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The local and global dimensions of metalliferous pollution derived from a reconstruction of an eight thousand year record of copper smelting and mining at a desert-mountain frontier in southern Jordan

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Abstract

This paper establishes an eight-thousand year history of anthropogenic metal pollution at one of the oldest, most important and longest sustained sites of the extraction and smelting of copper ores in the Old World: the Faynan Orefield in Jordan, which is located between the hyper-arid southern desert and the front of the wetter Mountains of Edom. The modern land surface is shown, in significant part, to be a complex palimpsest of archaeological sites, metal pollution of various ages and ore processing deposits. Quantitative and qualitative observations of the storage and cycling of heavy metals through the local natural and domestic systems have produced a body of information on processes with which the past has been interpreted. Heavy metal concentrations in semi-continuous sedimentary bodies indicate that over the last 1500 years, metals were removed by natural processes at a comparatively slow rate given the scale of the original anthropogenic metal burden: the proportions of lead with respect to copper have increased as the overall metal burden has been lowered. Distinctive anthropogenic metal-pollution signatures have been detected in ash- and charcoal-rich deposits that were discarded onto the banks

of a perennial stream in the late Neolithic. At present, the nature of the human activities that might have produced these pollution signatures is unknown. Substantial metal pollution from industrial – scale smelting activity was present from the Early Bronze Age. The intensity of heavy metal pollution produced in Classical Times locally exceeded that recorded at major European copper smelting centres in the nineteenth century A.D. The pollution evidence indicates that intensive copper smelting at the immediate area took place until approximately the end of the Byzantine period; with the exception of one further minor episode of smelting radiocarbon dated to cal. BP 530-330. An observed partial, but perhaps significant, parallelism is also noted between this local record and the records of the metal burdens of the northern hemisphere determined at mire or ice-sheet sites at high-altitudes or high-latitudes.

The paper discusses the extent to which this parallelism might be a geochemical indicator of the actual existence of “economic systems” of a geographical scope and scale to have both exercised substantial “pull” upon the resources of such isolated and difficult locations at the Wadi Faynan, and to have generated sufficient overall metal pollution to have materially altered the chemistry of the global atmosphere. The comparative absence of quantitative information upon the nature, spatial scale and impacts of human, industrial and natural processes affecting metal-pollution in other metal-rich arid lands significantly provides a significant to such approaches that seek to research “locally and think globally”.

Key Words: Ancient Economy, Atmospheric Circulation, Deserts, Geomorphology, Holocene, Jordan, Lead, Copper, Thallium, Mediterranean, Pollution, World-Systems Theory.

1. INTRODUCTION: THE LOCAL AND GLOBAL DIMENSIONS OF METALLIFEROUS AIR POLLUTION IN THE HOLOCENE

1.1 *Local and global dimensions of pollution*

This paper explores the local and the global dimensions of metalliferous pollution from the working and smelting of complex ores containing copper and lead over an eight thousand year period in the Faynan Orefield at the margins of the boundary between the hot and hyper-arid desert of the Wadi Araba(h) (58) and the ~1500 m high edge of the wetter *Mountains of Edom* in the Hashemite Kingdom of Jordan (Figures 1 and 2). This Orefield covers ~500 km² (8, 16, 17) and is one of the oldest and most important centres of sustained metal extraction in the ancient world. The metal content of ores varies with their source in either sedimentary or igneous bedrocks (40, 79, 99, 109); but ores with 35% copper can still be collected by hand (64). Concentrations of lead can reach 6% in copper ores from the “Burj Dolomite Shales” (“Dolomite Limestone Shales” – the DLS of 64), but they are typically below 1% in the other main copper source the “Umm ‘Ishrin Sandstones” (the “Massive Brown Sandstones”: the “MBS” of 40). The best known archaeological site in this orefield is the Khirbet Faynan (Figures 1-3) which is associated with extensive metallurgical activity in the Roman-Byzantine period (~2550-~1450 calendar years B.P: Table 1) and is identified with Roman Phaeno in the well-known contemporary descriptions by Eusebius of Caesarea (34, 44). To the north-west of Khirbet Faynan is the Iron Age copper-working site of Khirbet en Nahas (Arabic: *The Ruins of Copper*) and in the Wadi Fidan to the west, there is extensive evidence of ancient copper working including an Early Bronze Age site of mass copper production at Khirbet Hamra lfdan (Figure 2) with geochemical evidence for substantial copper manufacture in the Early Bronze Age (3; 83, 84). The immense geographical scope, human and palaeoenvironmental significance of

copper metallurgy over time in this and other adjacent areas is suggested by Figure 1.

This paper pioneers a process-based, biogeochemical-ecosystematic approach to determining the nature and reality of past large-scale economic activities through explorations of the origins, scale and distribution of ancient air-pollution signatures, rather than the more familiar documentary or archaeological-artifactual evidence of the geographical scale and intensity of past trade or exchange. The approach adopted is an extension of modern observations of the economic and political roles of important metals, which interpret the history of airfall-metal pollution in high latitudes and high altitudes over recent decades as a global environmental consequence of the changing power, nature, growth and inter-connection of various major and minor economies, variously linked, in different parts of the globe.

Frank (38, p. 131) articulated the role of the supply of important metals, in such global-scale economies of the historical period by explicit mention of the ideas of the eighteenth century Scottish economist Adam Smith who wrote "*The silver of the new continent* ("Spanish" America) *seems in this manner to be one of the principal commodities by which the commerce between the two extremities of the old one* (Old World) *is carried on, and it is by means of it, in a great measure, that those distant parts of the world* (China, India, the Philippines and South America) *are connected with one another*" (123, p. 312). In such a global model of political economy, it might be expected that the scale of silver metal production in the Spanish Americas and hence of air pollution related to the scale of smelting of silver ores corresponded with both the dimensions of the global trade; and if the scale of metal refining were sufficiently large, the global air-pollution record. In more detail, the approach in this paper has the following nested components in a chain of enquiry, evidence and argument.

- It explores the extent to which a long term air-pollution history can be reconstructed for an area of major and sustained economic importance – in the Wadis Dana and Faynan adjacent to the Khirbet Faynan (Figure 2).

- Field and laboratory investigations that determine the changing intensity of metalliferous pollution over time at a major ore-source location; information that is then compared with other independent evidence of changes in the intensity of metallurgical work in the area to determine if the pollution record accords with the archaeological and documentary evidence of the scale of ore-extraction and processing.
- Observational field research that considers the scale and geographical extent of the natural and human mechanisms that can be observed for the transport or storage of anthropogenic pollutant metals in the locality. In particular, several local, interpretative and taphonomic questions are addressed; are there mechanisms that are capable of spreading pollution over several kilometres as well as into the high atmosphere where it can be transmitted around the global atmosphere?
- Geochemical and geomorphic investigations address the representativeness and meaning of metal pollution concentrations determined in the geomorphic sequences in the study area through the following questions. Are the metal-pollution burdens determined for sediments of different ages in the Faynan area representative of the original air-pollution concentrations at the time of their deposition? To what extent have they been altered by post-depositional biological or geomorphic-pedological processes? To what extent are observed pollution concentrations the product of the recycling of older exposed pollutants into the aggrading deposits?
- The paper explores by simple comparison the extent to which the local record of air-pollution determined at the long-term site of sustained metal smelting corresponds with the global anthropogenic air-pollution record matches. The history of natural and the anthropogenic air-pollution of the global atmosphere throughout the Holocene is well-established from studies of the air-fall metal precipitation onto major ice-sheets at high-latitudes or mires at high altitudes (15, 19, 20, 21, 22, 23, 80,81, 92, 93, 119, 120, 121).

Finally, this enquiry examines the extent to which the air-pollution records – local and global - can be seen to be in part or wholly in parallel.

