“Red Ochre” Use by Early Neandertals

Wil Roebroeks a,1
Mark J. Sier a,b,c
Trine Kellberg Nielsen a
Dimitri De Loecker a
Josep M. Parés b
Charles E.S. Arps d
Herman J. Múcher e

a Faculty of Archaeology, Leiden University, P.O. Box 9515, 2300 RA Leiden, The Netherlands, b Centro Nacional de Investigación sobre la Evolución Humana (CENIEH), Paseo Sierra de Atapuerca s/n, 09002 Burgos, Spain, c Paleomagnetic Laboratory ‘Fort Hoofddijk’, Department of Earth Sciences, Faculty of Geosciences, Utrecht University, Budapestlaan 17, 3584 CD Utrecht, The Netherlands d NCB Naturalis, P.O. Box 9517, 2300 RA Leiden, The Netherlands e (retired from the University of Amsterdam) Prinses Beatrixsingel 21, 6301 VK Valkenburg, The Netherlands

1 To whom correspondence may be addressed. E-mail: w.roebroeks@arch.leidenuniv.nl

Author contributions: W.R. and T.K.N designed and carried out research; D.D.L took care of the site documents and sampling, M.J.S., J.M.P., C.E.S.A. and H.J.M. performed research and analyzed data; W.R., T.K.N. and M.J.S. wrote the paper.

Keywords: human evolution / paleolithic archeology / red ochre/ iron oxides
Abstract:
The use of manganese and iron-oxides by late Neandertals is well-documented in Europe, especially for the period 60-40,000 years ago. Such finds have often been interpreted as pigments even though their exact function is largely unknown. Here we report on significantly older iron-oxide finds which constitute the earliest documented use of red ochre by Neandertals. It concerns small concentrates of red material, retrieved during excavations at Maastricht-Belvédère (The Netherlands). These excavations exposed a series of well-preserved flint artifact (and occasionally: bone) scatters, formed in a river valley setting during a late Middle Pleistocene full-interglacial period. Samples of the reddish material were submitted to various forms of analyses for a study of their physical properties. All applied methods show that the red material was hematite. This is a non-local material, which was imported to the site, possibly over dozens of kilometers. The identification of the Maastricht-Belvédère finds as hematite pushes the use of red ochre by (early) Neandertals significantly back in time, to minimally 200,000 to 250,000 years ago; i.e. to the same time range as the early ochre use in the African record, created by Middle Pleistocene ancestors of modern humans.
Recent debates on Neandertal material culture have highlighted the fact that Middle Paleolithic sites occasionally contain pieces of manganese- and iron-oxides, interpreted as pigments, possibly for personal decoration (1, 2). Some have taken these findings an inferential step further and speculated on the “symbolic implications of body painting” and ochre use for our views on Neandertals (2). From the Upper Paleolithic record red ochre is indeed well-known for its use in cave paintings and in ritual burial contexts. More “mundane” or “domestic” use of red ochres (derived from hematite, Fe₂O₃) is known from the ethnographic record of modern hunter-gatherers, for instance as (internal and external) medication, food preservative, tanning for hides, and as insect repellent (3-9). In archeological studies ochre powder has been shown to be an ingredient in the manufacture of compound adhesives (10). Hence, the use of iron oxides for “symbolic” purposes should be seen as a hypothesis which needs to be tested, rather than simply assumed.

For Europe, a recent review (11) mentions more than 40 Middle Paleolithic sites with possible pigments from the Marine Isotope Stage (MIS) 6-3 time range. These concern mostly manganese oxide finds and almost all sites date to the very end of the Middle Paleolithic, between 60-40 ka (1 ka = 1000 years before present) (11). Some of these late sites yielded considerable quantities of these materials. From Pech de l’Azé I (France) for instance more than 450 small pieces of manganese dioxide are known, with a total weight of c. 750 grams; more than 250 of these finds showed traces of utilization (12). Hence, solid evidence for use of manganese- and iron-oxides by Late Pleistocene Neandertals is recorded from at least 60 ka onward. Claims for an earlier, Middle Pleistocene, use of “red ochre” in Middle Pleistocene archeological sites in Europe do exist, such as for Terra Amata (France), Becov (Czech Republic) (13) and Ambrona (Spain), but all of these have been contested for a variety of reasons, which include identification and dating issues (14).

