Analyses of patterns of copper and lead mineralization in human skeletons excavated from an ancient mining and smelting centre in the Jordanian desert: a reconnaissance study

J. Grattan^{1,*}, L. Abu Karaki², D. Hine¹, H. Toland¹, D. Gilbertson³, Z. al-Saad² and B. Pyatt⁴

¹ Institute of Geography and Earth Sciences, University of Wales, Aberystwyth SY23 3DB, UK

² Faculty of Archaeology and Anthropology, Yarmouk University, Irbid, Jordan

³ School of Geography, University of Plymouth, Drake Circus, Plymouth PL4 8AA, UK

⁴ Interdisciplinary Biomedical Research Centre, School of Biomedical and Natural Sciences, Nottingham Trent University, Clifton Lane, Nottingham NG11 8NS, UK

ABSTRACT

In this reconnaissance study, skeletal materials from people, dating from ~1500 B.P., who lived by or worked at the ancient copper mines and furnaces of the Wadi Faynan in southern Jordan, were analysed using atomic absorption spectrophotometry (AAS) to determine the intensities of accumulation of copper and lead in their bones. Many of the bones analysed contained concentrations of these metals which are comparable to those of modern individuals who are heavily exposed to metals through contemporary industrial processes.

Patterns of partitioning throughout the skeleton of a number of individuals were also studied. These AAS data suggest that within the human organism there may be some ability to influence the patterns of accumulation of copper within the skeleton. The humerus was frequently found to contain more copper than other bones studied. Within the humerus itself, the medial epicondyle frequently contained the highest concentrations, which may indicate a significant degree of organization or control of the process. These metal concentration data together with their toxicological consequences suggest that the health of the ancient human populations must have been adversely affected by exposure during life to copper in the environment. They also point to the need for further detailed studies of metal partitioning within the bones of the human skeleton.

Keywords: copper mining, smelting, biomineralization, pollution, copper, lead, human skeleton, partitioning, Roman, Byzantine, health, desert, palaeoecology, Jordan.

Introduction

This investigation of human skeletal remains and associated sediments-soils excavated from graves of Byzantine age, $4^{th}-7^{th}$ centuries A.D., in the Wadi Faynan in the Jordanian Desert, examines the extent to which the geochemistry of human skeletal remains, and in particular of long bones, may reflect exposure during life to metal pollution from intense industrial activity. The paper addresses issues of metal accumulation and

* E-mail: John.Grattan@aber.ac.uk DOI: 10.1180/0026461056950277 partitioning in human skeletal materials that are difficult to achieve in modern anatomical or pollution studies. This work is part of a multidisciplinary study of the human and environmental impacts of copper mining and smelting around the ancient metallurgical centre of the Khirbet Faynan – the ancient Roman Phaeno (Feinan, Phinon: Knauf, 1992; Schick, 1995): studies started in 1996 and are ongoing (Grattan *et al.*, 2002, 2003*a*,*b*, 2004; Hunt *et al.*, 2004; McLaren *et al.*, 2004). Whilst this paper deals in detail with aspects of the accumulation of copper and lead within human skeletal remains, it also presents this new information within an integrated wider environmental and archaeological framework that begins in prehistory. Recognizably, industrial scales of exploitation of copper in this hyper-arid desert began as early as 4000 B.P. (Adams, 2002; Hauptmann, 2000; Levy et al., 2002, 2004) and small-scale production appears to have begun several millennia earlier (Al-Najjar et al., 1990). This evidence makes the Wadi Faynan one of the few places in the world which can be genuinely described as a birthplace of the first industrial revolution. Copper was one of the most important metal resources of the Ancient World. Initially it was used for decoration and then weaponry, and it remained important for the production of both household and prestige items. Control of mineral resources was as important in the ancient world as it is in the modern. Hence the copper mines of Favnan Orefield fell within either the economic orbit or direct political control of many of the small Kingdoms of the Levant and the great Empires of the Ancient World; the last great powers to wield direct control were the Roman and Byzantine Empires.

Substantive copper mining and smelting appear to have ceased in the $6^{th}-7^{th}$ centuries A.D., but in the preceding 2500 years the degree of exploitation was intense, on a scale not seen again until the Eighteenth Century A.D. industrial revolution in Europe. There remain in the desert landscape of the Wadi Faynan >250 copper mines, adits and mining galleries, >250,000 tonnes of metal-working slags from various periods of time, as well as innumerable ore and metal processing sites which remain the subject of research (Fig. 1).

Unlike the situation in many familiar sites of ancient copper mining and smelting in Mediterranean and temperate regions of Europe, where greater rainfall, leaching, geomorphic and biological cycling can be expected to progressively dilute the significance of metal contamination of the environment, the Wadi Faynan region with an average annual rainfall of only ~63 mm (Rabb'a, 1992) is in a quite different geoecological situation. The resulting sparse vegetation of this desert environment has neither stabilized nor grown over the ancient spoil. In consequence, the Faynan is an ideal location in which to study the long-term impacts of metal contamination of landscapes (Grattan et al., 2003a.b).

