Article

Weathering intensity response to climate change on different timescales: A record of Rb/Sr ratios from Chaonaqiu Lake sediments, western Chinese Loess Plateau

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Abstract: The Rb/Sr ratio of lake sediments has been widely adopted as an indicator of weathering intensity in studies of past climate change, but the geochemical significance of this ratio varies with timescale. Here we present Rb/Sr data for the last 300 years for sediments collected from Chaonaqiu Lake in the Liupan Mountains of the western Chinese Loess Plateau as a decadal-scale record of weathering intensity. To validate the application of this weathering proxy, we correlated the record with those of other major elements, rock-forming minerals, and paleoclimatic proxies. We found that Rb/Sr ratios are influenced mainly by Sr activity within the lake catchment (where Sr is likely sourced from albite). In addition, higher (lower) Rb/Sr ratios of bulk sediments from Chaonaqiu Lake are correlated with lower (higher) fractions of terrigenous detritus (SiO_2, Ti, K_2O, Al_2O_3, and Na_2O). Lake bulk sediment Rb/Sr ratios are dominated by the input of terrigenous detritus over decadal timescales. Our data show that physical and chemical weathering in Chaonaqiu Lake catchment have opposing influences on bulk sediment Rb/Sr ratios, competing to dominate these ratios of lake sediments over different timescales, with ratios reflecting the relative importance of the two types of weathering. We infer that the Rb/Sr ratios of Chaonaqiu Lake sediments reflect climatic signals of precipitation and temperature over long timescales and of precipitation over short timescales.

Keywords: Rb/Sr ratios; Weathering intensity; Decadal timescales; Chaonaqiu Lake

1. Introduction

Numerous proxies based on ratios of elements have been used as indicators of past environmental and climatic change over various timescales [1–7]. For instance, Rb/Sr ratios have been utilized for reconstructing the degree of chemical weathering and changes in climatic conditions over geological timescales [8–11]. For example, Rb/Sr ratios in Chinese loess/paleosoil profiles have been used as an indicator of Asian summer monsoon intensity, as the correlation between this ratio and the degree of weathering indicates a correlation with monsoonal precipitation over long timescales [7,12–15]. Rb/Sr ratios of lake sediments have been increasingly applied in environmental studies, often as indicators of weathering intensity in catchments, where lower ratios are correlated with higher degrees of weathering [2–4,10,16,17]. For example, Liu et al. [10] applied Rb/Sr ratios in reconstructing chemical weathering variability over the past 1200 years in the Gonghai Lake catchment, north China. Rb/Sr ratios in lake sediments have also been adopted as an indicator of the evolution of the Central Asia westerly winds [18] and the Asian summer monsoon [10,19,20].

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However, the factors affecting the material load transported into lakes can be complex and include local climate, the chemical and physical properties of bedrock, vegetation cover. Such factors may add complexity to the environmental interpretation of Rb/Sr ratios of lake sediments. For example, with strong physical loads in a catchment, although accompanying chemical weathering may be enhanced, lake sediment chemical compositions may be dominated by terrigenous detritus rather than chemical/biogenic deposition. Rb/Sr ratios of lake bulk sediment would in such a case serve primarily as an indicator of physical rather than chemical weathering intensity [21–24]. Rb/Sr ratios have also been regarded as an indicator of glacial conditions and lake-water depth. For example, low (high) Rb/Sr ratios of sediments of the glacial Kalakuli Lake have been interpreted to indicate glacier advance (retreat) [25], and Rb/Sr ratios have been used as a proxy for water depth in lacustrine deposits in the Qaidam Basin [26]. It follows that an understanding of the controls on Rb/Sr ratios and their limitations is fundamental to their application and interpretation.

