3SV20	Raccoon	39.497	0.003	15.733	0.001	19.969	0.002	0.214
AN40	SW Arkansas	63-39-278	3LR49	Rabbit	38.922	0.006	15.678	0.002	19.130	0.003	0.152
AN41	SW Arkansas	63-39-33	3LR49	Raccoon	39.069	0.003	15.696	0.001	19.306	0.001	0.343
AN42	SW Arkansas	63-39-57	3LR49	Raccoon	38.893	0.005	15.672	0.002	19.038	0.003	0.124
AN43	SW Arkansas	63-39-51	3LR49	Opossum	38.776	0.007	15.660	0.002	18.913	0.002	0.134
AN44	SW Arkansas	63-39-40	3LR49	Rabbit	38.414	0.005	15.627	0.001	18.628	0.003	0.154
AN45+	SW Arkansas	63-39-45	3LR49	Deer	39.097	0.009	15.699	0.003	19.363	0.006	0.048
AN46	SW Arkansas	63-39-43	3LR49	Deer	38.961	0.004	15.682	0.002	19.149	0.002	0.096
AN47+	SW Arkansas	63-39-63	3LR49	Deer	38.933	0.006	15.689	0.002	19.175	0.003	0.040
AN48	SW Arkansas	63-39-49	3LR49	Deer	38.823	0.005	15.675	0.002	19.079	0.002	0.057
AN49	SW Arkansas	64-50-3196	3SV15	Deer	39.319	0.003	15.723	0.001	19.631	0.001	0.099
AN50	SW Arkansas	64-50-252	3SV15	Deer	38.719	0.004	15.655	0.001	18.947	0.001	0.090
AN51+	SW Arkansas	64-50-203	3SV15	Deer	38.971	0.005	15.682	0.002	19.201	0.003	0.047
AN52+	SW Arkansas	64-50-341	3SV15	Deer	38.811	0.003	15.665	0.001	18.960	0.002	0.073
AN53+	SW Arkansas	64-50-325	3SV15	Deer	38.349	0.008	15.623	0.004	18.572	0.004	0.054
AN54	SW Arkansas	64-50-231	3SV15	Opossum	38.922	0.003	15.670	0.002	19.123	0.002	0.179
AN55+	SW Arkansas	64-50-319a	3SV15	Rabbit	38.824	0.007	15.667	0.003	19.006	0.003	0.090
AN56	SW Arkansas	64-50-437	3SV15	Rabbit	38.434	0.003	15.625	0.001	18.690	0.001	0.412

AN57 B	NW Mississippi	F-944	22TU549	Raccoon	39.170	0.009	15.693	0.004	19.480	0.004	0.259
AN58	NW Mississippi	F-2300	22TU549	Opossum	39.047	0.002	15.679	0.001	19.372	0.001	0.382
AN59	NW Mississippi	F-2300	22TU549	Raccoon	39.384	0.002	15.724	0.001	19.806	0.001	0.190
AN60	NW Mississippi	F-2300	22TU549	Raccoon	38.988	0.003	15.667	0.001	19.290	0.001	0.336
AN61+	NW Mississippi	F-799	22TU549	Deer	39.168	0.005	15.695	0.002	19.548	0.003	0.015
AN62+	NW Mississippi	F-2300	22TU549	Deer	39.010	0.004	15.674	0.001	19.331	0.002	0.055
AN63	NW Mississippi	F-2300	22TU549	Deer	39.405	0.004	15.728	0.002	19.846	0.002	0.055
AN64	NW Mississippi	F-1611	22TU549	Deer	39.393	0.002	15.725	0.001	19.815	0.001	0.153
AN65	NW Louisiana	16NA70-41	16NA70	Deer	38.903	0.002	15.669	0.001	19.108	0.001	0.262
AN66	NW Louisiana	16NA70-63	16NA70	Deer	38.895	0.001	15.672	0.000	19.082	0.000	0.675
AN67	NW Louisiana	16NA70-77	16NA70	Deer	38.908	0.002	15.676	0.001	19.102	0.001	2.133
AN68	NW Louisiana	16NA70-Nat_F	16NA70	Deer	38.876	0.001	15.667	0.000	19.090	0.001	0.375
AN69+	NW Louisiana	16NA70-27	16NA70	Rabbit	38.741	0.007	15.662	0.003	18.980	0.003	0.062
AN70	NW Louisiana	16NA70-115	16NA70	Rabbit	38.902	0.001	15.673	0.000	19.099	0.000	1.315
AN71	NW Louisiana	16NA70-104	16NA70	Beaver	38.910	0.001	15.674	0.000	19.104	0.001	6.080
AN72	NW Louisiana	16NA70-404	16NA70	Rabbit	38.895	0.003	15.671	0.001	19.107	0.001	0.516
AN149	Ouachita (SW Ark.)	87-710-954	3CL418	Deer	38.778	0.011	15.663	0.004	18.904	0.005	0.043
AN150+	Ouachita (SW Ark.)	87-710-647	3CL418	Deer	38.653	0.020	15.649	0.008	18.840	0.011	0.057
AN151+	Ouachita (SW Ark.)	87-710-95	3CL418	Deer	38.516	0.032	15.613	0.015	18.932	0.018	0.044
AN152	Ouachita (SW Ark.)	87-710-417	3CL418	Squirrel	38.873	0.004	15.681	0.002	19.099	0.002	0.334
AN153	Ouachita (SW Ark.)	87-710-86	3CL419	Squirrel	38.389	0.002	15.655	0.001	18.757	0.001	1.020
AN154*	Ouachita (SW Ark.)	74-746-14	3HS60	Deer	38.676	0.156	15.556	0.052	18.935	0.094	0.040
AN155	Ouachita (SW Ark.)	74-746-27	3HS60	Deer	38.837	0.007	15.664	0.003	18.863	0.005	0.060
AN156	Ouachita (SW Ark.)	74-746-28	3HS60	Opossum	38.701	0.018	15.663	0.007	18.830	0.009	0.467

