Journal of Archaeological Science 34 (2007) 83–110

Journal of Archaeological SCIENCE

http://www.elsevier.com/locate/jas

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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Received 16 November 2005; received in revised form 21 March 2006; accepted 2 April 2006

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 provides a significant impediment to such approaches that seek to research “locally and think globally”.

Keywords: Ancient economy; Atmospheric circulation; Deserts; Geomorphology; Holocene; Jordan; Lead; Copper; Thallium; Mediterranean; Pollution; World-systems theory

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doi:10.1016/j.jas.2006.04.004

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 (Figs. 1 and 2). This Orefield covers ~500 km^2 [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 (Figs. 1–3) which is associated with extensive metallurgical activity in the Roman-Byzantine period (~2550–1450 calendar years BP: 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 Ifdan (Fig. 2) with geochemical evidence for substantial copper manufaction 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 Fig. 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

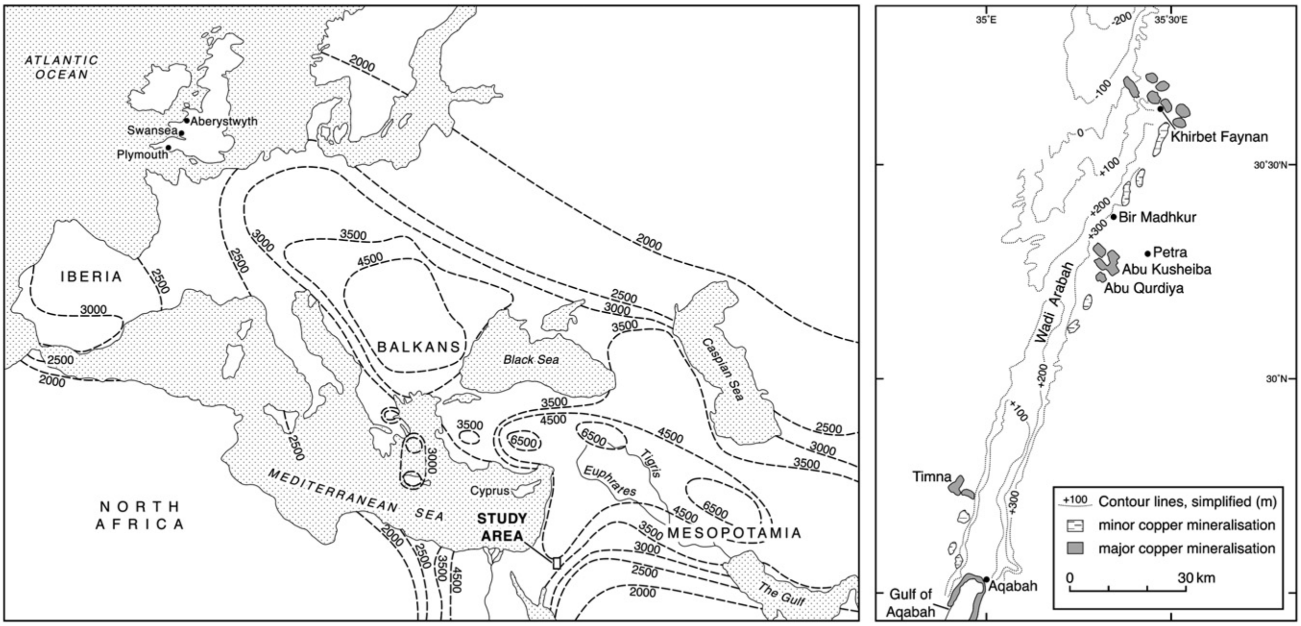

Fig. 1. The location of the Faynan Orefield in southern Jordan in relation to other European sites mentioned in the text. Contours reflect the dates of the introduction of metallurgy.

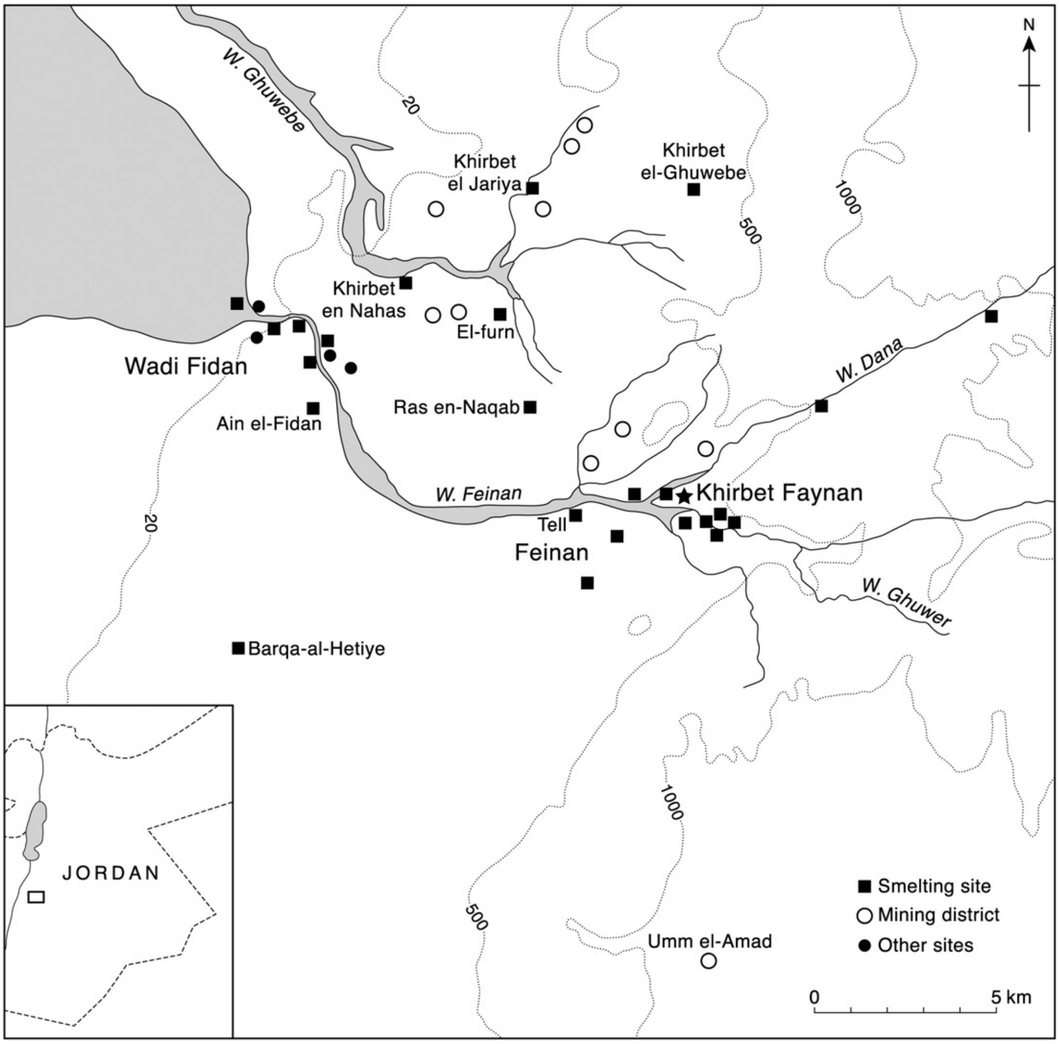

Fig. 2. The survey area of the Faynan Orefield in relation to the hyper arid desert to the west and the mountain front to the east of the ancient centre of the Khirbet Faynan at the confluence of the Wadis Khalid, Dana, Salawan, Ghuweir (Ghuweyr; Ghuwair) and Ashegair (Ashegar; Ushakir) adapted from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

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 (Fig. 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

Fig. 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.

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 [15,19–23,80,81,92,93,119–121].

- 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. [26,37–39,57,75,112,118]).

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 [9–14,61]. 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. [67–69] to be in the region of 2300 t/year. 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 5500 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; 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 obscure.

