The first anthropologically-contaminated river? Repeated heavy-metal enrichment of fluvial sediments associated with Late Neolithic human activity in southern Jordan

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Supplementary Information

1. PREVIOUS WORK

Initial ICP-MS work on <2 mm sieved residues (Grattan et al. 2007) on the Faynan Member - Upper Component found high concentrations of copper and lead in samples D, E, F, G and H. Samples were checked visually for minute ore fragments (Figures S1, 2a,b). (The local copper ores are highly visible: typically bright green-blue, or black-purple.) The sediments enriched in heavy metals comprised mostly overbank silts at the edge of a near-perennial meandering streamchannel, with reed-swamp, and other riparian/wetland habitats, bordered by dry steppic vegetation (Hunt et al. 2004; 2007a). Worked chert and unrolled fragments of pottery and bones demonstrated the immediate proximity of people. Nearby Holocene alluvium lacked charcoal and anthropogenic debris and generally had much lower concentrations of copper, lead and other heavy metals, often an order of magnitude lower. The single exception to this association was analysis I, with high copper but located in in-channel sediments lacking anthropogenic debris (Grattan et al. 2007). Grattan et al. (2007) concluded that the the high values of copper and lead were likely the result of contamination emitted by fires that had incinerated copper-rich materials.

In the occupation deposits at Tell Wadi Faynan, small clasts of green copper ores were reported *in-situ* by the excavator (Najjar et al. 1990) and were seen during the 2010 field season by JPG. The presence of lime-plasters on the site indicates the capability of its inhabitants to make and control very hot fires.

2. DATING OF THE ARCHAEOLOGICAL SEQUENCES AT TELL WADI FAYNAN

The discovery of Tell Wadi Faynan (TWF) by local Bedouin in 1986, led to the investigation of the site by the Deutsches Bergbau Museum (DBM) expedition to Faynan. The discovery of the site came about due to the active flooding in the Wadi Faynan which had exposed the site in the section of the wadi. It was called a tell (Arabic tall) because of the very obvious site shape in the stratigraphy of the wadi section, despite the fact that the site was not at all visible at ground level. It was clear to all who saw the site, that the occupational levels as exposed in the wadi were of a classic tell shape, with the edges of the site sloping down to what had presumably been the edge of the site. A short description of the site is outlined in Hauptmann's “Site Catalogue” (Hauptmann 2007, 109-110), but the most complete report on the site and excavations by Najjar (Najjar et al. 1990) outlines the first season of work in 1988 and provides a full description of the site, its stratigraphy and finds. Sadly, renewed excavation of the site in 1990 coincided with severe winter flooding which washed away parts of the section, making some areas of the site too precarious for continued work (RBA personal observation).

Najjar’s excavations of the site provide conclusive evidence of the long term occupation of the site, beginning in the Pottery Neolithic A, or Yarmoukian (Stratum 3, see the radiocarbon list), which continued uninterrupted into the Chalcolithic (Stratum 2), a period in which the vast amount of evidence for developed copper metallurgy in the Levant has come. In 1988 the wadi section at TWF measured 7.40 metres, with the upper 2.4 metres containing cultural material. In the centre of the site, Najjar’s Profile A indicated that the earliest occupation levels were founded on wadi gravels, with a pit cut into the uppermost gravel level, in which a large ceramic vessel was embedded, and below this vessel was found a piece of copper ore and a layer of ash of about 80 cm width (Najjar 1990, Figure 3). At Profile B the wadi section was 7.45 m, and the cultural sequences were 2.55 m in depth. In profile B, as in Profile A, the occupation levels were directly on top of wadi gravels, but at Profile B there were numerous examples of fire pits and ash deposits in association with grinding querns, occupation surfaces and a stone-built wall (Najjar 1990, Figure 4).

It is clear from the earliest excavation reports that the cultural deposits at TWF contained numerous pieces of copper ore (n=53), including ores from both the Umm Ishrin Sandstone and the Dolomite-Limestone-Shale levels from wadis further east and north of Wadi Faynan. Although Najjar found no direct evidence of copper metallurgy in the Neolithic levels, it is clear from the extensive list of copper ore finds from the excavation (Najjar 1990, Table 2) that the occupants of the site, both in the Neolithic and later Chalcolithic times were actively collecting the ores, which could not have been deposited in these levels any other way.

The question of the relationship of the enhanced levels of copper at TWF found by the authors and the origin of copper metallurgy are not clear. What can be said is that these populations in the region, as early as the Pottery Neolithic were actively utilizing these coloured ores. It has been demonstrated elsewhere, that from at least the Aceramic Neolithic (PPNB), that these copper silicates minerals of bright green and blue colour had been used for beads (Bar-Yosef Mayer and Porat 2008), pigments (such as on the Nahal Hemar mask, Bar Yosef et al. 1988), and other artifacts (Simmons and Najjar 2006, 89) and that eventually they were heated and used to extract metallic copper (Golden 2014; Shugar 2000; Shugar and Gohm 2006). Where in this long prehistoric sequence at Faynan this occurs, has yet to be determined, but the evidence of enhanced copper pollution levels in close association with cultural deposits cannot be ruled out as evidence of this working of copper (in whatever form), which has clearly lead to one of the earliest examples of enhanced “pollution” levels.