Chaonaqiu Lake is located in the Liupan Mountains on the western Chinese Loess Plateau in a zone that is transitional between semi-arid and semi-humid climatic conditions on the margin of the area affected by the modern Asian summer monsoon. An understanding of material transport from catchment to lake is critical in evaluating and predicting ecological and environmental changes, clarifying the environmental interpretation and significance of limnological indices, and elucidating weathering processes. Studies of Rb/Sr ratios of lake sediments may help to address these issues. Sediment cores recovered from Chaonaqiu Lake have yielded continuous varve sequences at high resolution with well-constrained chronology, and past changes in climate and vegetation have been reconstructed in detail on the basis of proxies such as total organic carbon (TOC) content [27], nitrogen stable isotope ratios ($\delta^{15}N$; Chen et al., 2018), n-alkanes [28], carbonate [29] and pollen records [30,31]. Recently, Zhang et al. [32] suggested that Rb/Sr ratios in Chaonaqiu Lake sediments may be dominated by chemical weathering intensity on centennial–millennial scales, although the exact weathering processes were not discussed. This study focuses on the geochemical significance of Rb/Sr ratios in Chaonaqiu Lake sediments on decadal timescales, based on the characteristics, geographical location, and significance of multiple proxies, and considers the relevant weathering process on different timescales.

2. Materials and methods

2.1. Setting and materials

Chaonaqiu Lake (latitude 35°16′N, longitude 106°19′E, altitude 2430 m a.s.l.) is located in the Liupan Mountains, ~30 km to the northeast of Zhuanglang County (Fig. 1a). It has a surface area of 0.02 km^2 with a maximum water depth of 10 m. Chaonaqiu Lake is a alpine-barrier freshwater lake with a seasonal outflow on its western margin (Fig. 1b), the lake water is supplied mainly by precipitation. The bedrock of the catchment basin of the lake is red sandstone. The lake waters have a pH of 7.83 and a salinity of 0.17 g L^{-1} [33], and total P and N concentrations of 40.2 and 1096.8 μg L^{-1}, respectively [34]. Meteorological records for 1960–2007 from the nearby Zhuanglang meteorological station indicate a mean annual temperature (MAT) of 8.1°C, with mean temperatures for January and July of −5.2°C and 20.1°C, respectively (Fig. 1c). The mean annual precipitation is 513 mm, with ~70% falling in summer (June–September; Fig. 1c).

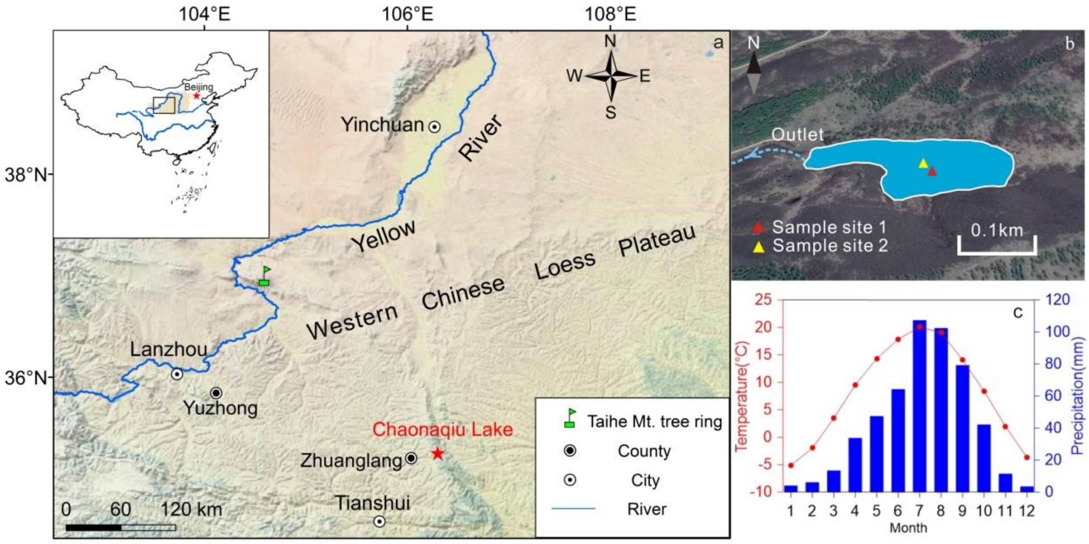

Figure 1. Overview of the study site. (a) Location of the study area on the western Chinese Loess Plateau, and other sites mentioned in the text. (b) Aerial view of Chaonaqiu Lake and sampling locations. (c) Yearly climatic trends for the Zhuanglang meteorological station, showing monthly mean temperature (red) and precipitation (blue) for 1960–2007.