An unremarkable economic explanation of the lack of such parallelism would be that unlike the historical case described by Adam Smith, there was either no or insufficient aggregate economic “pull” by any ancient metal-using economies (as a result of insufficient power, geographical scale or integration) capable of driving the exploitation of scattered and isolated ore-sources such as the Faynan Orefield, with the result that there was insufficient overall economic activity to create a sufficiently powerful aggregate pollution “push” to alter the chemistry of the entire northern hemisphere atmosphere. Parallelism might imply the contrary, or perhaps simply the effects of a chance co-incidence. Caution is needed in these analyses and inferences drawn from them. Evidently, a local and global history of metal-pollution stemming from the exploitation of copper ores over an eight thousand year period at the edge of a hyper-arid desert will have many different natural and human dimensions to its interpretation in comparison with the specific historical case of silver described above. The field investigations that determined the geomorphological and geoarchaeological sequences in the region (70, 71, 90) were limited in number and were at the reconnaissance level or, as is the case of the other sites of copper metallurgy in the immediate area such as the Wadi Khalid and Qalb el Ratiye, put to one side because of lack of time and resources. Indeed, the convincing demonstration of each linkage noted above is a fundamental and major research exercise in its own right, especially in terrain as arduous to operate in as the Wadi Faynan region. Nevertheless, this paper attempts to make a first, qualified step to examine the linkage of natural and human systems, at the local and the global scale, through the geochemical pollution manifestations of economic activity over the much of the Holocene.

This approach also has a partial resonance with the idea of viewing such large scale economic activity through the concept of “*world systems*”. Such an inferred system or a network of systems would have been vast regional or continental scale economic-social entities that changed over time, which had

economic power and significance, that were articulated by trade and exchange, of which the sustained ore-processing activities based in the Faynan Orefield could typically have been an important supply constituent. The concept of a “World-System” was originally envisioned by Wallerstein (125) and has been extensively debated by many researchers on historical, geographical or archaeological grounds, as well as in terms of utility, definition, continent-wide economic and political cycles and (anti)eurocentrism and geographical focus (e.g. 26, 37,38, 39, 57, 75, 112, 118).

1.2. The development and decline of copper working and smelting in the Faynan Orefield.

A pollution history for the Holocene in the study area has not been previously established. There has, however, been substantial recent archaeological, climatic and palaeoecological research in the area (52, 56, 70, 71, 90, 106, 107) and although the deposits are patchy and discontinuous it has proved possible to establish an outline environmental history for the Holocene based upon geomorphic, molluscan, pollen, and skeletal evidence. The published archaeological and historical evidence indicates that both the nature and especially the intensity of the copper working and smelting in this area have changed dramatically over time, with distinct episodes of industrial-scale activity, whilst other times saw substantial or total abandonment (9, 10, 11, 12, 13, 14, 61). The synthesis presented in columns 1-3 of Table 1 is used to make the predictions of the possible nature and intensity of past anthropogenic metal pollution in the area (column 4).

1.3. Global dimensions - copper and lead in the global atmosphere

The flux of anthropogenic copper through the atmosphere to the Greenland icesheet during the Roman-Byzantine period was substantial and estimated by Hong et al. (67, 68, 69) to be in the region of 2300 t/yr⁻¹. These authors proposed that the economic rise of “Athenian Civilisation”, the development

around the Mediterranean of a copper-based coinage, and the rise and decline of “Roman-Byzantine” mining and metallurgy were responsible for the distinctive heavy metal-pollution record detected in the Greenland ice cores. Anthropogenic lead production is of broadly similar antiquity (29,30) and the concentrations of lead accumulating in high altitude mires in Switzerland as a result of hemispheric atmospheric pollution were demonstrated by Weiss et al. (126) to have exceeded “natural levels” as long ago as 5,500 radiocarbon years ago. A subsequent and more general increase in lead pollution of the northern hemisphere is suggested to have taken place from approximately 2500 years ago. The ice-core record suggests that global atmospheric concentrations of lead in Roman-Byzantine times approached 4000 t/yr^{-1} ; 15% of the concentration reached during the peak use of leaded-alkyl additives in gasoline in modern times. Indeed, the broad conclusions from investigations of lead and copper from cores from the Greenland Ice Sheet, as well as mires at high latitudes or high altitudes in Europe (15, 47, 86, 111, 127) indicate that many of the primary contributions of metalliferous pollutants to the global atmosphere from late prehistory to the modern day reflected changes in both the manner and intensity of mining, ore processing and smelting in the ancient Classical World. However, early metal processing also took place in many other areas including China, India, Africa and Sweden; as well as non-Mediterranean regions of the Old World such as Mesopotamia, Anatolia and Egypt. All were dynamic metal-using entities several thousand years before the development of the Athenian State in Classical Antiquity (37).

1.4. Ancient copper working, smelting and environmental pollution: human and natural processes and the dispersion of heavy metals

Vital initial questions that have to be addressed in any study to establish the history of metalliferous air-pollution based upon geochemical studies concern the nature and meaning of the metal concentrations detected within the sediments. In particular, information is needed on the primary processes which

contributed to the present situation, as well as the spatial *representativeness* of the evidence of past sediment pollution determined at the various sites discovered. These questions have to go beyond the assumptions that the metal-polluted sediments reflect in a simple manner the changing intensity and frequency of ore-processing and smelting previously conducted at one or another location in the general area. Such issues concerning the past must start with an investigation of the manner in which modern processes transfer and modify pollution burdens from the past, since if such issues cannot be clarified with the evidence available at the present day, the meaning of any record of ancient pollution must be obscured.

Much recent research in the Faynan has placed the well-being of people and their ecosystem at its centre. In particular attention has focussed upon the possible health and agricultural impacts of past industrial activity mainly in terms of consuming, breathing, touching, local re-working and the bioaccumulation of dangerous substances, in the modern environment and estimated for the Roman-Byzantine period (53, 54, 56; 103, 104, 105, 106). These qualitative and quantitative observations made over eight separate field visits are combined with published findings from elsewhere and presented in Table 2, to suggest the many processes that might exist for the “release”, “transport” and “immobilisation” of heavy metals in this particular desertic environment.

Experimental studies by Merkel (91) at Timna, suggest that the “wind-powered” smelting processes applied in antiquity to copper ores in the Faynan area are unlikely to have been particularly efficient even when located on windy ridges, an observation that implies significant emissions of heavy metals to the local environment. Using reconstructions of ancient furnaces at Timna, Merkel showed that the greatest loss of the element being extracted – in this case copper - was in the slag that was typically discarded onto the desert soils, rather than as metals released into the atmosphere. At present, the “typical” air-pollution “footprint” from a “typical” single furnace is unknown. Initial recoveries of 50-60% of “easily recoverable” copper from the remnant slag were anticipated, and could reach 80-90% as a result of the further processing of discarded

copper-rich slag using hand-sorting to select the more copper-rich materials. As a result, significant burdens of heavy metals, including lead, eventually entered the local environment in such slag, on discarded furnace bricks and linings, residual charcoal and ash, as well as in dusts of charcoal, ash and gases blown out of furnace flues. The re-smelting in antiquity of earlier slags in the Faynan was reported by Hauptmann (61).

Attention is given occasionally in this paper to the release of thallium, this dangerous, volatile and poisonous metal ultimately has its origins in the ores that are abundant in the Orefield. Typically thallium may be released into the environment during “primitive” smelting both as metal-rich vapours and dusts that condense onto the surfaces of charcoal or hot ash and fall back to the surface where the metal can persist in the soil (76).

The transfer of substantial quantities of dust from the Wadi Arabah into global atmospheric circulation was observed using satellite and meteorological observations by Dayan et al. (31) and Singer et al. (122). In contrast, post-depositional vertical movements – upwards or downwards – of copper and lead through Holocene sediments by biological or hydrological processes were demonstrated by Pyatt et al. (102) and Condon (27) to be at rates that are sufficiently slow in relation to the observed metal burden of these sediments, to demonstrate that the observed metal loadings are relatively close to those produced by the original input of polluted materials – these conclusions are explored in this paper in greater depth using studies of Classical and post-Classical sediments impounded behind the Khirbet Faynan barrage (site 5512-Figures 2, 3; 6 and Table 4). The carbonate-rich nature of these sediments (94) also serves to inhibit post-depositional “vertical” movement of these metals.

The concentrations of copper and lead might also reflect their introduction by geomorphic processes or dilution as a result of the deposition of materials lacking heavy metals. This possibility is examined below through interpreting the abundance of strontium as an indicator of the movement allochthonous carbonate sediments. The most immediate and mobile sources of strontium are in carbonate clasts and aeolian dusts in Quaternary deposits in the area;

materials that have their origins in the widespread, thick Cretaceous Limestone to the east of the Faynan Orefield, and to a much lesser degree in the resistant Palaeozoic Limestones in the area.