3. COPPER ORES IN THE CATCHMENT

The bedrock copper ores lie in Cambrian formations: the Burj Dolomite-Shale and parts of the Umm ‘Ishrin Sandstone (Barjous 1992; Hauptmann 2000, 2007; Raab’a 1994). Archeometallurgical research is described in Barker et al. (2007a); Bender (1965, 1974); el-Rishi et al. (2007); French National Public Institute (1974); Gilbertson et al. (2007); Grattan et al. (2007, 2013, 2014); Hauptmann (1989, 2000, 2007); Hauptmann, Weisgerber (1987, 1992); Hauptmann et al. (1992); Hunt, el-Rishi (2010); Kind (1965); Overstreet et al. (1982); Van den Boom and Ibrahim (1965). There is minimal information on reworked ores in the Quaternary sequences (McLaren et al. 2004; Raab’a 1994), but heavy metals in the modern braidplain were examined in relation to bedrock sources (Saffarini, Lahawani 1992) and to the exposure of ancient copper smelting slags around the Khirbat Faynan (Grattan et al. 2007, 2013). In both, they are variable in concentration and composition, and patchy in distribution.

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Table S1: Possible causes of the heavy-metal anomaly in Holocene deposits at WF5021.

Explanations	**Supporting detail**	**Comments**
Contamination by pyrometallurgy	Late Neolithic hot fires and activity with copper ore was proved by the finds of green-blue copper ore within Late Neolithic occupation deposits at nearby Tell Wadi Faynan (Najjar et al. 1990) and still exposed in 2009, 2010, and 2013.	Too early for pyrometallurgy according to consensus of regional evidence.
Direct or indirect contamination by cold metallurgy (defined by Radivojević et al. 2010):	Gathering, crushing, use, heating and exchange of copper ores is established at Late Neolithic Tell Wadi Faynan, and in the region during the Pre-Pottery Neolithic (PPN) and Pottery Neolithic (PN); from the 10 - 11th millennia cal. BP to approximately the start of ~8th millennium cal. BP (Adams 1991; Barker et al. 2007a, b; Bar-Yosef and Porat 2008; Craddock 1995; Hauptmann 2000; 2007; Thornton et al. 2010; Weisgerber 2006).	Lack of visible metal ore particulates in the sediments. Deposition of microscopic ore fragments or metal vapours s is possible. There is no native copper in the region.
Contamination from pyrotechnological activities	Making of lime plasters (known from Neolithic Tell Wadi Faynan: Najjar et al. 1990), or burning of heavy-metal-rich biomass.	Sources of limestone are well-removed from outcrops of metal ores. Heavy-metal-rich biomass growing on ore outcrops would have been several km distant. The Zipf principle suggests that local biomass would have been burnt in preference.
Secondary depositional contamination	Changes in runoff and fluvial sedimentation caused by human impact on hillslope processes.	Contemporary sites (WF5015, WF5051) located close to ore outcrops are not contaminated. It is implausible that WF5021, located several km from ore outcrops, would be contaminated while these were not.
Geomorphological processes	The outcome of geomorphic processes and wildfires; which might include the reworking of ore-rich materials from Pleistocene and bedrock sources in an area which contains significant bodies of copper-lead ores.	Contemporary sites (WF5015, WF5051) located close to ore outcrops are not contaminated. It is implausible that WF5021, located several km from ore outcrops, would be contaminated while these were not.
Contamination from heavy-metal rich infiltration	Contamination infiltrated along desiccation cracks and fissures from the overlying Tell Loam Member, which contains smelting slags and metallurgical debris of Bronze Age to Byzantine age.	Desiccation cracks and fissures were avoided during sampling. On examination the few available cracks and fissures showed no sign of the downwash of materials (which would be evident as colours on fissure faces different from those on freshly-cleaned sediment surfaces, redox markings and/or illuvial clay skins)
Groundwater movement	Groundwater leaching, circulation and redeposition of metals from overlying deposits, preferential adsorption of metals on organic matter including charcoal.	The sediments seem to have been dry since very shortly after deposition, when wadi incision lifted them above regional and local watertables. There seems to be no trace of redox in the sections (ie no manganese or iron-oxide structures, discoloured joints etc. which might suggest active groundwater circulation). There are no iron pans. Charcoal and organic matter are cation sorbs, but this does not negate the fact that most high Cu figures are from slightly-organic clayey-silts without visible charcoal. Distribution of sample points with high concentrations of copper in the fluvial overbank deposits is not consistent with material moving down in that it is highest near the middle of the unit. Similarly, in the overlying Tell Loams, apart from occasional small root-$CaCO_3$ nodules, there is no trace of

