

RESEARCH ARTICLE

Red pigments and Boraginaceae leaves in mortuary ritual of late Neolithic China: A case study of Shengedaliang site

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Abstract

Investigation of mortuary ritual is an important method to reconstruct many aspects of past societies. Due to the lack of relevant analytical work, little evidence related to organic materials in a burial can be found in China. Here we report materials collected from a burial during the excavation of the Shengedaliang site. The recovered materials were analyzed using Raman spectroscopy and plant analysis: flotation, pollen and phytolith analysis. The red pigments found scattered over the human remains were identified as cinnabar. Extracted phytoliths associated with the burial are mainly leaves from the Boraginaceae family. This is the first time that a covering of leaves have been identified with a burial in Neolithic China. The presence of "special" leaves fossil may indicate a type of "plant worship" and the identification of an important plant used in traditional Chinese medicine. The finding of the two materials allows us to better identify indicators of funerary ritual and its relationship to social inequality.

KEYWORDS

pigments, leaves, mortuary ritual

1 | INTRODUCTION

Investigation of mortuary ritual is an important method to reconstruct many aspects of past societies. One symbolic practice associated with death and burial, the covering of human remains with red pigments, appears as early as the Middle Paleolithic (Zilhão, Angelucci, Badal-Garcco, D dal-Ga, Daniel, Dayet, . . . Zapata, 2010). The first known use of red pigments could be related to the appearance of anatomically modern Homo sapiens in Eurasia (Hovers, Ilani, Bar-Yosef, & Vandermeersch, 2003). Iron oxides were also sprinkled over burials recovered from the Shandingdong site (Norton, & Gao, 2008; Pei, 1939). Prehistoric humans also used plants to express their feelings towards the deceased as a part of their mortuary ritual. Plant remains recovered

during the excavation of Burial IV at Shanidar Cave in the Zagros Mountains of northern Iraq suggests that human of an adult Neanderthal was laid to rest on a bed of flowers (Leroi-Gourhan, 1975). However on the basis of taphonomic issues the association of the plant remains with the burial has been questioned (Sommer, 1999). Flowers from Lamiaceae or Scrophulariaceae were found in association with burials of terminal Pleistocene age from Raqefet (Hillman, Hedges, Moore, Colledge, & Pettitt, 2001; Munro, 2003; Willcox, 2008). The use of red pigments related to burial practices is well documented with the archaeological discoveries by a few scholars in the Chinese archaeological record. However, due to the lack of relevant analytical work, little evidence related to organic materials as a ritual practice can be found in China.

In May of 2013, Shaanxi Provincial Institute of Archaeology excavated a site covering some 1,800 square meters named Shengedaliang,

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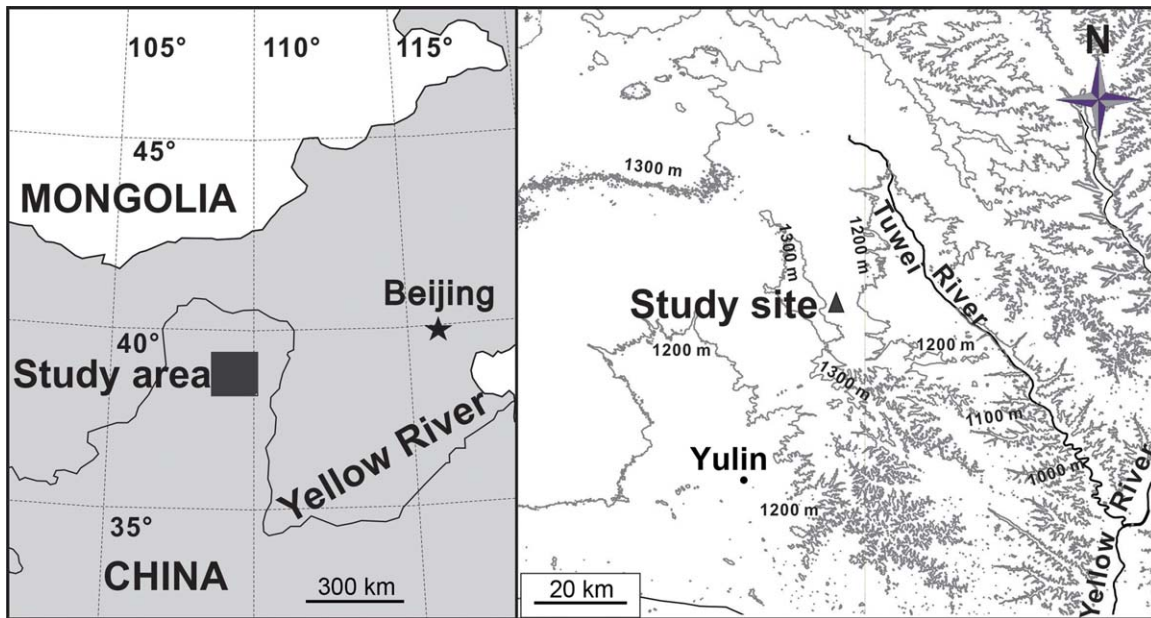