In September 2012, four surface sediment cores, designated CNQ12-1, CNQ12-2, CNQ12-3 and CNQ12-4, were retrieved from the centre of Chaonaqiu Lake using a gravity corer (UWITEC, Austria). Cores CNQ12-1 (~73 cm) and CNQ12-2 (~130 cm) were retrieved from Site 1, and cores CNQ12-3 (~70 cm) and CNQ12-4 (~138 cm) were retrieved from Site 2 (Fig. 1b). Sediment profiles were undisturbed, with a clear interface between sediment and water. Core CNQ12-1 was subsampled in the field at an interval of 1 cm to quantify the core mass depth for constructing an accurate chronological model. Core CNQ12-4 was subsampled in the laboratory at an interval of 1 cm. The core CNQ12-1 wae used for this study.

2.2. Methods and analysis

For cores CNQ12-1 and CNQ12-4, ^{137}Cs and $^{210}Pb_{ex}$ contents were determined by HpGe (GWL-250-15 detector; Ortec, USA) high-resolution gamma spectrometry, with an experimental error of <10% and a detection limit of 0.1 Bq kg^{-1} at a 99% confidence level [35]. Thirty-seven subsamples from core CNQ12-1 were selected for traditional XRF analysis by X-ray fluorescence spectroscopy (XRF; PW440, Axios advanced, Netherlands; [36]). For this, ~1 g of dried sample powder was calcined in a muffle furnace at 550°C to remove organic matter, and 0.6 g subsamples mixed with 6 g dry lithium tetraborate ($Li_2B_4O_7$) for fusion to glass in a platinum crucible at 1000°C. Calibrations involved four Chinese soil samples (GSS1~4) with analytical uncertainties of ±3% for major elements. Twenty-six representative sediment samples from core CNQ12-1 and one surface soil sample from the catchment were selected for X-ray diffraction (XRD) analysis (details see [27]) to obtain mineral assemblages and elemental geochemical characteristics of sediments. All analyses were undertaken at the Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, China.

3. Results

3.1. Chronology

The ^{137}Cs profiles for cores CNQ12-1 and CNQ12-4 display unimodal distributions with marked ^{137}Cs peaks (details see [27]), which similar to those of global atmospheric ^{137}Cs deposition [23, 37–39]. This establishes the reliability of the 1964 time-marker (all dates reported here refer to years AD) in core CNQ12-1. The $^{210}Pb_{ex}$ profiles for the two cores not only show differences in absolute values, but also display clear subordinate

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fluctuations superimposed on long-term logarithmic trends (details see [27]), which may indicate that biological activity had a leaching effect on $^{210}Pb_{ex}$ as observed in Lake Chenghai, Yunnan Province [40]. Considering the possible influence of such leaching on the accuracy of $^{210}Pb_{ex}$ age models, we chose to use ^{137}Cs to establish the chronology of core CNQ12-1, based on a constant mass accumulation rate (MAR) of 0.0852 $g{\cdot}cm^{-2}{\cdot}yr^{-1}$ over the period 1743–2012 (details see [27]). The ^{137}Cs–^{210}Pb ages of Guo et al. [31] and Chen et al. [34] from Chaonaqiu Lake are also plotted in Fig. 2 and correlate well with those of our dating model (Fig. 2). In addition, the ^{14}C age of 500 cal yr BP occurs at 100 cm depth [31] and 620 cal yr BP at 162 cm depth in Chaonaqiu Lake [41,42], consistent with our ^{137}Cs age model. The close correspondence of these various age constraints confirms that our presented chronology is reliable.