Archaeological research in the 1990s revealed one aspect of the human cost of these activities with the excavation of a late-Roman Christian cemetery reported by Findlater *et al.* (1998). The Wadi Faynan was known in the Roman world as the site of notorious copper mines and furnaces *'where even a condemned murderer may only live a few days'*. It was also the final destination of many Christians condemned to the mines during the times of the Roman persecution and of heretics after the Empire adopted Christianity (Eusebius, 1969).

The South Cemetery excavation

The excavation of the South Cemetery of the Wadi Faynan has provided the opportunity to study a human population engaged in, or indirectly exposed to, industrial activity in the ancient world. It allows us to gauge the skeletal consequences of this exposure during the life of the longer-term residents to metal contamination. The excavation was conducted as a rescue exercise by the British Institute for Archaeology and Ancient History in Amman and the Institute for Anthropology and Archaeology at the University of Yarmouk in response to the frequent professional looting of the site. By the time of the excavation, of 1200 graves in the cemetery, >700 had been looted.

The South Cemetery is located ~900 m to the southeast of Khirbet Faynan (Fig. 1), on a shallow slope 100 m above the Wadis Ghuweir (Ghuwayr) and Ashegar (Ashegair, Shayqar), which separate it from the main copper processing areas by the Khirbet (Freeman and McEwan, 1998). All the graves were found to be cut into gravelly-sandy loam of fanglomerates, derived entirely from the erosion of the Shara Mountains which lie immediately to the south and which were not an ore source. The graves excavated were all found to contain single inhumations, the skeletons recovered at depths between 1.20 and 2.40 m. All the bodies in the graves were laid out in a standard fashion, with the head to the west; the skeletal remains lying fully extended, on their backs, and with arms placed in the pelvic area. This combination of this alignment and the absence of significant grave goods suggested a Christian burial to the excavators. The bodies were usually placed within a carefully-cut slot in the base of the grave, which had been roofed with sandstone blocks. These stone slabs afforded the skeleton a degree of separation from the soil matrix of the area. This 'protection' together with the prevalent hyper-arid conditions, has led to excellent preservation of the skeleton, items of footwear and textiles, including clothing and

burial shrouds, as well as occasional body softtissues and hair (Findlater *et al.*, 1998). The field evidence indicated that the low rainfall and the shelter given by the stone-lined burial cists may have acted to inhibit post-mortem contact of bones with pore fluids and to have reduced the possibility of metal transfer from the soil to the skeleton.

Main characteristics of the ore deposit

The Faynan Orefield is mainly situated within the base of the distinctive mountain front that separates the hot and hyper-arid desert basin of the Jordanian Desert-Wadi Arabah to the west, from the wetter uplands and rugged terrain of mineralized granitic and sedimentary bedrocks of Early Palaeozoic and Precambrian age that crop out in the Mountains of Edom to the north and east (Barjous 1992). In total, the orefield covers an area of ~500 sq. km (Fig. 1) extending in the north from the Wadis el Ghuwēbe and el Jăriye,

through the Iron Age copper-working site of Khirbet en Nahas (Arabic: The Ruins of Copper), to the Wadis Faynan, Khalid, Ghuweir and Ashegar. To the west is the Wadi Fidan where there is extensive evidence of ancient copper working including an Early Bronze Age site of mass copper production at Khirbet Hamra Ifdan (Levy et al., 2002, 2004), whilst to the south is the very large copper mine of Umm el Amad (illustrated in Grattan et al., 2004). The Favnan Orefield appears to be the equivalent of the more widely known Timna Copper Ore field that is located ~90 km to the SSW, west of the Dead Sea Fault, from which the Faynan has been separated by strike-slip faulting (Klinger et al., 2000). The origin of the copper mineralization is still a matter of debate and is beyond the scope of this paper (cf. Barjous, 1992, pp. 61-62.)

Whilst copper is found in the Ghuwayr and Ahaymir Volcanic Suites these appear not to have been mined in either Roman or earlier times, the metal-working communities perhaps being



FIG. 1. Locations of mining and smelting centres and main settlements in the area of the Wadi Faynan, Hashemite Kingdom of Jordan (adapted from Hauptmann, 2000; Levy *et al.*, 2002).

unaware that copper could be extracted from these formations. In contrast, the copper-bearing Umm Ishrin Sandstone Formation (the 'Massive Brown Sandstones' of Hauptmann, 2000), and the Burj Dolomite-Shale Formation (the 'Dolomite Limestone Shales' of Hauptmann, 2000) were mined intensively. These were readily accessible using the mining technologies of the ancient world.

The copper ores in the Faynan Orefield are relatively pure. They consist mainly of malachite, chrysocolla, atacamite and chalcocite. Copper content varies with bedrock source, but ores with 35% copper can still be collected by hand (Barjous, 1992; Rabb'a, 1992; Hauptman and Weisberger, 1987; Hauptmann *et al.*, 1992; Overstreet *et al.*, 1982). Trace elements in the ore are typically <1%. Lead, however, reaches 6% in the copper ores from the Burj Dolomite/Shales, which was the ore source most heavily exploited in Roman times, but is much less in the Umm Ishrin Sandstones which were exploited by miners in earlier periods.