		any groundwater activity or translocation of material. All depositional evidence points to deposition of this unit in hyperarid conditions (Hunt et al. 2007; Grattan et al. 2007). These loams contain highly stratified copper and lead contamination concentrated well above the base of the unit in layers containing Late Bronze Age and Classical period artefacts (Grattan et al. 2007).
Grain-size or organic matter variations causing apparent anomalies	Ability of clay minerals and organic matter to scavenge heavy metals leading to systematic 'over-representation' of heavy metals in fine-grained and organic-rich materials.	Sampling strategy was aimed at sand-sized materials. Analysis of both organic and inorganic materials.
Chance – unrepresentative-ness	An anomaly produced by a small number of atypical samples which by co-incidence may have particular anthropogenic or geomorphic associations.	The present study was formulated to address this issue. The larger number of sample points and the stability of the patterns encountered makes this less likely
Unsuspected errors or limitations	Problems with ICPMS analysis (Grattan et al. 2007 gives the ICPMS working protocols used previously); poor observation of this difficult exposure.	The present study was formulated to address this issue. The larger number of sample points and the stability of the patterns encountered using an alternative technology makes this less likely

Table S2: Details of radiocarbon dates mentioned in the text

Site	Context	Material	Dates BP	Cal. BP (2 σ)	Cal. Dates BC (1 σ)	Reference
Wadi Dana WF5015	Neolithic pit fill overlying fluvial deposits with PPA biozone pollen	Charcoal	7240±90	8310 (3.0%) 8240 8220 (90.5%) 7920 7900 (2.0%) 7860	6160-5980	Hunt et al. 2004 (Beta-111121)
Tell Wadi Faynan	Wadi section (depth 2.5 m)	Charcoal	6370±42	7430 (92.4%) 7240 7210 (3.0%) 7170	5330-5265	Hauptmann 2000, 65 (HD 12335)
Tell Wadi Faynan	Wadi section (depth 4 m)	Charcoal	6408±114	7600 (95.4%) 7000	5430-5250	Hauptmann 2000, 65 (HD 10576)
Tell Wadi Faynan 5021	Wadi section 5021G at top of PPA biozone	Charcoal	6200±40	7250 (95.4%) 6990	5220 5060	Grattan et al. 2007 Beta-205964
Tell Wadi Faynan	Sq B, locus 6 (depth 0.8 m)	Charcoal	6132±50	7170 (95.4%) 6890	5195-4940	Hauptmann 2000, 65 (HD 13775)
Tell Wadi Faynan	Sq Fa, (depth 0.2 m)	Charcoal	6105±68	7170 (95.4%) 6790	5195-4930	Hauptmann 2000, 65 (HD 12338)
Tell Wadi Faynan	Sq A, locus 23 (depth 1.4 m)	Charcoal	5740±35	6640 (95.4%) 6440	4675-4575	Hauptmann 2000, 65 (HD 12337)

Table S3: Raw geochemical data

Analysis no.	Location	Original sample no.	Author	Analysis type	Copper ppm	Lead ppm
Modern braidplain transect						
B1	0 m from TWF	S36		pXRF	21	10
B2	23m from TWF	S37		pXRF	23	12
B3	43m from TWF	S38		pXRF	46	4
B4	63m from TWF	S39		pXRF	25	21
B5	89m from TWF	S40		pXRF	65	42
B6	104m from TWF	S41		pXRF	50	17
B7	143m from TWF	S42		pXRF	1	2
B8	150m from TWF	S43		pXRF	43	20
B9	170m from TWF	S44		pXRF	79	29
B10	218m from TWF	S45		pXRF	12	29
B11	270m from TWF	S46		pXRF	23	15