2. PATTERNS OF HEAVY METAL POLLUTION ACROSS TIME IN THE WADI FAYNAN

Khirbet Faynan and the other archaeological features and sites discussed in this paper are located upon Quaternary colluvial, aeolian, fan and fluvial sequences that were described by Hunt et al. (70) and McLaren et al. (90). The Pleistocene deposits are all relatively low in heavy metals compared to mineralised bedrocks in the area (Figure 4). The sedimentary properties, geomorphological origins and radiocarbon dates for the most important sedimentary sequences studied in this account are summarised in Table 1, and set out in detail in Tables 3 and 4.

All samples were from the <2mm fraction of largely carbonate materials from the matrix of recognisably water-lain, wind-blown sediments and some colluvial sequences; some samples were from ash with charcoal (e.g. 5021#G) in such sequences. No samples included pieces of smelting slag, although the presence of slag or ores in these materials, with some reservations, has been used as an indicator of ore-handling or processing at that location in the past. In each case this knowledge of sedimentary origins is based upon field observation and has been used to identify sediments affected directly or indirectly by air-fall metal pollution. The concentrations of metals reported here were determined using 5 g samples of sieved and ground fine-grained sediment matrix by ICP-MS analyses at the Institute of Geography and Earth Sciences at the University of Wales Aberystwyth. Full analytical details, protocols and quality control systems were set out by Pearce et al. (100). Concentrations have been converted to ppm for consistency of reporting. Radiocarbon dates are presented in summary in Table 3.

3 THE CONTEMPORARY LAND SURFACE

The results of the field reconnaissance and air-photography mapping of evidence of ancient copper exploitation, geomorphology and other archaeological features are presented in summary form in Figures 4 and 5. In practice, field experience suggests the map presented in Figure 4 under-emphasises the distribution of smelting slags but more appropriately represents the functional geomorphic relationships in the landscapes. Archaeological features that range in age from at least the Bronze Age to the present day are exposed at the surface. These include smelting sites, buildings, walls, barrage, aqueducts, reservoirs, water control features, wall networks and fields. Large “carpets” and “mounds” (5 – 8 m high) of fragments or slabs of vitreous black copper smelting slag are concentrated in three areas; adjacent to the Khirbet Faynan; and north and south of the confluence of the Wadis Ghuwayr (Ghuweyr, Ghuweir) and Ushakir (Ashegair, Ashegar, Shaygar) (Figures 3 and 4). Field mapping found innumerable remnants of black copper smelting slag that are too small to appear at these scales.

Investigations of surface aeolian and colluvial sediments through the ancient field systems and south overlying the large alluvial fans that debouch from the Mountains of Edom and the adjacent smaller massifs show that the concentrations of copper slag suggest a general decline in copper smelting from east to west across the wall network from samples SP9 to SP17 (Figure 5); the pattern is however, less clear for lead (Figure 5). The lateral movement of aeolian dust from east to west by katabatic winds has been observed in the field and is also indicated by the planforms of shallow sand dunes located in Figure 2 that have been shown to be inter-bedded with ancient wall systems of at least Bronze Age date. The distributions of these dunes confirm the field evidence that aeolian processes can also affect the braid-plains of the wadi floors. The lateral passage of substantial bodies of sediment with patchy distributions of heavy metals along wadi floors has been observed in winter floods. Whilst overland flow during winter rain has been seen in fieldwork to scavenge metal-

fallout and concentrate it in topographic lows. Such outcomes in arid-land fluvial systems, especially the heterogeneities in the observed concentrations of copper and lead in fluvial systems, have been previously identified in other (semi) arid wadi floors in mineralized bedrocks within the region and elsewhere (e.g. 7, 18, 85, 115, 116, 128).

Field observation also suggested that another potential anthropogenic mechanism of metal transport through this area in the past is the extraordinarily large quantities of broken pottery visible on the surface during field survey in the spaces between the walls of the ancient field systems. Often, but not always of Roman-Byzantine antiquity, these ceramics may be derived from vessels that could have been used to transport animal manures, plant wastes, and human night-soil to the fields in order to improve yields which were probably in decline as a result of metal accumulation in the topsoil (Table 2). A by-product would have been the introduction to the fields of material with high concentrations of metals as a result of bioaccumulation and the liberation as dust to the atmosphere of any metal-enriched topsoil by any associated ploughing (102, 103). The further complexities of processes and events, large and small, that have been determined from field observation of the modern natural and human environment that are likely to have influenced the rates of movement and the storage of metals in the past, are also indicated in Table 2. Many processes remain unquantified, but comparison between the data on the metal burdens of modern wood, plants and animals in the modern Faynan (52, 53, 56, 107) with those referenced in the caption of Table 2, as well as with the sediment metal concentrations presented in Figures 4-9, all serve to emphasize the over-arching importance for both people and the environment of the immediate consequences of metal pollution from immediate air pollution and the discard of waste from ore-quarrying, processing and smelting.

Several key points about process and environment emerge from this survey. This land surface is evidently a complex archaeological and geochemical palimpsest. Inspection of Table 1 indicates this is likely to have also been the case during the later prehistoric and the historic periods.

Evidently, heavy metals from a variety of sources and ages are evidently still being re-cycled laterally across it by wind, water, and biological processes and human activity, nearly sixteen hundred years after the cessation of nearly all ore-mining and smelting. The evidence on the distribution and concentrations of copper and lead on the modern land surface also make the further important points about the spatial representativeness of the evidence of past sediment pollution determined at the sites described below. These data suggest that substantially enhanced metal pollution took place in the immediate vicinity of the smelting sites and that these enhanced concentrations of metal did not extend more than few kilometres downwind. Beyond this distance, concentrations appear to approach those of the background, an outcome aided by the loss of metals as very fine-grained sediment is taken up into the global atmosphere (31, 122). The evidence shows that fluvial processes are less predictable in their effects upon the composition and abundance of heavy metal pollution. The concentrations of heavy metals determined in deposits at land surface are evidently also the result of complex, but more immediate deposition and discard by people. Overall, these observations on process and outcome suggest that any pollution metal-signature obtained from the Holocene sands and silts reported below is most likely to have resulted from past metallurgical that was in the immediate study area: typically within a few kilometres, and often reflecting activities that took place within a few tens of metres.

4. ANCIENT BARRAGE-RESERVOIR INFILL AND UNDERLYING DEPOSITS: >3710 CALIBRATED YEARS; AND LATE ROMAN-BYZANTINE BP TO THE PRESENT DAY.

Immediately north of the Khirbet Faynan, within the densest concentration of the copper slag there is a narrow and shallow wadi with ten *Acacia* trees, ~ 5m tall, which grow on fine grained sediment impounded behind a substantial barrage wall that has been constructed across the wadi (5512 – Figures 2, 3 and 6).

Radiocarbon dates suggest this barrage was constructed ~ 1800-1900 years ago (Tables 1-4). Behind this wall collected a sequence of pond and aeolian carbonate-rich sediments. Both the wall and the sediments impounded behind it rest unconformably upon ore-rich colluvium that is much older (Unit 6 - 3710 – 3450 calibrated years BP [Beta 203402] Tables 3 and 4). Palynological studies and sedimentological studies are reported in full in Mohamed (94) and Hunt et al. (71). The geochemical history of these sediments based upon ICP-MS studies is presented in Figure 6. To aid description and interpretation, the geochemical data (Table 4 and Figure 6) have been set out following the stratigraphic approach of Pyatt et al. (101).

The patterns of metal concentration and its association with particular lithologies and sedimentary structures through the sediment column set out in Table 4 suggest that the importance of different human and natural agencies affecting heavy metal concentrations changed over time. The high metal loads (geochemical zones KFB3-4) detected by ICP-MS analyses are clearly associated in unit 5 with other evidence of the intense local smelting, and the crushing and processing of metal-rich materials. Above this basal layer the geochemical patterns also reflect the character of the immediate local environment at the time of sediment deposition. The distinctive character of the relationships down the sediment column between the geochemical, sedimentological and stratigraphic evidence (Figure 6; Table 4) suggest that the changing abundances of lead, copper and thallium in these sequences primarily reflect the original abundance of these metals in the infill sediments. As a result, they appear to suggest the changing scale and intensity of metal processing activities over time, rather than any secondary post-depositional mobilisation of metals. Likewise, the lack of a relationship between the abundance of strontium and the other heavy metals throughout this entire sequence indicates that the concentrations of heavy metals are not primarily the result of the introduction of allochthonous metal-rich materials being derived from upwind or up-catchment. Biogeochemical studies of the organic component in units 1- 4 at this site, as well as in other similar desertic sites,

have indicated the rates of biological transfer of heavy metals are small in relation to original metal burdens present (6, 27, 106, 33). This suggests that the patterns of metal concentrations determined essentially reflect those of the sediments at the time of their deposition, rather than over-printing by post-depositional processes.

