



Marine carbonate records of terrigenous input into Paleotethyan seawater: Geochemical constraints from Carboniferous limestones

Ming-Yu Zhao, Yong-Fei Zheng*

CAS Key Laboratory of Crust-Mantle Materials and Environments, School of Earth and Space Sciences, University of Science and Technology of China, Hefei 230026, China

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Abstract

A combined study of trace elements, carbon and oxygen isotopes was carried out for Carboniferous marine limestones from the Lower Yangtze platform in South China. The results provide insights into the influence of terrigenous input to Paleotethyan seawater, from which pure and impure carbonates precipitated. In terms of the correlations between the concentrations of elements with different properties, variable extents of two-component mixing are evident between seawater and terrigenous detritus. This is quantitatively dictated by such parameters as the calcite–seawater partition coefficients (D_X) and the seawater–continental upper crust partition coefficients (K_X). Water-soluble elements with high and medium K_X values, such as Na, Mg, P, V, Cr, Ni, Cu, Zn, Sr, Mo and U, are susceptible to incorporation into carbonate lattices, and thus their concentrations represent the geochemical composition of paleoseawater. In contrast, water-insoluble elements with low K_X values, such as Be, Al, Sc, Co, Ga, Cs, REE, Hf and Th, cannot be incorporated into carbonate lattices, thus providing a proxy for contributions from terrigenous material.

The Carboniferous marine limestones from a stratigraphic profile exhibit three positive and three negative $\delta^{13}\text{C}$ excursions from the late Viséan to Gzhelian. These $\delta^{13}\text{C}$ excursions generally match those of contemporaneous carbonates elsewhere in the world, which can be linked to the alternative occurrences of cold and warm paleoclimates in the Carboniferous. The lowest $\delta^{13}\text{C}$ values in the profile are generally associated with the lowest values of Y/Ho and the highest values of (Nd/Yb)_{PAAAS}, Th, Sc and insoluble residue. This indicates a significant contribution from the terrigenous material to the negative $\delta^{13}\text{C}$ excursions. The effect from the warm paleoclimate is also indicated by high chemical indices of alteration for the insoluble residue from the three intervals of high terrigenous input. On the other hand, the positive $\delta^{13}\text{C}$ excursions are associated with the decreases of terrigenous input. Therefore, the incorporation of terrigenous material into the epicontinental seawater is temporally linked to the paleoclimatic change on continental margins adjacent to the Paleotethyan seawater.

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1. INTRODUCTION

The element and isotope compositions of chemical precipitates (e.g., carbonate, phosphate and chert) in

marine sediments can record the geochemical compositions of paleoseawater. The geochemical compositions of seawater are quantitatively dictated by a number of factors such as the physicochemical property of water-soluble and water-insoluble elements, biological activity and climatic change. Thus, the temporal and spatial variations of paleoseawater element and isotope compositions provide important information about the surficial processes on Earth.

* Corresponding author.

E-mail address: yfzheng@ustc.edu.cn (Y.-F. Zheng).

Elements and isotopes with known behaviors in modern geochemical cycles are appropriate to serve as ancient geochemical proxies. In particular, the geochemical behaviors of rare earth elements and yttrium (REE + Y) in sedimentary carbonates have been extensively utilized to decipher their depositional environments. This includes the use of Y/Ho ratios, La-anomalies and (Nd/Yb)_{PAAS} ratios to detect the terrestrial influence on chemical precipitates and to distinguish the marine from terrestrial environment (e.g., Webb and Kamber, 2000; Kamber and Webb, 2001; Van Kranendonk et al., 2003; Kamber et al., 2004; Zhao et al., 2009; Delvigne et al., 2012; Zhao and Zheng, 2013), Eu-anomalies to discern hydrothermal influence on seawater (e.g., Kamber and Webb, 2001; Chen et al., 2006; Alexander et al., 2008; Delvigne et al., 2012), and Ce-anomalies to investigate the redox status of paleoseawater (e.g., German and Elderfield, 1990; Shields and Stille, 2001; Ling et al., 2011; Song et al., 2012).