Metal cycling in the modern environment $- % \left({{\mathbf{x}}_{i}} \right)$ a paradigm for the ancient world

Although metal extraction and smelting effectively ceased at the site over 1500 years ago, the industrial waste by-products which remain in the landscape are largely unconsolidated and are available today for redistribution by aeolian, fluvial and biological processes. There is ample evidence that metals are indeed cycling through the modern environment (Pyatt et al., 1999, 2000, 2002a,b, 2005). One skeleton of a modern goat was shown to contain 163-405 µg of Pb/g of bone and 21-65 µg of Cu/g of bone; whilst modern vegetation contained 116-1025 µg of Pb/g of plant material and 71-603 µg Cu/g of plant material (Pvatt et al., 2000). Goat milk, urine and faeces also contained measurable copper, concentrations ranging from $3-10 \ \mu g/g$ (Pyatt et al., 1999). Up to 37 µg Pb/g of body tissue and 8 µg Cu/g of body tissue were also shown to have accumulated in both carnivorous and herbivorous invertebrates; snails, crickets, ants and spiders (Pyatt et al., 2002a).

The broadly contaminated nature of the contemporary environment is starkly illustrated in studies of metal particulate accumulation within Bedouin tents. Bedouin campsites are traditional, repeatedly occupied over hundreds of years. As a result of the combustion of metalcontaminated wood fuel and animal dung, copper in the dirt floors within studied campsites reached an upper value of 2480 μ g/g of sediment (Grattan *et al.*, 2003*a,b*), with inevitable consequences for the contamination of food cooked in such a domestic environment (Pyatt *et al.*, 2002*b*). It is clear that both copper and lead continue to cycle through the modern environment and that trophic accumulation of metals continues to occur today despite a gap of 1500 years since the copper smelters of the Wadi Faynan were last active. It is reasonable to hypothesize that all the pathways identified in the modern environment would have operated in ancient times, and perhaps at greater intensity at times of active metal production.

The Ancient environment and industrial contamination in Wadi Faynan

In addition to the trophic pathways noted as active in the modern world, the ancient population of Wadi Faynan would also have been exposed to a wide range of industrial processes ranging from direct involvement in mining, ore processing, smelting and transport; through to indirect exposure via smelting fumes, the deposition of metalliferous dusts on food and domestic surfaces, the contamination of water and the ingestion of sediment, food, crops or livestock, which had absorbed pollutant metals into their structures. Analyses of sediments/soils which have accumulated in archaeological features and at mine and smelting sites, suggest that by as early as 1000 B.C. the region was highly polluted: measured copper and lead concentrations reaching as high as 11961 µg Cu/g of soil and 15205 µg Pb/g of soil (Pyatt et al., 2000). These contaminant concentrations would invoke legislation to require intervention today in some countries (Canadian Council of Ministers of the Environment, 1991).

²²²Rn gas emanating from primary and secondary mineral sources in the granitic and metamorphic basement rocks has been measured in the mines (Grattan *et al.*, 2004) and an added problem for those involved in the mining could have been radon-induced cancers. However, this is likely to have been a problem only for the relatively longer-lived specialist mine overseers and engineers.

Many professionals of Roman times, and no doubt their Byzantine successors, were likely to be aware of the dangers posed by mining and smelting pollution: '*What stenches Scapentulum* (a mine) breathes out underground…How strange they make men's faces, how they change their colour. Have you not seen or heard how they are wont to die in a short time and how the powers of life fail'; 'they build their … furnaces with high chimneys, so that the fumes from the ore may be carried into the air for it is heavy and deadly' (Hughes, 1996). It is no wonder that those people the Roman State wished to kill, but not execute, were condemned to the mines of Faynan (Schick, 1995).

Previous analyses of human bone of Byzantine Age from the Wadi Favnan region have looked at either relatively small data sets (Grattan et al., 2003a) or have focused on maximum levels of accumulation (Grattan et al., 2002). In these studies, the Byzantine inhabitants of Wadi Faynan were shown to have concentrations of lead and copper in their skeletons which were comparable with, and often in excess of the modern studies of a population in Silesia described by Baranowska et al. (1995). The highest copper concentrations reported for Silesia were 5.17 µg Cu/g of bone, with an average of only 0.59 µg Cu/g of bone. These figures contrast with the average value determined from the Favnan material in these earlier studies of 52.6 µg Cu/g of bone, the highest being 296 µg Cu/g of bone. The maximum lead values for Silesia and Faynan were broadly comparable: 205 and 289 µg Pb/g of bone, respectively (Grattan et al., 2002). Human skeletal material from a Bronze Age cemetery elsewhere in the Wadi Fidan also contained high concentrations of lead and copper. Pvatt et al. (2005) reported lead values of 98 µg Pb/g of femur and 35 µg Pb/g of cranium and copper concentrations of 108 µg Cu/g of femur and 103 µg Cu/g of cranium.

All these analyses present a broad picture of the dramatic accumulation of lead and copper in human bone in antiquity, which is useful for drawing comparisons with contemporary industrial pollution, but which throws little light on the uptake of metals around the whole skeleton; this present paper seeks to address this issue.