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 [53,54,56,103–106]. 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 also 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 that 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 Condron [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

Table 1
Summary of information on the scale of mining and metallurgy and anticipated pollution outcomes in the Faynan area [1–3,10–14,25,41,42,50–65,70,78,83,84,89,90,94,96]

Approximate age; with lithostratigraphic member; and radiocarbon dating in the Wadi Faynan	Environment in the Wadi Faynan (after [71,90])	Nature and intensity of copper mining and smelting, largely after Hauptmann [61]	Predicted and *actual anthropogenic sediment metal-pollution, and related geomorphic outcomes
~9600–7500 BP. Pre-pottery Neolithic (8th-7th millennium BC). Faynan Member, upper component, including a palaeosol overlying a fluvial channel fill at 5015 dated to 7240 ± 90 BP; cal BP 8276–7868 (Beta 11121)	Trees and shrubs (*Quercus*, *Ostrya carpinifolia*, *Juniperus*, *Pinus*, *Tamarix*, with steppe and grasslands, perhaps affected by grazing) grow in wetter climate than prevails today which progressively desiccates, before wetter conditions in late Neolithic	The first use of local copper ores in the Wadi Ghuweir. 'Greenstone' beads and green powder for cosmetic purposes from Faynan became popular throughout Jordan and Palestine	Negligible anthropogenic pollution expected
~7500–6000 BP. Late Neolithic (6th–5th millennium BC). Faynan Member, upper component. Sample G in the fluvial-stream bank sequence of 5021 radiocarbon dates to 7240 ± 40 BP; 2σ cal. BP 7240–6990 (Beta 205964), the Faynan Member, upper component. Field relationships with Tell Wadi Faynan, TWF [96] adjacent to 5021; both are overlain by the deposits of exposure 5022. At TWF archaeological materials dated to 6110 ± 75 BP: cal BP 7235–6761 (HD12338): 5740 ± 35 BP; cal BP 6654–6412 (HD 12337): and 5375 ± 30; cal BP 6278–5995 (HD12336)	A wetter climate with perhaps ≥ ~150 mm/annum, rainfall which supports a perennial river in the wadi Faynan. This deposits epsilon cross-bedded sands and silts with anthropogenic wastes that form the upper part of the Faynan Member. Trees and shrubs (*Quercus*, *Ostrya carpinifolia*, *Juniperus*, *Pinus*, *Tamarix*, perhaps at stream sides, elsewhere with steppe including *Ephedra*, perhaps also grasslands), however, as a result of distinctive regional climatic aridification, the lowland landscape changes to steppe with some *Pinus* and *Pistacia*. Cultivation and pastoral farming present. During the latter stages of this period, aeolian processes become important depositing dunes of silt and sand with carbonate induration which form the basal part of the Tell Loam Member	Copper ores and some 'greenstone' beads at Tell Wadi Faynan; reflecting the use of copper, but not the deliberate smelting of ores. Ores from Umm 'Ishrin Sandstone (sometimes with significant lead) used from exposures in wadis and mountain front	Negligible anthropogenic pollution that is relatively lead-rich expected with corresponding minimal pene-contemporaneous re-cycling. *Occasional pollution of stream bank environment with small pockets of significant pollution in copper, and particularly of lead and thallium, when associated with ash and charcoal (5021; #B, C,D,G, H,L) that were discarded onto the banks of a perennial stream. The precise character of the human activities that produced these packets of polluted sediment is unknown, at present
~5500 BP. Chalcolithic. Tell Loam Member. 5022 dated by reference to 5021 below. Khirbet Member associated with Chalcolithic and Bronze Age at 5051 and 5518 [12]	Marked climatic aridification continues, with perennial water confined to spring-fed runoff water in the Wadis. Wind-blown water-washed silts, sands with carbonate induration and ped development at on low rise on the Tell Wadi Faynan produces Tell Loam Member (5022). A grass-dominated steppe occurs with no evidence of trees may have occurred at this time or slightly later. Essentially the modern pattern of climate, with its occasional winter storms and floods. Braid-plains incise, causing the older Holocene fluvial deposits to be buried, eroded, or abandoned in wadi-cliff edges	As at other Chalcolithic settlement in Jordan and Palestine, evidence of pyro-metallurgical activities in the second half of the 4th millennium BC indicating "household metallurgy", small-scale operations. Small pieces of Chalcolithic slag and copper prills at Tell Wadi Faynan, small scale mining and metal working took place in the region in the Faynan	Small-scale, local pollution, with corresponding pene-contemporaneous re-cycling of liberated metals. *Estimated to be represented by 5022, geochemical zone 4, sustained, distinctive, increase in copper and lead when compared to 5021; but with no proportional increase in lead to accompany copper (zone 4b); no peaks of thallium or other volatile elements associated with ash-charcoal. Minor re-cycling and/or dilution of metal pollutants in carbonate, as a result of wind and surface wash in an increasingly open arid environment

~5550–3950 BP. Early Bronze Age. Tell Loam Member at 5022 age estimated by reference to 5021 below and reference to adjacent TWF [96]. Atlal Member at 1491/5741, radiocarbon date of 4240 ± 40 BP; cal BP 6190–5940 (Beta 203414); which is overlain by 5690 ± 40 BP; cal. BP 6550–6400 (Beta 203413)	A grass-dominated steppe with no evidence of trees may have occurred at this time. Essentially the modern climate and geomorphic regime: represented by the Tell Loam Member of 5022. "Social collapse" or some form of move from urban settlements to rural settlement posited elsewhere in Early Bronze Age IV	The first extensive mining and pyro-metallurgical activities, peak in Early Bronze Age II and III; high-grade, copper-manganese ores from widespread but thin, secondary enrichment, including malachite and chryscolla, in Numayr Dolomite Limestone and elsewhere in Burj Dolomite Limestone-Shale exploited. Mines in Wadis Khalid and Dana. Mining and pyrotechnology improve. Large scale copper "manufactory" at Khirbat Hamra Ifdan (~4150–4650 calendar years BP)	Significant local pollution, relatively low in lead, with corresponding pene-contemporaneous re-cycling. *Estimated to be represented by 5022 (geochemical zone 4 b/a) with sustained increase in copper pollution, with no proportional increase in lead to accompany copper peak, i.e. relatively lead-poor ores; no peaks of thallium or other volatiles that could have been deposited onto ash and charcoal
~3950–3450 BP. Middle Bronze Age, Tell Loam Member at 5022 age estimated by reference to 5021 below and reference to adjacent TWF [96]	Essentially the modern climate and geomorphic regime: represented by the Tell Loam Member of 5022; but strong sustained drought evidenced to the west; perhaps with significant down-cutting by water in wadis? A grass-dominated steppe with no evidence of trees may have occurred at this time	Relatively little ore extraction or processing. Minimal pollution, only minor re-cycling	Minimal pene-contemporaneous anthropogenic pollution, minor re-cycling of existing metal pollution burden. *Sustained absence of pollution signatures (5022, zone 3)
~3650–2850 BP. Late Bronze Age to Early Iron Age; Tell Loam Member, at 5022 age estimated by reference to 5021 below and reference to adjacent TWF [96]. Atlal Member at 5739, radiocarbon dating: 2860 ± 40 BP; cal BP 2862–3140 (Beta 203411) and 3390 ± 40 BP; cal BP 3485–3816 (Beta 203402) at 5512	Trapping and management using walls of winter floodwaters to sustain runoff farming take place; development of the major network of walls to manage water, and provide crops and animal products east of the Wadi Faynan. A grass-dominated steppe with no evidence of trees may have occurred at this time. Essentially the modern climate and geomorphic regime: represented by the Tell Loam Member of 5022	Renaissance of metal production and smelting	Significant local metal pollution, relatively low in lead, with corresponding pene-contemporaneous re-cycling. *Progressive increase in copper pollution, a proportionately larger increase in lead pollution (5022 zone 2d), followed by minor reduction in heavy metal pollution (5022 zone 2c); (uncertain, inferred decline in input by wind of carbonate minerals?). Substantial metal pollution of clastic sediments within slags at 5739. Substantive metal burdens in ore-processing deposits at 5512 (unit 6)
~2850–2550 BP. Iron Age, 1st millennium BC. Tell Loam Member at 5022 where age estimated by reference to 5021 below and reference to adjacent TWF [96] and overlying Roman-Byzantine sherds. Atlal Member at base of 5512, radiocarbon date of 3390 ± 40 BP; cal BP 3485–3816 BP (Beta 203402)	A grass-dominated steppe with no evidence of trees may have occurred at this time. Essentially the modern climate and geomorphic regime: represented by the Tell Loam Member of 5022, and wadi floor and barrage infill deposits at 5512 and 5017	Second main period of copper production in region. Innovations in mining and smelting occur, together with further substantial expansions of copper mining and smelting. Original outcrops of high grade copper ores in Burj Dolomite Limestones Shale become exhausted. Mining reverts to the Umm 'Ishrin Sandstone, which contains less lead with re-processing of earlier prehistoric slag. Metallurgical activities on a new industrial scale. Mining and smelting, well-organised and sophisticated scale with improved geological understanding. Exploitation of deep mineralization by shafts 60 m deep. Faynan region has largest copper production of the entire Near East beside Cyprus. 100,000 tons of black, copper slag, still often unvegetated, left at the Khirbet Faynan and Khirbet en-Nahas	Substantial metal pollution, relatively lead-rich anticipated, with corresponding extensive pene-contemporaneous re-cycling. *Sustained increase in sediment pollution in copper; with significant pollution in lead that fluctuates slightly in its intensity (5022 zones b and a). Notable pollution associated with ore-processing at 5512

