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The effect of depositional conditions on mineral transformation, chemical composition, and preservation of organic material in archaeological Hg-enriched bone remains

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ABSTRACT

Archaeological bone remains are exposed to depositional conditions that profoundly affect their preservation. This manuscript reports on the mineral transformation, chemical composition, and preservation of organic material in pre-Hispanic archaeological Hg-enriched bone remains exposed to different depositional conditions, namely, open and confined environments. Here, we studied 2 vertebrae specimens collected inside Temple XIII of the archaeological site of Palenque, Mexico. Specifically, the 2 vertebrae were collected inside and outside a sarcophagus (VIS and VOS) located in a burial chamber and belonged to a 30-yr-old woman and an 11-yr-old boy, respectively. Subsections of the vertebrae specimens were characterized by radiocarbon dating, high-resolution electron microscopy (HRSEM), and energy-dispersive X-ray spectroscopy analyses (EDS). VIS and VOS showed conventional radiocarbon ages dating of 5050 ± 30 BP and 5440 ± 30 BP respectively. HRSEM-EDS analysis for VIS showed a high content of Hg (~30%) in the upper layer and below the detection limit in the lower layer. In contrast, HRSEM-EDS analysis for VOS confirmed the lack of a layered structure, for which the accumulation of Hg was as high as 30%. Furthermore, VIS was the only vertebrae that showed the presence of S. This was because VIS belonged to a pre-Hispanic character exposed to HgS, typically used in Mayan royal rituals; VOS did not contain S because the Hg source in this case was probably from cooking utensils, the elaboration of paintings or murals, or the ingestion of contaminated food or water. In addition, VOS contained a particularly low content of P, which was attributed to hydroxyapatite undergoing higher rates of transformation in the open environment. Moreover, the incorporation of Hg in hydroxyapatite *via* Hg-for-Ca substitution further accelerated the mineral transformation pathway(s). Finally, the pre-Hispanic archaeological Hg-enriched bones preserved the genetic material, regardless of the depositional environments (exposed or unexposed). Therefore, the prevailing preservation mechanisms differ from those underpinned by physical protection by collagen fibrils or natural antibacterial mechanisms.

1. Introduction

Archaeological human remains accumulate mercury (Hg), an element intrinsically associated with bactericidal and fungicidal properties (Geier et al., 2007) as well as with high toxicity in living organisms (Tchounwou et al., 2012). The accumulation of Hg in remains has been shown to occur in many regions of the world (Yamada et al., 1995; Gazzola, 2009; Cervini-Silva et al., 2013; Emslie et al., 2015). Early

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evidence in human remains belonging to two different periods, centuries 6 to 7 and 12 to 17, collected from Tokushima and Matsuyama, Japan, showed accumulation of Hg in concentrations as high as 1700 μg g^{-1} (Yamada et al., 1995). In particular, cinnabar (HgS), a natural source of Hg used in funerary rituals or ceremonials and as a prime ingredient for paintings or murals (Gazzola, 2009), has been identified in archaeological sites located in different geographical regions (Emslie et al., 2015). An X-ray study of HgS-enriched archaeological remains recovered from Jaina, Monte Alban, and Ranas, Mexico showed the incorporation of Hg within the hydroxyapatite mineral lattice (Ávila et al., 2014). Meanwhile, HgS has been proven to contribute to the preservation of ancient organic materials by acting as an antibacterial agent (Cervini-Silva et al., 2013). However, to the authors' best knowledge, little information is available on the preservation mechanisms prevailing in archaeological Hg-enriched specimens, and little is known about the effects of depositional conditions on such preservation mechanisms. In this work, we studied two vertebrae specimens recovered from inside (VIS) and outside (VOS) a sarcophagus inside a burial chamber located in Temple XIII, which belonged to a 30-yr-old woman and an 11-yr-old boy, respectively. The specimens were analysed using radiocarbon dating, high-resolution electron microscopy, and energy-dispersive X-ray spectroscopy techniques.

2. Materials and methods

2.1. Sample collection and carbon dating analysis

2.1.1. Sample collection

The 2 vertebrae specimens were collected from Temple XIII, Palenque Chiapas, in southern Mexico (Fig. 1, panels A) at the coordinates 17° 29′ 2.32″ N, 92° 2′ 46.78″ W (Fig. 1, panel B), in the archaeological site of Palenque (Fig. 1, panel C). Specimen VIS was inside the sarcophagus (Fig. 1, panel D) and specimen VOS was outside the sarcophagus (Fig. 1, panel E) in the same burial chamber. VIS belonged to a 30-yr-old woman and VOS to an 11-yr-old boy (Fig. 1, panels D and F, respectively).