Analytical methodology

The skeletal remains of individuals from a series of graves dating from the Byzantine Era, $4^{\text{th}}-7^{\text{th}}$ century A.D., were excavated in Wadi Faynan in 1996 (Fig. 1 and Findlater *et al.*, 1998, Karaki, 1999). These are stored in the University of Yarmouk, at Irbid in the Hashemite Kingdom of

Jordan. Associated sediment/soil samples excavated with the skeletons were also investigated from this archive in order to establish a background metal content of the grave soils/sediments: a source which could serve as one measure of the possible post-mortem, diagenetic contamination of the bone within the ground. The skeletons were sub-sampled in the University of Yarmouk, where possible following the idealized sampling procedure presented in Fig. 2, which was designed by Dr Jason Maher of the University of Bradford to ensure a broad coverage of bone groups and types. In practice, the entire bone/joint was removed from the skeleton and then dispatched to Abervstwyth for analysis. The vascularity of each sample was not controlled in this study. The analyses took place in three stages. In Stage 1, bones from 25 individuals have been analysed in order to establish the broad concentrations of lead and copper in each individual skeleton. In Stage 2, building on the ideas presented recently by Skinner (2000), further skeletal material from 11 individuals, was selected for a multi-bone study designed to explore whether or not patterns of partitioning could be established within the skeletal material of each individual. Subsequently, in Stage 3, the lead and copper contents of the humerus of 17 individuals were examined in more detail.

The bones selected for analysis were pre-cleaned with a soft brush to remove any remaining sediment/soil or other detritus. Sub-samples of the sectioned bones were removed and placed in an ultrasonic bath for 5 min. These sub-samples inevitably included both the exterior surface and the interior bone matrix. The cleaned bone samples were then placed under protective covers in a dustfree atmosphere and allowed to air-dry for 48 h. The samples were weighed, bagged and re-labelled ready for analysis. These were crushed in a clean, sterile mortar to give a fine powder, which was then transferred to sterile resealable plastic bags. From these bags 1±0.0005 g of powder were weighed out precisely. The measured quantity of powder was then added to individually-labelled 50 ml glass beakers, to which 5 ml of Milli-Q distilled water and one measure of 1 M strength HCl were added in a fume cupboard. Watch glasses were used to cover the beakers to stop any vapours escaping. The beakers with their watch glass lids were then transferred to a hotplate set at ~80°C. Care was taken not to allow these solutions to boil. Once all the bone powder had been digested, the beakers were removed from the



J. GRATTAN ET AL.

- 1 Section taken from the centre of the parietal bone, close to the parietal boss.
- 2 Section taken at the gonial angle of the mandible (corner of the jaw).
- 3 Section taken from the centre of the humeral head proximal articulation.
- 4 Section taken from the inferior portion of the bicipital groove.

5 Section taken from medial margin of the distal articular surface of the humerus.

- 6 Sample taken from the most superior portion of the iliac crest.
- 7 Sample taken from interosseus crest at mid-shaft of radius.

8 Sample taken from the centre of the femoral head, superior to the fovea capitis.9 Sample taken from the most superior portion of the greater trochanter of the femur.

- 10 Sample taken from the linea aspera at the midshaft of the femur.
- 11 Sample taken from the most lateral portion of the lateral margin of the lateral condyle of the femur.
- 12 Sample taken from the medial margin of the medial condyle of the tibia.
- 13 Sample taken from the mid-shaft of the tibia, anterior portion.

14 Sample taken from the posterior portion of the calcaneus, calcaneal tuber, insertion of the Achilles, tendon.

15 Sample taken by removing spinous process at junction of laminae from C3-6 (one sampled, which ever was present and could be identified).

16 Sample taken by removing spinous process at junction of laminae from T2-11 (one sampled, which ever was present and could be identified).

17 Sample taken by removing spinous process at junction of laminae from L2-4 (one sampled, which ever was present and could be identified).

18 Sample taken from anterior margin of the superior, articular surface on S1 (sacral articulation with Lumber vertebra 5)

FIG. 2. Idealized bone sampling strategy adopted.

hotplate and allowed to cool. During this process, blank solutions were made up at random intervals, to regulate any possible errors or contaminations. After the solutions had cooled they were passed through Whatmann filters into volumetric flasks. 40 ml of Milli-Q distilled water was used to help flush the samples through the filters, creating a final ~10% HCL (v/v) 50 ml solution. After filtration, each solution was transferred from the volumetric flask to a plastic bottle with a screw-top lid and labelled accordingly.

Samples and blanks were then analysed by flame atomic absorption spectrophotometry using a Perkin-Elmer 3110 instrument. After every 10 analyses the standard solutions were used to check for instrument drift and error and to ensure the precision and accuracy of the analyses. On the basis of this, the estimated error in our dataset is 2.41 µg/g for Cu and 5.39 µg/g for Pb and the detection levels are 0.1 µg/g for Cu and 0.5 µg/g for Pb.

Results

The general characteristics of the exposure of ancient people to lead and copper demonstrated in

these new analyses (Fig. 3a,b) are comparable to those reported by Grattan *et al.* (2002) and suggest exposure to lead and copper on a scale greater than detected in other ancient and modern benchmarks (Ahlgren *et al.*, 1976; Ahlgren and Mattsson 1979; Baranowska *et al.*, 1995; Drasch, 1982). It is clear that there is considerable variability in the present Wadi Faynan data set. This may reflect the degree to which individuals were directly involved in metal processing and perhaps is inevitable given that the personal histories of the individuals concerned is unknown.