(*continued on next page*)

Table 1 (*continued*)

Approximate age; with lithostratigraphic member; and radiocarbon dating in the Wadi Faynan	Environment in the Wadi Faynan (after [71,90])	Nature and intensity of copper mining and smelting, largely after Hauptmann [61]	Predicted and *actual anthropogenic sediment metal-pollution, and related geomorphic outcomes
~2550–1450 BP. Roman-Byzantine Period. Tell Loam Member through field relationships with Roman-Byzantine remains at (5022 zone 1). Atlal Member at 5731, 5738, 5741; Khirbet Member at 5512–5017. Excavation, archaeological associations, pollen-biostratigraphy; basal radiocarbon date 1800 ± 40 BP; cal BP 1611–1858 (Beta 203401). Disaggregated charcoal fragments in borehole 5017 ~225 cm 2630 ± 50 BP; cal BP 2543–2859 (Beta 110840) suggest recycling of old charcoal	Steppic and desertic vegetation and geomorphic regime, with a wetter climate; perhaps but not certainly local exhaustion of wood supplies. Extensive pressures on wooded vegetation for fuel and for cultivation and grazing may have caused the vegetation to remain open and steppic. In Byzantine Period, Khirbet Faynan became an even more significant location: it became the Seat of a Bishopric, with three other churches. Lower parts of barrage infill deposits at Khirbet Faynan (5512, 5017, and ash-rich smelting waste (e.g. 5738, 5741)	Extensive use of shafts and deeper underground passages in Mn-rich ores in Umm. 'Ishrin Sandstone, together with new methods of smelting countered the local exhaustion of the thin seams of DLS. Large scale mining and smelting from the first century BC to fifth century AD, much exploitation in the third and fourth centuries AD. At Umm el-Amad, mines first worked 2500–3000 years earlier during the Chalcolithic/Early Bronze ages, are significantly re-used; old mines serve as entrances for the new larger underground mines driven through shafts with deep connected underground passages. 1.5 m high galleries interpreted to indicate animals transport the ore inside the mines as well from the mines to the central smelting works at Faynan. Spring and stream water was supplied to the Khirbet by a series of aqueducts and stored in a large reservoir. 50–70,000 tons of copper slag at Faynan (e.g. 5052, 5053). Metal working with professional mine engineers. Reports that forced labourers on occasion receive brutal treatment	Substantial metal pollution, relatively lead-rich anticipated, with corresponding extensive pene-contemporaneous re-cycling. *Substantial and dangerous concentrations of copper and lead, initially rich in lead, with change from relatively lead-poor to lead-rich pollution in wastes reaching >40k ppm Pb; 16k ppm Cu, and substantial 90 ppm Tl where there are ash-charcoal from smelting (5738, 5741); transition from relatively rich to lead-poor pollution at dangerous, poisonous and toxic concentrations with substantial pollution in crushed ores in colluvium, then in impounded water by smelting areas. Disposal of massively contaminated wastes from smelting as waste piles and onto braid plain where rapid geomorphic removal and/or dilution down-wadi (5731, unit 3), occurs suggesting significant geomorphic re-recycling indicated by fluvial deposits producing heterogeneity and undated wind-induced distance-decay effects to south SP8-2; copper and lead appear to behave differently. Many smaller sites (SP1,6,18, 31, 32-35), display intense local pollution with copper and lead with associated suites of dangerous volatile metals sporadically preserved with ash-charcoal (5022 zone1)
~1450–650? calendar years BP. Early Arab period; Khirbet Member at 5512, 5017 through biostratigraphic correlation with radiocarbon dated deposits nearby; and underlying radiocarbon dates; ?Atlal Member at 430 ± 40 BP; cal BP 350–330 BP (Beta 203412)	Steppic and desertic vegetation and geomorphic regime, in climate similar to that of the present day, but which became increasingly arid. Represented by barrage infill deposits at 5512 and 5017, and Upper and Lower Dana Wadi Members	Mining and smelting activity around Faynan decline rapidly after the Roman-Byzantine period. After ~500 AD the Faynan ceases to be as a major copper supplier in the Levant. Small-scale smelting, during the early medieval Islamic periods at el-Furn, Faynan, Ain Fidan and probably in the Wadi Dana. Minor local pollution expected, with local re-working of exposed smelting wastes from preceding smelting. In late Ayyubid/early Mamluk period of the thirteenth century AD, possibility that in addition to copper, lead was deliberately smelted	Loss of anthropogenic metal pollution input, with progressive decrease metals at the aggrading sediment surface. *Initial rapid decline in concentrations of copper and lead, and associated metals after peak of Byzantine activity, followed by progressive reduction over ~1300 years to present, during periods in which the geomorphic environments altered with one sustained episode of aridity; input of strontium increases; lead reduces in concentration in surface deposits much less rapidly than copper. One episode detected of the deposition of copper slag?

~650? to present; Khirbet Member. ? Atlal Member at 5450 dated to 430 ± 40 BP; cal BP 350–330 BP (Beta 203412)	Steppic and desertic vegetation, become fully desertic with marked aridification that lasts from about ~550–100 years ago, when fully modern conditions occur, with wind-blown silts entering the global atmosphere	No metallurgical activity: re-cycling of surface deposits anticipated. Modern survey of ore resources by the Natural Resources Agency clears some entrances to mines and adits	Progressive decrease in re-cycled metals, minor input of new anthropogenic metals in sediments about ~500 years ago. *One episode detected of the deposition of copper slag? Recycling of copper and lead continued at comparatively low rate through biological processes to a few hundred ppm; fluvial and aeolian processes on wadi floor produce great heterogeneity in heavy metal burdens; mix of aeolian and overland flow, recent ploughing and clearance, and the sporadic nature of many sites, the surface heavy metal loads beyond the land around the Khirbet Faynan and the wadi-floor are also heterogeneous

behind the Khirbet Faynan barrage (site 5512, Figs. 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 of 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 (Fig. 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 <2 mm 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 Figs. 4 and 5. In practice, field experience suggests the map presented in Fig. 4 under-emphasises the distribution of smelting slags but more appropriately represents

Table 2
An inventory of the natural and human processes that interact with each other as well as with events to 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 [4,5,7,9,18,28,31–33,35,36,43,46,49,73,74,82,87,88,98,99,110,113–117,122,124,128])

Vegetation

Composition, form, cover; interception capability; soil drainage; soil biological, physical & chemical properties; bioaccumulation in food chains and decomposer chains; soil binding and protection; long-term changes & responses to climate, colonisation & recolonisation, fuel gathering, wood for construction, grazing and browsing, clearance and burning; selective uses, susceptibility to toxins especially riparian vegetation.

Regional & local climatology

Global & regional climate changes, wetter early Holocene, mid-Holocene aridification, arid later Holocene. Short term fluctuation & extreme events, droughts, floods, storms, local climatology. Katabatic winds, wildfire, bioclimatology, shelter & shade, updraughts.

Sources of ore metals in the environment.

Bedrocks:	Lower Palaeozoic Proterozoic
"Natural" Quaternary sediments	Polluted sediments and soils Colluvium Fluvial sediments

Bioaccumulation and human activity

Differential recycling by deep rooted trees, pasture & crops, grazing & geophagy by invertebrates and vertebrates; defecation by herbivores. Burning wood and manure as fuel that releases or absorbs metals in ash. Human inhalation, ingestion, consumption, contact, washing and cleaning; medicines; dyes and cosmetics, cooking; use of manure as fertiliser; night-soil as fertiliser; waste disposal; open water storage and transport; washing, handling and contact with tents, clothing, textiles, artefacts, animals, fuel, tents and materials. Metal slags as building materials.

Land surface changes

Inter-connected changes in surface geomorphology & soils, especially soil & slope stability from mass-movement, mining, quarrying of building stone, rock breaking and crushing, smelting; trampling by people, animals, wheeled and sledge vehicles; ploughing, tillage, sowing, harvesting, rain and floodwater collection, transport and use of plant wastes, animal manures and night-soil as fertiliser; changes in infiltration capacity, overland flow, in surface and stream runoff rates; differential erosion, transport and deposition by wind and water, deposition of dust, changes in the size, erodibility, floods and windstorms on the floodplain. Sub-surface differential leaching, translocation and accumulation, and biological concentration and transport by soil fauna and vegetation. Accumulation behind walls, within and from natural and constructed water courses & water storage facilities; upon and within tents, clothing, artefacts.