Here we report on “red material” of considerably higher antiquity than the Late Pleistocene, minimally 200-250 ka old. This material was recorded during the 1980-s excavations in the Maastricht-Belvédère loess- and gravel pit (The Netherlands; 50°52'09.40"N, 5°40'27.33"E). Fieldwork there focussed on an interdisciplinary study of an early Middle Paleolithic site complex of flint (and occasionally: flint and bone) scatters, preserved in primary archeological context in fine grained sediments of the Middle Pleistocene Maas river. From
these sediments eight archeological sites were excavated as well as a series of test pits, which in total created an excavated surface of 1577 m² (15-17). The Middle Pleistocene river deposits yielded a full interglacial vertebrate fauna with 26 species (18) and a mollusc fauna containing more than 70 land and freshwater species (19). Terrace and loess stratigraphy as well as mammal and molluscan biostratigraphical evidence indicate an age prior to the next-to-last glacial phase, i.e. prior to MIS 6 (20). Radiometric techniques included TL dating of heated flint artifacts which yielded an age of 250±20 ka (21) and ESR-dating of shells (220±40 ka), all converging in an MIS 7 age for the Maastricht-Belvédère interglacial (20). Amino Acid Racemisation dating of *Corbicula* shells from the interglacial deposits as well as biostratigraphically important elements of the mollusc fauna itself however suggest an earlier, i.e. MIS 9, age for the Belvédère interglacial and its associated archeology (22).

In the course of the archeological excavations one of the sites, Site C (excavated between 1981-1983), yielded 15 small concentrates of red material, ranging in maximum size from only 0.2 to 0.9 cm and 0.1 to 0.3 cm thick, with sharp boundaries to the sedimentary matrix (Fig 1-2). The contrast in colour between the bright red concentrates and the yellowish-brown (Munsell Soil Color 2.5Y5/3) to greyish-olive (5Y5/3) sediment was striking, enabling the recovery of a number of these small and friable pieces at this site, excavated over an area of 264 m² (Fig 3). Although the red material has been interpreted as hematite (15, 23), these finds never played a role in the history of ochre use, even though Maastricht-Belvédère became one of the flagship sites of Middle Paleolithic archaeology, reviewed extensively in various textbooks (24). Improvements in identifying methods and the importance given to ochre use in current debates in paleoanthropology justified a new, systematic study of the Maastricht-Belvédère material.

**Results**

Binocular microscope investigation of three of the the largest red concentrates at Site C (23) revealed that a red staining agent surrounded the larger quartz grains of the sedimentary matrix, as a very thin coating (Fig 1; see also SI Text Fig 8). The boundary between the red concentrates and the surrounding matrix is sharp, macroscopically (Figs 1-2) as well as in thin sections (SI Text Fig 9). Across this boundary a sharp decrease in grain size was observed from the matrix towards the red concentrates, with the matrix richer in silt sized mineral particles than the red concentrates. Occasionally the fine red material had clotted together with the clay and silt particles of the sediment. Importantly, individual reddish crystal grains,
e.g. hematite, were not visible. Earlier attempts at identifying this red material (23) had focused on the largest pieces recovered from Site C, with X-ray Diffraction (XRD) analysis suggesting the presence of hematite (23). For this study only the small concentrates were available, preserved in their sedimentary matrix (SI Fig 4). Two pieces came from Site C (Cz 11-1, max. dimension 3 mm; WW10-8, max. dimension 5 mm), one piece was retrieved during the excavations at Site F (20/23-1, max. dimension 3 mm), one of the other sites in the finegrained fluvial deposits (see below). Study of these samples by XRD, Environmental Scanning Electron Microscopy (ESEM), Energy-dispersive X-ray spectroscopy (EDX) and several rock magnetic studies clearly indicate the presence of hematite in the samples, alongside a strong quartz component of the sediment matrix of the red material (see SI Text). These studies confirm the results of the previous XRD analyses of three samples from Site C, mentioned above (23).