In more detail, this is the only site in the present survey with a relatively simple geomorphic history that includes sedimentation up to and including the present day. As a result, it is possible at this location to begin to understand the characteristics of the uppermost deposits in terms of our knowledge of the modern landscape summarised in Figures 4 and 5. As a result, the following analysis begins with the uppermost deposits with the visible that reflect sediment and metal-cycling in the modern, relatively empty, non-industrial environment and then extends down the sediment column to the heavily industrialised ancient past.

Geochemical zone KFB1 has accumulated from the post-Byzantine period to the modern day. From the base of this zone (1610 ± 40 BP; cal. BP 1403–1601 (Beta 203399) to the surface, these deposits indicate the scale of the significant and progressive reductions in the concentrations of copper and lead deposited that have occurred in a (mainly) non-industrial environment as a result of re-working. Within this overall decline, copper has evidently been removed from the pool of re-cycling metals more rapidly than lead, with the result that proportions of lead increases up the sediment column; a property previously identified in geomorphic, biological and human systems in this area (52, 56).

The quantities of allochthonous carbonate-rich materials, suggested by strontium, that were being introduced by wind and water through much of this period of time remained relatively constant. There was however a notable sustained increase in strontium input which took place from ~600-700 calendar years ago, suggesting an increase in the processes of erosion and deposition at about that time. There are no indications in this series of samples taken at 2 cm intervals of the presence of metal smelting nearby in the historical period noted by Hauptmann (61), and which has been by radiocarbon dating of charcoal in

nearby slag (5450 Atlal Member : 430 ± 40 BP; cal BP 350-330 BP [Beta 203412]). It may, however, have been recognised in analyses of the adjacent borehole samples (5017) at 65 cm depth at 5017 (14).

Geochemical zone KFB 2 is quite different. It is complex and is composed of sediments that appear to reflect both industrial activity and local geomorphic processes. The zone is characterised by a marked increase and then decline in the concentrations of copper, together with a number of distinctive peaks and troughs. The ratio of copper to lead has a broadly similar pattern. Two sustained peaks of local metal pollution at ~200 cm and 170-185 cm are separated by a marked “trough” at ~185-195 cm. Whilst the concentrations of lead are relatively low, there is a sustained increase in the proportions of copper to lead through time. Thallium concentrations are low. The presence of such fluctuations in an uncomplicated impounded carbonate-rich depositional environment suggest that these distinctive fluctuations, which date to between ~1403 - 1610 cal. BP to ~1712 – 1890 cal BP, are more readily interpreted in terms of distinct episodes of industrial activity rather than as the result of fluctuations in surface geomorphological processes.

The situation within geochemical zone KFB2 from 205 to its base at 230 cm differs slightly from that above. It is characterised by a sustained increase through time in the concentrations of copper, whilst the abundance of lead remains comparatively high. This pattern of change might indicate the progressive release of ever larger quantities of copper pollutants into the local environment. The evident sedimentary structures indicate a more active and complex geomorphic in this ponded environment than elsewhere in the sequence (unit 5 – Table 4). Pollution concentrations were often high where clasts of smelting slag, with ash, charcoal and grains of ore, were observed in the excavation. The geochemical data clearly demonstrate the close proximity and increased efficacy of copper working and smelting, but it is not yet possible to distinguish clearly the effects of geomorphological processes in and around the barrage-pool in these metal-concentration data from those that reflect the changing scale, location and intensity of the industrial activities. These

observations on geomorphic processes might suggest that the radiocarbon-dated charcoal might also have been re-worked: this could be the case. The calibrated radiocarbon dates are however, very close in suggested age in what was a rapidly accreting sedimentary environment.

Geochemical zone KFB3 is characterised by sustained declines over time in the concentrations of copper, and especially lead from earlier high levels. The relatively low overall concentrations of metals in the upper parts of this zone have a number of industrial and geomorphological explanations – either singly or in combination. At present there is insufficient evidence at this site to evaluate these further.

- (i) The adoption and sustained use of relatively lead-poor ores from the “Umm Ishrin Sandstones” replacing ores previously exploited from the relatively lead-rich “Burj Dolomite/Shales”.
- (ii) The sustained re-use of previously smelted materials at this site from which lead had been previously extracted.
- (iii) Changes in the quantity, nature and intensity of ore-processing-smelting that had the overall effect of increasing the release of copper.
- (iv) Differences in the geomorphological behaviour of copper as opposed to lead as a result of variations in the movement of different fractions in fluvial sediments have been noted previously elsewhere in the world, but such effects would appear insufficient to explain the magnitude and character of the changes observed at this site.
- (v) The consequences of the types of mass-movement, sediment deformation and fluvial processes evidenced by the types of sedimentary structures (Table 4)
- (vi) The progressive re-working of the materials below – unit 6 – geochemical zone KFB4.

Geochemical zone KFB4 is characterised by the substantial concentrations of lead, copper and thallium, with concentrations notably higher above 275 cm. One important piece of evidence is that geochemical zone KFB4

is ~ thirteen hundred years older than the overlying materials. The lithological properties, sediment chemistry and the dating evidence (3390 ± 40 BP; cal BP 3485-3816 (Beta 203402) at 270 cm depth also point to a distinct stratigraphic unconformity between the geochemical zones KFB3 (unit 5) and KFB4 (unit 6). The dominance of lead over copper in geochemical zone 4 is especially clear. There are several possible explanations of these properties. They might represent either the composition of crushed and graded ores or smelting products from copper ores that were proportionally rich in lead, such as the “Burj Dolomite Shale”. The stratigraphic and sedimentological observations for Unit 5 in Table 4a indicate that the two metal pollution peaks and intervening trough may be closely related in age and are the result of local human activity on a dry wadi floor. The concentrations of thallium which are high at the top of this zone and increase further into geochemical zone KFB 3 above, suggest local accumulation of thallium condensing or accumulating on the land surface, especially on ash. It is equally evident that interpretations of the geochemical records in KFB4 may also relate to the changing character of the geomorphological environment and to the nature and intensity of past human activities.

Overall, the pollution record in the sediments that collected before and after the construction of the Khirbet Faynan barrage are distinctive and represent the complex interactions of both natural processes and human activities. Intense industrial activity from ore-processing and smelting led to intense metal pollution at the site during the Bronze Age; and there is further clear evidence of its continuation in the Roman-Byzantine period. The site was initially dry but became wet, before becoming a ponded-lacustrine environment created by the construction of a cross-wadi barrage. The character of pollution altered dramatically over time – initially with substantial pollution from lead and thallium, then with increasing copper – variations that are attributed provisionally to changes in the composition of the materials smelted, perhaps the technologies employed, and the effects of geomorphic processes, including re-working. The changing proportions of copper to lead in the pollution signatures

would correspond with a simple shift from the use of ores from the “Burj Dolomite Shale” to the “Massive Brown Sandstone”. Sometime at or about the end of the Byzantine dominance of the area, there was a clearly defined and rapid decline in smelting activity and pollution input. A distinctive sedimentary lag deposit (Unit 3) corresponds with this cessation.