In modern oceanography, the trace element compositions of marine carbonates are also widely used to reconstruct the geochemical composition of seawater and to trace the oceanic environment and climatic change. This includes the use of coral skeleton Mg/Ca, Sr/Ca and U/Ca ratios to estimate the variability of seawater surface temperatures (e.g., Shen and Dunbar, 1995; Shen et al., 1996; Wei et al., 2000; Armid et al., 2011), foraminifera V/Ca ratios to decipher the areal extent of anoxic and suboxic sediments (Hastings et al., 1996a; Morford and Emerson, 1999), and coral skeleton P/Ca ratios to estimate the phosphate concentration of seawater and to trace the upwelling (e.g., Montagna et al., 2006; LaVigne et al., 2008; Chen and Yu, 2011; Anagnostou et al., 2011). Other than REE + Y, however, trace elements in marine carbonates have rarely been used to reconstruct the composition of pre-Quaternary seawater. To the best of our knowledge, only one study used I/Ca ratios to constrain the I concentrations of Mesozoic seawater (Lu et al., 2010).

Sedimentary carbonates can host various trace elements in their crystal lattices, with concentrations dependent on the geochemical compositions of seawater and their partition coefficients between carbonate and seawater. On the other hand, fine grains of terrigenous detritus may coprecipitate with carbonate from seawater to form impure carbonates. Diagenesis can also facilitate the mixing between terrigenous detritus and carbonate through dissolution and recrystallization. These processes result in a significant variation in the trace element composition of marine carbonates, principally depending on the solubilities of these elements in surface water and on the intensity of chemical weathering of continental crust. For instance, water-insoluble elements (e.g., REE, Hf and Th) generally exhibit much higher abundances in impure carbonates than in pure carbonates (Webb and Kamber, 2000; Kamber et al., 2004; Frimmel, 2009; Zhao et al., 2009). This is because these elements are resistant to dissolution in surface water and thus inherit the composition of detrital material from weathered rocks. In contrast, water-soluble elements (e.g., Na, Mg and U) are susceptible to dissolution in surface water, providing a proxy for the composition of seawater if thermodynamic equilibrium is achieved. Therefore, the trace element

composition of marine carbonates is a useful means to trace the terrigenous input into the paleoseawater. Furthermore, to link the carbonate trace elements to paleoseawater geochemistry, it is necessary to determine which elements are diagnostic of terrigenous detritus, and how the geochemical composition of paleoseawater is recorded by pure and impure carbonates.

The carbon isotope composition of marine carbonates is one of the most useful tools in sedimentary geochemistry because nearly all of the major changes in global environments are associated with large fluctuations in carbonate $\delta^{13}\text{C}$ values (e.g., Veizer et al., 1999; Zachos et al., 2001). For instance, positive $\delta^{13}\text{C}$ excursions of as much as 4–7‰ have been revealed for Paleozoic carbonates and thus linked to the glacial events in this period (Bruckschen et al., 1999; Kump et al., 1999; Saltzman et al., 2000). In this regard, a combined study of carbon isotopes with trace elements in marine carbonates is capable of providing robust constraints on paleoclimatic changes. However, only a few studies (Fio et al., 2010; Loope et al., 2013) have combined these two different types of geochemical variables to decipher their paleoenvironmental significance.

In this paper, we present a combined study of major-trace elements and carbon–oxygen isotopes in the marine limestones of Carboniferous age from the Lower Yangtze platform in South China. The results provide insights into the two-component mixing between terrigenous detritus and paleoseawater, shedding light on the reconstruction of sedimentary environment in the geological history. By doing so, we have established geochemical parameters to trace the changes of terrigenous input and have linked them to paleoclimatic changes.

2. GEOLOGICAL SETTING

The South China Block was located near the equator on the eastern edge of the Paleotethyan ocean during the Carboniferous (Qie et al., 2011; Zheng et al., 2013). It is primarily composed of two landmasses of Yangtze and Cathaysia (Fig. 1A), with small intracontinental basins in the southwest but a unified platform in the Lower Yangtze area with carbonate and detrital sediments in the northeast. The study area is located in the central part of the Lower Yangtze platform that was connected to the Paleotethyan ocean during the Carboniferous (Qie et al., 2011; Zheng et al., 2013). The Carboniferous sequence in this region is predominated by limestones (Lin et al., 2002; Sheng et al., 2013).

The present study focused on the Carboniferous marine limestones in the Lower Yangtze platform. The selected sampling profile is located at East Fenghuangshan (N31°37'46", E117°51'27") in the suburb of Chaohe city, Anhui province (Fig. 1B). The East Fenghuangshan profile contains seven sedimentary formations (Fig. 2), including, in ascending order, the Wutong, Jinling, Gaolishan, Hezhou, Laohudong, Huanglong and Chuanshan formations (Feng et al., 1998; Lin et al., 2002). The formations found here are comparable with Carboniferous sequences found elsewhere in South China (Feng et al., 1998; Lin et al., 2002). The study profile begins with the Leigutai member

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