B12	268m from TWF	S47		pXRF	43	7
B13	288m from TWF	S48		pXRF	29	12
B14	376m from TWF	S49		pXRF	77	26
B15	1m from TWF	twf31		pXRF	46	25
B16	25m from TWF	twf32		pXRF	147	0
B17	33m from TWF	twf33		pXRF	51	4
Tell Loam Member at 5021 lowest 1m						
T1	Overlies Faynan Mbr U	twf20		pXRF	104	10
T2	Overlies Faynan Mbr U	twf21		pXRF	107	24
T3	Overlies Faynan Mbr U	twf22		pXRF	138	44
T4	Overlies Faynan Mbr U	10i		pXRF	164	25
T5	Overlies Faynan Mbr U	10h		pXRF	238	21
Tell Loam Member at 5022						
T6	Overlies TWF & Faynan Mbr U	twfUnit2 75cm		pXRF	131	11
T7	Overlies TWF & Faynan Mbr U	twfUnit2 80cm		pXRF	70	8
T8	Overlies TWF & Faynan Mbr U	twfUnit2 85cm		pXRF	57	21
T9	Overlies TWF & Faynan Mbr U	twfUnit2 90cm		pXRF	46	5
T10	Overlies TWF & Faynan Mbr U	twfunit2 95cm		pXRF	52	10
T11	Overlies TWF & Faynan Mbr U	twfUnit2 100cm		pXRF	71	12
T12	Overlies TWF & Faynan Mbr U	twfUnit2 105cm		pXRF	88	11
T13	Overlies TWF & Faynan Mbr U	twfUnit2 110cm		pXRF	99	24
T14	Overlies TWF & Faynan Mbr U	twfUnit3 115cm		pXRF	43	20
T15	Overlies TWF & Faynan Mbr U	twfUnit3 120cm		pXRF	47	18
T16	Overlies TWF & Faynan Mbr U	twfUnit3 125cm		pXRF	124	19
T17	Overlies TWF & Faynan Mbr U	twfUnit3 130cm		pXRF	180	35
T18	Overlies TWF & Faynan Mbr U	twfUnit3 135cm		pXRF	118	29
T19	Overlies TWF & Faynan Mbr U	twfUnit3 140cm		pXRF	120	35
T20	Overlies TWF & Faynan Mbr U	twfUnit3 145cm		pXRF	48	19
T21	Overlies TWF & Faynan Mbr U	twfUnit3 base		pXRF	332	51
5	Overlies Faynan Mbr U	5 cm	Grattan et al. 2007	ICPMS	271	144
10	Overlies Faynan Mbr U	10 cm	Grattan et al. 2007	ICPMS	227	68
15	Overlies Faynan Mbr U	15 cm	Grattan et al. 2007	ICPMS	241	90
20	Overlies Faynan Mbr U	20 cm	Grattan et al.	ICPMS	238	97

			2007			
25	Overlies Faynan Mbr U	25 cm	Grattan et al. 2007	ICPMS	152	44
30	Overlies Faynan Mbr U	30 cm	Grattan et al. 2007	ICPMS	1166	147
35	Overlies Faynan Mbr U	35 cm	Grattan et al. 2007	ICPMS	170	50
40	Overlies Faynan Mbr U	40 cm	Grattan et al. 2007	ICPMS	90	36
45	Overlies Faynan Mbr U	45 cm	Grattan et al. 2007	ICPMS	96	56
50	Overlies Faynan Mbr U	50 cm	Grattan et al. 2007	ICPMS	236	243
55	Overlies Faynan Mbr U	55 cm	Grattan et al. 2007	ICPMS	180	120
60	Overlies Faynan Mbr U	60 cm	Grattan et al. 2007	ICPMS	91	37
65	Overlies Faynan Mbr U	65 cm	Grattan et al. 2007	ICPMS	62	16
70	Overlies Faynan Mbr U	70 cm	Grattan et al. 2007	ICPMS	38	12
75	Overlies Faynan Mbr U	75 cm	Grattan et al. 2007	ICPMS	34	13
80	Overlies Faynan Mbr U	80 cm	Grattan et al. 2007	ICPMS	18	7
85	Overlies Faynan Mbr U	85 cm	Grattan et al. 2007	ICPMS	22	8
90	Overlies Faynan Mbr U	90 cm	Grattan et al. 2007	ICPMS	23	8
95	Overlies Faynan Mbr U	95 cm	Grattan et al. 2007	ICPMS	27	10
100	Overlies Faynan Mbr U	100 cm	Grattan et al. 2007	ICPMS	18	9
105	Overlies Faynan Mbr U	105 cm	Grattan et al. 2007	ICPMS	25	6
110	Overlies Faynan Mbr U	110 cm	Grattan et al. 2007	ICPMS	25	5
115	Overlies Faynan Mbr U	115 cm	Grattan et al. 2007	ICPMS	67	21
120	Overlies Faynan Mbr U	120 cm	Grattan et al. 2007	ICPMS	51	15
125	Overlies Faynan Mbr U	125 cm	Grattan et al. 2007	ICPMS	50	16
130	Overlies Faynan Mbr U	130 cm	Grattan et al. 2007	ICPMS	667	17
135	Overlies Faynan Mbr U	135 cm	Grattan et al. 2007	ICPMS	44	12
140	Overlies Faynan Mbr U	140 cm	Grattan et al. 2007	ICPMS	44	15
Anthropogenic-Fluvial Lithofacies 5021						
A1		twf11		pXRF	101	30
A2		twf10c ii		pXRF	226	30
A3		twf10d		pXRF	157	14
A4		twf10e		pXRF	94	12

