



# Primary and secondary carbonate in Chinese loess discriminated by trace element composition

Gaojun Li\*, Jun Chen, Yang Chen

MOE Key Laboratory of Surficial Geochemistry, Department of Earth Sciences, Nanjing University, Nanjing 210093, China

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## Abstract

Carbonate in the eolian deposits on the Chinese Loess Plateau carries a significant amount of information on the evolution of paleo-environment. However, the paleo-proxies based on total carbonate are largely complicated by the coexistence of primary and secondary carbonate. This work provides a new method to quantify the relative proportion of primary and secondary carbonate in these eolian deposits according to the Mn/Ca and Mg/Ca ratios of bulk carbonate. The carbonate associated Mg and Mn, which can be extracted by selective dissolution using diluted weak acid, are mostly derived from primary carbonate because the secondary carbonate contains very little Mg and Mn. The amounts of Mg and Mn associated with carbonate could thus serve as proxies for the pedogenic dissolution of primary carbonate, which reflect clear glacial–interglacial changes of summer monsoon. A positive correlation between the Mn/Ca and Mg/Ca ratios of bulk carbonate has been observed, indicating binary mixing between secondary carbonate with the low Mn/Ca and Mg/Ca ratios and primary carbonate with high Mn/Ca and Mg/Ca ratios. The high Mn/Ca and Mg/Ca ratios of primary carbonate endmember also indicate Alxa arid lands as the main source of eolian deposits on Chinese Loess Plateau. The Mn/Ca and Mg/Ca ratios of carbonate could be employed as a quick method to check the purity of the secondary carbonate used for paleo-environmental studies and help to interpret the paleo-proxies based on the bulk carbonate. The results reveal significant amount of primary carbonate included in the traditionally believed pure authigenic carbonate phases such as carbonate concretion, rhizolith, and the fine grain sized carbonate. The new method also helps to appraise the control of the relative portion of primary and secondary carbonates on the stable carbon and oxygen isotopes of bulk carbonate. High Mg/Ca ratio and low Mn/Ca ratio of the carbonate in red clay formation is observed, which confirms the existence of authigenic dolomite.

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## 1. INTRODUCTION

Soil carbonates carry information on the paleo-environmental condition under which they formed (e.g., Cerling, 1984; Gocke et al., 2011). Being preserved by continuous dust accumulation, soil carbonate in the eolian deposits on the Chinese Loess Plateau provides a rare opportunity to explore the evolution of terrestrial environments during the late Cenozoic Era (e.g., Han et al., 1997). A band of loess deposits covers large areas in the south of Chinese arid and semi-arid lands (Fig. 1a). It has been shown that the dust transported

from the arid lands in North China and neighboring Mongolia by prevailing northwesterly winds are responsible for the accumulation of loess (Liu, 1985; Li et al., 2009). The thick eolian deposits on the Chinese Loess Plateau (Fig. 1b) are arguably the longest terrestrial archives for late Cenozoic environmental changes back to the late Oligocene (Heller and Liu, 1982; Sun et al., 1998; Guo et al., 2002; Qiang et al., 2011). The upper portion of the eolian deposits, which is commonly referred as Chinese loess, is a typical loess-paleosol sequence consisting of more than ten alternations of loess-paleosol layers (Heller and Liu, 1982). Alternation of loess and paleosol layers is believed to be a reflection of glacial–interglacial climate changes since about 2.6 Ma (Liu, 1985). Loess layers, which have higher carbonate content and low magnetic susceptibility, were formed during

\* Corresponding author.

E-mail address: [ligaojun@nju.edu.cn](mailto:ligaojun@nju.edu.cn) (G. Li).

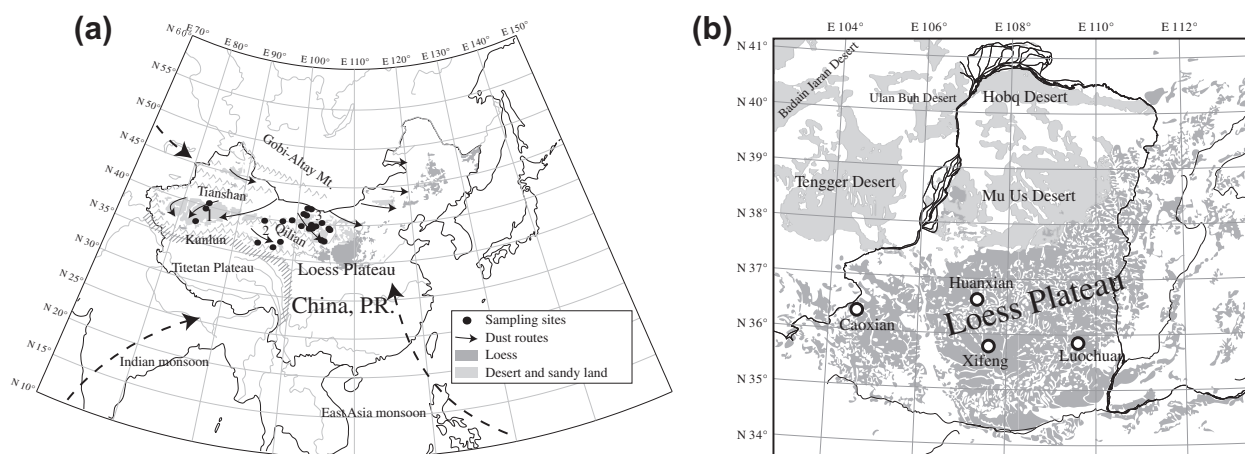


Fig. 1. Maps show the study sites. (a) Geographical setting of North China and the sampling sites in the potential source regions: (1) Taklimakan Desert (Tarim Basin); (2) Qaidam Desert (Basin); (3) Alxa arid lands. (b) Chinese Loess Plateau and the sites of eolian sequences.