5. COPPER SMELTING-DEPOSITS: ~CHALCOLITHIC TO POST-MEDIAEVAL

Three pits ~1- 1.6 m deep (sites 5738, 1491/5741 and 1491/5741b) were excavated in sands and fine silts containing slag attributed to the Atlal Member (Table 3) associated with a widespread surface scatter of black copper smelting waste adjacent to Khirbet Faynan. The lithologies, stratigraphy, and calibrated radiocarbon dates obtained from the pits are summarised in Figure 7. The compacted ash and clay layers observed in 1491/5741b were interpreted in the field as “hardened working surfaces”. The radio-carbon dated fragments of charcoal, like those of pottery, suggest maximum ages for the deposits which are sufficiently toxic and visibly well stratified to suggest that the downward movements of materials is unlikely. The radiocarbon dates obtained from these sections reveal a long history of metalliferous activity, ranging from 4240 ± 40 BP; cal BP 6190-5940 (Beta 203414); which is overlain by 5690 ± 40 BP; cal. BP 6550 – 6400 (Beta 203413), a sequence that suggests re-working of charcoal. Both of these were overlain by the early historical date of 430 ± 40 BP; cal. BP 350 – 330 (Beta 204412) which is locally important and might reflect a brief phase of copper smelting activity attributed to a period of Mameluk control from Egypt. There are marked discontinuities in metal concentrations approximately at the boundary drawn between Units 3 and 2 at each site that may suggest a significant break in the sequence.

The concentrations of heavy metals detected in sands and silts at all three sites were high and clearly hazardous: ~45,000 ppm lead and ~17,000 ppm copper in site 5738; and ~14,000 ppm lead and ~15,500 ppm copper in site 1491/5741; concentrations in site 1491/5741b are even higher. Concentrations of thallium are likewise dangerously high reaching ~90, ~35 and 145 ppm respectively. The highest metal concentrations are associated with ash rather than clasts of slag. The lower concentrations of copper, lead and thallium in the basal 20 cm of exposures 5738 and 1491/5741b may be the result of the introduction of unpolluted sediments from up-wadi and the evacuation down-wadi of these polluted wastes. These lithologies also demonstrate that these polluted waste materials were discarded by people onto an active wadi braid plain. These residual concentrations of lead and copper in exposed surface slags near the Khirbet Faynan equal or exceed those that remain in European locations, such as the Lower Swansea Valley of South Wales (Figure 1); areas that were world-centres of smelting in the nineteenth century (24).

Together, these sites demonstrate the poisonous extremes of much earlier metal pollution that can be detected in the areas that are visually dominated by evidence of Roman-Byzantine ore processing and smelting. The intensity of contamination revealed at the earliest dates are a firm indication that the intensity of activity hereabouts in the early Chalcolithic Age-Bronze Age was industrial in its scale and impact, Khirbet Faynan appears to be the site of one of the world's first metal factories, comparable with those recently discovered down wadi in the Wadi Fidan (83, 84).

6. COPPER MINE IN THE WADI DANA

Details of the location, dimensions and antiquity of the deposits in this mine entrance (5740) attributed to the Hamman Member and the Nabatean-Byzantine period are given in Grattan et al. (55) where it is mine 2 of that survey. The

lower part of this mine's infill deposits are slumped post-abandonment mining and ore wastes overlain by surficial material that has slumped back into the mine from higher ground outside. Total concentrations of copper and lead are shown in Figure 7. The distinction between the observed sediment types noted in the mine entrance is reflected clearly in the metal concentrations. Total copper concentrations range from ~6500 to ~16000 ppm; lead varies from ~3000 to 5000 ppm. Copper concentrations in the post-mining abandonment infill deposits are typically in about 1000 – 1500 ppm, with one sample at ~3500 ppm. Lead concentrations are typically in the range 800 – 2000 ppm. A later influx of sediment with lower metal contents through wind, water and animals' inputs of sediments is evident in the lower concentrations recorded in the uppermost samples. Again, the proportion of lead in relation to copper appears to have increased over time as the accumulating sediments become younger.

7. WATER-WASHED AEOLIAN SILTS AND THIN PALAEOOLS FROM LATE NEOLITHIC TO ROMAN-BYZANTINE

Site WF5022 is a 1.4 metre deep section in exposed in a low mound, the Tell Wadi Faynan, eroded by the modern wadi approximately 1.2 km west of Khirbet Faynan (Figures 8-11). The geoarchaeological sequence is the type site of the Tell Loam Member and interpreted as an episodic aggradation of wind-blown clay-silts and silts that were often water washed, with occasional episodes of skeletal soil formation, soil ped development, and surface scour-and-fill (Table 3). The presence of fragments of pottery and worked chert, charcoal and ash indicate the presence of people throughout the development of the sequence from its base. This lowest deposit can be followed through the exposure, as well as by study of pottery, charcoal, ash and worked chert, to the adjacent Neolithic structure named Tell Wadi Faynan which was excavated by Najjar et al. (96). This stratigraphic level provided calibrated radiocarbon dates: 5740 ±

35 BP; cal BP 6654-6412 (HD 12337) and 5375 ± 30 ; cal BP 6278-5995 (HD12336) (Figures 9 and 10). The evidence provided by distinctive sedimentary parting planes with thin lag deposits in the exposure suggests that the sequence provides a relatively straightforward record of episodic deposition and temporary soil development. The common absence of any notable increases in the concentrations of heavy metals at six observed temporary surfaces-palaeosols observed suggests that the abundance of heavy metals does not simply reflect hiatuses or changes in the rate of aeolian deposition, rather the concentrations of metals reflect the low metal content of the local environment (Figure 10).

The base of the sequence, contemporaneous with the Neolithic settlement, has a distinctive peak of copper. There is a higher proportion of copper to lead at 130 cm (geochemical zone TWF4b). There is no correlation between the trends in strontium – taken here as an indicator of the input of allochthonous materials – mainly aeolian carbonate dusts – and the abundances of copper and lead. The metal concentrations detected in the base of this sequence between 140 – 115 cm, and in particular the spike at 130 cm, may represent small scale smelting of copper ores, with minimal lead, in domestic hearths, rather than any natural process. This situation ends at a distinctive sedimentary parting plane which is evident at 110 cm – this might be a non-sequence or even a period of erosion and weak pedogenesis. It corresponds with a particularly clear break in all the geochemical profiles – especially that of strontium. These features suggest that the sediments attributed to geochemical zone TWF3 are separated by a discontinuity of unknown significance from those of TWF4a beneath. Between 110 and 80cm in TWF3, there is no evidence of enhancement in any heavy metals. Concentrations of strontium also fall at these depths suggesting less soil disturbance. One interpretation of zone TWF3 based upon extrapolating from the available radiocarbon dates and extant stratigraphy is that it may have accumulated in the so-called Bronze Age “Dark Age”, when the area appears to

have been effectively abandoned and that the sharpness of its lower boundary at this location reflects geomorphic processes.

Between 80 and 10 cm (geochemical zone TWF2) there is evidence for three episodes of lead and copper deposition; between 60-45 cm; ~30 cm; ~20 cm (TWF 2d, 2b, 2a). There was no geomorphological evidence in the exposed face to suggest these were primarily the result of geomorphological processes, as inferred for the “event” at 110 cm depth. Overall, the combination of stratigraphic and geochemical information suggest that within geochemical zone TWF 2 there was a gradual increase in the scale of smelting activity; this then accelerated to produce the higher proportions of lead being liberated at 50 cm. This was followed by a marked decline or disappearance of smelting at ~40 cm. Another peak in smelting activity followed at ~30 cm – this time releasing greater proportions of copper. A further decline occurred at ~25 cm, followed by a smaller peak at ~15-20 cm; and another decline at ~10 cm. The concentrations of strontium follow those of lead and copper to only a limited degree. In this particular geomorphological situation this partial relationship suggests that episodes of pollution-smelting tend to be associated with increased deposition of carbonate dusts that reflect the intensity of human and agricultural activity in the area. The precise antiquity of these events in TWF2 is unclear. Information from the site and comparison with Hauptmann (61) suggests geochemical zones TWF2d–2a correspond with the middle –late Bronze Age and/or the Iron Age-Roman period. The uppermost peak of geochemical zone TWF1 (which includes the present land surface) is most readily explained by the abundant smelting slag and debris of Roman-Byzantine age (Figure 10). Once again these high concentrations are associated with the visible presence of ash in the deposits.

Overall, the combination of geomorphic and geochemical evidence evident in the sedimentary sequence through site 5022 indicates initial substantial increases then significant falls in the intensity of deposition of metal pollutants at the site that are distinct from changes in the sedimentary environment. These features imply that the notable fluctuations primarily reflect changes in the abundance of smelting and other human activities in the region.