Note: * Sample AN154 had high standard error and was generally excluded from analysis. + Blanks were under 3‰, but over 1‰, of these samples' concentrations.

Table 3. Pb Isotope Ratios and Pb, Cu, and Zn Concentrations of Soil Weak-acid Leachates (2N HCl) and Water Leachates (Ultra-pure Water) from Crenshaw.

Lab Number	Type	Context	Catalog Number	$^{208}Pb/^{204}Pb$	2*Std. Err.	$^{207}Pb/^{204}Pb$	2*Std. Err.	$^{206}Pb/^{204}Pb$	2*Std. Err.	Pb ppm	Cu ppm	Zn ppm
SO1	Weak-acid Leach.	Rayburn Cluster - soil outside cranium	69-66-589-6	39.165	0.001	15.718	0.000	19.494	0.000	1.539	0.386	1.791
SO1 Dup	Weak-acid Leach.	Rayburn Cluster - soil outside cranium	69-66-589-6	39.177	0.001	15.721	0.001	19.496	0.001	1.358	0.326	1.426
SO2	Weak-acid Leach.	Rayburn Cluster - soil inside cranium	69-66-589-6	39.332	0.001	15.730	0.001	19.632	0.001	1.482	0.557	1.614
SO3	Weak-acid Leach.	Rayburn Cluster - soil outside cranium	69-66-589-7	39.033	0.001	15.707	0.000	19.389	0.001	2.114	0.441	1.938
SO4	Weak-acid Leach.	Rayburn Cluster - soil inside cranium	69-66-589-7	39.208	0.001	15.724	0.000	19.545	0.000	1.683	0.453	1.695
SO5	Weak-acid Leach.	WSA Cluster 1 - soil	83-377-2-1	38.990	0.001	15.690	0.000	19.202	0.001	1.794	1.042	2.972
SO6	Weak-acid Leach.	WSA Cluster 1 - soil	83-377-2-2	39.052	0.001	15.695	0.000	19.235	0.000	2.010	0.975	1.707
SO7	Weak-acid Leach.	WSA Cluster 25 - soil	83-377-61-3	39.028	0.001	15.693	0.000	19.221	0.001	1.254	0.504	1.020
SO8	Weak-acid Leach.	NSA Cluster 8 - soil	83-377-41-4	39.164	0.001	15.710	0.000	19.400	0.000	2.569	1.222	2.697
SO9	Weak-acid Leach.	NSA Cluster 2 - soil	83-377-25-1	39.451	0.001	15.746	0.000	19.791	0.001	1.755	0.704	1.850
SO10	Weak-acid Leach.	Mound C - Center Block, Stratum A, soil	62-40-21a	39.967	0.001	15.813	0.001	20.500	0.001	5.841	2.076	4.892
SO10 B	Water Leach.	Mound C - Center Block, Stratum A, soil	62-40-21a	39.885	0.001	15.804	0.000	20.369	0.001	0.012	0.130	0.169
SO11	Weak-acid Leach.	Crenshaw Core 6 - 25cmbs, soil		39.017	0.001	15.690	0.000	19.186	0.000	3.607	1.896	2.526
SO12	Weak-acid Leach.	Crenshaw Core 6 - 50cmbs, soil		39.067	0.001	15.697	0.000	19.258	0.001	2.791	1.724	2.018
SO12 B	Water Leach.	Crenshaw Core 6 - 50cmbs, soil		39.067	0.001	15.691	0.000	19.241	0.000	0.012	0.069	0.120
SO13	Weak-acid Leach.	Crenshaw Core 6 - 125cmbs, soil		39.006	0.001	15.691	0.000	19.194	0.000	1.177	0.493	1.046
SO14	Weak-acid Leach.	Crenshaw Core 7 - 25cmbs, soil		39.063	0.001	15.697	0.001	19.257	0.001	2.941	1.519	2.540
SO15	Weak-acid Leach.	Crenshaw Core 7 - 50cmbs, soil		39.035	0.001	15.693	0.000	19.199	0.000	4.126	2.225	2.802
SO15 B	Water Leach.	Crenshaw Core 7 - 50cmbs, soil		39.052	0.003	15.684	0.001	19.201	0.001	0.004	0.073	0.384
SO16	Weak-acid Leach.	Crenshaw Core 17 - 25cmbs, soil		38.966	0.001	15.686	0.000	19.124	0.001	3.838	2.152	3.063
SO17	Weak-acid Leach.	Crenshaw Core 17 - 50cmbs, soil		39.003	0.002	15.688	0.001	19.139	0.001	2.918	2.171	2.700
SO18	Weak-acid Leach.	Crenshaw Core 17 - 125cmbs, soil		38.990	0.001	15.688	0.000	19.139	0.000	3.494	2.585	2.888
SO19	Weak-acid Leach.	Crenshaw Core 8 - 25cmbs, soil		39.202	0.002	15.715	0.001	19.447	0.001	4.527	2.042	3.814
SO20	Weak-acid Leach.	Crenshaw Core 8 - 50cmbs, soil		39.671	0.001	15.773	0.000	20.073	0.001	5.793	2.992	4.829
SO20 Dup	Weak-acid Leach.	Crenshaw Core 8 - 50cmbs, soil		39.671	0.001	15.773	0.001	20.071	0.001	5.176	2.656	4.196
SO20 B	Water Leach.	Crenshaw Core 8 - 50cmbs, soil		39.560	0.002	15.754	0.001	19.925	0.001	0.009	0.037	0.279