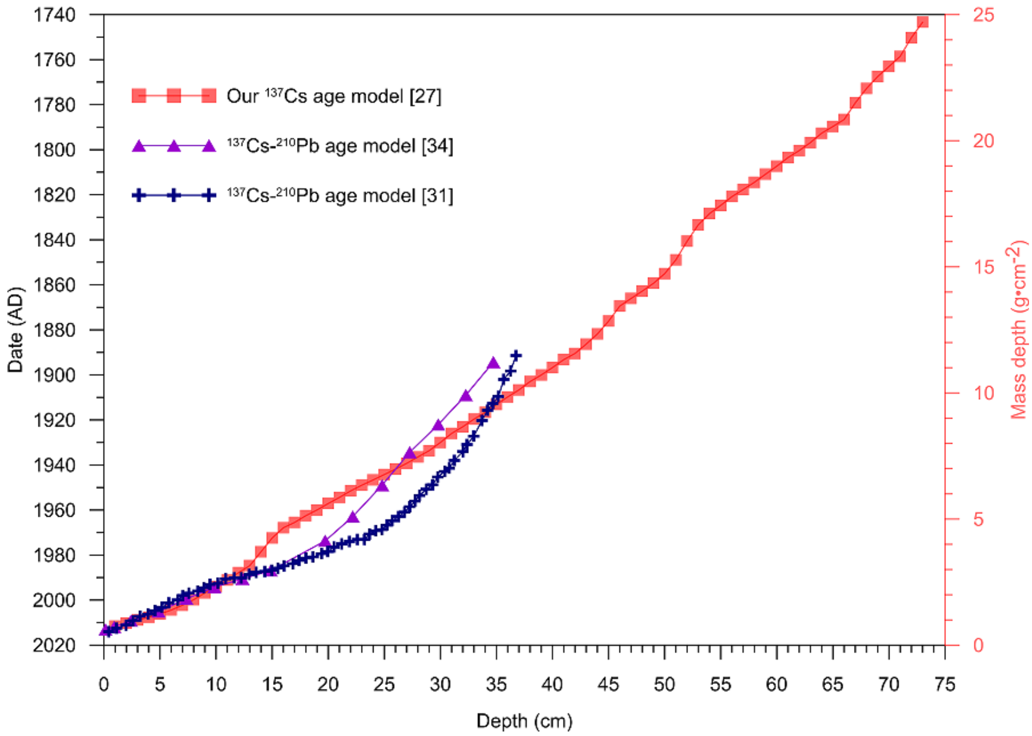

Figure 2. Chronologies for cores CNQ12-1 [27], GS14A [34], and that reported by Guo et al. [31] from Chaonaqiu Lake.

3.2. Elemental geochemical and mineral assemblages

The Rb contents of core CNQ12-1 are in the range of 176.1–246.5 ppm with an average of 200.8 ppm, while Sr contents are 175.6–236.6 ppm with an average of 203.58 ppm. Rb/Sr ratios range from 0.78 to 1.29 with an average of 0.99 (Fig. 3). Rb and Sr display generally opposing trends at 1–15 cm and 55–73 cm depths in the profile, but are consistent at 15–55 cm. Rb/Sr ratios are negatively correlated with Sr (Pearson correlation coefficients r = -0.703; Table 1) and positively correlated with Rb (Pearson correlation coefficients r = 0.623; Table 1) contents (Fig. 3). This suggests that the variation of Rb/Sr ratios depends primarily on Sr elements activity during weathering.

Table 1. Pearson correlation coefficients between species in sediments from core CNQ12-1.