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) (Figs. 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 (Fig. 5); the pattern is however, less clear for lead (Fig. 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 Fig. 2 whose predecessors 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 suggested by 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

Fig. 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.

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 Figs. 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 the 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 activity that was in the immediate study area: typically

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 Figs 2–5, and the analytical results shown graphically in Figs. 7–12 (the table is based upon [10–14,55,61,70,71,90,94,96,108])

Deposits, sequence	Summary, location, lithology, properties
Modern land surface. Palimpsest of deposits and materials of different ages	Surface samples SP1-38 obtained along transects across and through the modern braid-plain and onto Holocene deposits of the Wadis Dana and Faynan. These include modern and late Holocene, wind blown and water-washed silts and sands, and deposits of copper smelting slag that range in age from the Bronze Age to Byzantine. The modern surface associated with in the ancient wall networks is visibly a palimpsest of *in situ* and re-worked materials reflecting its modern geomorphic situation and its episodic development from the Bronze Age to Byzantine, parts of the wall network continue to concentrate and re-direct overland flow in times of storm (see Figs. 4 and 5)
Ancient barrage, impounded sediments and earlier land surface deposits. ∼3700 calibrated BP; and ∼1800 calibrated BP to the Present Day	Khirbet Member overlying Atlal Member. The Khirbet Faynan Barrage, immediately north of Khirbet Faynan, sites 5017 and 5512. The Khirbet Member of aeolian, storm and pond deposits from the present to the base of lithological unit 5 at 260 cm near to a radiocarbon date 1800 ± 40 BP; cal BP 1611–1858 (Beta 203401). Disaggregated charcoal fragments in borehole 5017–225 cm 2630 ± 50 BP; cal BP 2543–2859 (Beta 110840) suggest recycling of old charcoal. The basal 70 cm of exposure is of the Atlal Member (unit 6) comprises colluvium and crushed ores, radiocarbon dated to 3390 ± 40 BP; cal BP 3485–3816 (Beta 203402) at 5512. The lower 70 cm of crushed ores and colluvium accumulated rapidly as a result of metallurgical and colluvial processes. The overlying sediments plus a mix of lacustrine, fluvial, aeolian and colluvial material accumulated behind the well-constructed barrage until about 1400 years ago; after which time, aeolian sands, colluvium and fluvial sediments have infilled the basin (see Fig. 6)
Chalcolithic to Mediaeval metal-working sediments. ∼5450–500 calibrated BP	Atlal Member, intrusive inter-bedded fluvial sand lenses. Three accumulations of metal slags with ash; radiocarbon dates exist at two sites located between the Khirbet Faynan barrage and the braid plain of the wadi Dana. In the basal 20–30 cm at 5738 and 1471/5741b, copper slag and ash were interbedded with fluvial braid-plain sands (see Fig. 7). Atlal Member at 1491/5741, radiocarbon date of 4240 ± 40 BP; cal BP 6190–5940 (Beta 203414); which is overlain by 5690 ± 40 BP; cal. BP 6550–6400 (Beta 203413) which suggests re-working; and which is in turn overlain by 430 ± 40 BP; cal. BP 350–330 (Beta 204412). Atlal Member at base of 5512, radiocarbon date of 3390 ± 40 BP; cal BP 3485–3816 BP (Beta 203402). Atlal Member at 5739, radiocarbon dating: 2860 ± 40 BP; cal BP 2862–3140 (Beta 203411); and 3390 ± 40 BP; cal BP 3485–3816 (Beta 203402) at 5512
Copper mine, Nabatean, Byzantine	Hamman Member. Wadi Khalid, 5740 (mine 2 in [55]): 1.1 m of post mining colluvial and aeolian slumped back into mine entrance covering 1 m of slumped mining debris that is attributed to the Nabatean to Byzantine times (see Fig. 7)
Neolithic, Byzantine environmental sequence. ∼6200–1400 calibrated BP	Tell Loam Member. A quasi-continuous sequence of water-washed wind-blown silts/sands without pits or cuts exposed in a cliff at the southern edge of the braid-plain of the Wadi Faynan at a location that is 5–10 m higher than all the immediate surrounding area. This extends at site 5022 from surface copper smelting slag with ash and pottery attributed to the Roman-Byzantine (∼2000–1600 years ago) down through similar water-washed aeolian deposits Iron Age, Bronze Age, Chalcolithic to the upper parts of the Late Neolithic site of Tell Wadi Faynan 6110 ± 75 BP; cal BP 7235–6761 (HD12338): 5740 ± 35 BP; cal BP 6654–6412 (HD 12337): and 5375 ± 30; cal BP 6278–5995 (HD12336). The site overlies 5021 radiocarbon dates to 7240 ± 40 BP; 2σ cal. BP 7240–6990 (Beta 205964). Detailed field examination of the excavation face that produced site 5022 is summarised in Fig. 9
Early-Mid Holocene perennial stream deposits. ∼7470–6525 calibrated BP	Faynan Member, upper component. A quasi-continuous sequence of fluvial, stream bank and anthropogenic deposits of a perennial meandering stream in a wetter climate with evidence of channel fill, erosion and non-sequences at site 5021. 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). These deposits can be traced lithostratigraphically in the cliff exposure to the lower components of the Late Neolithic site at Tell Wadi Faynan at 5022 ([96]; their plate II: 1-2) where the appropriate part of the sequence provided the following radiocarbon dates: 6110 ± 75 BP; cal BP 7235–6761 (HD12338): 5740 ± 35 BP; cal BP 6654–6412 (HD 12337): and 5375 ± 30 BP; cal BP 6278–5995 (HD12336 (see Figs. 10 and 11)
List of radiocarbon dates quoted	430 ± 40 BP; cal. BP 350–330 (Beta 204412) 1610 ± 40 BP; cal. BP 1403–1601 (Beta 203399) 1800 ± 40 BP; cal BP 1611–1858 (Beta 203401) 1870 ± 40 BP; cal BP 1712–1890 (Beta 203400) 2630 ± 50 BP; cal BP 2543–2859 (Beta 110840) 2860 ± 40 BP; cal BP 2862–3140 (Beta 203411) 3390 ± 40 BP; cal BP 3485–3816 (Beta 203402) 4240 ± 40 BP; cal BP 6190–5940 (Beta 203414) 5690 ± 40 BP; cal BP 6550- 6400 (Beta 203413) 5375 ± 30 BP; cal BP 6278–5995 (HD12336) 5740 ± 35 BP; cal BP 6654–6412 (HD 12337) 6110 ± 75 BP; cal BP 7235–6761 (HD12338) 7240 ± 40 BP; cal BP 7240–6990 (Beta 205964)