A5		twf10f		pXRF	446	17
A6		twf10g		pXRF	277	48
A7		twf10j		pXRF	140	17
A8		twf10k		pXRF	193	17
A9		twf10L		pXRF	209	25
A10		5021D	Grattan et al. 2007	ICPMS	1459	109
A10		5021E	Grattan et al. 2007	ICPMS	105	12
A11		5021F	Grattan et al. 2007	ICPMS	104	10
A12		5021G	Grattan et al. 2007	ICPMS	49	14
A13		5021H	Grattan et al. 2007	ICPMS	138	178
Fluvial-Clastic Lithofacies 5021						
F1		twf10		pXRF	59	16
F2		twf12		pXRF	135	96
F3		twf13		pXRF	124	29
F4		twf14		pXRF	119	21
F5		twf15		pXRF	120	6
F6		twf16		pXRF	143	56
F7		twf17		pXRF	89	16
F8		twf18		pXRF	87	65
F9		twf19		pXRF	125	54
F10		twf10a		pXRF	78	19
F11		twf10b		pXRF	62	9
F12		5021A	Grattan et al. 2007	ICPMS	26	9
F13		5021B	Grattan et al. 2007	ICPMS	129	22
F14		5021C	Grattan et al. 2007	ICPMS	318	14
F15		5021I	Grattan et al. 2007	ICPMS	62	16
F16		5021J	Grattan et al. 2007	ICPMS	60	16
F17		5021K	Grattan et al. 2007	ICPMS	49	14
Surfaces of pottery fragments 5021						
C1		NP1		pXRF	258	23
C2		NP2		pXRF	224	13
C3		NP3		pXRF	296	30
Holocene river Wadi Dana 5015						
D1		0cm		ICPMS	27	18
D2		2.5cm		ICPMS	21	9
D3		7.5cm		ICPMS	24	11
D4		15cm		ICPMS	26	12
D5		25cm		ICPMS	26	12
D6		31.5cm		ICPMS	22	11
D7		34.5cm		ICPMS	32	19

Holocene river Wadi Ghuwayr 5510						
G1		5510gh1		pXRF	52	18
G2		5510gh2		pXRF	38	12
G3		5510gh3		pXRF	49	6
G4		5510gh4		pXRF	49	10
G5		5510gh5		pXRF	46	7
Faynan Member [Lower Component] Pleistocene fluvial 5021						
P1		twf8		pXRF	134	0
P2		twf9		pXRF	86	5
P3		twf23		pXRF	34	0
P4		twf24		pXRF	128	15
P5		twf25		pXRF	34	0
P6		twf26		pXRF	73	9
P7		twf27		pXRF	92	30
P8		twf28		pXRF	52	16
P9		twf28		pXRF	28	10
P10		twf30		pXRF	8	9
P11		twf10a		pXRF	67	16
P12		twf10p		pXRF	82	9
Lisan Marls Barq'a Gully below Bronze Age						
L1		BG827		pXRF	44	23
L2		BG828		pXRF	44	2
L3		BG829		pXRF	58	3
L4		BG830		pXRF	55	0
L5		BG831		pXRF	36	17
L6		BG832		pXRF	304	26
L7		BG833		pXRF	90	10
L8		BG834		pXRF	61	16
L9		BG835		pXRF	426	18
L10		BG836		pXRF	55	3
L11		BG837		pXRF	138	28
L12		BG838		pXRF	31	27
L13		BG839		pXRF	13	4
L14		BG840		pXRF	45	6
L15		BG841		pXRF	46	0
L16		BG842		pXRF	39	10
L17		BG843		pXRF	37	30
L18		BG844		pXRF	75	21
L19		BG845		pXRF	47	8
L20		BG846		pXRF	28	5
L21		BG847		pXRF	21	25
L22		BG848		pXRF	72	29
L23		BG849		pXRF	36	4
L24		BG850		pXRF	56	26
L25		BG851		pXRF	105	16
L26		BG852		pXRF	77	18
L27		BG853		pXRF	61	25
L28		BG854		pXRF	67	20

Table S4: Mann-Whitney test for difference between Methodologies

			ICPMS			ICPMS	
			Tell Loams			Faynan M Upper	
		Mean	156.1786			227.1818	
			Mann-Whitney significance	Difference significant		Mann-Whitney significance	Difference significant
pXRF	Tell Loams	113.1905	0.293	no			
pXRF	Faynan M Upper	149.2				0.197	no

Table S5: Mann-Whitney test statistics for difference between Anthropogenic-Fluvial Lithofacies and other units

Mann-Whitney test for difference between Anthropogenic-Fluvial Lithofacies (mean= 174.64) and			
	Mean	Mann-Whitney significance	Difference significant
Lisan Fm	87.000	0.000	yes
Faynan Member Lr	94.130	0.000	yes
Ghuweir E Holocene	67.000	0.001	yes
Dana E Holocene	23.000	0.000	yes
Faynan U FluvClastic	131.380	0.012	yes
Tell Loams	121.540	0.013	yes
Modern braidplain	59.150	0.000	yes