	Ti	Al_2O_3	K_2O	Na_2O	Rb	Sr	Rb/Sr
SiO_2	**0.686****	**0.134**	**0.163**	**0.817****	*-0.095*	**0.340***	*-0.303*
Ti		**0.659****	**0.630****	**0.409***	**0.449****	**0.210**	**0.153**
Al_2O_3			**0.896****	*-0.258*	**0.919****	**0.034**	**0.633****
K_2O				*-0.094*	**0.920****	**0.385***	**0.353***
Na_2O					*-0.407**	**0.543****	*-0.698***
Rb						**0.111**	**0.623****
Sr							*-0.703***

Note: *Significant at $p = 0.05$ (Pearson correlation; 2-tailed). **Significant at $p = 0.01$ (Pearson correlation; 2- tailed). Bold and italic fonts indicate positive and negative correlations, respectively. SiO_2 data are from [27].

Overall, element and oxide contents are strongly correlated in core CNQ12-1 (Fig. 3; Table 1). Rb contents are positively correlated with those of Ti, K_2O, and Al_2O_3, especially with K_2O and Al_2O_3 ($r > 0.90$; Table 1); Sr contents are significantly and positively correlated with SiO_2, K_2O, and Na_2O contents; and Rb/Sr ratios are likewise with Al_2O_3, K_2O, and Na_2O contents (Table 1). Si, Al, K, and Ti contents of lake sediments are generally considered to be associated with the influx of exogenous clastic materials [43,44] and are positively correlated with each other in Chaonaqiu Lake sediments (Fig. 3; Table 1), reflecting their similar characteristics, sources, and transport and sedimentation processes [45].

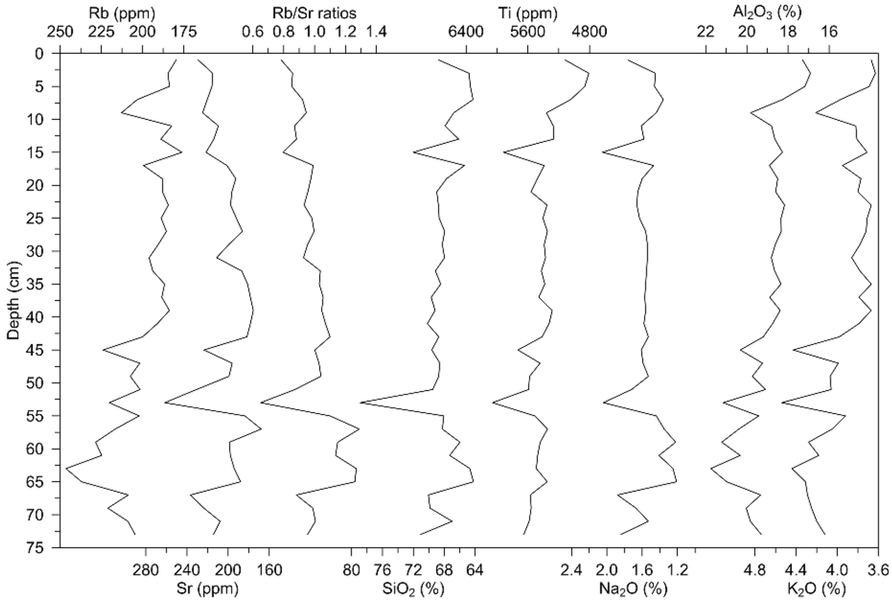

Figure 3. XRF analysis results for core CNQ12-1. SiO_2 data are from [27].

Our XRD analyses reveal that core CNQ12-1 has a mineralogic composition that is dominated by *quartz*, *biotite*, *albite*, *chlorite*, and *calcite* (Fig. 4). Except for calcite, the close correspondence of mineralogic compositions and primary peaks between surface soils and lake sediments implies that sediment in Lake Chaonaqiu is sourced primarily from terrestrial clastic material. The element compositions of these minerals in sediment are similar to the elements listed in Table 1, indicating that these elements have similar characteristics, sources, transportation and sedimentation processes [45].