FIGURE 1 Diagrams showing the study area in Shaanxi Province, China [Color figure can be viewed at wileyonlinelibrary.com.]

under the direction of Wei-Lin Wang and Xiao-Ning Guo. The Shengedaliang site (Figure 1) is located 4.8km away from Shenmu city in Shaanxi Province. According to the preliminary investigation of the pottery, the presence of amphora and Dakou-type jars unearthed from the

site indicates that the excavated portion of Shengedaliang was occupied during the of Dakou II period from roughly the later Longshan (4,200–3,900BP) to the early Xia period (4,000–3,800BP).Thirteen tombs (from M1 to M13), eleven habitation structures, fifty-eight trash

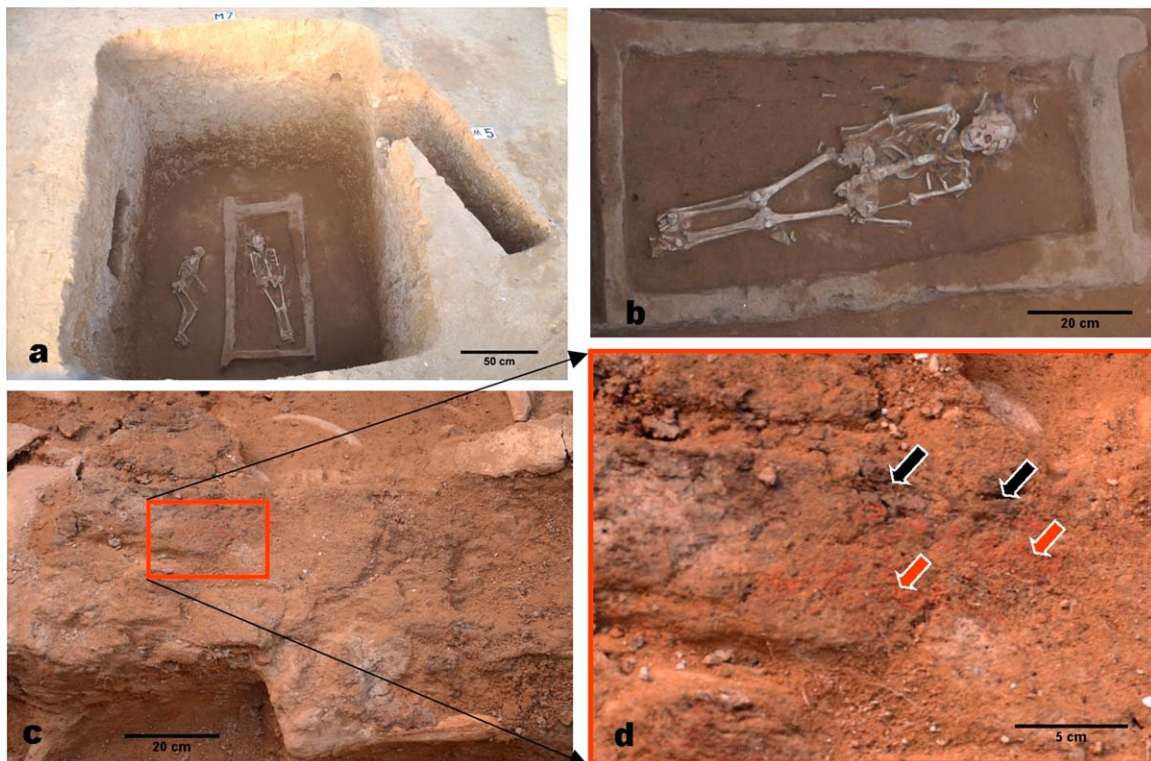


FIGURE 2 (a) Burial showing an intact coffin with a female sacrificial victim on the right side of the senior male (b) The details of an intact coffin with a female (c) Some red pigments and some black residues covered the coffin (d) The details of some red pigments and some black residues [Color figure can be viewed at wileyonlinelibrary.com.]

