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Formation of calcareous nodules in loess–paleosol sequences: Reviews of existing models with a proposed new "per evapotranspiration model"

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ABSTRACT

Loess is a product of aeolian deposition during Quaternary glaciation cycles. Loess–paleosol sequences are rich in calcareous nodules (CNs). In the literature, two models are widely cited for the formation of CNs, namely "per descendum" and "per ascendum". However, there has been no direct testing or monitoring to support either of these contradictory models. This paper reviews a large number of multidisciplinary literature to evaluate the consistency, reliability and rationality of these two models. Three main conclusions are drawn: (1) the causative factors (variation of pH value along loess–paleosol sequence, decrease of CO_2 partial pressure, and reduction of solvent water) that are used to support the per descendum model do not completely explain the supersaturation of infiltration solution with $CaCO_3$, thereby making this model questionable; (2) the per ascendum model explains the formation of CNs along narrow horizons through upward evaporation; however, it fails to produce sporadic distributions and irregular shapes of nodules on loess slope faces and the frequent appearance of nodules around plant roots. In order to reconcile these deficiencies, we conducted an extensive field survey in various parts of Shanxi province. Based on this new set of observations, it was concluded that the "per ascendum" model can be extended to explain all occurrences of CNs. This extended model is called "per evapotranspiration".

1. Introduction

Loess (loess–paleosol sequence) is a product of aeolian deposition and soil forming processes during Quaternary glacial–interglacial cycles and is therefore an invaluable record of continental paleo-climatic and -environmental conditions (Pye, 1995; Rost, 2001; Cilek, 2001; Smalley and Marković, 2014). Loess accounts for approximately 6% of the Earth's land surface and is mainly distributed in the Central and Northwestern United States, Alaska, Argentina, Europe, China, Russia, Central Asia, and New Zealand, and sporadically in Africa and the Middle East (Gallant et al., 2014; Muhs, 2013). Containing carbonates (approximately 10%; Cilek, 2001) is one of the important characteristics of loess (Pesci, 1990). Loess possesses two types of carbonates, primary and secondary (Wen, 1989). The primary carbonates are the detrital carbonates transported from a source area (Wen, 1989) and distributed evenly in loess (Smalley, 1971), whereas the secondary carbonates are the product of dissolution–migration–recrystallization of primary carbonates during loess evolution (Smalley, 1971; Han et al., 1996; Smalley and Marković, 2014; Pesci, 1990) and are unevenly distributed in loess (Xue et al., 2011).

The geochemical characteristics, morphology and accumulation depth of secondary carbonates are used to evaluate paleoclimate (e.g., rainfall and temperature), vegetation, CO_2 partial pressure (pCO_2) and age of soil layers (Monger et al., 1991; Diaz et al., 2016; Jiang et al., 2001; Wang and Zheng, 1989). The C and O isotopic compositions of secondary carbonates are controlled by soil's CO_2 and meteoric water, respectively; thus, the ratios of these stable isotopes are considered as indicators of paleo-climate and -environment, for example, for the evolution of land vegetation types (C3/C4 ratio) and precipitation-evaporation cycles during formation of soil horizons (Cerling, 1984; Breecker et al., 2009; Huang et al., 2003; Sheng et al., 2002).

The common forms of secondary carbonates in loess-paleosol sequences include: (a) coating (Fig. 1a; on the surfaces of soil peds and voids with a few millimeters to centimeters thickness; Becze-Deàk et al., 1997; Barta, 2011; Pye, 1995), (b) hypocoating (Fig. 1b; penetrating into soil matrix around pores with a few millimeters thickness; Becze-Deàk et al., 1997; Zamanian et al., 2016), (c) pseudomycels (Fig. 1c; cotton-like accumulations of fine $CaCO_3$ needles, several microns in diameter and tens of microns in length; Kovda et al., 2009), (d) earthworm biospheroids (Fig. 1d; in biochannels or in the soil matrix,

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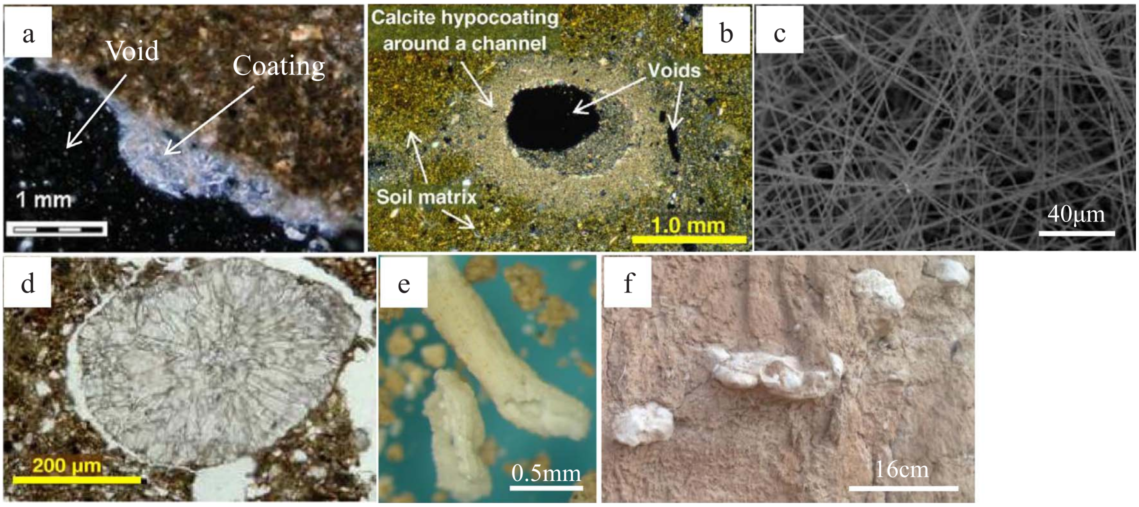