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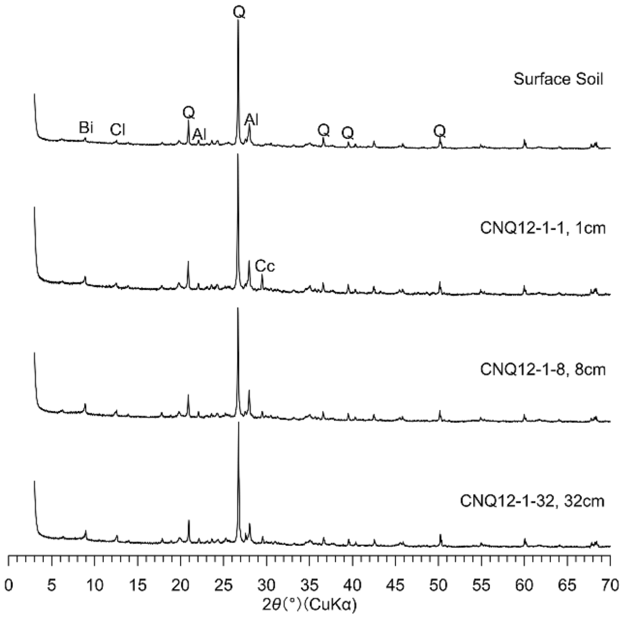

Figure 4. Results of XRD analysis for core CNQ12-1 and surface soils. Q – Quartz; Bi – Biotite; Al – Albite; Cl–Chlorite; Cc – Calcite.

4. Discussion

4.1. Rb/Sr ratios of sediments and physical weathering

Under natural conditions, Sr-bearing minerals in lake sediments are derived from two sources: terrigenous detritus (directly brought in by physical erosion) and dissolved materials (produced by chemical weathering). If the Rb/Sr ratios in bulk sediments are controlled mainly by physical loads, then more intense physical weathering should be correlated with higher contents of terrigenous detritus, with lower Rb/Sr ratios [21,22]. In contrast, if the Rb/Sr ratios in bulk sediments are mainly determined by chemical weathering, more intense chemical weathering is expected to be correlated with higher Sr contents and lower Rb/Sr ratios [3,4,8,10,16,17]. Therefore, Rb/Sr ratios of lake sediments reflect the competition between (i.e., the relative importance of) physical weathering and chemical weathering [23]. Where Sr in sediments occurs mainly in coarse terrigenous clastic minerals (e.g., feldspar minerals), the Rb/Sr ratio reflects the intensity of physical transport within the basin [3,21]. The minerals in lake sediments and surface soil are dominated by *quartz*, *albite*, *biotite*, and *chlorite* (Fig. 4), with the *calcite* in sediments being authigenic [29], and the trends in Sr content are consistent with those of *albite* (Fig. 5), indicating that Sr is likely enriched in *albite*. Rb/Sr ratios are also strongly correlated with the content of the terrigenous fraction (SiO_2, Ti, K_2O, Al_2O_3, and Na_2O; Fig. 3), implying that the lake sediments are likely dominated by terrigenous detritus and therefore that physical weathering predominated within the catchment of the lake [21–23].