The Sedimentary Context of the Finds

With the red material identified as hematite, how did it enter the sediments? Our null-hypothesis is that the hematite concentrates were part of the sedimentary environment of the archeological sites, i.e. part of a natural background scatter of such finds. The Site C matrix consisted of well-sorted, fine to very fine silty sands, with a silt and clay content of at least 15% by weight. Micromorphological studies of the Site C sediments indicated low-energy deposition, burying the archaeological remains very calmly and gradually (25). Laterally, the Site C sediments developed into loams. Study of the rich mollusc fauna retrieved from such loamy deposits adjacent to the excavated area shows the presence of stagnant water, and suggests that the Site C area was protected from the main channel of the river Maas: a densely vegetated lacustrine niche in a predominantly fluviatile environment, during the climatic optimum of the interglacial (19). Site C contained flint knapping scatters of mainly Levallois debitage, from which large amounts of artifacts could be refitted (21.5% of the 3D recorded flint artifacts at Site C, and 70.4% by total weight), including a Levallois recurrent reduction sequence (15, 26). The excavations entailed a 3D-recording of all identifiable finds, including small (<0.5 cm) flint chips and pieces of bone and >5800 charcoal fragments. The size distribution of the flint material is dominated by small (<2 cm) flint artefacts, which comprise c. 75% of the total 3D-recorded material. Spatial studies showed an absence of winnowing patterns, again underlining the primary context character of the assemblage.
The Site C “red ochre” finds constituted a very strong “search profile” during the subsequent 1984-1989 excavations and geological fieldwork in the quarry, during which another 1200 m² were excavated, distributed over various locales (Sites F-N). Excavators were explicitly instructed to look for “red material”. At all sites but one (see below), excavation and documentation procedures were comparable to the Site C ones, but despite of this no ochre finds were recorded during subsequent excavations at Site G (excavated over 50 m²) or Site K (370 m²). At Site N 765 m² were meticulously excavated and recorded with the explicit goal to study the “background scatter” of flint artefacts, bones and other finds present in the interglacial river deposits; the low density distribution yielded 450 flint artefacts, partly conjoinable, but again no find of red ochre concentrates (16). Part of the excavations of the large and rich Site K (10,192 flint artifacts) proceeded faster than at the other sites (17), so there the absence of comparable finds cannot be interpreted as a “real” absence. We did discover three more pieces of red material during the 1984 excavations at Site F, c. 300 m SE of Site C. One of the pieces from this site was analyzed in this study and proved to contain hematite too. Site F was excavated over an area of 42 m². Its excavation yielded 1215 flint artefacts, of which we refitted 12,8% by numbers and 67% by weight. Apart from these 3 pieces from Site F, no other hematite finds were made during the extensive archeological excavations in the quarry. Furthermore, during the multidisciplinary studies of the exposures in the quarry over the 1981-1989 period thousands of metres of sections of the interglacial Maas river deposits were cleaned and examined for geological studies and for the potential occurrence of various types of finds. Hundreds of meters of such sections were sampled and drawn in detail, while dozens of thin sections were made and studied (27), with no traces of hematite recorded during any of these activities. On the basis of these observations we reject the null hypothesis that the hematite fragments were part of the sedimentary environment of the Maastricht-Belvédère archeological assemblages. Data independently supporting this interpretation comes from the structure of the concentrates themselves, mentioned above: the hematite staining surrounded the quartz particles of the sedimentary matrix, with the hematite concentrate itself more fine-grained than the matrix. This implies that the red material entered the sediments after their formation.

Discussion
The combined evidence of on-site observations, studies of the non-archeological deposits as well as the character of the concentrates themselves concur in our inference that the presence of these small fragments of non-local hematite was related to hominin activities at Sites C and
Site F. With the null-hypothesis rejected and given the data presented above, we need to explain the presence of this hematite material surrounding the sediment particles in the Site C matrix. The best explanation, we hypothesize, is that the fine hematite material was originally concentrated in a liquid solution, and that blobs of this ochre-rich substance became embedded in the sediments during usage of the liquid, spilled on the soil surface. In order to test this interpretation we performed an experiment to observe the impact of drops of a “hematite rich liquid” on the Site C sediment (see SI Text). Despite the limitations of the experiment, the similarity of the experimentally produced concentrates with the archeological ones, both on a macro- as well as a microscopic level, are remarkable (see SI Text) and lend support to our interpretation of how the material entered the sediment.

What may have been the context of use of the hematite-rich liquid substance at Maastricht-Belvédère? The Site F assemblage contained only one formal tool, whereas its unmodified flakes did not yield any microscopic signs of use. The absence of several larger flakes in conjoining groups indicates that it was a site where large blanks were produced for use elsewhere in the landscape. 15 heated flints and some charcoal particles suggest the former presence of a fire.