TABLE 1 AMS ^{14}C radiocarbon age of M7 male occupier

Sample	Laboratory number	Dated Material	Measured radiocarbon age	$^{13}\text{C}/^{12}\text{C} \delta^{13}\text{C}(\text{‰})$	Conventional radiocarbon age	2 Sigma calibration
SGDL-M7	Beta - 403918	bone collagen	3180 ± 30 BP	-9.0	3440 ± 30 BP	Cal BC 1875 to 1840 (Cal BP 3825 to 3790) and Cal BC 1820 to 1795 (Cal BP 3770 to 3745) and Cal BC 1780 to 1685 (Cal BP 3730 to 3635) and Cal BC 1670 to 1665 (Cal BP 3620 to 3615)

pits, and two graves were uncovered from the excavation. All tombs were scattered among house foundations and trash pits of the site. However there were extensive differences in the types of material culture recovered from the various excavated contexts across this site. Clear evidence of social ranking had been indicated by variation in the amount of labor and the materials that were associated with the different burials.

M7 (Figure 2a) was the largest tomb (416 cm length, 318 cm width and 325 cm height) located in the eastern of Trench No.1115, which consists of the burial chamber and the niche. In M7 the remains of a senior male (personal communication from Xiao-Ning Guo) were recovered in an intact coffin with a female sacrificial victim (personal communication from Xiao-Ning Guo) with lashed hands and feet on the right side of the senior male (Figure 2b). M7 is the largest tomb discovered in that time in northern Shaanxi area, and also the only one with sacrificial victim. Moreover, red pigments and some black residue were merely found on the senior male human from M7 as shown in Figure 2b-d. This paper aims to elucidate the composition of the red pigments and black residues used in the context of the burial practices.

2 | MATERIALS AND METHODS

2.1 | Dating

Phalange of the M7 male occupier (Figure 2b) was dated by accelerator mass spectroscopy (AMS) ^{14}C by the Beta Analytic Radiocarbon Dating Laboratory. The dates were then calibrated based on the dataset INTCAL13 (Reimer, Bard, Bayliss, Beck, Blackwell, Bronk Ramsey, ... van der Plicht, 2013).

2.2 | Sampling

Samples were analyzed from sediments derived from three different contexts. Firstly, three soil samples not associated with red pigments and three soil samples not associated with black residues were chosen to represent the background reference samples. The second sample type was the sediment directly from red pigments itself. In this process, two samples (M7-1 and M7-2) associated with red pigment were selected for analysis. The third sample type was taken directly from three black residues samples possibly mixed with a small amount of sediment.

2.3 | Micro Raman spectroscopy analysis for red pigments

A XploRA confocal Raman scope spectrometer was employed to determine the phase structure of red pigments. Pigments of red were illuminated using the 785 nm line of argon. The average spectral resolution in the Raman shift range of $100\text{--}1200\text{ cm}^{-1}$ 2 cm^{-1} within 20X microscope was prepared for the specimen surface. [AQ] Fixed-point sweep time (RTD) 2s, 5-10 s in a single scan, scan cycles 10 times. Calibration of the instrument was carried out on a daily basis with the 520.7 cm^{-1}

TABLE 2 Leaves of 21 species from modern Boraginaceae, Ulmaceae and Moraceae

Family	Genus	Species
Ulmaceae	<i>Celtis</i>	<i>Celtis koraiensis</i> Nakai
	<i>Celtis</i>	<i>C. bungeana</i> Blume
	<i>Ulmus</i>	<i>Ulmus macrocarpa</i> Hance
	<i>Ulmus</i>	<i>U. davidiana</i> Planch.
	<i>Ulmus</i>	<i>U. lamellosa</i> C. Wang & S.L. Chang
	<i>Ulmus</i>	<i>U. pumila</i> L.
	<i>Pteroceltis</i>	<i>Pteroceltis tatarinowii</i> Maxim.
Moraceae	<i>Cannabis</i>	<i>Cannabis sativa</i> L.
	<i>Humulus</i>	<i>Humulus scandens</i> (Lour.) Merr.
	<i>Humulus</i>	<i>H. lupulus</i> L.
	<i>Morus</i>	<i>Morus alba</i> L.
Boraginaceae	<i>Anchusa</i>	<i>Anchusa officinalis</i> L.
	<i>Cynoglossum</i>	<i>Cynoglossum divaricatum</i> Stapf . ex Lehm.
	<i>Eritrichium</i>	<i>Eritrichium borealisinense</i> Kitag.
	<i>Eritrichium</i>	<i>Eritrichium rupestre</i> (Pall. ex Georgi) Bunge
	<i>Lithospermum</i>	<i>Lithospermum zollingeri</i> A. DC.
	<i>Microula</i>	<i>Microula sikkimensis</i> (C.B. Clarke) Hemsl.
	<i>Microula</i>	<i>M. tibetica</i> Benth.
	<i>Onosma</i>	<i>Onosma paniculatum</i> Bureau & Franch.
	<i>Sinojohnstonia</i>	<i>Sinojohnstonia plantaginea</i> Hu
	<i>Trigonotis</i>	<i>Trigonotis peduncularis</i> (Trevir.) Steven ex Palib.