Evidence of the differential uptake of metals within 11 skeletons studied in greater detail is suggested in the data presented in Figs 4 and 5. This reconnaissance study has not been able to analyse each bone in the skeletons of the individuals studied, but within these data, the medial condyle of the humerus appears noteworthy for its uptake of copper (Fig. 4a,b). In nine of the 11 individuals studied, the humerus has a greater uptake of copper than any other bone analysed, whilst in the remaining two skeletons it had the second greatest uptake. This pattern of enhancement is clear regardless of the overall concentration detected in each individual;

CU AND PB MINERALIZATION IN ANCIENT HUMAN SKELETONS, JORDAN





FIG. 3. (*a*) Maximum Cu concentrations detected in 25 individuals including those presented in detail in Fig. 4*a,b*. The dotted line indicates the highest bone Cu content of a modern comparative population (after Baranowska *et al.*, 1995). (*b*) Maximum Pb concentrations detected in 25 individuals including those presented in detail in Fig. 5*a,b*. The dotted line indicates the average Pb content of a Roman population (after Drasch, 1982).



FIG. 4. (*a*) Skeletal partitioning of Cu. The error bars displayed are calculated on the basis of the difference between original and blind reanalysis of material from each grave. Where not visible, the error bars are smaller than the symbols used to show data points. (*b*) Skeletal partitioning of Cu. The error bars displayed are calculated on the basis of the difference between original and blind reanalysis of material from each grave. Where not visible, error bars are smaller than the symbols used to show data points.



CU AND PB MINERALIZATION IN ANCIENT HUMAN SKELETONS, JORDAN

FIG. 5. (*a*) Skeletal partitioning of Pb. Error bars displayed are calculated on the basis of the difference between original and blind reanalysis of material from each grave. Where not visible, error bars are smaller than the symbols used to show data points. (*b*) Skeletal partitioning of Pb. Error bars displayed are calculated on the basis of the difference between original and blind reanalysis of material from each grave. Where not visible, the error bars are smaller than the symbols used to show data points.

particularly in graves 74, 86 and 102. Systematic partitioning trends for lead are not as clear (Fig. 5a,b). The humerus contained the maximum concentrations of lead in only two of the individuals analysed. These trends and their implications are discussed in more detail below.

Trends in metal partitioning within the humerus can also be seen in Tables 1*a* and 1*b*. Of the 17 individuals for whom these data are available, the highest copper concentrations occur in the medial epicondyle in 16 individuals. In contrast, the medial epicondyle contains the highest concentrations of lead in only seven of the 17 skeletons analysed.

Discussion

Life in a contaminated landscape

It is clear from these metal concentrations in human skeletal remains that the Late-Roman inhabitants of Wadi Faynan lived and were buried in an environment which was profoundly contaminated by copper and lead derived from the preceding 1700 years of intensive mining and smelting. Future studies of this material will seek to investigate further any differences related to the age, sex and status of the person represented by the bone sample. Importantly, at present there are no means by which to assess the extents to which individuals were associated with industrial tasks Nevertheless, the copper content of these individuals' bones significantly exceeds the values reported by Baranowska et al. (1995) for a modern human population living in Silesia that is thought to be at substantial risk from local metal extraction and working processes. The highest copper value reported for Silesia was 5.17 ug Cu/g bone, which contrasts with the highest value in this study, 119 µg Cu/g bone (Fig. 3a, Grave 101). Drasch (1982) measured the lead uptake in the bones of a late-Roman population between 0.9 and 13.0 μ g/g bone; this concentration being the result of drinking and eating from lead utensils and using water carried by lead-lined conduits. The inhabitants of the arid industrial area and 'penal colony' of Wadi Faynan

TABLE 1. (a) Cu partitioning within the humerus ($\mu g/g$ of bone). (b) Pb partitioning within the humerus ($\mu g/g$ of bone).

Grave			(a)				(b)			
	Medial Epicondyle	Mid shaft	Head	Grave	Medial Epicon- dyle	Mid shaft	Head			
10	22.5	14.0	16.5	10	49	45.5	39.5			
11	8.68	6.10	no data	11	14.4	14.8	no data			
12	23.6	no data	20.4	12	21.6	no data	22.8			
63	22.0	no data	18.0	63	52.5	no data	40.5			
71	20.0	18	19.0	71	51.5	50	51.0			
72	10.1	8.90	no data	72	31.2	36.8	no data			
73	20.6	11.6	10.8	73	4.35	2.48	3.11			
74	17.5	13.5	17.5	74	49.5	45.5	49.0			
77	17.2	18.0	18.4	77	16.8	26.8	26.8			
80	11.6	8.64	no data	80	18.4	22.0	no data			
81	9.7	5.96	no data	81	20.4	20.4	no data			
84	14.5	no data	14.5	84	48.0	no data	50.5			
86	20.5	no data	15.5	86	51.5	no data	52.5			
95	20.8	no data	17.2	95	25.6	no data	18.8			
97	17.7	7.41	12.2	97	8.22	6.85	5.86			
102	79.1	60.0	50.4	102	61.8	56.4	42.5			
110	24.0	16.0	17.5	110	49.5	44.5	51.5			
Max	79.1	60.0	50.4	Max	61.8	56.4	52.5			
Min	8.68	5.96	10.8	Min	4.40	2.50	3.10			
Median	20	12.6	17.5	Median	31.2	31.8	40.5			
Average	21.2	15.7	19.1	Average	33.8	31.0	35.0			
Standard	15.8	14.7	9.77	Standard	18.6	17.9	17.6			
dev.				dev.						