As for Site C, two-third of the Site C hematite particles clustered in the NW part of the site, partially around the concentration of small (< 0.5 cm) charcoal particles. Five hematite pieces were found in the southern part of the site, amongst the flint artefacts recovered there, many of which were heated (15), possibly reflecting a former fire place (28).

Site C yielded some (use-) retouched tools, including three scrapers (15). Faunal remains were badly preserved (15). Some butchering activities may have taken place there according to use wear analysis of the flint artifacts, whereas the presence of scrapers suggests that some hide working may have occurred (29), an activity in which hematite can play a role (30, 31). No traces of hematite were recorded on these artefacts though. In sum, the spatial and the functional context of the Site C hematite finds offer no entries into the former use of the hematite.

What we are able to state is that ochre use is now documented in an early Middle Paleolithic context, minimally dating to MIS 7 – even though the application of the ochre is unknown. Our interpretation of the Maastricht-Belvédère material predicts that more traces of hematite use will turn up in future excavations of Middle Paleolithic sites in comparable archeological as
well as geological settings, i.e. with a sedimentary matrix which guarantees the survival and visibility of such small pieces and a research context which allows careful excavation.

The nearest hematite sources known are at c. 40 km from the site, in the Ardennes and Eifel area (32) (Fig 5). The Ardennes sources, in the Liège-Dinant-Namur area, are in the catchment of the Maas river, but despite of this hematite has thus far not been recorded in stone counts of the river gravels (15) in the Maastricht region. One cannot rule out though that very small quantities of hematite were collected from river bars in the late Middle Pleistocene. Hematite is - albeit very sporadically- present in Paleozoic rocks in the Ardennes-Rhine Massif, especially in quartz veins with hematite crystals. Stones and boulders of these rocks are present in Maas deposits, transported from their source areas on ice rafts. Hypothetically, Neandertals may have stumbled upon such hematite in a large quartz boulder, though this would in all probability have been a very rare encounter.

For a possible connection to the sources in the northern part of the Eifel (32) it is interesting to note that two early Middle Paleolithic (MIS 6) sites in the Eifel area yielded small numbers of artifacts made out of flint from Cretaceous deposits from the Maastricht area (Schweinskopf site: n=5, Wannen site: n=8 (33). These flints were discarded in the Eifel at distances of c. 100 km from their geological sources near Maastricht (33, 34). The hematite material may have traveled in the opposite direction, from the Eifel to the Maastricht area, but it would take better (i.e. significantly larger) samples to test such a hypothesis by establishing a solid provenance for the Belvédère material. The occasional transport of stone artifacts over distances comparable to the ones discussed here is well documented for the European Middle Paleolithic (34-37) and a hypothetical import of hematite over such a distance does fit with our data on Neandertal movements through Pleistocene landscapes.

In Africa pieces of red ochre became a common phenomenon in Middle Stone Age (MSA) rock shelter sites from about 160 ka onward (8, 9, 38), but there are a few earlier occurrences, in the same time range as the ones reported here for Maastricht-Belvédère (39). Site GnJh-15 in the Kapthurin Formation, Baringo, Kenya (40) yielded hematite fragments too friable to preserve traces of grinding, ranging from pulverized granular material weighing less than 3 g to large chunks more than 250 g. The early MSA site Twin Rivers, thought to date to 200-300 ka, likewise produced pieces of ochre (41). The Maastricht-Belvédère material dates to the same time range as these early cases of ochre use in the African MSA, produced by ancestors.
of modern humans. The Maastricht-Belvédère material shows that for those workers who see iron oxide use as an archaeological indication of “symbolic behaviour”, there is now also early data for red ochre manipulation by members of the Neandertal lineage to take into account (42). But again, in our view there is no reason to assume that the mere presence of iron oxide at an archeological site, whether Neandertal or modern human, implies symbolic behaviour.