Fig. 1. Common forms of secondary carbonates: (a) coating on the surface of a void (Kovda et al., 2009); (b) hypocoating around a pore (Zamanian et al., 2016); (c) pseudomycelium (Kovda et al., 2009); (d) earthworm biospheroid (Zamanian et al., 2016); (e) calcified root cell (Barta, 2011); and (f) calcareous nodules on slope surface.

with 350–1000 µm long and 200–600 µm wide ellipsoidal shapes; Becze-Deàk et al., 1997; Barta, 2011; Zamanian et al., 2016), (e) calcified root cells (Fig. 1e; pore infillings with a diameter of several millimeters; Barta, 2011), (f) calcareous nodules (CNs) or concretions (Fig. 1f), and (g) calcrete or continuously cemented bedding (a few centimeters to a few meters; Hu et al., 2000; Alonso-Zarza and Wright, 2010). Among these, CNs are the most abundant in loess (Yang et al., 2014).

A CN is a special mineral aggregate (Teng et al., 1990) formed by impregnation and cementation of soil matrix with $CaCO_3$. Though impregnation often tends to diffuse the external boundary of CNs (Durand et al., 2010), some nodules may have sharp boundaries (Chandra et al., 2016). CNs have columnar, branching, conical, spherical, ellipsoidal, disk-like, or other irregular shapes (Fig. 2; Liu, 1966; Cao, 1983; Barta, 2011). They generally are 15–25 cm long and 5–10 cm wide. Large CNs can be up to 50 cm long and 30 cm wide, whereas the small ones are only a few millimeters. CNs appear as distinct horizons within or as dispersed across loess layers (Fig. 3; Cao, 1983).

The $CaCO_3$ content in a CN ranges between 30 and 70% (Liu, 1985). Ca originates from calcium minerals (e.g., mainly calcite and some calcium feldspar), rainwater, soluble calcium salts, exchangeable calcium ion in soil colloid minerals, groundwater, and plant tissues (Yu, 1990; Pecsi, 1990; Chen et al., 2002; Monger, 2002; Candy and Black, 2009; Cilek, 2001; Wu and Liu, 1987). Two models are widely cited in the literature for the CN formation in loess, namely, “per descendum model” and “per ascendum model” (Goudie and Pye, 1983; Monger, 2002; Zhu, 1965; Guo and Fedoroff, 1990). In the per descendum model, three factors contribute to the reprecipitation of $CaCO_3$: variation of pH along loess–paleosol sequence (Wen, 1989; Galović, 2014), decrease of pCO_2 with depth along loess–paleosol sequence (Han et al., 1995) and downward reduction of solvent water in infiltration solution (Arkley, 1963; Zhao, 2002; Gocke et al., 2012). However, mechanisms of these factors are yet to be investigated in laboratory or field. Moreover, neither of the two models explains the common appearance and sporadic distributions of CNs on loess slope surfaces (Fig. 3).

In this study, the consistency, rationality and reliability of per descendum and per ascendum models are analyzed based on a critical review of a large number of relevant literature. The causative factors (i.e., variation of pH, decrease of pCO_2, and reduction of solvent water) in the infiltration solution are discussed with regard to the per descendum model. The conditions required for the realization of the per ascendum model are discussed in the light of new observations. These discussions along with the latest observations lead to a new model for CN formation, namely, “per evapotranspiration model.”

2. Per descendum model

Fig. 4 illustrates the mechanism of CN formation according to the per descendum model. During the development and evolution of paleosol, biological activities within soil produces large amounts of CO_2, which results in an increase of pCO_2 in soil air (Gocke, et al., 2010). In the process of downward infiltration of rainwater or surface water, CO_2 in soil air is dissolved into water, resulting in an increase in acidity of infiltration solution. Calcium minerals (mainly calcite) are continuously leached by the infiltration solution, and the solution with calcium bicarbonate $Ca(HCO_3)_2$ is formed as expressed in Eq. (1):

$$CaCO_3 + CO_2 + H_2O \Leftrightarrow Ca^{2+} + 2HCO_3^- \quad (1)$$

Other sources (e.g., soluble calcium salts and exchangeable calcium ions) also contribute to the increasing Ca^{2+} concentration in the infiltration solution (Yu et al., 1990; Wu and Liu, 1987). The solution with $Ca(HCO_3)_2$ migrates downward, and the $CaCO_3$ in infiltration

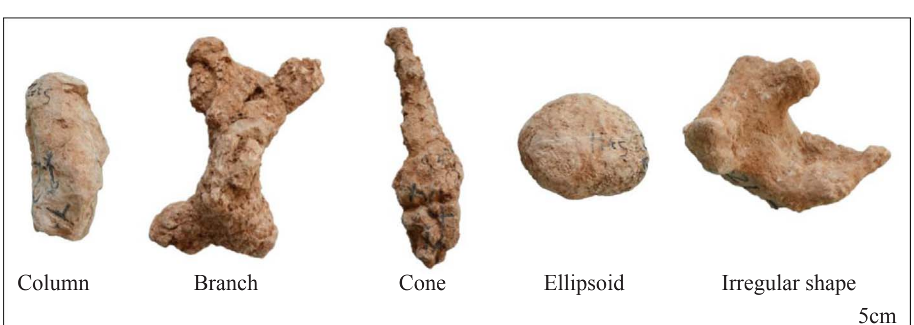

Fig. 2. Common shapes of calcareous nodules.

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