Figure S1: Summary of the Late Quaternary stratigraphy at WF5021 in the Wadi Faynan before the major flood in May 2014. The exposure is located beneath and immediately west of Tell Wadi Faynan (WF5022), and adjacent to the braidplain of the Wadi Faynan (after el Rishi et al. 2007; Grattan et al. 2007; Hunt et al. 2004, 2007b; McLaren et al. 2004). The exposure includes the Late Pleistocene Faynan Member – Lower Component (OSL dated to 15.8±1.3ka: Aber18/JA8), overlain by the early Holocene Faynan Member - Upper Component which is overlain by mid to late Holocene Tell Loams. The Faynan Member - Upper Component is divisible into the interbedded Fluvial-clastic Lithofacies and the Anthropogenic-fluvial lithofacies. Charcoal at G is cal. BP 2σ 7245-6994 (Beta-205964; Hunt et al. 2007b; McLaren et al. 2004). The upper boundary of the Faynan Member - Upper Component is marked by a palaeosol that was not disturbed by pits, desiccation cracks, or bioturbation. This section is a composite constructed from drawings made in 1995-1999, reconciled on site with the extant lithostratigraphy and sample locations during July 2009, January 2010 and May 2013. Points A to K mark the samples analysed by ICP-MS studies reported by Grattan et al. (2007). V marks large voids. pXRF analyses in 2009 are designated 1 to 30; those in 2010 as 10a to 10n. NP1 to NP3 are surfaces of separate and stratigraphically distinct, and partially exposed fragments of Late Neolithic pottery examined by pXRF in 2010. Modern surface sediments on the braidplain of the Wadi Faynan are designated 31w to 33w, sampled at 15m, 25m and 35m distant from the exposure. Analyses S36 to S49 are pXRF measurements at the surface and at 15 cm depth, made at regular intervals northwards across the full ~400m width of this modern braidplain (Table 2).

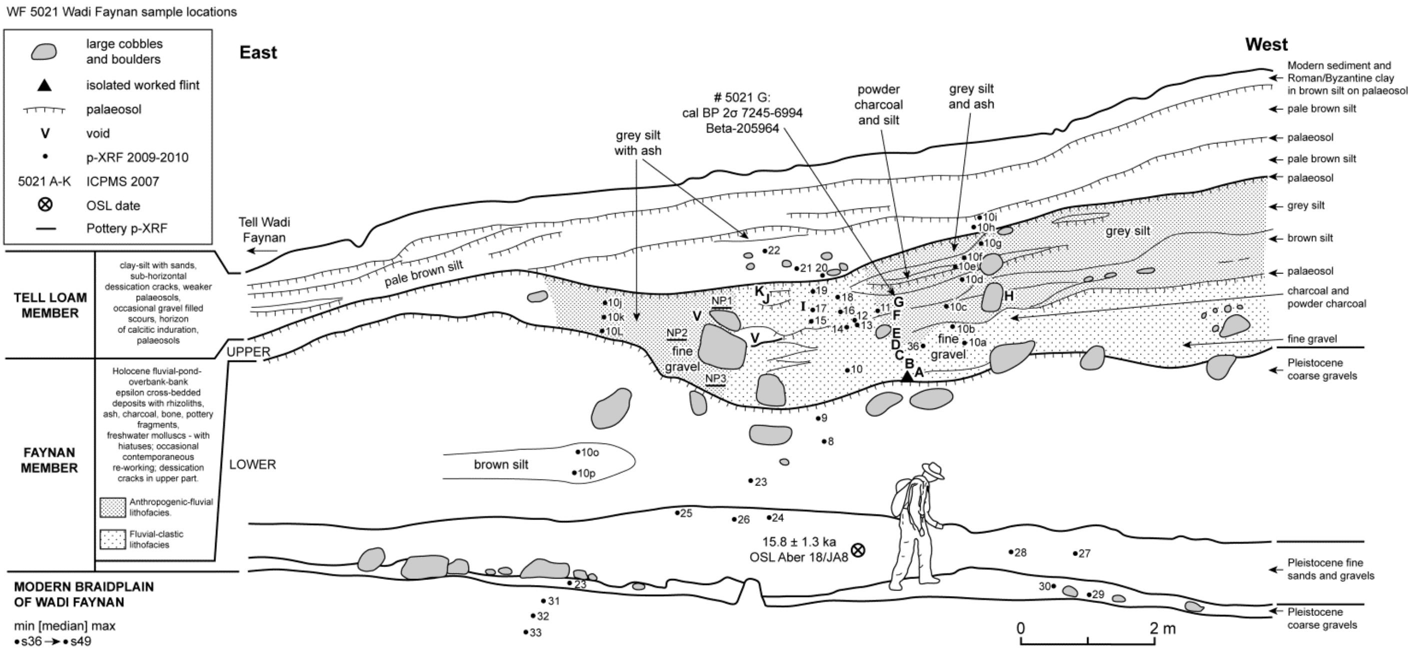

Figure S2: Exposure WF 5021 in August 2009 of the eroding cliffed edge of the modern braidplain of the Wadi Faynan upon which is a rucksack is at the base of the image. The lithostratigraphy is described in Figure S1. The hand-held PXRF device is close to and about to be used to take a measurement at point 21 on Figure 2. This exposure was largely destroyed during violent floods in May 2014. The elevation of the braidplain here has risen and fallen by ~1m since 1996 (D. Gilbertson).

Figure S3. Fluvial in-channel and overbank, and colluvial deposits of the Faynan Member - Upper Component at site WF5015 in the gorge of the Wadi Dana. Sample depths, codes, and measured concentrations in ppm of copper and lead by ICP-MS in Unit 5 of Hunt et al. (2004, 2007a,b,c). Redrawn with minor modifications after el-Rishi et al. (2007); Hunt et al. (2004, 2007a).