Conclusion
The small hematite concentrates reported here constitute the earliest unambiguous case of use (and possibly transport over several dozens of kms) of this material in the Neandertal archeological record. The finds probably entered the matrix of the site as drops from an ochre-rich liquid substance during unknown application activities. The finds provide only a very limited window into manipulation of red ochre by early Neandertals, certainly when compared to the unique and detailed information recently published for Blombos Cave, South Africa (38). But importantly, with the Maastricht-Belvédère material identified as hematite, the use of red ochre by early Neandertals has been pushed back in time to at least 200 – 250 ka (MIS 7), i.e. to the same time range as documented for the African record, produced by Middle Pleistocene ancestors of modern humans. It is to be expected that future studies will yield comparable finds from early Middle Paleolithic settings, either during fieldwork or as the result of the re-analysis of old finds. The evidence currently available suggests a sporadic use of red ochre by early Neandertals, minimally from MIS 7 onward.

Methods
In the first study of the largest pieces from Site C, one of us (C.E.S.A.) produced a concentrate of the red crusty material of find Dz23-16 by carefully grounding it to release the reddish material. The sample was then placed in a concave glass dish filled with alcohol and the finest fraction of the reddish powder could be separated from the bulk sample by panning. After further grinding to obtain a suitable grain-size this concentrate was used for XRD, which showed that the red stain was caused by the presence of hematite (23). For the present study 2 small samples of reddish material from the Maastricht-Belvédère excavations at Site C (Cz 11-1 and WW10-8) and a fragment from Site F (20/23-1) (see SI Fig 4) were submitted to various types of analyses in order to study their physical properties: Environmental Scanning Electron Microscopy (ESEM), Energy-dispersive X-ray
spectroscopy (EDX), X-ray Diffraction (XRD) and several types of rock magnetic analyses. All measurements were taken at the laboratories of the CENIEH, Burgos, Spain (See SI text for more details), where also thin sections of sample Bz 13-6 (Site C) were produced.

Acknowledgements

We acknowledge the large number of colleagues involved with the Maastricht-Belvédère project, in particular the late Paul Hennekens. We are grateful to various colleagues including two anonymous PNAS-referees, Alain Turq (Les Eyzies), Paola Villa (Boulder), Mark Dekkers (Utrecht), Ian Watts (Athens), Francesco d'Errico (Bordeaux) and Alexander Verpoorte and Adam Jagich (Leiden) for comments on earlier versions of the paper and/or advice on aspects of the study. Herman Mûcher (Valkenburg), was especially helpful with the experiment and the thin section study. We thank Jan Paupit and Joanne Porck (Leiden) for their work on the figures, Annelou van Gijn and Erik Mulder (Leiden) for help with the microscopic work, Silvia Gonzalez Sierra (Burgos) for her work with the ESEM and EDX, Ana Isabel Alvaro Gallo (Burgos) for collecting the data with the XRD and Carlos Saiz Domínguez for work on the thin sections. The Royal Netherlands Academy of Arts and Sciences supported the study with an Assistant Grant for T.K.N.


Fig 1 The two largest hematite concentrates from Site C at Maastricht-Belvédère. On the left concretion (Dz23-16) against the background of the fine-grained fluvial deposit from which it was recovered. Dessication of the matrix caused breakage of the concentrate. On the right is concretion no. Bv-894, c. 2 mm thick, shown during excavation in Fig 2.

Fig 2 Site C, square Fz14, find Bv-894 (see Fig 1) during excavation, January 28th 1982. Next to the hematite fragment is a flint flake. Scale bar is 5 cm.
Fig 3 Distribution map of the Site C finds (see lower left hand corner for captions), with superimposed the position of the 15 hematite dots. Measuring grid in metres. The south-eastern part of the site, indicated in gray, was disturbed by karst. Source: Fig 27 from (15).
Fig 4 Maastricht-Belvédère Site C: refitted group of 162 flint flakes, comprising the remains of a flat disc-like core, recovered from the southern part of the site. This composition is one of the many refits demonstrating the primary context character of the Site C assemblage (15). Scale bar is 5 cm (conjoining study by Paul Hennekens and Wil Roebroeks).
Fig 5 Map indicating the location of Maastricht (-Belvédère) and in red the Ardennes iron ore sources of the Liège-Dinant-Namur area (1) as well as the Eifel sources (2-3) (modified after (32). The arrow indicates the transfer of flint artefacts from the Maastricht Cretaceous chalk area to the East Eifel sites of Wannen and Schweinskopf, as discussed in the text. Terrain >500 m above sea level is indicated in dark green.
**Physical Properties Study**

**Methods**

In order to study the physical properties of the possible hematite fragments from the Maastricht-Belvédère excavations, three samples (Cz11-1, WW10-8, 20/23-1) were analyzed using several methods including Environmental Scanning Electron Microscope (ESEM), Energy-dispersive X-ray spectroscopy (EDX), X-ray Diffraction (XRD) and rockmagnetic measurements. Samples were measured and analyzed at the Geochronology Laboratory of the CENIEH, Burgos, Spain. Additionally, sample Bz13-6 (Site C), was submitted to thin section analysis, likewise at Burgos.