Note : SO1 and SO1 Dup were not homogenized as soil prior to sampling. SO20 and SO20 Dup were homogenized as soil prior to sampling.

Table 4. Lead Isotope Ratios of Whole Rocks from Granite Mountain and Magnet Cove.

Lab Number	Type	Location	$^{208}Pb/^{204}Pb$	2*Std. Err.	$^{207}Pb/^{204}Pb$	2*Std. Err.	$^{206}Pb/^{204}Pb$	2*Std. Err.
GM1	Whole Rock	Granite Mountain	40.002	0.001	15.634	0.000	19.681	0.001
GM2	Whole Rock	Granite Mountain	39.468	0.001	15.603	0.000	19.519	0.001
GM4	Whole Rock	Granite Mountain	39.384	0.004	15.601	0.002	19.564	0.002
GM5	Whole Rock	Granite Mountain	39.314	0.001	15.603	0.001	19.487	0.001
GM6	Whole Rock	Granite Mountain	39.361	0.002	15.606	0.001	19.538	0.001
GM7	Whole Rock	Granite Mountain	39.398	0.001	15.609	0.001	19.567	0.001
GM8	Whole Rock	Granite Mountain	39.381	0.001	15.609	0.000	19.555	0.001
GM9	Whole Rock	Granite Mountain	39.404	0.002	15.608	0.001	19.570	0.001
GM10	Whole Rock	Granite Mountain	39.381	0.002	15.606	0.001	19.571	0.001
GM11	Whole Rock	Granite Mountain	39.374	0.001	15.602	0.001	19.547	0.001
GM12	Whole Rock	Granite Mountain	39.325	0.002	15.600	0.001	19.520	0.001
MC2	Whole Rock	Magnet Cove	39.081	0.002	15.606	0.001	19.453	0.001
MC3*	Whole Rock	Magnet Cove	39.148	0.045	15.646	0.013	20.261	0.010
MC4	Whole Rock	Magnet Cove	39.297	0.001	15.618	0.000	19.721	0.000
MC6	Whole Rock	Magnet Cove	39.057	0.002	15.602	0.001	19.428	0.001
MC7	Whole Rock	Magnet Cove	39.306	0.002	15.619	0.001	19.572	0.001
MC8	Whole Rock	Magnet Cove	39.125	0.002	15.630	0.001	19.565	0.001
MC9	Whole Rock	Magnet Cove	39.031	0.001	15.627	0.001	19.450	0.001

Note : * Sample MC3 had high standard error and was generally excluded from analysis.

Table 5. Pb Isotopes and Trace Element Concentrations (ppm) of Duplicate Human Tooth Samples from Crenshaw (3MI6).

Lab Number	Catalog Number	$^{208}Pb/^{204}Pb$	2*Std. Err.	$^{207}Pb/^{204}Pb$	2*Std. Err.	$^{206}Pb/^{204}Pb$	2*Std. Err.	Pb	V	Mn	Fe	La	Ce	Nd	Dy	Yb	Th	U
HU4 B	62-40-49	40.369	0.005	15.860	0.002	21.072	0.003	0.240	0.722	2.855	16.581	0.020	0.034	0.028	0.003	0.001	0.001	0.000
HU7 B	62-40-121	40.734	0.002	15.902	0.001	21.538	0.001	2.792	1.962	10.442	11.310	0.052	0.024	0.034	0.005	0.002	0.001	0.011
HU13 B	83-376-1	40.587	0.004	15.883	0.002	21.353	0.002	0.351	0.384	4.084	19.585	0.024	0.027	0.026	0.002	0.002	0.001	0.007
HU15 B	83-376-3	40.111	0.008	15.824	0.003	20.737	0.004	0.121	0.243	2.000	10.510	0.012	0.016	0.013	0.002	0.001	0.001	0.002