present little evidence of a population enjoying a middle-class lifestyle and may have had little domestic exposure to lead, yet in most cases the lead concentration in their skeletons comfortably exceeds the concentrations identified by Drasch (Fig. 3b). A more immediately comparable population was studied by Ahlgren et al. (1976) and Ahlgren and Mattsson (1979), who determined the lead contents in the bone of modern Swedish metal workers and found that the concentrations ranged between 40 and 100 ug of Pb/g of bone. In the industrial Wadi Faynan in Byzantine times, the lead concentrations in these ancient human skeletal remains range between 2.48 and 63 ug of Pb/g of bone. This suggests a wider range of exposures, but it should be noted that it is unlikely that all the inhabitants of Wadi Favnan studied here, would necessarily have been directly engaged in industrial activity and represent a wider cross-section of a community than those investigated in Ahlgren's studies. The copper and lead concentrations reported here for the Faynan are also likely to have been associated with a wide range of debilitating sickness and possibly early death; discussed in Grattan et al. (2002, 2003*a*.*b*: 2004).

The uptake of copper and lead within the skeleton

The combination of overall aridity of soils in the area and the particular design of each grave with capping and lining stones may both have acted to reduce contact between the skeleton and pore waters in adjacent materials. Sediment and bone data provide another approach to this important issue. Taking the metal concentrations for the cemetery soils as copper: 1.70 µg of Cu/g of soil; and lead: 1.40 µg of Pb/g of soil (Grattan et al., 2002), it is possible to determine an average bonemetal/soil-metal ratio for Faynan cemeteries, which are for copper, 31:1; and for lead, 30:1. These ratios are far greater than that of 3:1 used by Oakberg et al. (2000) to suggest the presence of early copper smelters in Shigmim, Israel across the Wadi Arabah from the Faynan. Whilst the possibility that the metal burdens of co-eval sediment/soil conditions have influenced the chemistry of these ancient bones (Pike et al., 2002) cannot be entirely discounted, the burial environment, soil conditions and the emerging patterns of differential uptake presented here indicate significant ante-mortem uptake of these metals. On the assumption on the grounds described above that there has been only either

limited or no major post-mortem modification of the metal content in the skeletal material studied. these new geochemical data suggest organized uptake of copper in the human skeleton has taken place to an extent which has not been noted before. Clearly the trends apparent in Figs 4 and 5 and Tables 1a and 1b should be treated with caution until a larger sample of material can be obtained and analysed, nevertheless, the emerging results are striking. In 80% of the individuals studied, copper concentrations were highest in the humerus and in the remaining 20% the humerus contained the second highest concentrations detected. In contrast, no such trends are readily apparent in respect of lead concentrations (Fig. 5a,b; Table 1b). These patterns of metal enhancement are apparent whether the individual skeleton contains large or small body burdens of lead and copper. The implications of these patterns of metal partitioning are considerable. Copper is a necessary trace element, whereas lead serves no useful biological function (Alloway and Avres, 1993; Scheinberg, 1979; Wittmers et al., 1988). As a result, following Skinner (2000), it is suggested that human physiology has better control over the ultimate skeletal destination of copper in the human body than lead; perhaps it is more readily and usefully incorporated into the inorganic (apatite) or organic parts of the bone such as collagen (Skinner, 2000 and pers. comm.). Differential uptake of lead and copper in vertebrate skeletons was also observed by Pyatt et al. (2000, Table 3) in their study of modern goats from Wadi Faynan. Similar patterns have been likewise observed in the bones of ancient goats retrieved from archaeological excavations in the Wadi Faynan area (Table 2 in Pyatt et al., 2005). Why the humerus is the most common destination for copper in the bones analysed in this study is not clear. This question is not resolvable using solution analysis techniques and a further study by the authors will attempt to identify and map the location of metal within each bone.

Partitioning within the humerus

In many respects the patterns of partitioning of copper and lead within the humerus (Table 1a,b) display similar trends to those discussed above for the wider cross-section of human skeletal material presented in Figs 4 and 5. In each humerus analysed, copper was shown to be partitioned in a similar fashion from one individual to the next,

whilst lead was more chaotically distributed. These analyses show that the medial condyle contained more copper than other parts of that bone. Why should this be so? It might be suggested that this area of the bone displays these effects because it has a rich blood supply since it anchors important muscle groups and perhaps also recycles and builds bone more frequently than other studied bone locations. But if this is the case why are the similar areas of similar bones such as the femur and tibia not similarly enriched? Further analyses by the authors of the bone matrix using electron microprobe mapping techniques which focus on more than just the two metals discussed will be undertaken to answer these questions, as are more extensive sampling and analysis of each individual bone of the skeleton.

Conclusions

These analyses have confirmed earlier studies which suggested that the skeletal remains of inhabitants of Wadi Faynan in Byzantine times contained concentrations of lead and copper that are comparable to and in fact higher in some cases than concentrations found in modern industriallycontaminated populations. The burial environment is such that these metal enrichments are believed to have happened mainly ante-mortem.

