

Acetic acid-leachable elements in pedogenic carbonate nodules and links to the East-Asian summer monsoon



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ABSTRACT

Carbonate nodules at the base of paleosols within Chinese loess are products of pedogenic neof ormation and thus have great potential to document the summer monsoon history. Here we present mineralogical and elemental compositions of carbonate nodules from a north–south loess transect of northern China for the last and penultimate interglacials. X-ray diffraction patterns show that the nodules are composed mainly of calcite, together with some silicate minerals such as quartz, plagioclase, orthoclase, illite, and kaolinite. Elemental composition of the acetic acid-leachable fraction is characterized by extremely high Ca concentration and modest K and Mg content, followed by Al, Si, Sr, Mn, Fe, P, Na, Ba, Pb, Th, and U. Based on the mineralogical data and relationship between carbonate and the leachable element content, we suggest that K, Mg, Sr, Ba, and U mainly come from clay minerals, Na from highly soluble salts trapped by calcite particles, and Al and Si from amorphous phases of pedogenic origin. For both the last and penultimate interglacials, the Na content decreases from north to south, while the Al, Si, and U concentrations show a southward-increasing pattern. These characteristics all suggest a southward decrease in aridity across the Chinese Loess Plateau for both interglacials, consistent with the pattern of a southerly increase in pedogenic development for the two interglacial soils, and with the pattern of the present north–south rainfall gradient. Therefore, Na/Al and Na/Si ratios in the acetic acid extract are efficient proxies for aridity and are ultimately controlled by the summer monsoon precipitation, i.e. high ratios indicate weakened summer monsoon intensity. In addition, the significant content of highly soluble Na salts in carbonate nodules highlights soil dewatering as an important mechanism for the formation of pedogenic carbonate, at least in the case of the northern Loess Plateau.

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1. Introduction

Chinese loess deposits, with an area of 440,000 km², were transported by the East-Asian winter monsoon from the arid regions in northwestern China (An et al., 1991a; Liu, 1985; Liu and Ding, 1998; Yang and Ding, 2008). On the Chinese Loess Plateau, complete loess sequences consist of over 30 loess–soil couplets, which date back to ~2.8 Ma (Ding et al., 1993; Rutter et al., 1991; Xiong et al., 2003; Yang and Ding, 2010). The yellowish loess beds were deposited during cold, dry glacial periods, whereas the brownish or reddish soils developed in warm, humid interglacials (Ding et al., 1993; Kukla, 1987; Liu, 1985). From glacial to interglacials, the East-Asian winter monsoon weakened, while the summer monsoon strengthened (An et al., 1991b; Liu and Ding, 1998; Rutter and Ding, 1993; Yang and Ding, 2008). Thus the alternation of loess and soils documents large-amplitude oscillations of the winter and summer monsoons on orbital timescales.

The East-Asian summer monsoon is the main moisture carrier for the Loess Plateau, with over 70% of annual rainfall occurring in summer.

Therefore, chemical weathering intensity of loess deposits is widely used as a proxy for the East-Asian summer monsoon variations, and it is mostly quantified by a suite of elemental ratios such as Ba/Sr (Ding et al., 2001a; Gallet et al., 1996), Th/U (Gallet et al., 1996; Gu et al., 1997), Rb/Sr (Chen et al., 1999), Na/Al (Gu et al., 1999), Chemical Index of Alteration (CIA) (Liu et al., 1995; Xiong et al., 2010), Fe₂O₃(free)/Fe₂O₃(total) (Ding et al., 2001b; Guo et al., 2000), and (CaO + Na₂O + MgO)/TiO₂ (Yang et al., 2006). However, chemical weathering intensity reflected by most of these proxies is a combined result of chemical weathering in dust sources and sinks, and that caused by transport-induced particle size differentiation (Eden et al., 1994; Gu et al., 1997, 1999; Yang and Ding, 2004; Yang et al., 2006). It is only the signature resulting from post-depositional weathering that can be linked to summer monsoon intensity. In this regard, authigenic clay and carbonate minerals, products of soil-forming processes, are ideal materials for the reconstruction of summer monsoon. However, many of the clay minerals in loess and paleosol units are mainly detrital, rather than pedogenic origin (Ji et al., 1999; Liu, 1985), as the Chinese loess–paleosol deposits have experienced only the incipient and early stages of chemical weathering (Chen et al., 1998; Han et al., 1998). This makes it difficult to distinguish and separate authigenic from detrital clay minerals.