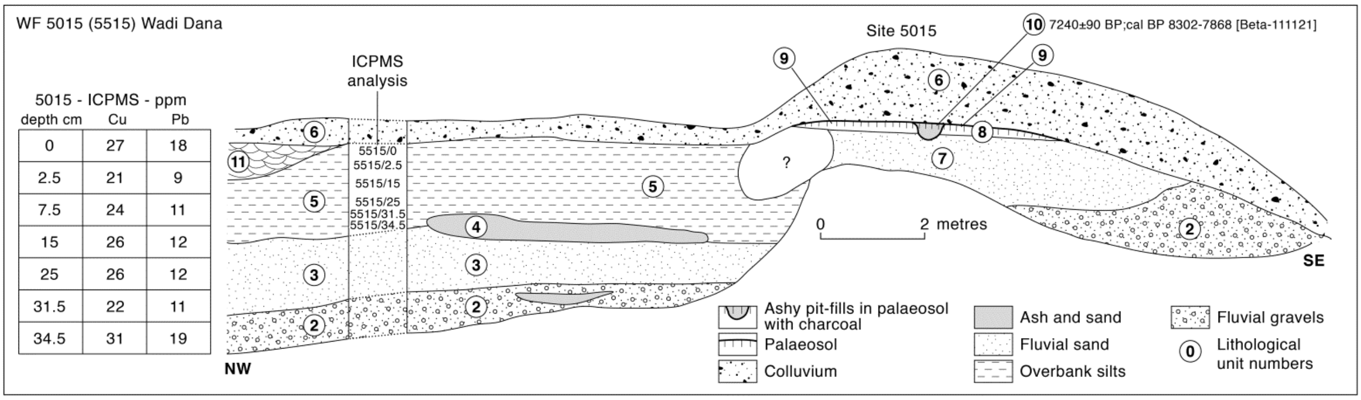

Figure S4. Exposure of Faynan Member - Upper Component at site WF5510 in the gorge of the Wadi Ghuwayr. Lithological unit 2 comprises fossiliferous marls and clays deformed by differential loading and was largely removed by erosion by a flood in May 2014. The concentrations in ppm of copper and lead listed in the table are for the profile marked by gh1 to gh5 through unit 2 and measured by pXRF in 2010. Redrawn with minor modifications (after el-Rishi et al. 2007; Hunt et al. 2004, 2007a).