The quantity of pollutant released varies with the ratio of copper to lead in a systematic manner that may point to differences in the ores smelted, and/or changes in technologies. The earliest pollution episode inferred at the exposure in the late-Neolithic appears to have resulted from the exposure to fire of relatively lead-free ores. The pollution record at this site may point to the transition from small scale smelting activity, to the processing of ores and metals at an industrial scale.

9. INTERBEDDED FLUVIAL-STREAM BANK AND ANTHROPOGENIC DEPOSITS OF LATE NEOLITHIC AGE

The stratigraphy, geochemistry, sample-lithologies and dating of this fluvial and stream-bank sequence of the Faynan Member – upper component – at site 5021 (Figures 9-12) are exposed in a cliff-like wadi edge of the modern braid-plain of the Faynan are set out in Figures 12 and 13. These fluvial-stream bank sediments underlie the Tell Loam Member which rests conformably upon them and adjacent Pleistocene fluvial deposits of the Faynan Member – lower component (see 5022 above). These exposures of stream-infill / stream-bank deposits are aligned approximately along the centre of an ~E~W aligned channel of a river that is estimated to have been ~10-12 m wide (Hunt et al. 2004, in press). The presence of Neolithic pottery fragments, layers of ash, charcoal, radiocarbon dates and the worked flints, inter-bedded with this quiet-water fluvial deposits, all record human activity discarding materials on to the (sometimes dry) banks of this essentially (often) quiet, perennial stream during the Late Neolithic; a location on the opposite bank of the stream channel to the site of Tell Wadi Faynan (5021). This sequence is not a palimpsest of a stable dryland soil, nor an aggradation of dryland soils upon which a series of *in situ* archaeological remains accumulated. Close examination of this sequence failed to detect any evidence that the charcoal and ash deposits studied had been

significantly reworked as a result of their environment of deposition on the banks, perhaps under shallow water: distinct intra-formational lenses of ash were visible in the field. Water conditions must have been comparatively quiet at or before the burial of these fine grained materials. All this sequence is underlain by Late Pleistocene fluvial deposits of the Faynan Member – lower component (90).

The antiquity of this channel infill sequence and hence the geochemical samples studied from it are known directly from a radiocarbon date of charcoal taken within a lens of ash and charcoal at sample point G that is located in the mid-point of the sequence. Sample G gave a radiocarbon date of 6200 ± 40 BP; 2σ cal. BP 7240-6990 (Beta 205964). This AMS date is supported by the pollen biostratigraphy (71) that depend upon radiocarbon dates in adjacent wadis; as well as the lithostratigraphic relationships (Figures 3, 10-13 and Tables 1 and 3) of other radiocarbon dates associated with earlier studies at nearby Tell Wadi Faynan immediately to the north. The antiquity of the geochemical samples and the geomorphic sequence within which they are found are therefore well-constrained by a variety of lines of evidence.

Copper and lead concentrations through these fluvial deposits are broadly constant with three notable exceptions - samples 5021 D, H and L (Figure 12), which were mainly charcoal, ash, fragments of pottery and fluvial silt. The concentrations of copper and lead in these fluvial sediments were high, reaching ~ 1500 ppm. Neither clasts of ore-bedrocks nor fragments of refined metals were noted in these analysed samples – they were no clasts of ores observed in the fluvial sequence. Samples L and H contained ash and/or charcoal and also a significant suite of other metal pollutants such as Be, Cu, As, Hg, Tl and Pb (Figure 13). In particular, the metal assemblage in sample L suggests the heating of copper-lead ores which also contained substantial quantities beryllium (which is known in the area (109) rather than the treatment of only pure copper and very pure copper ores. The concentrations of metals and their geomorphic associations in these three particular samples imply both the metal-enrichment of the ash and charcoal (and perhaps mineral residue),

followed by the discard of these polluted materials onto the stream bed-bank, where they were buried without significant disaggregation by quiet water clastic deposits. The notable concentrations of metals in these particular samples ultimately point to the profound heating of copper ores that were rich in copper and lead. The associated presence of ash and charcoal similarly points to the related use of fire. Indeed, it appears that the presence of charcoal and ash is associated with the highest levels of metal contamination through the sequence. The relatively high proportions of lead and arsenic in comparison with copper suggest the pollution signature might derive from the processing of the “Burj Dolomite Shales”. The sedimentary structures, sedimentary texture and composition, as well as the wider stratigraphic evidence and geomorphic situation elucidated provide no support for an explanation that these episodes of charcoal-ash accumulation reflect some form of wildfire – rather the opposite. Nor do they indicate that the elevated metal concentrations in the samples 5021 D, H and L reflect the chance of wildfire scorching clasts ores occurring on the stream bank without the aid of people. The properties of the stream bank sediments at 5021 point to the natural deposition of finer-grained muds. These infill channel deposits and their margins were not like the slag-rich, ore-rich, clasts-rich conglomerates described in Figures 3-5 that constitute the modern braid-plain of the Wadi Faynan. Overall, this suite of contaminants and deposits in samples 5021 D, H and L indicates that (perhaps fortuitously or deliberately broken) ore-rich materials were substantially heated by people on the nearby stream bank using strong heating from wood fires. The resulting ash and wood fuel were contaminated by metal-rich fumes and dust from this process and the debris cast onto the geomorphologically-quiet margins of the stream at ~7240-~6990 cal. BP (Beta 205964), at a time when the adjacent Tell Wadi Faynan was occupied (Table 1).

It is not yet possible to answer the immediate archaeological question concerning intent - whether this geochemical and geomorphic evidence points to the deliberate and purposeful practice of copper smelting at perhaps small, domestic hearths; or merely chance-strong heating of some crushed ores that

have produced a metal pollution and ash signatures that fortuitously mimic the outcome of deliberate smelting. There were no finds of slag, ore, or crucibles in the excavation of the site. Uncertainties about interpretation also stem from the procedural and evidentiary differences that exist between the more familiar archaeological evidence used to recognise ancient metallurgy and those which stem from investigations of the local production and deposition of heavy metal pollution as determined in this study. Pieces of copper and copper ore in archaeological contexts are known in this immediate area at about these times (Table 1; Najjar et al. 1990) whilst the conventional archaeological record indicates copper metallurgy did not begin in the Araba(h) region until ~5500 year ago (61). The archaeological evidence for the smelting of copper ores in the Neolithic was discussed by Adams (1998) and it has yet to be demonstrated for the Neolithic. Craddock (1995) discussed “smelting slag” excavated from Çatal Hüyük which was attributed to the 6th millennium BC, following Neuninger et al. (97); however, Muhly (95) regarded this reported “slag” to be material produced by the melting of native copper.

10. DISCUSSION

10.1. Processes and patterns of heavy metal pollution

Field mapping, geochemical analysis, photographic interpretation and radiocarbon dating, combined with observations of archaeological structures show that the modern landscape is a palimpsest that reflects past human, industrial and agricultural activities and bio-geomorphological processes. The surviving geomorphological and geoarchaeological deposits from the Holocene in the Wadi Faynan are demonstrably patchy, manifested in the episodic and discontinuous nature of the data presented here. The recovery of a record of this nature is, nevertheless, unique in an arid zone far from the peat bogs and lakes of higher latitudes. A detailed examination of the geomorphic processes and the depositional environment together have shed important light on the spatial and

temporal interpretations of geochemical records of pollution and the human and industrial histories that they reflect.

The intensity of pollution observed in exposures of the Atlal Member dated from the Early Bronze Age indicates that palimpsests of sediments with distinctive pollution signatures are likely to have existed from that time. The area within 1 km of the Khirbet Faynan contains the greatest number of slag deposits of the Atlal Member (Figures 2-4), but in addition field survey has revealed innumerable small scale episodes of smelting in Roman-Byzantine times at sites ranging from small rocky ledges in the wadi Ghuwayr to the open plains of the Wadi Faynan.