**Environmental Scanning Electron Microscope (ESEM) Energy-dispersive X-ray spectroscopy (EDX)**

Samples were photographed using a Fei Quanta 600 Environmental Scanning Electron Microscope system (ESEM) while micro-analyzes were performed with an Oxford, INCA 350, Energy-dispersive X-ray spectroscopy micro-analysis system (EDX). EDX is a non-destructive analytical technique used for elemental analysis or chemical characterization of samples, which measures the energy of the scattered X-rays when an electron beam impacts on a sample. The ESEM photographs were used to observe the samples and their relation with the surrounding matrix, if present. Observations were made both before and after samples were cut with a diamond hair cutter in order to get a smooth surface.

**X-Ray Diffraction (XRD)**

The mineralogical composition of the samples was analyzed by XRD. Patterns were collected with a Panalytical X’Pert PRO MPD diffractometer with CuKα radiation (\( \lambda = 1.5406 \text{ Å} \)) and a solid-state multichannel detector. Samples were analyzed as bulk powder sample with a reflection-transmission spinner stage and as solid-samples, by placing specimens on a multi-purpose sample stage. The following analysis parameters were used: 45kV, beam current of 40mA, scan range between 5 and 70° 2θ in continuous scan of 0.02° 2θ steps with 100-700
seconds per step. The diffraction patterns were compared with the International Centre for Diffraction Data (ICDD) database using X’Pert High Score Plus software.

Rockmagnetism
Vibrating Sample Magnetometer MicroMag Model 3900 (VSM, Princeton Measurements Corporation), was used to measure hysteresis loops, single moment versus temperature and acquisition curves of the Isothermal Remanent Magnetization (IRM). Measurements were taken at room temperature and single moment versus temperature measurements within the range of room temperature and to a maximum of 750° C. Data acquired from the IRM measurements were imported in a specially developed Excel worksheet (3) which can be downloaded at http://www.geo.uu.nl/~forth. The base function fitting of the IRM acquisition curves is based on Cumulative log Gaussian analysis developed by Robertson and France (2) and extended by Kruiver et al (3). This method makes it possible to differentiate magnetic mineral phases from the IRM data.

Results
Environmental Scanning Electron Microscope (ESEM) Energy-dispersive X-ray spectroscopy (EDX)
Two of three samples were retrieved from the sites situated in their sedimentary matrix. We observe that these two samples (Site C: Cz11-1 and Site F 20/23-1) have sharp boundaries with their matrix. For sample 20/23-1 we observed a separation from the matrix (see SI fig 1a) which is not present in sample Cz11-1 (see SI fig 2a). All EDX data shows the samples to have an increased iron content and for two samples with matrix an increased iron content in relation to their matrix (see SI fig 1b and c).

X-Ray Diffraction (XRD)
All XRD measurements on the three samples clearly identify several minerals including quartz and hematite (See SI fig 4). Sample 20/23-1 also contains clearly identifiable albite and goethite. Cz11-1 contains rutile and an additional iron oxide with low crystallinity which could not be identified. Sample WW10/8 contains, besides the hematite and quartz, a second form of quartz, muscovite and zircon. It is important to note that the quantification given (See SI fig 4) for the different minerals is meaningless, as the beam of the XRD is larger than the size of the samples, resulting in an underestimate of the amount of hematite. Our results confirm the previous analyses by Arps (4), who used a Guinier De Wolf Quadruple Focussing Camera (Enraf Nonius, Delft) for an XRD study of three of the hematite finds from Maastricht-Belvédère. His analysis indicated that the samples consisted of a mixture of quartz
(the strongest signal) and hematite, which was considered responsible for the red stains in the silty-fine sands deposits at Maastricht-Belvédère.

**Rockmagnetism**

Sample Cz11-1 was of insufficient size for rock magnetic measurements. Sample 20/23-1 and WW10-8 give similar results, with the latter (Site C: WW10-8) giving the clearest data, due to sample size. The hysteresis curves show (See SI fig 5) both paramagnetic and ferromagnetic contributions and saturation not is reached at 1.3 Tesla revealing the presence of a high coercivity magnetic phase.

