



## Carbonate composition and its impact on fluvial geochemistry in the NE Tibetan Plateau region



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### ABSTRACT

Using co-variations of Sr/Ca and Mg/Ca, we examined the carbonate compositions of various bedrocks (silicate and carbonate rocks) and sediments (eolian and fluvial sediments, sand, and topsoil) found in the NE Tibetan Plateau (TP) region. A combined carbonate composition dataset based on our results and other reported data shows that bedrock carbonate composition on the NE TP displays a much broader range of Sr/Ca and Mg/Ca ratios than restricted source carbonate endmembers reported upon in previous studies. This has clear implications for modern weathering studies in addition to paleo-reconstructions in this tectonically active and climatically variable area during the Late Cenozoic. Bedrock carbonate compositions are characterized by disseminated carbonates with higher Sr/Ca and Mg/Ca ratios, and sedimentary carbonates (mostly marine) with lower Sr/Ca, but variable Mg/Ca, ratios. The mostly authigenic carbonates found in sediments show similar trends, with a gradient  $\sim 0.97\text{--}1.00$  in a plot of  $\log(\text{Sr}/\text{Ca})$  versus  $\log(\text{Mg}/\text{Ca})$ , suggesting that 'calcite precipitation' processes – *i.e.* the sources of the dissolved cations in the water – control their chemistry. Based on observations and modeling, we conclude that the mixing of authigenic and bedrock carbonate endmembers, plus the incongruent dissolution of bedrock carbonates, accounts for the bulk carbonate composition of sediments (*e.g.* loess, sand and topsoil). A comparison of bedrock and sedimentary carbonate composition with reported fluvial water data in the NE TP suggests that weathering of carbonates in terrigenous sediments, rather than in bedrock, is mostly responsible for the changes in fluvial Sr, Mg and Ca compositions. Our study suggests that interactions between carbonates and water occur widely during the exposure, transport and deposition of sediments, significantly modifying regional carbonate compositions and fluvial geochemistry.

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### 1. Introduction

Carbonates in bedrock and sediments can play a unique role both in the study of modern chemical processes and in paleo-reconstructions (*e.g.* Gaillardet et al., 1999; Dettman et al., 2003; Han and Liu, 2004; Yokoo et al., 2004; Wu et al., 2005; Fan et al., 2007; Jin et al., 2011; Zhang et al., 2013a; Li and Li, 2014). Soil carbonates, as typical products of the interactions between carbonates and water in eolian deposits or floodplain environments, provide a powerful tool for paleo-reconstructions (*e.g.* Cerling, 1984; Dettman et al., 2003). Proxies derived from soil carbonates, *e.g.*  $^{87}\text{Sr}/^{86}\text{Sr}$ ,  $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$ , carbonate-clumped isotopes and trace elements, can be extensively used to explore paleoclimate, paleoenvironment and paleoelevation (*e.g.* Cerling et al., 1993; Rowley and Garzzone, 2007; Quade et al., 1989, 1997, 2011, 2013; Li and Li, 2014). Unfortunately, such paleosol-based

reconstructions are always hampered by the presence of detrital carbonates in sediments (Fan et al., 2007; Li et al., 2013). Isolating the authigenic and detrital carbonates in sediments has therefore long been a challenge.

Mn/Ca ratios in bulk carbonate have been proposed as discriminatory indicators of detrital carbonate signatures in Chinese loess–paleosol sequences, where a binary mixing between secondary carbonate with low Mn/Ca and Mg/Ca ratios and detrital carbonate with high Mn/Ca and Mg/Ca ratios is found (Li et al., 2013). However, an abundance of Mn oxide coating during soil development and possible Mn oxide dissolution by weak acetic acid (Chester and Hughes, 1967; Gourlan et al., 2010) complicates the valence and source of acid-exacted Mn, and thus limits the wider application of Mn/Ca ratios to carbonate analysis. Sr and Mg partition coefficients are  $<1$  in carbonates, and the co-variation of both elements (as opposed to Mn) is generally a more robust tool for describing the interactions between carbonates and water (*e.g.* Plummer and Mackenzie, 1974; Fairchild et al., 1994; Reeve and Perry, 1994; Galy et al., 1999; Tipper et al., 2008). Sinclair

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(2011) and Sinclair et al. (2012) demonstrated that an analysis of Sr/Ca versus Mg/Ca in water and carbonate, is ideal for diagnosing any interactions between carbonates and water in karst and speleothems. These relations can, in turn, be expressed using mathematical models. Li and Li (2014) followed this approach to develop a novel proxy for paleo-precipitation in authigenic microcodium in the Holocene paleosols of the Chinese Loess Plateau (CLP). Such a quantitative approach implies the possibility of distinguishing authigenic and detrital carbonate in sediments. Nevertheless, such an approach requires a well-defined composition of detrital carbonate in order for the proxy to be applied to carbonate compositions retrieved from terrigenous sediments.

The thick eolian and fluvial–lacustrine deposits on the NE TP provide fertile ground for an exploration of the interactions between carbonate and water, with regard both to paleo-reconstructions and studies of modern chemical weathering processes. This is because these deposits are embedded with abundant paleosol carbonate and thus present excellent Cenozoic climatic and topographic archives (e.g. Fan et al., 2007; Li and Li, 2014). Further, inversion model examining stream geochemistry in the headwaters of the Yellow River draining into the NE TP (Fig. 1) suggests that carbonates and evaporite dissolution dominate the fluvial water bulk alkalinity budget (Wu et al., 2005). Detailed investigations of the fluvial geochemistry of rivers draining into Qinghai Lake (Fig. 1) have highlighted the importance of carbonate weathering and eolian dust *vis-à-vis* seasonal hydrochemical variations (Jin et al., 2011; Zhang et al., 2013a). To date, except for several localized investigations of carbonate compositions conducted during studies of modern

weathering and paleo-reconstructions, variations in carbonate composition and its variability in bedrock and related sediments on the NE TP remain undefined, hindering any regional exploration of the interactions between carbonates and water. In this study, in combination with published carbonate data (e.g. Liu et al., 2002; Yokoo et al., 2004; Jin et al., 2010a; Li and Li, 2014), we have compiled a comprehensive carbonate composition dataset for the NE TP covering bedrock, loess–paleosols, red clays, modern topsoils, sand and fluvial sediments, aiming: 1) to reveal the carbonate composition of various bedrocks and sediments; 2) to provide a database with a better definition of authigenic and detrital carbonates in the sediments; and 3) to look further at the link between carbonate weathering and fluvial geochemistry in this region.