Investigations of patterns of metal partitioning around the human skeleton have suggested that for these ancient human skeletal remains, copper is preferentially incorporated into the humerus and within the humerus to the site of the medial epicondyle. It appears that the human organism may have some limited control over the uptake of copper within the skeleton, but no such pattern was evident in the uptake of lead. Further analyses will seek to more fully examine these initial conclusions by mapping the concentration of all metals in a wider range of skeletal material, rather than the limited selection of bones available for use in this present study.

Acknowledgements

The authors would like to acknowledge the help and assistance of the Bedouin of Wadi Faynan. This paper could not have been written without the support and advice of the editor, Eva Valsami-Jones, and I am indebted to the reviewers, Catherine Skinner and Clive Trueman, whose comments considerably improved this paper.

References

- Adams, R. (2002) From farms to factories: The development of copper production at Faynan, southern Jordan, during the Early Bronze Age. Pp. 21-32 in: *Metals and Society* (B.S. Ottaway and E.C. Wagner, editors). British Archaeological Reports, International Series. Archaeopress, Oxford, UK.
- Ahlgren, L. and Mattsson, S. (1979) An X-ray fluorescence technique for in vivo determination of lead concentration in a bone matrix. *Physics Medicine Biology*, 24, 136–145.
- Ahlgren, L., Liden, K., Mattsson, S. and Tejning, S. (1976) X-ray fluorescence analysis of lead in human skeleton in vivo. Scandinavian Journal of Work, Environment and Health, 2, 82–86.
- Al-Najjar, M., Abu Dayya, A., Suleiman, E., Weisburger, G. and Hauptmann, A. (1990) Tell Wadi Feinan. The first Pottery Neolithic tell in the south of Jordan. *Annals of the Department of Antiquities of Jordan*, 34, 27–56.
- Alloway, B.J. and Ayres, D.C. (1993) *Chemical Principles of Environmental Pollution*. Blackie Academic and Professional, London, 291 pp.
- Baranowska, I., Czernicki, K. and Aleksandrowicz, R. (1995) The analysis of lead, cadmium, zinc, copper and nickel content in human bones from the Upper Silesian industrial district. *Science of the Total Environment*, **159**, 155–162.
- Barjous, M.O. (1992) The Geology of the Ash Shawbak area. Map Sheet no. 3151 III. Amman, Geology Directorate, Geological Mapping Division, Bulletin 19, 79 pp.
- Canadian Council of Ministers of the Environment (1991) Canadian Environmental quality criteria for contaminated sites. Report CCME EPC-CS34. Winnipeg, Manitoba, Canada.
- Drasch, G.A. (1982) Lead burden in prehistorical, historical and modern bones. *The Science of the Total Environment*, 24, 199–231.
- Eusebius (1969) *The History of the Church from Christ to Constantine*. Harmondsworth, Penguin Books, London, 469 pp. (translated by G.A. Williamson).
- Findlater, G., El-Najjar, M., Al-Shiyab, A., O'Shea, M. and Easthaugh, E. (1998) The Wadi Faynan project: the south cemetery excavation, Jordan 1996. *Levant*, **30**, 69–83.
- Freeman, P.W.M. and McEwan, L.M. (1998) The Wadi Faynan Survey, Jordan: a preliminary report on Survey in area WF2 in 1997. *Levant*, **30**, 61–68.
- Grattan, J.P., Pyatt, F.B., Toland, H.T. and Huxley, S. (2002) 'Death … more desirable than life'? The human skeletal record of ancient copper mining and smelting in Wadi Faynan, South Western Jordan. *Toxicology and Industrial Health*, **18**, 297–307.
- Grattan, J.P., Condron, A., Taylor, S., Karaki, L.A.,

Pyatt, F.B., Gilbertson, D.D. and Saad, Z. (2003*a*) A legacy of Empires? An exploration of the environmental and medical consequences of metal production in Wadi Faynan, Jordan. Pp. 99–105 in: *Geology and Health: Closing the Gap* (H.C.W. Skinner and A. Berger, editors). Oxford University Press, UK.