The residual concentrations of copper and lead in the Bronze Age detected near the Khirbet exceed those at one of the world's largest and most notorious centres of smelting in the nineteenth century at Swansea in the United Kingdom. These new data show that the landscape of the Khirbet Faynan is, in significant part, one of profound industrial dereliction in which metal pollutants continue to be re-cycled at a variety of hill-side, slope and wadi-floor locations studied through a number of human, biological and geomorphic systems, these processes operating in modern times much as they must have done when the smelters were active more than sixteen hundred years ago. Such metal recycling and re-working is evident in the local pollution records detected. In particular in the sediments studied at the Khirbet Faynan barrage, the overall pattern of its decline through time from the post-Byzantine period is clear. Evidently, much but far from all, of the decline in the (re) deposition of recycled heavy metals, appears to have been complete within time periods measured in hundreds of years, with the consequence that the local and global pollution records at the start of an episode of intense metal working is likely to be in close correspondence at its beginning, but less so during its decline or end.

Field observation and some measurements reveal a significant number of human and other natural mechanisms that can lead to the movement and concentration of heavy metals at the modern land surface (Table 2). Overall, these observations about the processes and outcomes affecting heavy metals in

geoarchaeological contexts in the study area correspond with those determined previously in desert lands elsewhere. Of these mechanisms, the mix of fluvial and aeolian processes in the modern braided wadi floor produced the most complex results. The pattern of metal pollution in the present surface materials points to, but does not prove, the importance of aeolian processes carrying material 1-2 km from the immediate sources of pollution in the study area. If this result were replicated it would suggest the importance of the local geographical nature of the air-fall metal pollution record in the area, but this needs to be demonstrated in each case. At the land surface, in this area as in other arid regions, copper is lost by surface biogeochemical re-cycling more rapidly than lead, leading to increased proportion of lead in a diminishing overall recycling metal burden. Nevertheless, consideration of the scale of all these natural biogeochemical processes as well as many other human activities identified in the study area and listed Table 2 suggests that the concentrations of heavy metals determined in the particular ancient sediments investigated on the ground in the Faynan region are likely to be in much the same order as those present originally at the time of deposition; in brief, they do not primarily record post-depositional processes.

10.2. *Industrial activity and pollution over time*

The broader overview of metal deposition illustrated in Figure 12 focuses upon the generalised patterns of pollution through time, and not upon detection of any particularly high concentrations of pollutant metals that took place at one former moment in time. Such a comparison reveals many notable similarities and a few differences that also have to be understood in terms of the patchy and fragmented nature of the surviving sedimentary sequence. The former include the following:

- (a) a distinct increase in pollution in the Chalcolithic;

- (b) notable fluctuations in the intensity of pollution through the Bronze Age with a notable episode of actual cessation of metallurgical activity in the Middle Bronze Age, as opposed to an absence of evidence. This appears to have been a time of crisis or change (72) and included a time of sustained aridity which is suggested by the character of some of the deposits and palaeosols recognised within the Tell Loam Member;
- (c) confirmation of the factory-scale copper smelting in the region during the Early Bronze Age discovered by Levy et al. (83, 84) from their excavations in the Wadi Fidan. These findings are supported by the magnitude of concentrations of metal pollutants discovered in the sediments up-wadi of the Fidan, adjacent to Khirbet Faynan where these new data also point to production on a substantial scale that produced intense local metal pollution;
- (d) further overall rises in pollution concentrations in parts of ~Bronze Age, Nabatean-Iron Age,
- (e) profound levels of pollution affecting the non-slag sediments of the Roman-Byzantine periods;
- (f) the notable decline in environmental pollution following abandonment about sixteen hundred years ago,
- (g) a “brief” episode of further copper smelting, perhaps about ~five hundred years ago.

Examination of Table 1 indicates that overall, this is a remarkable correspondence between the pollution record determined in the Dana-Faynan and that worked out independently by the archaeometallurgical field research.

There is one notable difference between the predicted pollution history shown in Table 1 and that established in this research. This concerns the Late Neolithic sequence at Tell Wadi Faynan. Near this site, there was a clear episode of local anthropogenic metal pollution, caused by the discard of ash and charcoal used by smelting on to a stream bank (5021) in the Late Neolithic.

The balance of evidence from published artefact-based, archaeological evidence suggests that the distinctively-polluted deposits were *not* the result of intended, deliberate and purposeful metallurgical activity by people in the Late Neolithic. Nevertheless, this securely dated geomorphic and geochemical evidence is the oldest record yet detected of the oldest anthropogenic metal pollution associated with the use of fire by people.

10.3. *Global dimensions*

The combination of field observation, meteorological and satellite observation of the modern environment and its occupants (Table 2) have demonstrated that heavy metals have not only moved across and been re-worked, but have also been removed by natural processes from this desert margin. The possible wider significance of this export of heavy metals into the global atmosphere can be examined to a limited degree in Figure 13. This compares the overall record of observed metal pollution inferred through time from the Faynan area, bearing in mind the patchy nature of this record, with that indicated by heavy metal records from ice cores in Greenland and high altitude mires in Europe (51, 66, 67, 68, 77, 119, 120, 121).

There are both general differences and similarities between the record of pollution inferred in the Wadi Faynan and the information presented on the inferred production in metric tons per year of copper and lead over the last 5000 years; and in particular with the metalliferous air pollution record attributed to industrial activity in the Classical World (Figure 12). Before about ~3000 year ago, the Faynan pollution record is episodic in character, with distinctive intense local peaks. There were also episodes of little or no pollution; as well as a more general, if episodic, increase in overall heavy metal production through the Bronze Age. The hemispheric record of lead hints at such developments from about 5320 calibrated years before present. Essentially, before ~3-4000 years ago, the global hemispheric pollution record is seen to be largely dominated by the background airborne flux of metals produced essentially by “natural” erosion

and deposition. As proposed above, a simple explanation of this difference between the local and global is that there before the Bronze Age there was insufficient *aggregate* economic “pull” upon such scattered and isolated producing ore-sources such as the Faynan Orefield by any sufficiently large scale or integrated metal-using economies that were capable of generating the massive economic activity needed to create the substantial pollution “push” sufficient to alter the chemistry of the northern hemisphere atmosphere. Analysis of figure 12 also indicates that is a no correspondence at all between the local and the global pollution records for the period from the Industrial Revolution to the modern day, during which the Faynan has been a remote and isolated part of the desert: in contrast, local heavy-metal air-pollution events from roads, transport vehicles and settlements during the twentieth century were detected in the desert at Azraq in the east of Jordan (45, 48).

From the Bronze Age, comparisons between the new Faynan metal pollution data and the existing atmospheric chemical data suggest that there may sometimes be parallelisms annual fluxes of lead and copper (and the closely-related scandium) determined for the northern hemisphere atmosphere. Bearing in mind the patchiness of the Faynan record, this overall parallelism in the apparent intensities of metal pollution is especially marked in Iron Age-Roman-Byzantine times; the period that first attracted the initial interest of Hong et al. (66) amongst others. It is also particularly evident in the decline and subsequent low pollution concentrations that took place after about sixteen hundred years ago in both local and global records.

These broad parallelisms that last over about 4.5 millennia might be the result of chance. Equally, it might be argued that in the absence of quantitative knowledge of the actual air-pollution yield of heavy metals to the global atmosphere over time from the Faynan, and as well as each and every one of the other major sources where metal-rich ores have been exploited, it is premature to offer a more precise diagnosis analysis of the scale of economic drivers that linked local and global air pollution global air-pollution record. Nevertheless, given the archaeological and documentary evidence presented

above, that the Faynan Orefield was a major long-term source for economies in Southwest Asia and the Mediterranean, it appears possible that that degree of correspondence observed does actually reflect the reality of the large scale economic “pull” by large-scale economic entities, that was experienced by such isolated locations as the Faynan, and hence there were global-scale atmospheric pollution consequences.

In our judgment, this degree of parallelism does suggest that at both the local scale and global scale, these two data sets reflect the changing aggregate demand for the geological resources of the Faynan as the scale, integration and power of ancient metal-using economies emerged, changed, interacted or declined at local, regional and continental scales. As a result, it may be useful, more widely to view pollution records as a potential record of economic linkages over thousands of years between such marginal areas such as the Faynan with what were to become the substantial economies of higher latitudes.