2. Materials and methods

2.1. Sampling and data collection

Rocks exposed in the NE TP range from Precambrian, to Early Cretaceous, to Quaternary (Yuan et al., 2013; Jian et al., 2013). The composition of exposed rocks varies, but intermediate and acid crystalline and metamorphic rocks predominate (Jian et al., 2013). They are mainly grey gneiss, siliciclastic rocks, dolostone, quartzite, phyllite, marble, carbonatite, peridotite, serpentinite, augite peridotite, diorite and granite (Wu et al., 2005; Jin et al., 2010a; Li et al., 2010). A total of 162 bedrock samples collected were taken from freshly-exposed rocks in the

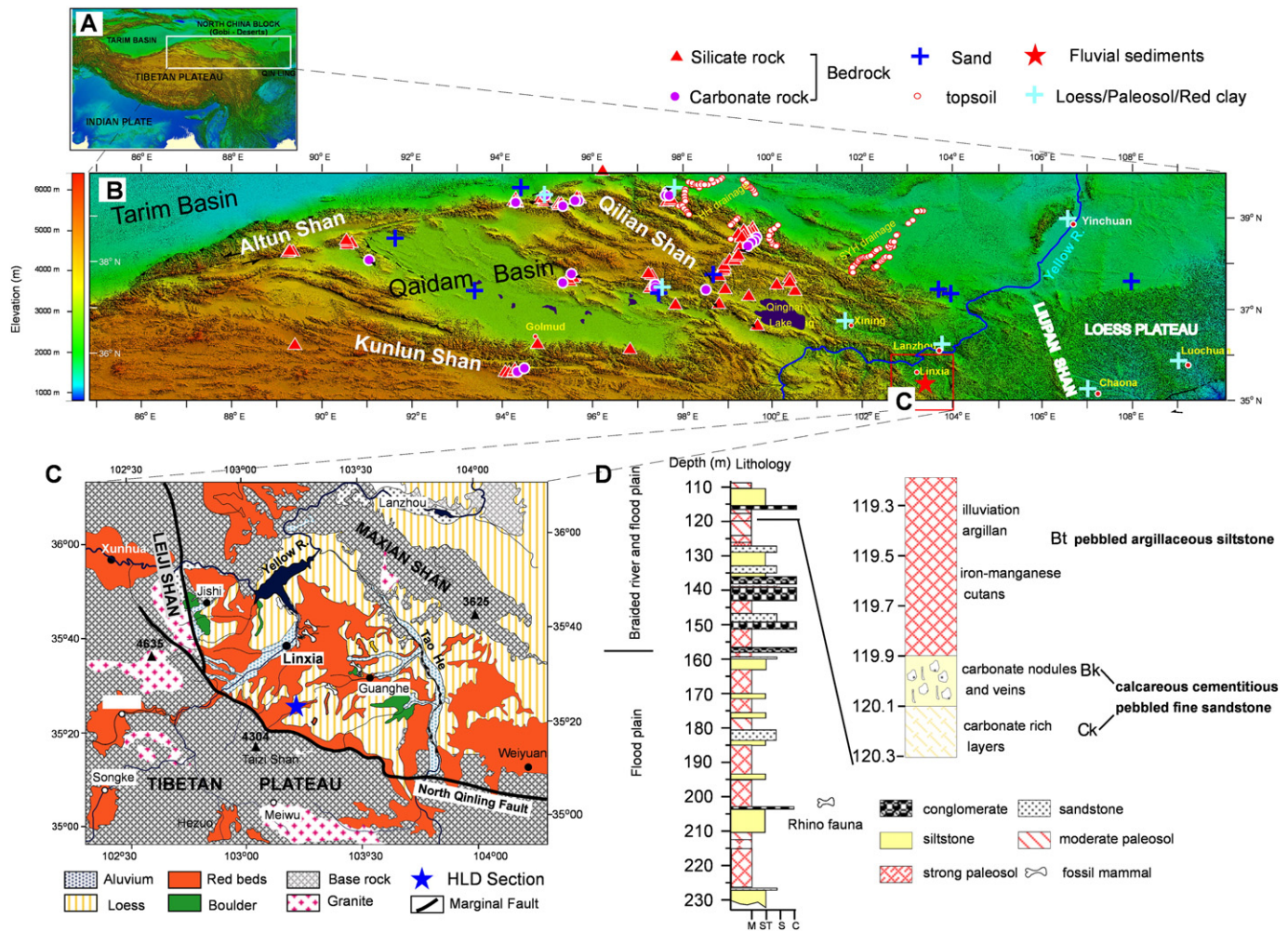


Fig. 1. (A–B) DEM presentation of the geomorphology of the NE TP, showing the sampling sites in this study and locations of reported carbonate composition data in other studies; (C) simplified surface geology map of the Linxia Basin and adjacent regions showing the location of the studied HLD Section (redrawn from Fan et al., 2006); (D) Lithologic stratigraphy of the Lower HLD Section and its representative paleosol profile.

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