The IRM acquisition curves (See SI fig 3) are in agreement with the hysteresis curves, in that a high coercivity component is the main magnetic component in these samples. IRM line fitting using the Kruiver et al. (3) method actually shows two magnetic components. Component 1 is an artifact (5, 6) and is not discussed further. The artifact is a result of the log Gaussian base function way of fitting which can only fit distributions that are symmetric in the log-field space. Left-skewed distributions (a consequence of time-dependent magnetic behaviour and/or magnetic interaction, must be fitted in the Kruiver et al. (3) package with an extra component with log B1/2 of around 1.0. The amount (SIRM value) of the skewed artifact component has to be added to the SIRM of the dominant coercivity component (with log B1/2 of 1.4 – 1.8) to give the true intensity of this dominant component. The component 2 and 3 with log B1/2 values of 2.45, DP 0.25 and log B1/2 values of 2.95, DP 0.29 do get physical interpretation.

The magnetic moment versus temperature data of sample WW10-8 shows (See SI fig 7) a slightly declining trend until 450 degrees C after which the magnet moment increases to return to pre 450 degrees C level around 650 degrees C.

**Discussion**

All data obtained by the different methods clearly indicate the presence of hematite in the three different samples. The EDX shows an increased percentage of iron oxide in the samples in relation with their matrix, while the XRD has positively identified hematite as a compound in the sample. Unfortunately the small sample size of the samples prevents the semi-quantification of the minerals components.

The rock-magnetic data, like the XRD data, reveals hematite in all studied samples. The hysteresis curve and IRM show a high coercivity component generally interpreted as hematite or goethite. Line fitting of the IRM data identifies two components with high log B1/2 values. Log B1/2 of 2.45 we interpret as hematite while the second component with a value of log
B1/2 of around 2.95 can be either hematite of different grain size or goethite. Goethite has a low Curie temperature of around 120° C. The single moment versus temperature curve does not show a drop in magnetic moment at this temperature suggesting that goethite is not present in sufficient amount in the sample. Due to thermo-chemical reactions (see (7)) in the sample starting at 450 degrees, most likely due to formation of magnetite preventing further identification of magnetic minerals after this temperature.

**Experimentally produced ‘hematite” concentrates in the Site C matrix**

Microscopic examination of the hematite concentrates by one of us (C.E.S.A.) had revealed that a red staining agent surrounded the quartz grains of the sedimentary matrix, as a very thin coating, and that occasionally the fine red material had clotted together with the clay and silt particles of the sediment (SI fig 8). Importantly, individual reddish crystal grains, e.g. hematite, were not visible. The boundary between the sedimentary matrix and the concentrates was sharp, also in thin section (SI fig 9), with the hematite concentrate itself more fine-grained than the matrix. This seems to imply that the red material entered the sediments after their formation. We hypothesized that the red material was originally concentrated in a liquid solution/paste, and that blobs of this substance became embedded in the sediments during usage of the liquefied substance, spilled on the soil surface. To test this interpretation we performed a small experiment to observe the impact of drops of a “hematite liquid” on the Site C sediment.

Air dried very fine sand from the matrix of the Site C finds (still available from sediment samples from excavation square Pz15) was packed in plastic Petri dishes (SI fig 10). The sediment was then submitted to droplets of a liquid consisting of a mix of hematite powder and rain water (1 gram of hematite for 20 ml of water), produced at 20 degrees Celsius. The hematite powder was obtained by grinding a hematite nodule on a quartzitic sandstone Maas river pebble.

The drops, 0.3 mm in diameter, were produced by means of a 2.5. ml plastic pipette, and launched manually from a height of 50 cm. The impact of the drops created well delimited hematite dots in the dry sediment, all smaller than 1 cm (SI fig 10A-B). Increasing the launch height to 150 cm led to somewhat larger dots, as did increasing the size of the drop from c. 0.3 to c. 0.5 cm (SI fig 10C-D). In both cases concentrates of around 1 cm in diameter were
produced. As the pictures show, the drops impact pressure created both a compression pit and a compensatory bulge around the perimeter of the pit – a phenomenon well known from experimental studies of the impact of rain drops on sediments (8-9). It is at the bottom of these “impact craters” that the red material was contained. The resulting hematite concentrates consisted of small semi-circular “discs” with a thickness of 1-2 mm, sometimes with a thin “rim” around their perimeter (the compensatory bulge mentioned above).