Overall, there is insufficient information at present to decide whether the degree of correspondence between one very local pollution record and the global air pollution of the northern hemisphere might actually point to the ancient version, from approximately the Bronze Age to the start of the modern period, of what has been termed a “*world system*”. Many more such studies with detailed knowledge of the scale and significance of pollution-transporting processes are needed. Such an argument does not necessarily imply that the hemispheric air pollution record during the Holocene is only or primarily the product of industrial activity in Southwest Asia and/or the Mediterranean regions, rather it suggests the need to examine the identity and integration of past economic or political entities at the spatial and temporal scales that are appropriate to explaining the magnitude of geochemical pollution effects to which they contributed. Such observations would serve to integrate into local and global scale systems framework, an approach to the history of human activity that would incorporate fundamental (geo)ecological processes such as biogeochemical cycling, into a more traditional archaeological approach based upon “civilisation”, “trade” and “economic activity”.

11. CONCLUSIONS

The study area is seen to be a complex palimpsest of archaeological sites and metal pollution of various ages that is being cleansed by natural processes at a very slow rate given the scale of the original anthropogenic metal burden. A record of heavy metal pollution in copper and lead attributed to the mining, processing and extraction of ores in the remote desert landscape around the Khirbet Faynan in southern Jordan has been established and shown to match many components of the known archaeological evidence of the development, intensification and cessation of mining and copper smelting activity over a period of eight thousand years in the area. This analysis derived from field and air-photograph mapping, detailed geomorphological and stratigraphic analyses, and ICPMS-based geochemical studies has emphasised the local complexities of biogeochemical cycling and re-working, as well as human activities that have existed over time and operated at different spatial and temporal scales. Many complex human and biogeochemical pathways and stores of liberated pollutant metals have been identified, and the scale and significance of a few have been quantified. Determinations of the nature and significance of contemporary natural agencies of pollutant heavy-metal transport confirm findings made in other metal-rich desert lands.

With one exception, the overall pattern of pollution during the Holocene in the study area corresponds surprisingly well with that predicted from published previous archaeometallurgical surveys. Comparisons with records of anthropogenic pollution in the Faynan Orefield and records of the changing geochemistry of the global atmosphere suggest that before about ~3-4,000 years ago, the demand and nature of metal working activity on the ground was too small in scale and scope to have a significant global atmospheric signature. This situation progressively changed after this time: levels of atmospheric pollution from remote sites such as the Faynan made substantial changes to the

geochemical characteristics of the global atmosphere. The local pollution record supports the contention that substantial copper manufactories were present and also affected their local environment in the Early Bronze Age. The modern land surface and landscape of great drama and wildness is an industrial metal-polluted wasteland, much of which stems from intense mining and smelting from the Iron Age to Classical times. After this time, with one brief exception, the liberation of metal pollutants ceased. One exception to this correspondence between the predicted and observed local metal pollution record is the discovery of several “brief” episodes of anthropogenic metal pollution from the heating of copper-bearing ore-materials and the subsequent discard of polluted materials in ash and charcoal; events that took place in a notably different climatic and ecological environment in the Late Neolithic.

There are further parallels between this local pollution record and that observed in the geochemical record of the entire global atmosphere over the Holocene. Such correspondences may be just chance, or perhaps are one local manifestation of the scale and geographical reach (as well as the subsequent later collapse) of the demand for ores and consequent metal production that large-scale, powerful economies made upon ore-rich but difficult landscapes such as the Faynan orefield.

The approach adopted in this paper serves to integrate systems-ecological analyses with standard archaeological approaches in a study that also links the past and present.

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Captions

Table1 Summary of information on the scale mining and metallurgy and anticipated pollution outcomes in the Faynan area (1, 2, 3, 10, 11, 12, 13, 14, 25, 41, 42, 50, 59, 60, 61, 62, 63, 64, 65, 70, 78, 83, 84, 89, 90, 94, 96).

Table 2. An inventory of the natural and human processes that interact with each other as well as with events influence the past and present concentrations of heavy metals in the Holocene in the desert regions of the Faynan Orefield (based upon numerous sources, especially 4, 5, 7, 9, 18, 28, 31, 32, 33, 35, 36, 43, 46, 49, 73, 74, 87, 88, 98, 99, 110, 113, 114, 115, 116, 117, 122, 128).

Table 3. Summary of the Holocene sequences and radiocarbon dates in calibrated years BP that are combined to provide evidence from about eight thousand years to the present day of anthropogenic copper and lead pollution in the vicinity of the Khirbet Faynan: the sites are located in Figures 2 to 5, and the analytical results shown graphically in Figures 7 to 12. (The table is based upon 10-14, 55, 61, 70, 71, 90, 94, 96, 108).

Table 4. Summary of geochemical, sedimentological and lithological properties and dating evidence in wadi-floor sediments at pit WF 5512, illustrated in Figure 7.

Figure 1. The location of the Faynan Orefield in southern Jordan in relation to other European sites mentioned in the text.

Figure 2. The survey area of the Faynan Orefield in relation to the hyper arid desert to the west and the mountain front to the east of the ancient centre of the Khirbet Faynan at the confluence of the Wadis Khalid, Dana, Salawan, Ghuweir (Ghuweyr; Ghuwair) and Ashegair (Ashegar; Ushakir) adapted from 61, 83,84.

Figure 3. Oblique air-photograph looking east of the Khirbet Faynan and its immediate hinterland; wall networks, cones and carpets of copper smelting slag, the Khirbet Faynan to the left (north) of the confluence of the Wadi Ghuweir (centre) and Ashegair; and trees of *Acacia* growing in a small sedimentary basin created by a barrage immediately north of the Khirbet Faynan. Reproduced by permission of Prof. David Kennedy

Figure 4. The distribution of ancient walls and larger areas of ancient copper smelting slag, ancient walls, Khirbet Faynan and Tell Wadi Faynan, and surface samples SP1-38, in relation to the location of the braid-plains of the major wadis, and distributaries at the toe of the alluvial fan to the south. The patterns of Holocene aeolian and stream deposits in relation to the distribution of ancient copper smelting slag and the Khirbet Faynan.

Figure 5. Total concentrations of HNO₃ acid-extracted ICPMS analysis of copper and lead in ppm in the fine fraction sediment matrix of samples SP 1-38 from the surficial materials of Holocene age over alluvial fans, bedrock, braid-plains, within wall-networks, and on ancient slag in the Wadis Dana and Faynan.

Figure 6. Summary of the geochemical record obtained by ICPMS analyses from sediments beneath or impounded by a barrage immediately north of the Khirbet Faynan at site 5512; total concentrations in ppm based upon HNO₃ extraction. The lithological units and geochemical zones are defined in Tables 4 and 4b; radiocarbon dates are calibrated years B.P.

Figure 7. Summary of the total concentrations of HNO₃ extracted copper lead and thallium in ppm and defined geochemical zones from three excavations in copper smelting slag (5738, 5741, 5741), near the Khirbet Faynan and the mine entrance 5740.

Figure 8. The stratigraphic relationships between surface Roman-Byzantine smelting slag; wind blown-water washed materials and soils of the Tell Loam Member and the neolithic site exposed at site 5022, overlying a late Neolithic settlement of Tell Wadi Faynan, and associated meandering fluvial deposits of Holocene age that are exposed at 5021 (figure 10); radiocarbon dates are calibrated years B.P.

Figure 9. Summary of the lithostratigraphy and geochemistry of the Tell Loam Member at exposure 5022 through aeolian-water washed silts and thin palaeosols: in ppm,

Figure 10. Summary of the lithological properties, stratigraphic relationships and sample locations in the fluvial-stream margin sequence at site 5021 near Tell Wadi Faynan; radiocarbon dates are calibrated years B.P. Sample G of ash, as well as charcoal; the latter gave a radiocarbon date of 7240 ± 40 BP; cal BP 2σ 7240-6990 cal. (Beta-205964).

Figure 11. Summary of sample properties and geochemical properties of Neolithic age from an exposure of aeolian, water washed silts and temporary soils (5021), Tell Wadi Faynan; in ppm. Sample G of ash, as well as charcoal; the latter gave a radiocarbon date of 7240 ± 40 BP; cal BP 2σ 7240-6990 cal. (Beta-205964).

Figure 12. Comparison between the pollution record for copper and lead over 8,000 years for the vicinity of the Khirbet Faynan, in ppm HNO_3 extraction ICPMS ; with estimates of copper and lead production in the Classical World; and the calculated rate of metal fluxes to high level mires in Europe and the Greenland icesheet (after 15, 19, 20, 119-121).