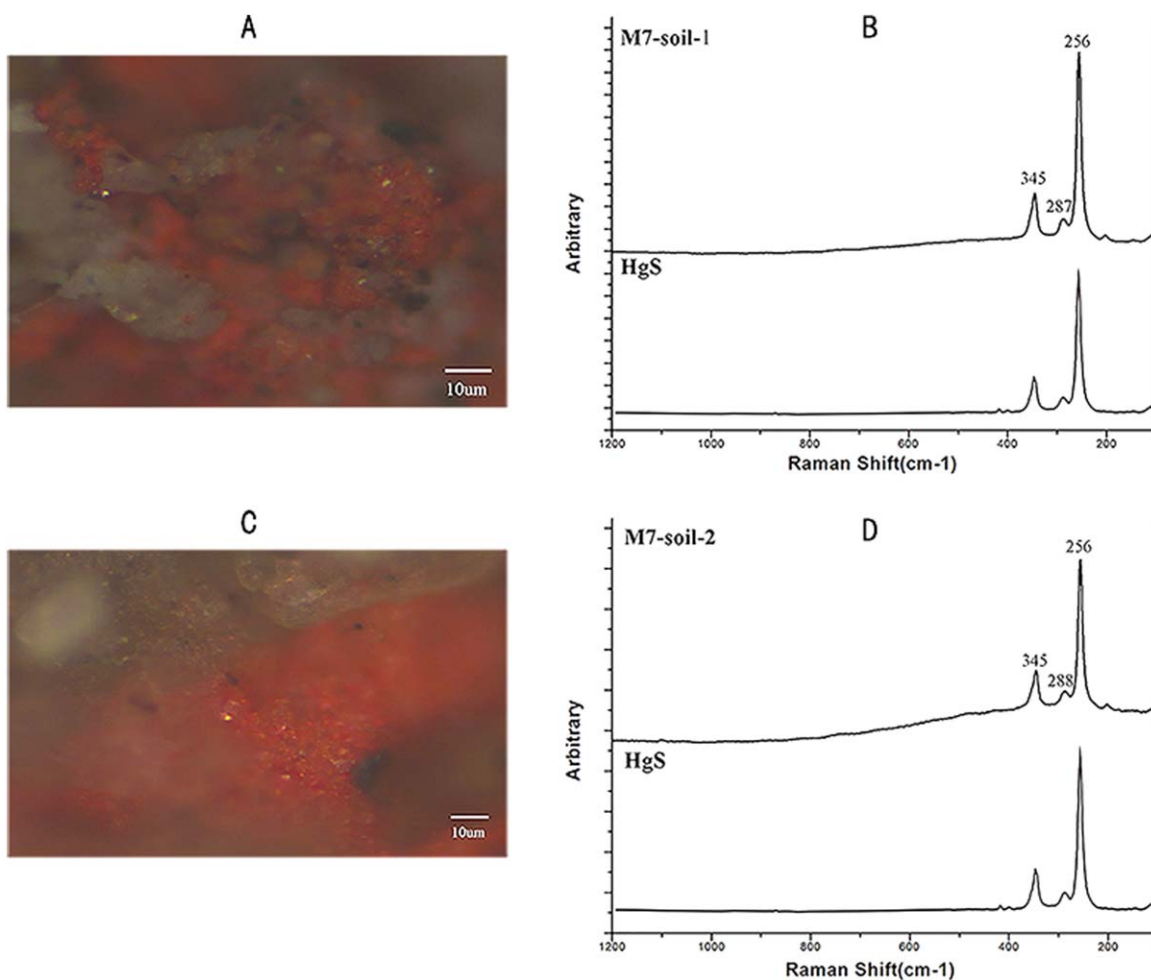


FIGURE 3 (a), (c) Optical images of the Raman measurements areas of the different samples (b), (d) Raman spectra of cinnabar associated with the red pigments [Color figure can be viewed at wileyonlinelibrary.com.]

silicon Raman band (Mio_C, Colombar, Sagon, Stojanovi_C, & Rosi_C, 2004). The Raman spectra were recorded by in situ optical microscopy, which assisted in the selection of sampling points on the specimen.