These experiments simulated the impact of drops under very dry conditions. To simulate more humid (and probably: more realistic) conditions in a separate run, 2ml of rain water was poured in the Petri dish, with the air dried sediment gently poured in subsequently. The sediment-filled Petri dish was then allowed to dry at room temperature for 24 hours, resulting in a more humid as well as more compacted sediment compared to the previous experiments. The resulting depressions containing the hematite liquid lacked the “rim” usually produced on the dry sediments (SI fig 11A-B). To simulate a larger “spill” of the liquid, 2 ml of the 1:20 solution was poured on the sediment from a test tube, c. 5 cm above a sediment filled disc. This created a well-delimited hematite concentration in the sediment, 2 cm in maximum dimension and with a thickness of c. 2 mm (SI fig 12A-B).

The experiments produced concentrates of hematite material which showed good similarities to the archeological finds (compare SI fig 8 to SI fig 11), both macroscopically and under the microscope. The results with the humid sediments provided the strongest similarities, as here the characteristic “rims” around the perimeter were less pronounced or absent. One difference between the experimental and the archeological ‘specimens’ consists in the fact that the archeological specimens showed denser concentrations of the red material than the experimental ones. This may be related to the fact that the hematite powder to liquid ratio was smaller in the past (i.e. more hematite powder used than in the experiment) and that in the small experiment only water was used as a liquid, leading to a more diluted solution in the matrix than when fatty substances would have been added/used as a “carrier” for the hematite powder. Apart from this obvious difference between the composition of the experimental liquid mixture and the (unknown) past one, there are more limitations to the value of this experiment. We have no idea about the humidity of the sediment matrix during creation of the archeological concentrates and only very indirect indications about the vegetation of the surface during hominin presence (certainly not as barren as in our experiment). After their creation the archeological specimens were covered by fine grained fluvial sediments and
subjected to various chemical and compaction processes during the 250,000 years they spend in their matrix. Despite of this, they were still visible enough to be recovered during archeological excavation.

SI References

Figure Captions
SI figure 1: a) ESEM image of sample 20/23-1 (Site F) on the left - on the right side of the image the sediment matrix b) mineralogical composition of the matrix. c) mineralogical composition of sample 20/23-1

SI figure 2: a) ESEM image of sample Cz11-1 (Site C). b) mineralogical composition of sample Cz11-1
Figure 3: IRM component analyses of sample WW10-8 (Site C)
SI figure 4: XRD analyses of sample Cz11-1, WW10-88 (both Site C) and 20-23/1 (Site F) from top to bottom.
SI figure 5: Hysteresis, uncorrected for slope, curve of sample WW10-8 (Site C)

SI figure 6: normalized IRM curve of WW10-8 (Site C)
SI figure 7: single moment versus temperature curve of sample WW10-8 (Site C)
SI figure 8 Close up of archeological hematite concentrate Dz20-56 (Site C), with its surrounding sandy matrix
SI figure 9 Thin section of part of sample Bz13-6 (Site C), showing a sharp boundary between the sedimentary matrix and the hematite concentrate, as well as a decrease in grain size from the sedimentary matrix (lower left) into the hematite concentrate. A: Plane Polarized Light, B: Cross Polarized Light.
SI figure 10 A-D Experimental hematite dots created on a “smoothed” (A) and an irregular (B) dry surface (50 cm height, drops 0.3 cm). The concentrates are within small craters produced during impact of the drops on the dry sediment. C: Concentrates created by 0.5 mm drops, launched from a height of 50 cm. D: Close up of the inner part of an experimentally produced hematite dot shown in SI Fig 9C (0.5 cm drop, 50 cm height).
SI figure 11A-B Hematite concentrates created on moist sediment by a 0,5 cm drop launched from a height of 50 cm (see text for explanation). B: close up of the experimental hematite concentrate on “moist” sediment, boundary with sand matrix – See A for position of the close up.
SI figure 12A-B A Hematite concentration experimentally produced by pouring 2 ml of the described hematite liquid from a test tube onto the dry sediment, from a height of 5 cm. B: Close up of the experimental hematite concentrate “spill” – see A for position of the close up