- Grattan, J.P., Huxley, S. and Pyatt, F.B. (2003b) Modern Bedouin exposures to copper contamination: an imperial legacy? *Ecotoxicology and Environmental Safety*, 55, 108–115.
- Grattan, J.P., Gillmore, G.K., Gilbertson, D.D., Pyatt, F.B., Hunt, C.O., McLaren, S.J., Phillips, P.S. and Denman, A. (2004) Radon and King Solomon's miners, Faynan orefield, Jordanian Desert. *The Science of the Total Environment*, **319**, 99–113.
- Hauptmann, A. (2000) Zur frühen Metallurgie des Kupfers in Fenan/Jordanien. Der Anschnitt Beiheft 11. Deutsches Bergbau Museum, Bochum, Germany, 239 pp.
- Hauptmann, A. and Weisburger, G. (1987) Archaeometallurgical and mining-archaeological investigations in the area of Fainan, Wadi 'Arabah (Jordan). Annals of the Department of Antiquities of Jordan, 31, 419–437.
- Hauptmann, A., Begemann, F., Heitkemper, E., Pernicka, E. and Schmitt-Steker, S. (1992) Early copper produced at Feinan, Wadi Araba, Jordan: the composition of ores and copper. *Archaeomaterials*, 6, 1–33.
- Hunt, C.O., Elrishi, H.A., Gilbertson, D.D., Grattan, J.P., McLaren, S., Pyatt, F.B., Rushworth, G. and Barker, G.W. (2004) Early Holocene environments in the Wadi Faynan, Jordan. *The Holocene*, **14**, 921–930.
- Hughes, J.D. (1996) *Pan's Travail*. Johns Hopkins University Press, Baltimore, Maryland, USA, 276 pp.
- Karaki, L.O.A. (1999) Skeletal Biology of the People of Wadi Faynan. A Bioarchaeological Study. Unpublished M.A. dissertation, Yarmouk University, Jordan, 114 pp.
- Klinger, Y., Avouac, J.P., Abou Karaki, N., Dorbath, L., Bourles, D. and Reyss, J.L. (2000) Slip-rate on the Dead Sea transform fault in northern Araba Valley (Jordan). *Geophysical Journal International*, **142**, 755–768.
- Knauf, E.A. (1992) Feinan, Wadi. Pp. 780–782 in: *The Anchor Bible Dictionary* (D.N. Freedman, editor). Doubleday, New York.
- Levy, T.E., Adams, R.B., Hauptmann, A., Prange, M., Schmitt-Strecker, S. and Najjar, M. (2002) Early Bronze Age metallurgy: a newly discovered copper manufactory in southern Jordan. *Antiquity*, **76**, 425–437.
- Levy, T.E., Adams, R.B., Najjar, M., Hauptmann, A., Anderson, J., Brandl, B., Robinson, M.A. and

Higham, T. (2004) Reassessing the chronology of Biblical Edom: new excavations and ¹⁴C dates from Khirbat en-Nahas (Jordan). *Antiquity* **78**, 865–879.

- McLaren, S.J., Gilbertson, D.D., Grattan, J.P., Hunt, C.O., Duller, G.A.T. and Barker, G.A. (2004) Quaternary palaeogeomorphic evolution of the Wadi Faynan area, southern Jordan. *Palaeogeography Palaeoclimatology Palaeoecology*, 205, 131-154.
- Oakberg, K., Levy, T. and Smith, P. (2000) A method of skeletal arsenic analysis, applied to the Chalcolithic copper smelting site of Shiqmim, Israel. *Journal of Archaeological Science*, 27, 895–901
- Overstreet, W.C., Grimes, D.J. and Seitz, J.F. (1982) Geochemical orientation for mineral exploration in the Hashemite Kingdom of Jordan. USGS OFR 82–791, 255 pp.
- Pike, A.W.G. and Richards, M.P. (2002) Diagenetic arsenic uptake in archaeological bone. Can we really identify copper smelters? *Journal of Archaeological Science*, **29**, 607–611.
- Pyatt, F.B., Barker, G.W., Birch, P., Gilbertson, D.D., Grattan, J.P. and Mattingly, D.J. (1999) King Solomon's miners – starvation and bioaccumulation? An environmental archaeological investigation in southern Jordan. *Ecotoxicology and Environmental Safety*, **43**, 305–308.
- Pyatt, F.B., Gilmore, G., Grattan, J.P., Hunt, C.O. and McLaren, S. (2000) An Imperial legacy? An exploration of ancient metal mining and smelting in Southern Jordan. *Journal of Archaeological Science*, 27, 771–778.
- Pyatt, F.B., Amos, D., Grattan, J.P., Pyatt, A.J. and Terrel-Nield, C.E. (2002a) Invertebrates of ancient heavy metal spoil and smelting tip sites in southern Jordan: their distribution and use as bio indicators of metalliferous pollution derived from ancient sources. *Journal of Arid Environment*, **52**, 53–62.
- Pyatt, F.B., Pyatt, A.J. and Grattan, J.P. (2002*b*) A public health problem? Aspects and implications of the ingestion of copper and lead contaminated food by Bedouin. *Environmental Management and Health*, **13**, 467–470.
- Pyatt, F.B., Pyatt, A.J., Walker, C., Sheen, T. and Grattan, J.P. (2005) Environmental toxicology: heavy metal content of skeletons from an ancient metalliferrous polluted area of Southern Jordan with particular reference to bioaccumulation and human health. *Ecotoxicology and Environmental Safety*, **60**, 295–300.
- Rabb'a, I. (1992) The Geology of the Al Qurayqira (Jabal Hamra Fadda). Map Sheet 3051 II. Geology Directorate, Amman, Geological Mapping Division, Bulletin 28, 55 pp.
- Scheinberg, I.H. (1979) Human health effects of copper. Pp. 17–39 in: Copper in the Environment: Part II,

Human Health (J. Nriagu, editor). John Wiley, London.

- Schick, R. (1995) The Christian Communities of Palestine from Byzantine to Islamic Rule: An Historical and Archaeological Study. Darwin Press, London, 583 pp.
- Skinner, H.C.W. (2000) In praise of phosphates, or why vertebrates chose apatite to mineralise their skeletal

elements. Geology International, 42, 232-240.

Wittmers, L.E. Wallgren, J., Allich, A., Aufderheide A.C. and Rapp. G. (1988) Lead in bone. IV. Distribution of lead in the human skeleton. *Archives of Environmental Health*, **43**, 381–391.

[Manuscript received 12 January 2005: revised 15 June 2005]