2.4 | Plant analysis for black residues

Flotation and pollen analysis were first conducted to identify these black residues. Unfortunately few macroremains and pollen were recovered from these black residues. However, phytoliths are more commonly preserved (Piperno, 2006) when macro-remains are unavailable or uninformative from these black residues. Therefore, phytolith analysis was employed as an important research method to identify black residue during this project.

The process of phytolith extraction followed the slightly modified methods outlined by Pearsall (2000) and Wu (2014). The samples were dried and ground into powder, then 10% HCl, and 30% H₂O₂ were added to each sample to remove carbonates and organic matter. To concentrate phytoliths and the other organic morphs, heavy liquid flotation is carried out using a ZnBr₂ at a density of 2.4 by centrifuging at 3000 rpm for 10 minutes. Identifica-

tion of phytoliths was performed through the examination of least 500 grains under a Nikon Eclipse LV100POL microscope using the published keys and modern reference collections in Key Laboratory of Vertebrate Evolution and Human Origins of Chinese Academy of Sciences, Institute of Vertebrate Paleontology and Paleoanthropology (IVPP), Chinese Academy of Sciences (CAS).

2.5 | Phytolith analysis for modern reference sampling

To compare the phytolith morphology, leaves of 21 species (Table 2) of main plants in the western region of China including modern Boraginaceae, Ulmaceae and Moraceae were sampled from the Laboratory of Systematic and Evolutionary Botany, Chinese Academy of Sciences and the Botany Laboratory of Beijing Normal University, China. Phytoliths were extracted by dry ashing method for analysis. The effect of dry ashing procedures for the extraction of phytoliths from modern plant samples had been discussed by several authors (Parr, Lentfer, & Boyd, 2001; Piperno, 2006; Sun, Wu, Wang, & Hill, 2012). The materials were scored for hair phytolith characters (Figure 4).