Each specimen was placed in a sterile tube to avoid contamination during handling. In addition, the laboratory was equipped with a high-pressure air system in the form of laminar flow hoods to filter the incoming air; UV light irradiation and bleach were used to clean all working surfaces (Muñoz et al., 2012); protective clothing was used at all times.

2.1.2. Radiocarbon age

Radiocarbon dating of the specimens by the Beta Analytical Radiocarbon Dating Laboratory (Miami, Florida) was performed according to the company procedure and included a pretreatment with alkali to extract collagen (Oeschger et al., 1975; Stuiver and Braziunas, 2006; Reimer et al., 2009; Heaton et al., 2011). A full description of specimen radiocarbon measurement is provided by the Beta Analytical Radiocarbon Dating Laboratory (Introduction to Radiocarbon

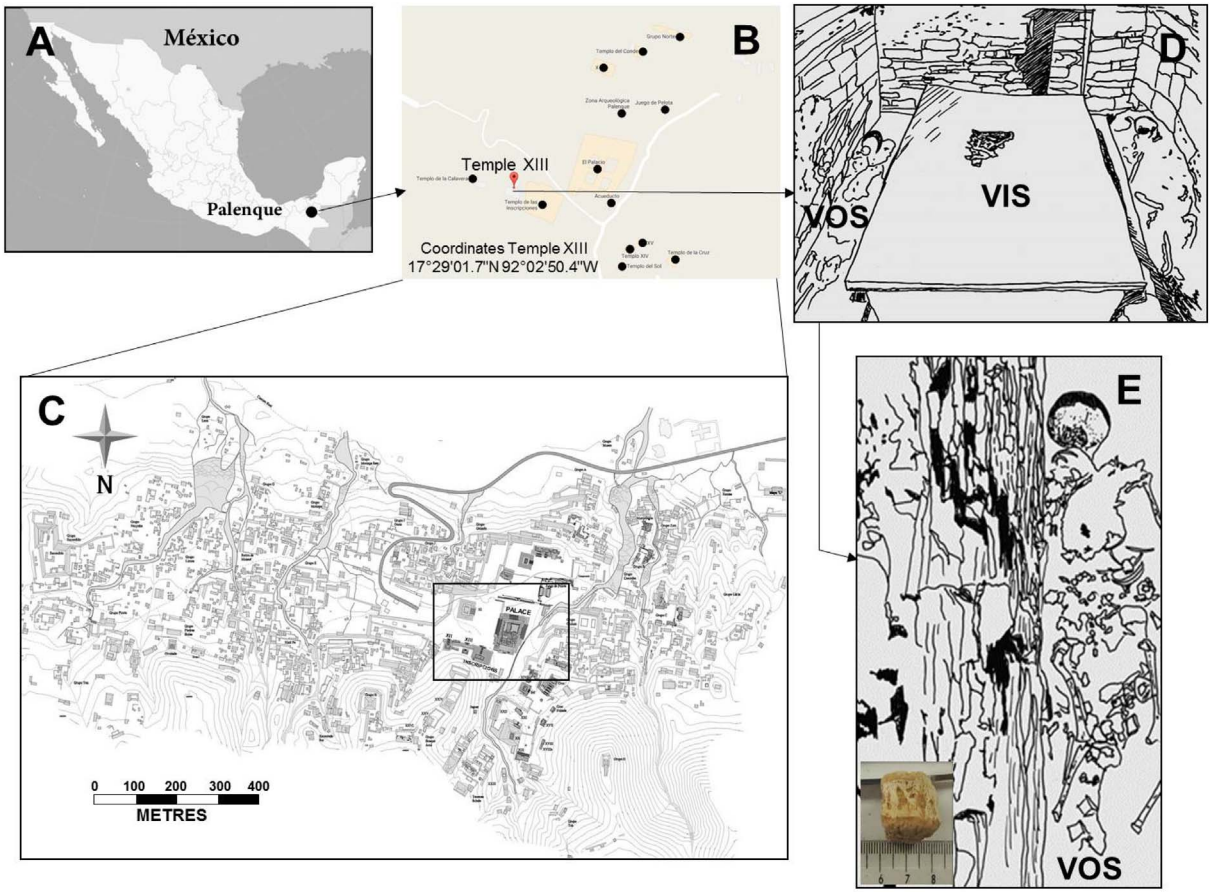

Fig. 1. Location of the vertebrae specimens in Temple XIII in the archaeological site of Palenque, Mexico. (A) Location of Palenque in southern Mexico; (B) location of Palenque in the north of the state of Chiapas, Mexico with coordinates; (C) scale map of the archaeological site Palenque and Temple XIII location; (D) remains belonging to a 30-yr-old woman (VIS) found inside the sarcophagus in the burial chamber; (E) remains belonging to an 11-yr-old boy (VOS) found in an extended position on the floor between the west wall and the sarcophagus in the burial chamber inside Temple XIII. On the left side is a picture of the VOS sample.