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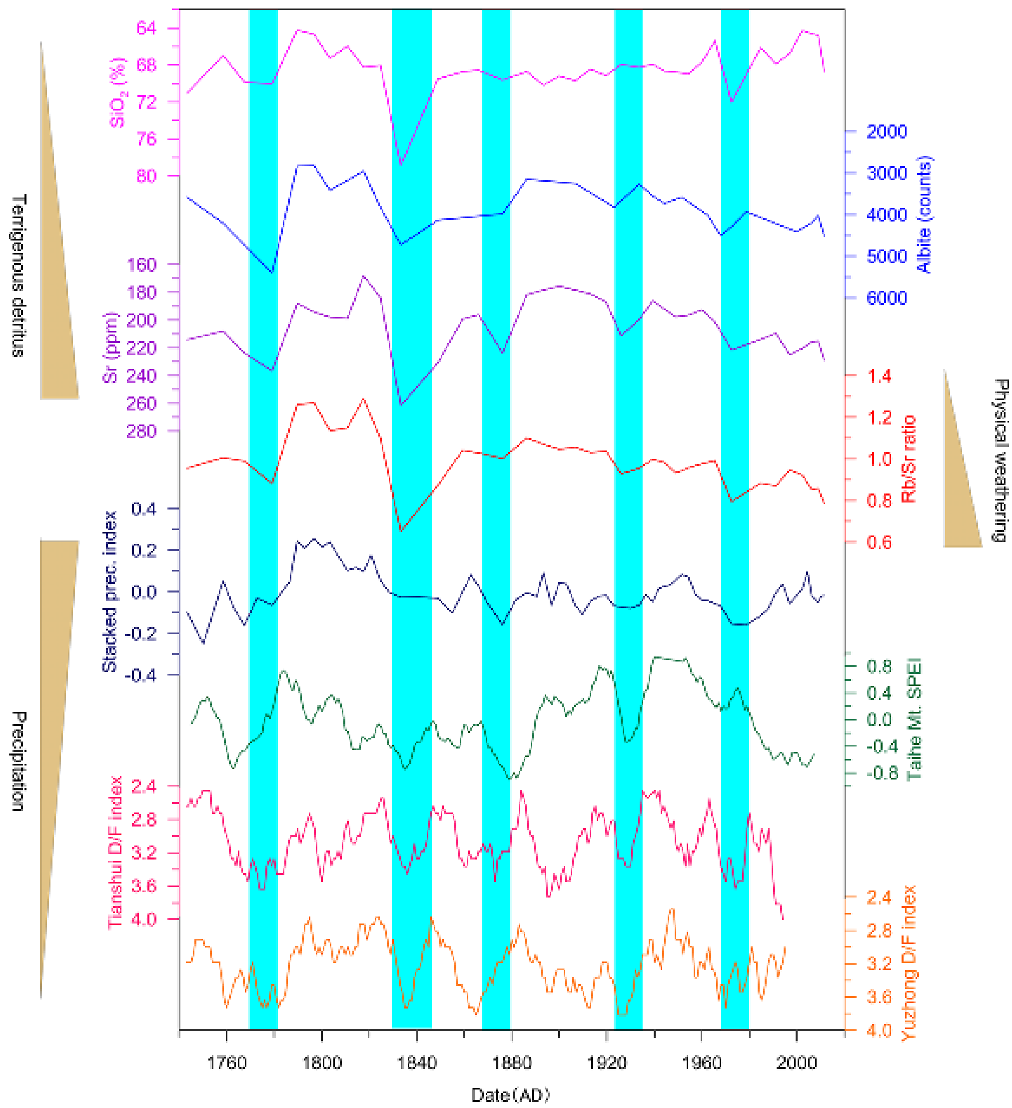

Figure 5. Comparison of the Rb/Sr ratio with other indices of core CNQ12-1 in Chaonaqiu Lake and other precipitation records in neighboring regions. The stacked precipitation index is the average of the normalized total organic carbon (TOC), detrended C/N ratios and $\delta^{13}C_{org}$ (see [27] for details). The smoothed curve of Taihe Mt. SPEI (Standardised Precipitation Evapotranspiration Index, higher value indicates a wetter climate condition; [46]), and the D/F index [47–49] are all 11-year running means. The cyan shadings indicate dry intervals.