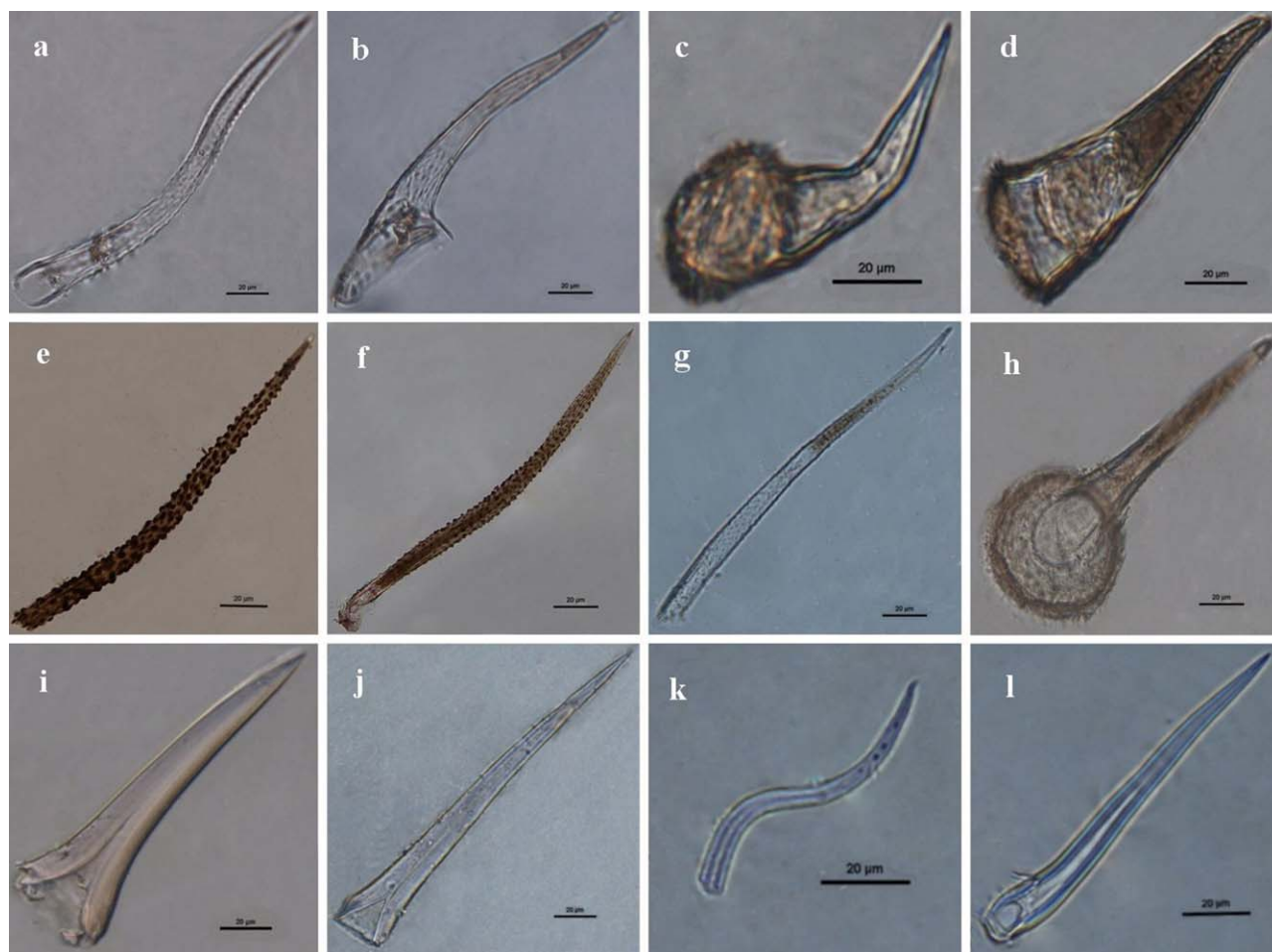


FIGURE 4 Hair phytoliths recovered from modern Boraginaceae, Ulmaceae and Moraceae. (a) *Cannabis sativa* L. (b) *Humulus scandens* (Lour.) Merr. (c) *Morus alba* L. (d) *H. lupulus* L. (e) *Eritrichium pauciflorum* (Ledeb.) A.DC. (f) *Eritrichium pauciflorum* (Ledeb.)A.DC. (g) *Eritrichium borealisinense* Kitag. (h) *Microula sikkimensis* (C.B. Clarke) Hemsl. (i) *Ulmus lamellosa* C. Wang & S.L. Chang (j) *Ulmus macrocarpa* Hance (k) *Ulmus parvifolia* Jacq. (l) *Pteroceltis tatarinowii* Maxim [Color figure can be viewed at wileyonlinelibrary.com.]

3 | RESULTS

3.1 | 14C date

In this study, AMS 14C dating directly on phalange of the M7 male occupier suggests an age (3,825 to 3,790 BP) based on the dataset INTCAL13 and archaeological evidence. Detailed information on the dated material, measured result and calibrated age (2 Sigma) were presented in Table 1.

3.2 | Red pigments

As can be seen from the Raman spectra of red particles in the two soil samples of M7-1 and M7-2 (Figure 3), the strong peaks at 256, 288, and 345 cm^{-1} are in accordance with the feature peaks of cinnabar (HgS) at 253, 283, and 343 cm^{-1} which described in the literature (Figure 3). Such peaks did not occur in the analysis of the 3 reference samples without red pigments. In particular, the peak at 253 cm^{-1} from red pigments was typically assigned to Hg-S bond stretching and vibration, while peaks at 285 and 343 cm^{-1} were assigned to transverse optical

phonon signal (optical phonon symmetrical signal) (Mio_c et al., 2004; Batta et al., 2013). As a result, three peaks of cinnabar (HgS) can be determined from Raman spectra in the two samples when the results of the analysis were compared with published literature. However, a displacement of approximately 2–5 cm^{-1} occurred in our samples with respect to the position of the peaks in the literature, which is likely due to the different measurement instruments used in experiment or differences in the spectral calibration process. Regardless of the sources of variation the displacement is within the error range. The red particles associated with the burial of M-7 were determined to be composed of cinnabar (HgS) by Raman spectroscopy analysis.

3.3 | Black residues

As shown by phytolith analysis, a large number of special hair phytoliths were found in three black residues samples and no such special hair phytoliths were found in the 3 reference samples. Based on current phytolith publications and reference collection, (Iriarte, & Paz, 2009; Morris, Baker, Morris, & Ryel, 2009; Tsutsui, Sakamoto,