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

Depth (cm)		
Lithofacies unit 5512: Sedimentological properties and dating		
1	0–86	Alternating layers (1–2 mm thick) of clay and silt; sometimes distinctly organic with leaf remains; interbedded with layers of fine and medium sand, often well sorted; individual layers bioturbated; pale brown; occasional roots; occasional large boulders; transitional lower boundary. *Interpretation: wind blown and water-washed and barrage-pool deposit of reworked sand with boulders from catchment, boulders introduced by or fallen from adjacent archaeological remains; clay-silt layers and organic debris are intermittent periods of pond-sedimentation*
2	86–157	Clay, silt and sand; sands often well sorted often in couplets of coarse to fine sands ~3 mm thick, but ranging from 1 mm to 8 cm thickness; pale brown; slightly irregular surfaces. Biostratigraphic correlation with arid episode of the Little Ice Age [71]. *Interpretation: aeolian deposits with episode of barrage-pool sedimentation at times of storm, some events with large sediment load*
3	157–158	Fine sand with grit; forming irregular laminae, pale brown. *Interpretation: distinctive flood-lag deposit in the barrage-pool*
4	158–205	Pale brown fine quartz and limestone sand; distinctive thin (1–4 mm thick) laminations; typically well-sorted; marked fining-upward in laminae with granite-derived grit and fine pebbles of lag deposits at the base of each lamina. Greater % sand than unit 5 below; brown hues. No evidence of any of the following were found: ash, charcoal, copper ores, copper slag, colluvial activity, local turbidity currents, mass-movement or sediment deformation, intra-sequence desiccation cracks, induration, or mineral deposition. Roman potsherds at 1.95 m. Dating: charcoal fragments between 174 and 176 cm, 1610 ± 40 BP; cal. BP 1403–1601 BP cal. (Beta 203399); between 204 and 206 cm 1870 ± 40 BP; cal BP 1712–1890 years (Beta 203400). *Interpretation: runoff and storm deposits into a perennial pool behind the barrage-pool; moving water re-working of surface materials in the immediate area*
5	205–260	Fine sand, often with much clay, some silt; irregularly laminated with laminae between 1 and 3 mm thick; lenses of sorted sands, including lenses of sorted sand-sized, green copper ores; comminuted charcoal present throughout; some large clasts of charcoal but less comminuted charcoal than unit 6 beneath; cobbles and boulders are common; the bases of several have deformed underlying laminae producing distinctive ''bird's eye'' deformation-loading structures; comparatively high %LOI and very high magnetic susceptibility. Localised variant between 240 and 260 cm clay-rich deformed sediment; overall poor sorting; occasional lenses of well sorted, sand-sized grains of copper ore and ash, colour varies: sand matrix, pale brown; ore sands, green; ash, grey. No evidence found of intra-sequence desiccation cracks, induration, or mineral precipitation. Roman potsherd at 2.28 m. Dating: Nabatean potsherd at 2.35 m; charcoal fragments at 224–226 cm 1800 ± 40 BP; cal BP 1611–1858 (Beta 203401). *Interpretation: a complex of individual mass flow deposits and local turbidity currents, and deposits of ponded-water and moving-water that accumulated rapidly in a perennial pool on a wadi-floor. Frequent local use of hot fires producing ash and charcoal hereabouts and some smelting nearby; at the site there was mass-movement and deformation of wet sediment and re-working, in perennial water; where crushed ores had been crushed, graded, and size-sorted (perhaps sorted in a flume or sluice), but not yet smelted. High clay content reflects erosion of exposed surfaces and profiles. No evidence of dryland colluvial activity or dry wadi floor*
6	260–285	Diamicton; matrix supported; abundant sand-sized matrix of different materials; some silt and clay; with numerous angular clasts angular slag-clinker; angular cobbles of limestone, no evidence of abrasion; much comminuted charcoal but no charcoal clasts seen; overall very poorly sorted; stratification not clear; rests upon a very hard, impenetrable layer of clay-sand located on a bedrock surface; black to dark brown colours. No evidence of intra-sequence desiccation cracks, induration, mineral deposition, slip planes, ponded or moving water. Comparatively raised % LOI and magnetic susceptibility. Dating: charcoal between 267 and 276 cm was 3390 ± 40 BP; cal. BP 3485–3816 (Beta 203402). *Interpretation: product of ore-smelting involving fire, ''anthropogenic colluvium'' and mass movement, with minor impact of deposits of airfall ash and silts from catchment, and overland flow, in a wadi floor that was dry, suggesting a hot and arid climate. The abundant sand-sized materials related to disintegration of the bedrock and ore materials. No perennial water, no barrage*
Geochemical zone 5512 KFB: geochemical properties and trends		
1	0–160	Typically copper less than 1000 ppm, lead typically 300–500 ppm; minimal fluctuations after an overall decline in concentrations upwards from the initial copper concentrations of ~2100 ppm at 159 cm, strontium 320–450 ppm and thallium ~1.5–2.5 ppm. *Interpretation: recycling by wind and runoff of natural materials and exposed smelting products*
2	160–230	Four or more fluctuations in concentrations of copper and lead; thallium concentrations low; overall a long-term decline in the input of heavy metals. Copper concentrations vary from ~1800 ppm to ~4500 ppm; lead concentrations are low, typically ≤200 ppm, with minimum ~75 ppm; strontium 200–450 ppm; thallium low ~1.5–2.5 ppm. Transitional change below to geochemical zone KFB3. *Interpretation: an uncertain combination of the following: incorporation and sorting of polluted earth surface materials from the area by wind and perhaps overland flow into a perennial water body with abundant moving water in an area of overall declining metal processing activities; from metallurgy nearby but not at this particular wadi-floor site; and-or the pattern of loss of residual pollutants on adjacent land surfaces by natural processes from a landscape from which metallurgy had already ceased*
3	230–260	Up-profile from base; copper declines to minimal ~470 ppm at 250 cm, then increase to ~4500 ppm in concentrations of copper. Lead concentrations at ~500 ppm and essentially stable with minor trough at 250 cm; strontium ~50–300 ppm; thallium typically <3.5 ppm, but 26 ppm at 250 cm. *Interpretation: pollution from ore processing and smelting, fluctuates in intensity according to type and intensity of ore processing and smelting, and according to input by geomorphic processes of metal-poor clays from adjacent surficial deposits*
4	260–282	Two peaks and a trough with lead concentrations in range from ~2400 to ~7825 ppm; and always significantly exceeding those of copper in range ~1000–2230 ppm; high thallium, 9–13 ppm at base; maximum 25–33 ppm at top of zone; strontium ~320–450 ppm. *Interpretation: lead-rich copper ore accumulating on dry wadi floor during the processing and smelting of copper ores*

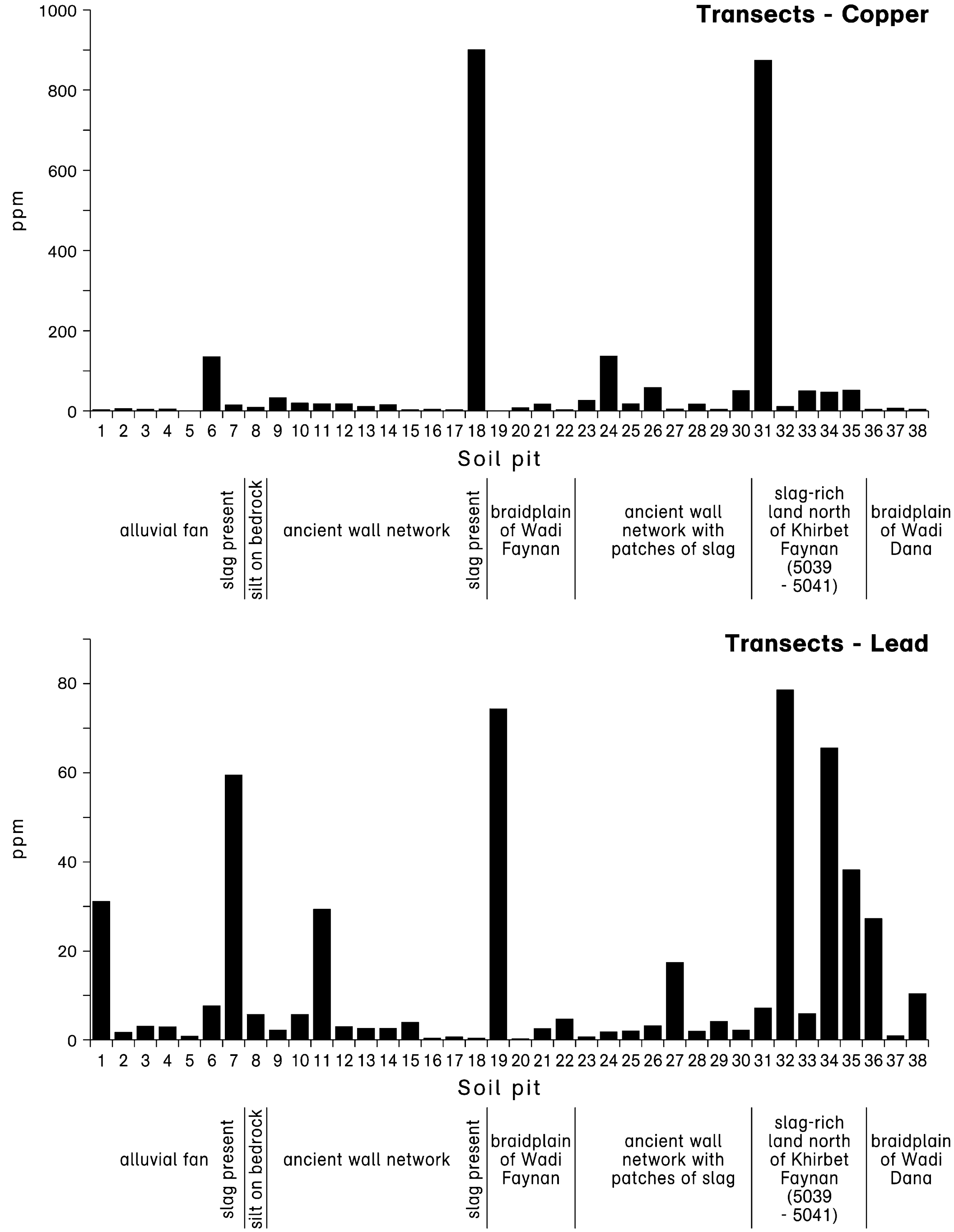

Fig. 5. Total concentrations of HNO_3 acid-extracted ICPMS analysis of copper and lead in ppm in the fine fraction sediment matrix of samples SP 1-38 from the surficial materials of Holocene age over alluvial fans, bedrock, braid-plains, within wall-networks, and on ancient slag in the Wadis Dana and Faynan.