In our previous study, we found that variations in $\delta^{13}C_{org}$ values, C/N ratios, and TOC contents of sediments in Chaonaqiu Lake are associated mainly with local variations in precipitation [27]. Fig. 5 shows the temporal trends in Rb/Sr ratios and other indices (e.g., SiO_2, *albite*) in core CNQ12-1 are generally similar to those of precipitation indices (e.g., C/N ratios, TOC and $\delta^{13}C_{org}$; [27]) and records from Taihe Mt. tree rings [46], Drought/Flood (D/F) index [47–49] in neighboring regions on decadal scales, more precipitation is correlated with higher Rb/Sr ratios, and less precipitation is correlated with lower Rb/Sr ratios. These patterns likely reflect the physical and chemical properties of the lake catchment, as well as local-scale environmental processes. Although the mean annual precipitation (513 mm) in the Chaonaqiu Lake basin reflects a semi-humid climatic environment, the precipitation distribution is uneven (~70% occurs during summer, often in heavy rainstorms), as the climate zonation of the Loess Plateau has not changed [50,51]. Therefore, regardless of whether total annual precipitation increases or decreases, local precipitation characteristics do not change. During wetter intervals, dense vegetation cover in the catchment stabilizes soil and also reduces the sediment-carrying capacity of runoff, resulting in less terrigenous detritus input into the lake and higher Rb/Sr ratios of lake bulk sediments. During drier periods, the sparse vegetation cover results in more soil being exposed, producing a less cohesive soil such that even low-intensity rainfall may generate a higher runoff sediment-carrying capacity with increased terrigenous detritus transport and lower Rb/Sr ratios of lake sediments.

4.2. Response of weathering intensity to climate change on different timescales.

As physical weathering decreases in the Chaonaqiu Lake catchment over short (decadal) timescales, the chemical weathering intensity likely increases with precipitation, but water–rock reactions may weaken with less terrigenous material being involved. This implies that during wet intervals, although the intensity of chemical weathering increases, the amounts of Sr-bearing minerals chemically/biogenically deposited in sediments would decrease, not increase. Increased chemical weathering would thus be correlated with higher Rb/Sr ratios over short timescales. The Rb/Sr ratios of lake sediments are affected mainly by terrigenous detritus input, reflecting environmental conditions on short timescales [3]. This effect is recorded with high resolution because the terrigenous detritus sinks rapidly to the lake bottom.

In comparison, over long timescales (millennium scales), chemical weathering is likely to be more important than physical weathering in regulating long-term trends in sediment Rb/Sr ratios. Our previous study found that the carbonate in the Chaonaqiu Lake sediments is authigenic and derived mainly from *calcite* [29]. The rate of autogenic/biogenic carbonate deposition in Chaonaqiu Lake is influenced by temperature-controlled water salinity, and long-term records of Sr contents (and hence Rb/Sr ratios) in lake sediments should include climatic signals of precipitation and temperature. Lake sediment Rb/Sr ratios are dominated by chemical/biogenic deposits [3], reflecting environmental trends with low resolution over long timescales, because chemical weathering is a continuous and gradual process affected by both climatic factors and the lake-water hydrochemical environment. Therefore, with respect to the relationship between precipitation and Rb/Sr ratios, a "higher precipitation ~ lower Rb/Sr ratio" relationship thus applies over long timescales, and a "higher precipitation ~ higher Rb/Sr ratio" over decadal timescales (Fig. 5). Hence, interpretation of the geochemical significance of Rb/Sr ratios of lake sediments must consider the timescale involved.

5. Conclusions

The geochemical significance of Rb/Sr ratios in bulk sediments of Chaonaqiu Lake was considered over different timescales. Rb/Sr ratios are controlled predominantly by Sr activity within the lake catchment, where Sr is likely sourced from *albite*. Decadal variations in Rb/Sr ratios of the lake sediments are strongly correlated with the content of the terrigenous fraction (SiO_2, Ti, K_2O, Al_2O_3, and Na_2O), suggesting that the Rb/Sr ratios are dominated by terrigenous detritus, reflecting variations in precipitation in the lake catchment over short timescales. However, long-term records of Rb/Sr ratios of the lake sediments are influenced by both precipitation and temperature. Therefore, physical weathering and chemical weathering in the Chaonaqiu Lake catchment have opposing influences on sediment Rb/Sr ratio and may compete on different timescales as the dominant control. Investigation of temporal trends in Rb/Sr ratios is necessary, especially on short timescales, before their application in studies of catchment weathering intensity.

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Conflicts of Interest: The authors declare no conflicts of interest.

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