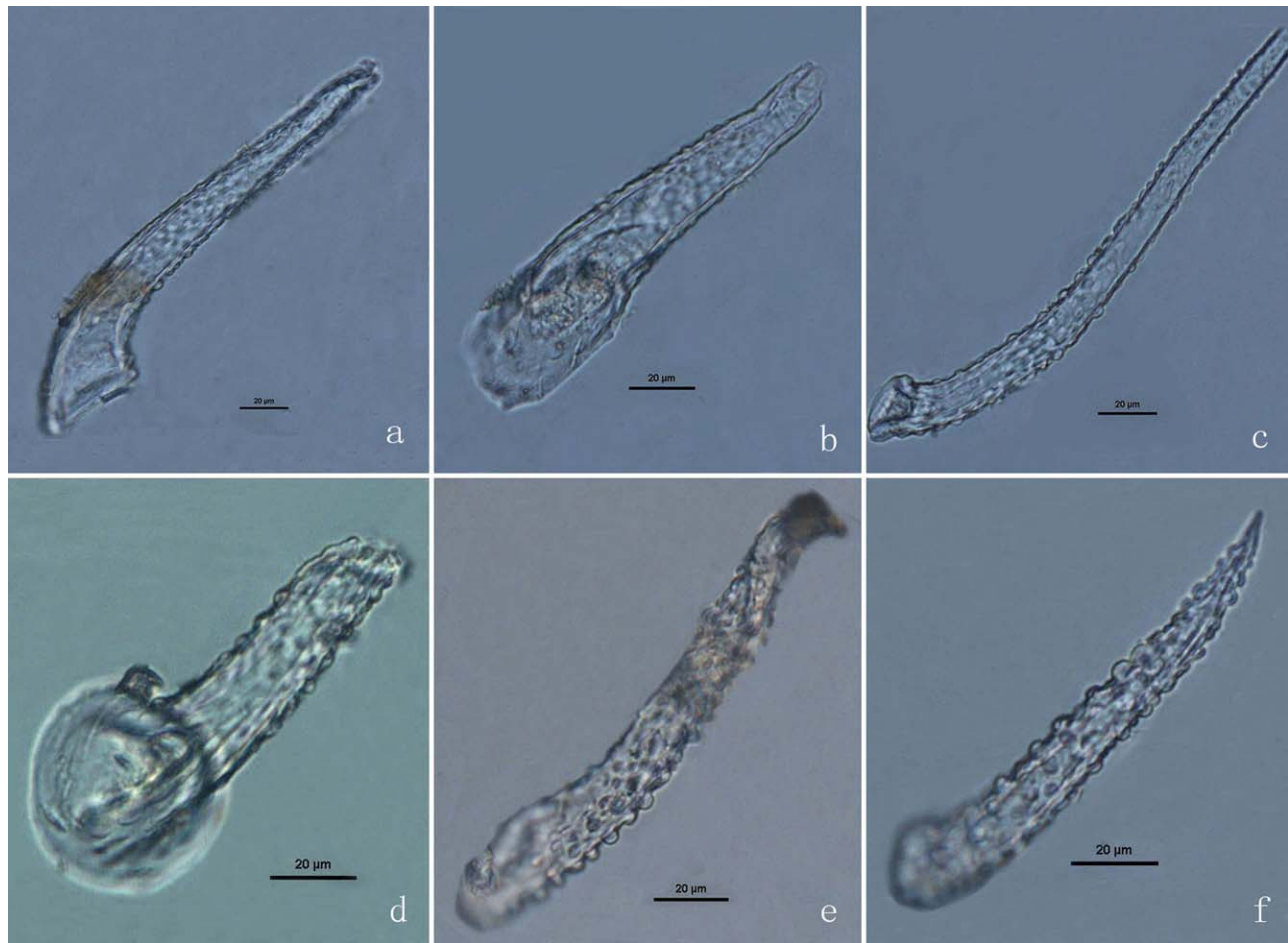


FIGURE 5 Hair phytoliths recovered from black residues [Color figure can be viewed at wileyonlinelibrary.com]

Obayashi, *et al.*, 2015; Wallis, 2003) this special hair phytoliths could be described as long, armed and unicellular hairs which are similar those recovered from the coffin of M-7 have been reported only from dicot leaves. These kinds of hairs phytoliths are possibly found in Boraginaceae, Ulmaceae and Moraceae families based on current phytolith publications. Besides, in the “Phytoliths in the Flora of Ecuador: the University of Missouri Online Phytolith Database (Pearsall, 2011),” we observed hairs of this general type in the Boraginaceae and Moraceae families. Thus, we might examine comparative plant specimens in those families to begin with Boraginaceae, Ulmaceae and Moraceae families.