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, ~5 m tall, which grow on fine grained sediment impounded behind a substantial barrage wall that has been constructed across the wadi (5512, Figs. 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 Fig. 6. To aid description and interpretation, the geochemical data (Table 4 and Fig. 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 and 6 with other evidence of the intense local smelting, and the crushing and processing of metal-rich materials. Above the 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 (Fig. 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 Figs. 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

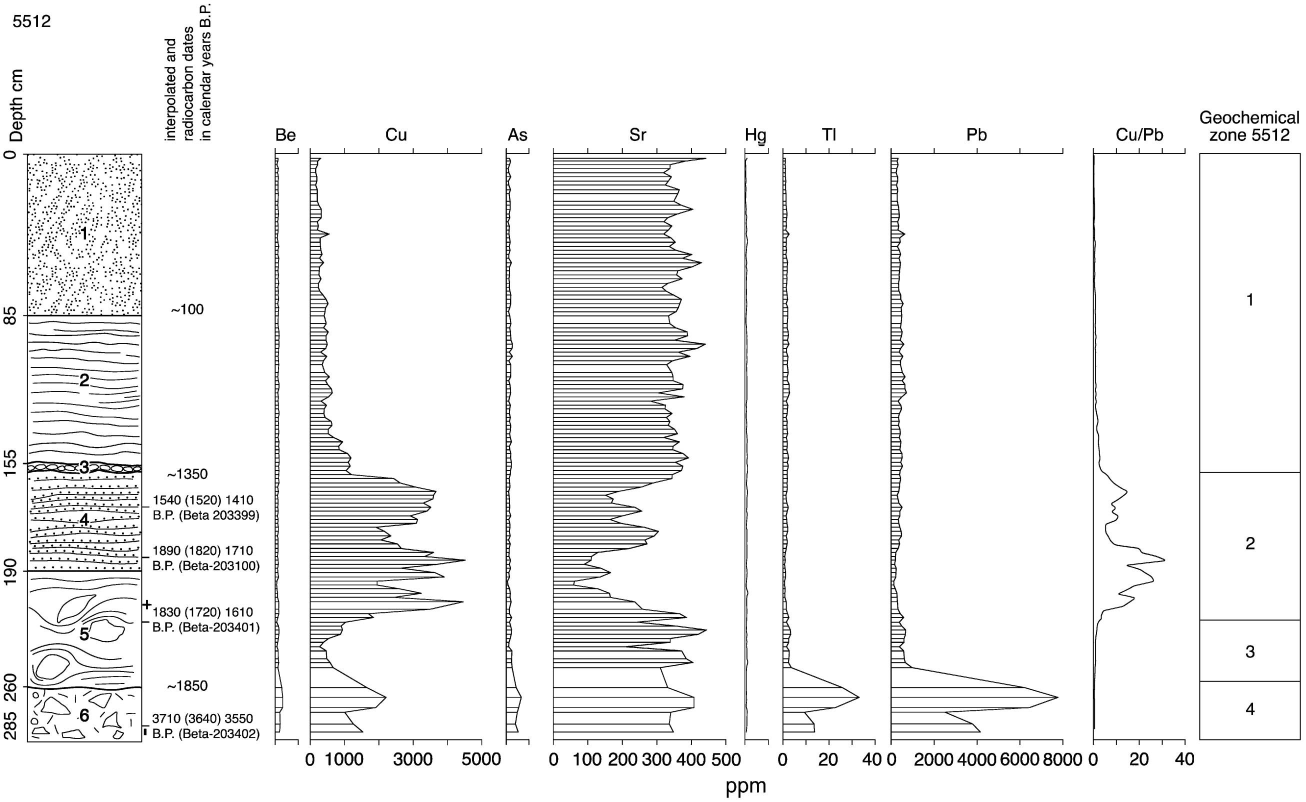

Fig. 6. Summary of the geochemical record obtained by ICPMS analyses from sediments beneath or impounded by a barrage immediately north of the Khirbet Faynan at site 5512; total concentrations in ppm based upon HNO_3 extraction. The lithological units and geochemical zones are defined in Table 4; radiocarbon dates are calibrated years BP.

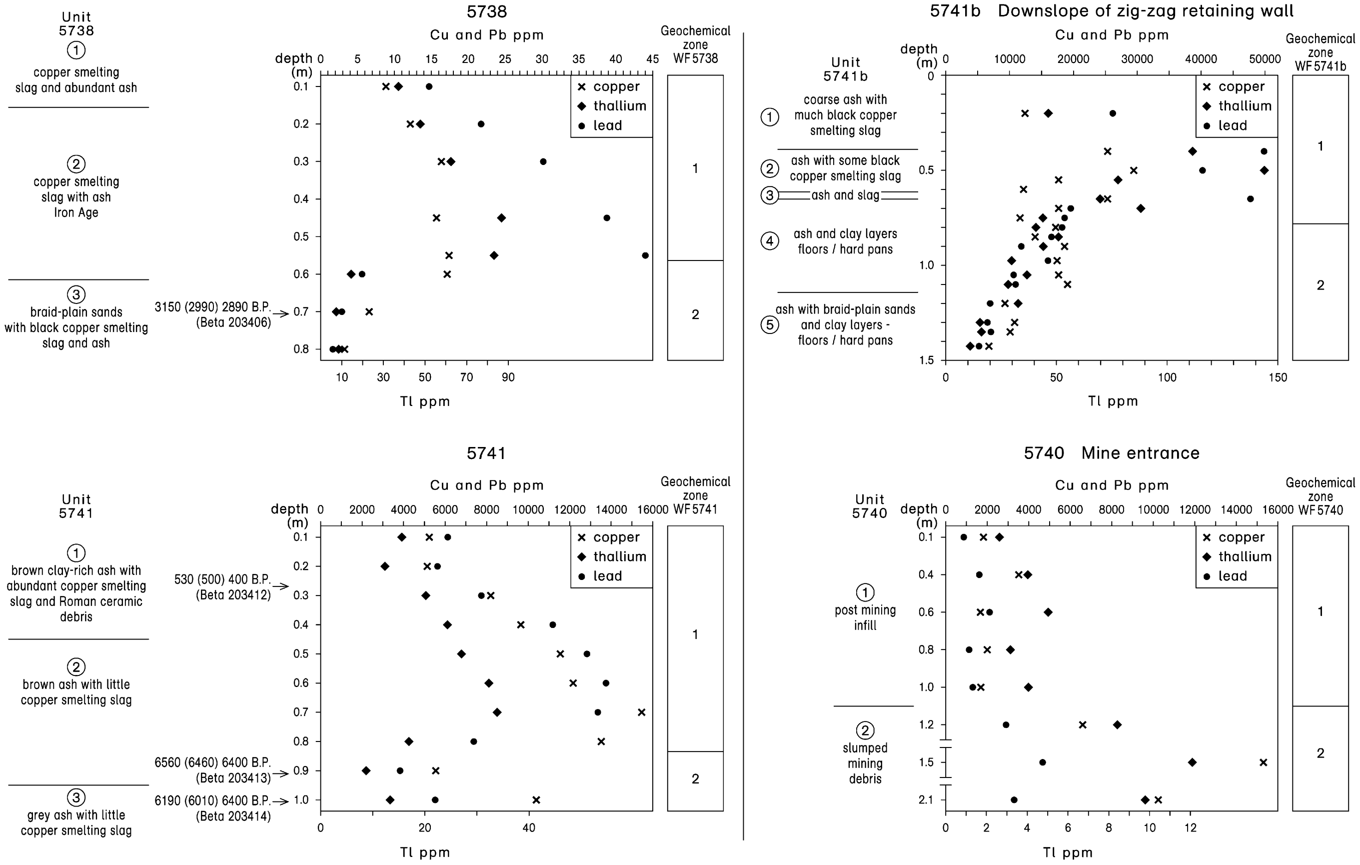

Fig. 7. Summary of the total concentrations of HNO_3 extracted copper lead and thallium in ppm and defined geochemical zones from three excavations in copper smelting slag (5738, 5741, 5741), near the Khirbet Faynan and the mine entrance 5740.