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During pedogenic processes, Ca^{2+} , released during dissolution of Ca-bearing minerals (mainly detrital calcite), are transported downward by precipitation-recharged soil water, and eventually reprecipitate as pedogenic carbonate at depth. Thus elemental and isotopic compositions of pedogenic carbonate have great potential to document local climate conditions. Carbonate nodules, a common form of pedogenic carbonate, are abundant at the base of most interglacial paleosols. To date, numerous isotopic studies of carbonate nodules from Chinese loess have been performed to investigate the history of vegetation and paleomonsoon (Ding and Yang, 2000; Han et al., 1997; Jiang et al., 1995, 1998; Wang and Zheng, 1989; Yang et al., 2012). However, little is known about the elemental composition of the carbonate nodules. On the Loess Plateau, there is a pronounced north–south climatic gradient in both present and past interglacials (Derbyshire et al., 1995; Yang and Ding, 2003; Yang et al., 2012). This provides a unique opportunity to evaluate the relationship between elemental composition of carbonate nodules and the summer monsoon.

This study presents the carbonate content, mineral and elemental compositions of carbonate nodules from a north–south loess transect for the last and penultimate interglacials. The aim is to identify the spatial pattern of elemental composition of carbonate nodules and to address its paleoclimatic implications.

2. Setting and stratigraphy

Our study transect runs north–south from Hongde near the Mu Us desert margin, to Yangling in the southernmost part of the Loess Plateau (Fig. 1). Four loess sections located at Hongde, Baimapu, Binxianbei, and Yangling were studied (Fig. 1). At present, the mean annual temperature and precipitation are 8.2 °C and 380 mm at Hongde, 8.5 °C and 500 mm at Baimapu, 10 °C and 560 mm at Binxianbei, and 13 °C and 630 mm at Yangling. Both mean annual temperature and rainfall increase southward along the loess transect (Fig. 1).

All loess sections consist of the loess(L)–soil(S) sequence S0, L1, S1, L2, and S2 (Fig. 2). The Holocene soil, S0, is characterized by a massive structure, relatively abundant dark organic matter and a few white-colored secondary carbonate pseudomycelia. The loess units L1 and L2

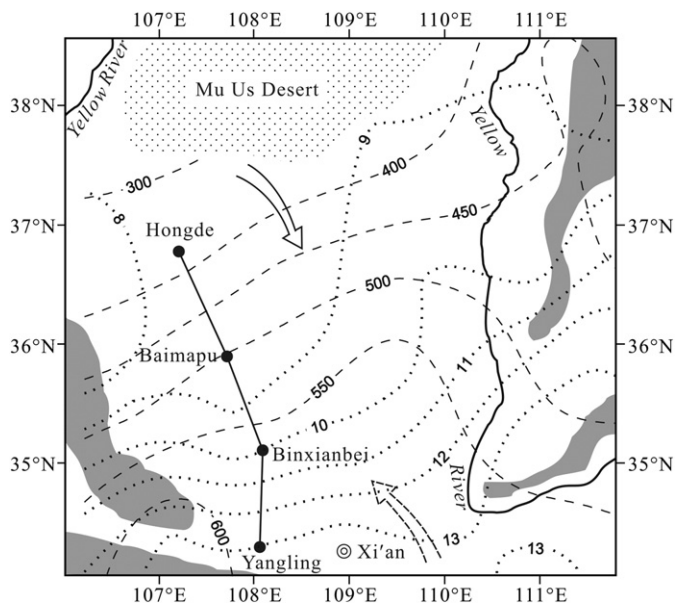


Fig. 1. Schematic map showing the location of the sites along the north–south transect on the Loess Plateau. Also shown are the Mu Us desert (dotted) and mountains (shaded) around and within the Plateau. The isohyets (mm, dashed lines) and isotherms (°C, dotted lines) are averaged values over 32 years (1970–2001). The solid and dashed arrows indicate the direction of the winter and summer monsoonal winds, respectively.