In this paper, more reference work has examined in greater detail hair phytolith characteristics from Boraginaceae, Ulmaceae and Moraceae families (Figure 4). Compared all of these hair phytoliths, we found Boraginaceae with a characteristic feature of the family being the presence of unicellular bristly hairs. Ornamented bristly hair phytoliths recovered from black residue look very much like phytoliths from a member of the Boraginaceae family (Figures 4 and 5). Given the demonstrated widespread production of phytoliths in members of the Boraginaceae, it is probable that at least some of the other members of this family present produce phytoliths of a similar type to those observed (Figure 5). 90% of special hair phytoliths were concentrated in black residues, which indicate that remains of Boraginaceae family

leaves were consciously brought into the burial. We believe possibly Boraginaceae family leaves were also used to cover the body, which cause many Boraginaceae family phytoliths to be recovered.

4 | DISCUSSION

The red pigments are cinnabar and black residues are mainly leaves phytoliths possibly from the Boraginaceae family. Cinnabar is a red colored mineral composed of mercury sulphide (HgS). It is scarce in nature and requires a great investment of time to obtain. The earliest cinnabar has been sought by humans since the Neolithic Age in China and Western Country (Marttryict, Marttryict, Delibes-de-Castro, Zapatero-Magdaleno, & Sarabia-Herrero, 1995). As a pigment, cinnabar has been used to obtain the bright red color known as vermilion, widely used by artists throughout history in the world of cultural convergence. As an exotic and uncommon mineral, the presence of cinnabar has also been used as an indicator of elevated social status. For example, in the tomb of the ruler of Calakmul Yuknom Yich'akK'ak also known as Jaguar Paw, located in Campeche, cinnabar was scattered across the tomb including his burial shroud and grave goods (Batta *et al.*, 2013).

Since c.6,000BC in China, cinnabar was used in ornate burials by spreading it on the body. In the Taosi Culture (c.4,600–4,000 BP) of the Longshan Period, the ceremonial procedure of putting cinnabar under the human or coffin was established, and passed on to the following Erlitou Culture (commonly equated with the Xia Dynasty). The association of cinnabar with elite burials became a key part of funerary practice during the subsequent the Shang and Zhou Dynasties. However, Cinnabar remains were only found in high social rank burials, and never excavated in low social rank burials from the prehistoric and historical sites (Fang, 2015; Gao, 2011). The presence of the female sacrifice found in association with the burial at the Shengedaliang site, along with the presence of cinnabar leads us to hypothesis that cinnabar is not only a ritual element but also an indicator of social rank.

Phytoliths from Boraginaceae leaves were found from the corpse and cinnabar layers in the tomb. According to our present knowledge, no leaf burial has been reported before in China. Due to the lack of comparative material it is difficult to determine when exactly humans started to use leaves in ceremonial contexts in China. Leaves symbolize life in many modern human societies. Leaves of species are used to symbolize various aspects of human life, e.g. love, joy, loyalty, fertility and sympathy from birth to death at religious and social occasions such as funerals or weddings (Heilmeyer, 2001). The presence of “special” leaves fossil also may indicate the strong sense of life in the otherwise desert environment that made people to treat leaves differently as a sign of energy and life and may also be an additional indicator of social inequality. Thus, the association with leaves fossil from a member of the Boraginaceae family may indicate a type of “plant worship.”

Moreover, Boraginaceae plants have always been one of most important and commonly used herbal medicine in Chinese history. It was described in a few famous Chinese medical books, like ‘Sheng Nong’s herbal classic’ (Shennong Bencao Jing) which was considered the oldest book on Chinese herbal medicine (Sun, & Sun, 1991). It is especially effective for diseases involving respiratory system, mostly including cough, common cold, flu, asthma, hay fever, allergy, and so forth. These diseases are generally caused by the negative factor of “wind” according to theory in traditional Chinese medicine (Cai, Luo, Sun, & Corke, 2004; Li, Zheng, Bukuru, & Kimpe, 2004). The association with Boraginaceae leaves from the M-7 burial may help to identify an important plant used in traditional Chinese medicine.

The social position of an individual is directly reflected by the mortuary treatment received by the individual upon death. In the case of M-7 a late Neolithic burial two types of natural materials were used as part of the final burial ritual for this individual. The red pigment is cinnabar and black residue is possibly residue from the leaves of Boraginaceae. This is a unique case of cinnabar and leaves associated with a prehistoric burial in China, we have no examples for comparison.

5 | CONCLUSION

This study has demonstrated that red pigments and Boraginaceae leaves collected from a burial have been identified in Shengedaliang site. Specially, this is the first time that a covering of leaves fossil have

been found in Mortuary ritual of late Neolithic China. These two materials provide indications of funerary ritual and its relationship to social inequality.

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