glacial periods under strong winter monsoon and thus a high dust accumulation rate, while the paleosols with low carbonate content and high magnetic susceptibility were pedogenic products under a relatively slow dust accumulation and strong influence of summer monsoon (Liu, 1985; An et al., 1990, 1991a,b). The eolian deposit underlying the loess-paleosol sequence is red clay formation deposited about 7–2.6 Ma (Sun et al., 1998). This red clay deposit is characterized by more than 10% carbonate content on average (Chen et al., 2007b). In the western Chinese Loess Plateau, loess and red clay aging back to the late Oligocene can be found with abundant carbonate nodules (Guo et al., 2002; Ge and Guo, 2010; Qiang et al., 2011).

Paleo-proxies related to the soil carbonate in the eolian deposits on the Chinese Loess Plateau include carbonate content, stable C and O isotopes and recently ‘clumped’ isotopes of bulk and/or secondary carbonate (Chen et al., 1996; Han et al., 1997; Wang and Follmer, 1998; Ding and Yang, 2000; Rao et al., 2006; Li et al., 2007b; Liu et al., 2011; Suarez et al., 2011). It is believed that carbonate content simply reflects the degree of pedogenic dissolution, which reflects the climate changes associated with the alternation of loess and paleosol layers (Liu, 1985; Chen et al., 1997). Recent studies show that the content of carbonate in red clay seems to be tightly coupled with the long-term evolution of Asian monsoon (Chen et al., 2007b; Sun et al., 2010). However, the usage of carbonate content as indicator for pedogenic leaching is largely limited by the precipitation of secondary carbonate. High content of carbonate in particular layers, especially those with abundant carbonate nodules, is caused by reprecipitation of the carbonate leached from overlain layers (Han et al., 1997; Rowe and Maher, 2000).

Carbon isotopes of bulk carbonate or carbonate nodules in eolian deposits on the Chinese Loess Plateau are generally interpreted as a reflection of the types of C3/C4 vegetation (Han et al., 1997; Wang and Follmer, 1998; Ding and Yang, 2000; Rao et al., 2006), or as a mixing signal between primary and secondary carbonate (Liu et al., 2011). It has been shown that oxygen isotopes of secondary carbonate in loess and paleosol may have recorded the  $\delta^{18}\text{O}$  value

of paleo-rainwater (Han et al., 1997; Li et al., 2007b). Depending on the relative portion of primary carbonate and the geochemical difference between the secondary and primary carbonates, paleo-environmental reconstruction based on the geochemistry of bulk carbonate might be biased by the contribution of primary carbonate in different degrees (Rao et al., 2006).

Microscopic observation has confirmed the coexistence of both primary and secondary carbonates in Chinese loess (Sheng et al., 2008). Primary carbonates in Chinese loess are mainly calcite and dolomite to a lesser extent, while the secondary carbonates are mainly calcite (Li et al., 2007a; Sheng et al., 2008). Secondary dolomite only can be found in the red clay formation underlying the typical loess and paleosol layers (He et al., 2012). A small fraction of the secondary carbonate in Chinese loess is derived from land snail aragonite, which is gradually transformed into calcite in aged layers (Sheng et al., 2008).

To avoid the influence of primary carbonate, pure secondary carbonates such as rhizoliths, carbonate nodules, calcified pseudomycelia and land snail shell are preferred for paleo-environmental reconstructions (Chen et al., 1996; Han et al., 1997; Ding and Yang, 2000; Li et al., 2007b; Sheng et al., 2008). However, the occurrence of these secondary carbonates is very rare (Sheng et al., 2008). For example, carbonate nodules are mostly situated below or in the lower part of paleosols (Han et al., 1997; Ding and Yang, 2000). Recent studies even show that the carbonate nodules and rhizoliths, once regarded as having a purely secondary origin, occlude significant amounts of primary loess grains (Liu et al., 2010; Gocke et al., 2011). To obtain a continuous record of secondary carbonate, Sheng et al. (2008) proposed that carbonate in the  $<2\ \mu\text{m}$  grain size fraction is mainly of pedogenic origin because the production of pedogenic calcite is mainly in the fine grain size range and the primary carbonates with fine grain size are less resistant to pedogenic dissolution.

In this work, we attempted to establish a quick method to discriminate primary and secondary carbonate using the trace element composition of bulk carbonate. The rationale is that secondary carbonate should contain very little Mg

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