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 proportion 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

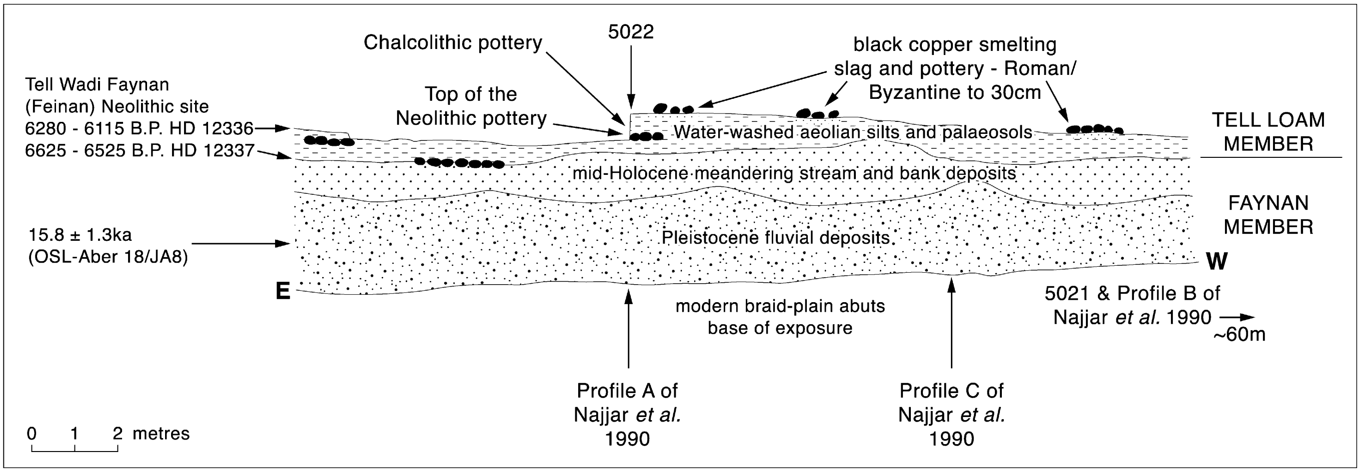

Fig. 8. The stratigraphic relationships between surface Roman-Byzantine smelting slag; wind blown-water washed materials and soils of the Tell Loam Member and the neolithic site exposed at site 5022, overlying a late Neolithic settlement of Tell Wadi Faynan, and associated meandering fluvial deposits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

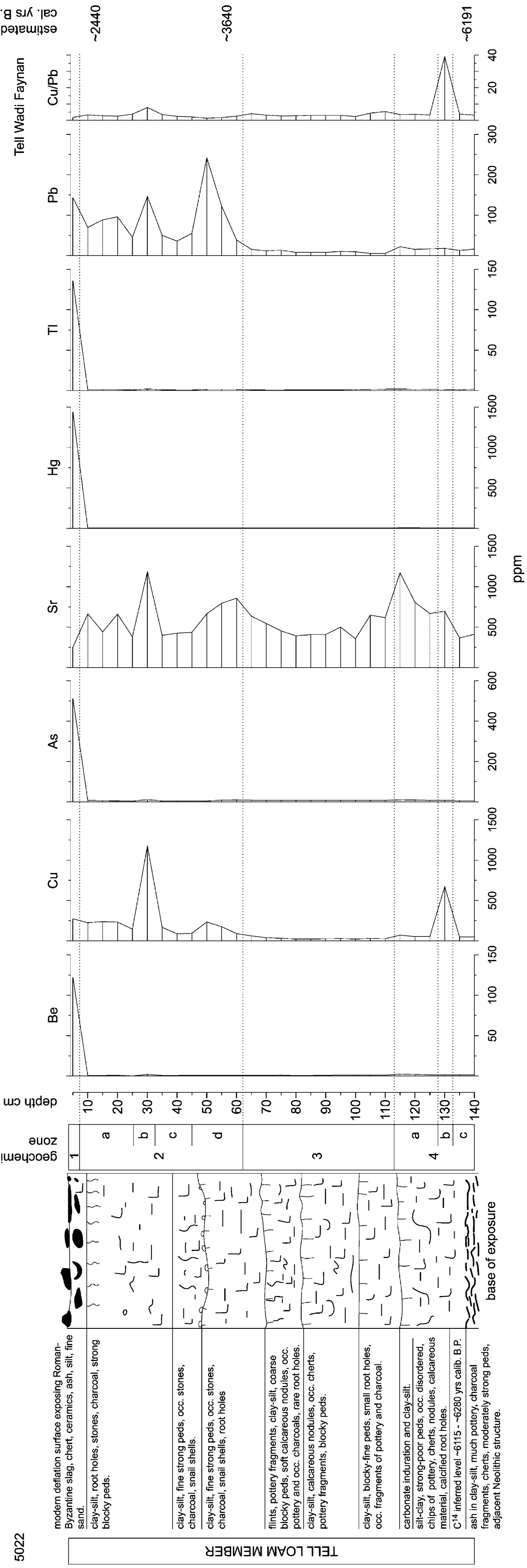

Fig. 9. Summary of the lithostratigraphy and geochemistry of the Tell Loam Member at exposure 5022 through aeolian-water washed silts and thin palaeosols: in ppm.

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 found by radiocarbon dating of charcoal in a 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 more active and complex geomorphic activity 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 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 proportional 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 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 approximately 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 4 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 might 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 Fig. 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 (Fig. 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 Fig. 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 ~16,000 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 palaeosols from Late Neolithic to Roman-Byzantine

Site WF5022 is a 1.4 m deep section exposed in a low mound, the Tell Wadi Faynan, eroded by the modern wadi approximately 1.2 km west of Khirbet Faynan (Figs. 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) (Figs. 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 (Fig. 10).

The base of the sequence, perhaps 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 and 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 80 cm 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 and 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

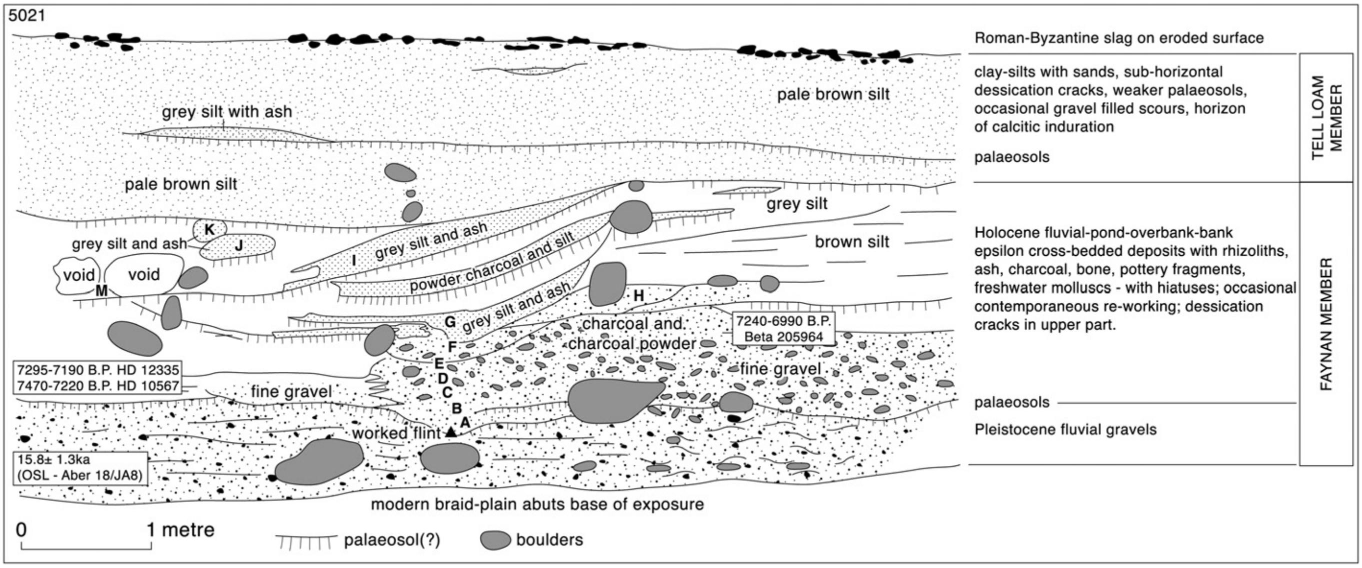

Fig. 10. Summary of the lithological properties, stratigraphic relationships and sample locations in the fluvial-stream margin sequence at site 5021 near Tell Wadi Faynan; radiocarbon dates are calibrated years BP. Sample G of ash, as well as charcoal; the latter gave a radiocarbon date of 7240 ± 40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

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 (Fig. 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 accidental smelting activity, to the deliberate processing of ores and metals at an industrial scale.