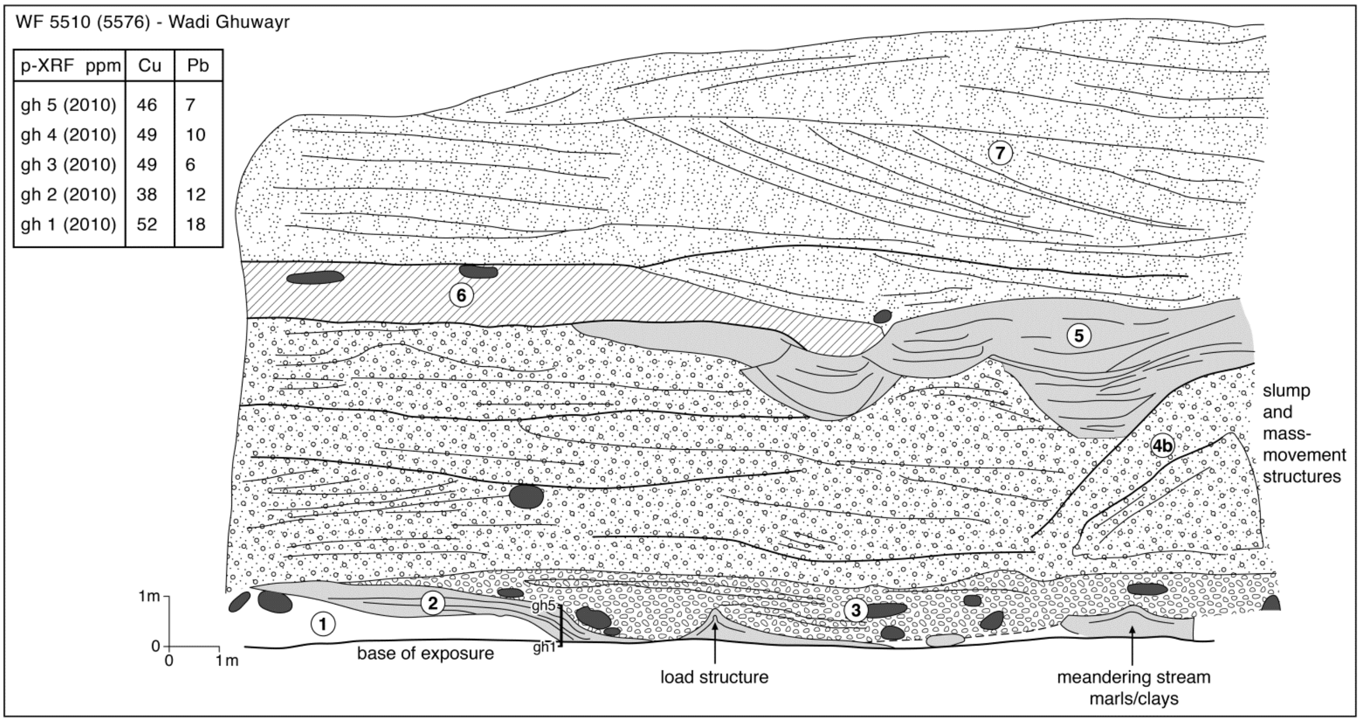

Figure S5: Exposure of Faynan Member - Upper Component visible at the base of the section to the left of the figure at site WF5510 in the gorge of the Wadi Ghuwayr, photographed in 2014 (D. Gilbertson).

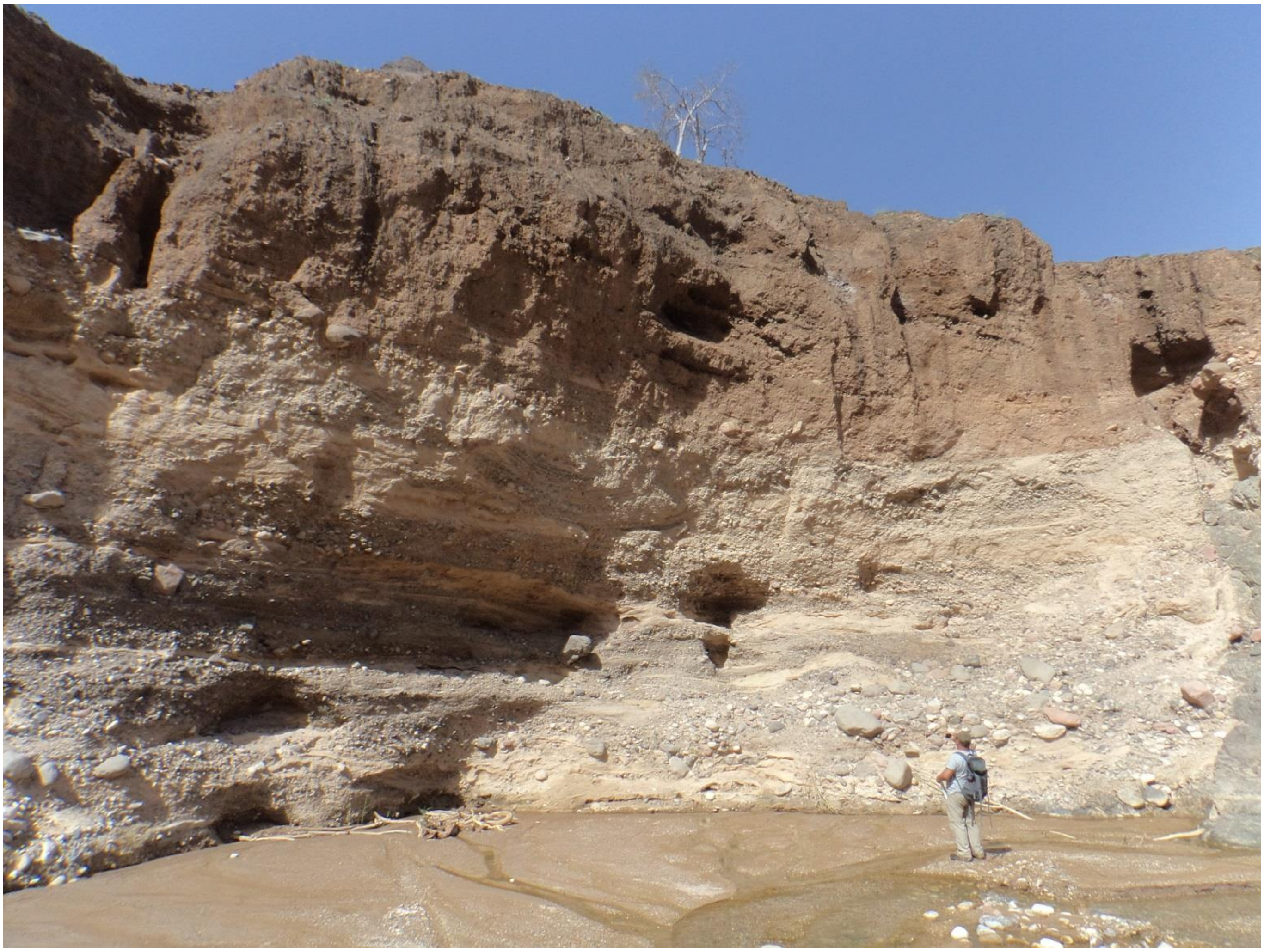

Fig. S6: Camp fire at the expedition camp showing brightly-coloured flames after the addition of fragments of copper ore (R. Adams).