were deposited during the last and penultimate glacial periods, respectively. Both L1 and L2 are yellowish in color and massive in structure. Previous studies (Ding et al., 2002; Kukla, 1987; Lu et al., 2007) have shown that L1 is correlated with marine oxygen isotope stages (MIS) 2 to 4, and the L2 loess unit with MIS 6. The soil units S1 and S2 developed in the last and penultimate interglacial periods and correlate with MIS 5 and 7, respectively (Ding et al., 2002; Kukla, 1987; Lu et al., 2007). Both are brownish or reddish in color, and have an A-Bw-Bk-C or A-Bt-Bk-C horizon sequence. Soil unit S2 is composed of two soils (S2-1 and S2-2) and a thin intervening loess horizon. Only the upper soil (S2-1) was sampled in this study, as it is better developed than the lower one (S2-2). From Hongde southward to Yangling, the thickness of the loess units L1 and L2 decreases from over 20 m to several meters, while that of the soil units S1 and S2-1 decreases from 3–5 m to ~1.5 m. Based on a stacked orbital timescale of Chinese loess (Ding et al., 2002; Fig. 2), the soil units S1 and S2-1 formed in the periods 128–73 ka and 219–190 ka, respectively.

Carbonate nodules, with grayish color and sub-spherical or irregular shape, are commonly found at the base of paleosols but are rare within loess units. Generally, nodules at the base of the Holocene soil (S0) are sporadic and small (<1.5 cm) and can be found only in the central and southern Loess Plateau. For the soil units S1 and S2-1, nodule sizes range from a few centimeters in the north to over 10 cm in the south. At each transect site, carbonate nodules were taken from the base of soil units S1 and S2-1.

3. Materials and methods

For all the sections, a total of 2512 samples were collected at 5 cm intervals. Bulk magnetic susceptibility and grain size were measured for all samples with a Bartington MS2 susceptibility meter and a SALD-3001 laser diffraction particle analyser. The particle analytical procedures were detailed by Ding et al. (1999). Eight carbonate nodules (~2 g) taken from the base of S1 and S2-1 were ground to powder and split into three parts for analyses of carbonate content and mineralogical and elemental compositions. Carbonate content was determined using the gravimetric method (Soil Conservation Service, 1972) and has an error of $\pm 5\%$. The mineralogical composition was determined by X-ray diffraction (XRD) using a PANalytical X'Pert PRO diffractometer with a Ni-filtered $\text{CuK}\alpha$ radiation (40 kV, 40 mA). A 0.0625° divergence slit and a 0.125° antiscatter slit were used. Powder samples, smeared on a glass slide with distilled water, were scanned from 2.5° to $70^\circ 2\theta$ with a step size of $0.0167^\circ 2\theta$ and a measuring time of 0.3 s per step.

Elemental composition of samples was analyzed by inductively coupled plasma mass spectrometry (ICP-MS) at the University of Arizona. Ground samples (~0.1 g) were first treated with 0.2 M NH_4 -acetate with a pH 8.2 for 4 h to minimize the effects of adsorbed cations, and then digested for at least 12 h in 0.2 M ultrapure acetic acid at room temperature to dissolve the carbonate fraction. After dissolution, carbonate digest was centrifuged, decanted, and rinsed with 18 M Ω H_2O . The supernatant was then dried down on a hotplate, redissolved in 1.5% ultrapure HNO_3 , and analyzed using a PerkinElmer Elan DRC II ICP-MS. Analytical uncertainties are $\pm 5\%$ for Ca content, and $\pm 10\%$ for content of other elements. This acetic acid leaching method has been widely used to measure the elemental and isotopic compositions of carbonate fraction in carbonate/silicate mixtures such as pedogenic carbonate (Quade et al., 1995; Van der Hoven and Quade, 2002), carbonate rocks (English et al., 2000), and ocean sediments (Lyle et al., 1984), and the acetic acid-leachable elements have been used to reconstruct paleoenvironmental conditions.

4. Results

4.1. Magnetic susceptibility and grain size

Paleosols are characterized consistently by higher susceptibility values and finer particle sizes compared with the loess horizons above

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