8. 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 (Figs. 9–12) are exposed in a cliff-like wadi edge of the modern braid-plain of the Faynan are set out in Fig. 12. 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

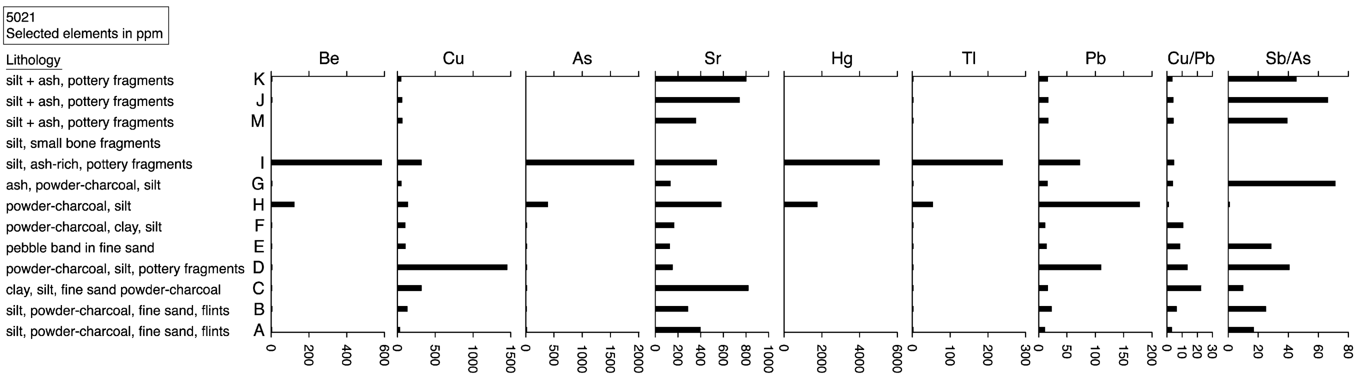

Fig. 11. Summary of sample properties and geochemical properties of Neolithic age from an exposure of aeolian, water washed silts and temporary soils (5021), Tell Wadi Faynan; in ppm. Sample G of ash, as well as charcoal; the latter gave a radiocarbon date of 7240 ± 40 BP; cal BP 2σ 7240–6990 cal (Beta-205964).

(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 [71]. The presence of Neolithic pottery fragments, layers of ash, charcoal, radiocarbon dates and the worked flints, inter-bedded with this quiet-water fluvial deposit, 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 depends upon radiocarbon dates in adjacent wadis; as well as the lithostratigraphic relationships (Figs. 3, 10–12 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 (Fig. 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; there 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 (Fig. 12). In particular, the metal assemblage in sample L suggests the heating of copper-lead ores which also contained substantial quantities of 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 Figs. 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 probably 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 the 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 this 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 [96]) 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 [1] and it has yet to be demonstrated for the Neolithic. Craddock [30] 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.

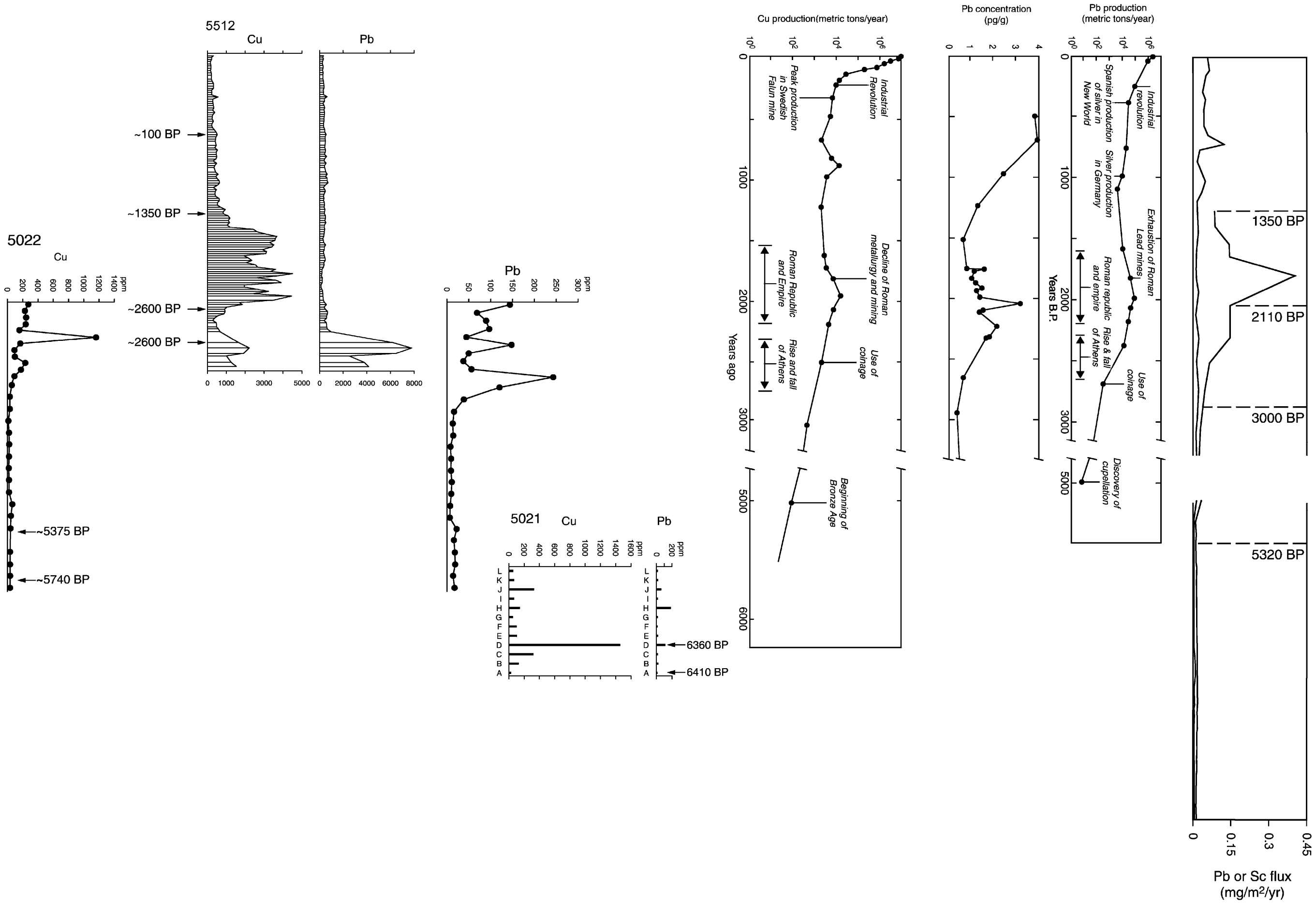

Fig. 12. Comparison between the pollution record for copper and lead over 8000 years for the vicinity of the Khirbet Faynan, in ppm HNO_3 extraction ICPMS; with estimates of copper and lead production in the Classical World; and the calculated rate of metal fluxes to high level mires in Europe and the Greenland icesheet (after [15,19,20,119–121]).

9. Discussion

9.1. 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 (Figs. 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 re-cycling 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 suggest 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.

9.2. Industrial activity and pollution over time

The broader overview of metal deposition illustrated in Fig. 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 contaminated ash and charcoal 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.

9.3. 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 Fig. 12. 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 (Fig. 12). Before about ~3000 years 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 Fig. 12 also indicates that is 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 of 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 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 and 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 the 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".

10. 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 ~3–4000 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 industrial 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.

Acknowledgements

The natural and archaeological features were mapped and interpreted in the field from the original high-quality black and white, vertical air photographs taken by the Royal Air Force in the 1930s which pre-date any modern (pre-1996) changes in the landscape. These photographs were provided by Professor D.L. Kennedy of the University of Western Australia from *The Archaeological Air Photograph Archive for Jordan* (catalogue references 41.018-20; 42.059-61; 42.114-6; 43.017-22). This project has been part of the wider study of the past landscapes of the Wadi Faynan directed by Graeme Barker, David Gilbertson and David Mattingly. We acknowledge the support and advice given over many years by our many colleagues in the field, especially Dr Sue McLaren and Professor Brian Pyatt; the British Institute in Amman for Archaeology and History, and the invaluable help and guidance from the Jordanian Department of Antiquities, especially Dr M Najjar who directed the earlier excavation of Tell Wadi Faynan and the good natured assistance of the Bedouin of Wadi Faynan. We are grateful for the helpful comments made by the reviewers of the earlier draft of this paper. The funding for this field and laboratory project stemmed primarily from the University of Wales Aberystwyth, the British Academy; The Council for British Research in the Levant; the Arts and Humanities Research Board. We are indebted for the analytical work in the Aberystwyth ICP-MS facility and are grateful for the laboratory determinations by Alan Condron, Sharon Taylor, Harry Toland and Charlotte Sykes reported here.

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