

Article

Relative Sea-Level Fluctuations during Rhuddanian–Aeronian Transition and Its Implication for Shale Gas Sweet Spot Forming: A Case Study of Luzhou Area in the Southern Sichuan Basin, SW China

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Abstract: Most scholars believe that transgression events can contribute positively to organic matter enrichment and shale gas sweet spot development, while whether or not regression events are conducive to shale gas sweet spot development remains to be further discussed. Variations in organic carbon content (TOC), and major and trace elements at the Rhuddanian–Aeronian stage in the Luzhou area, the southern margin of Sichuan Basin, were analyzed in this paper. We discuss differences in paleoenvironment organic matter enrichment and shale sweet spot development during transgression and regression. A transgressive system tract (TST) occurred during the early Rhuddanian stage, while early highstand system tracts (Ehst-1 and Ehst-2) occurred during the late Rhuddanian stage and Aeronian stage, and a late highstand system tract (LHST) developed during the late Aeronian stage. A rise in sea level during the TST in the upper Yangtze resulted in an anoxic environment, where a continuous upwelling current brought about a large number of nutrients in the seawater, significantly increasing paleoproductivity. Strong tectonic subsidence, weak chemical weathering, and a rising sea level together led to a low terrigenous debris supply in the catchment area. Therefore, paleoproductivity and redox conditions were the primary controlling factors of organic matter enrichment at the TST stage, with a clastic supply of secondary importance. With the advance of the Guangxi orogeny, the organic matter enrichment at the EHST-1, EHST-2, and LHST stages was mainly controlled by redox conditions and debris supply. A comparison of the key physical parameters and geochemical indicators of shale reservoirs from these four system tracts suggests that two shale sweet spot types (type I and II) were developed during the Rhuddanian–Aeronian stage, occurring in the TST and EHST-2 stages. High TOC and high microcrystalline quartz content are key to developing type I sweet spots, while enhanced anoxic conditions in the bottom water caused by ephemeral, small-scale sea level rises are the main determinant of class II sweet spots in the later EHST stage.

Keywords: Rhuddanian–Aeronian stage; paleoproductivity; organic matter enrichment; paleoredox conditions; sea level variation

1. Introduction

The late Ordovician to the Early Silurian (~455–430 Ma) was characterized by dramatic environment changes and a series of globally geological events, e.g., species extinction, the Hirnantian Glaciation, rapid global sea level variation, and oceanic anoxic events [1–4]. The

global transgression during this period deposited a large number of organic-rich shales around the world, including the Wufeng–Longmaxi Shale in South China [5,6]; the “hot shales” in North Africa and Arabia, e.g., Tanf Formation in Syria and Batra Formation in Jordan [7]; and Vinini Formation in Nevada, the United States [8,9]. They are commonly considered as important source rocks, as well as significant unconventional targets for oil and gas exploration [6,8,10,11]. The marine shales from the Ordovician Wufeng Formation to the Silurian Longmaxi Formation are widely distributed around the Yangtze River, comprising high-quality source rocks of upper Paleozoic detrital reservoirs in South China with considerable hydrocarbon generation potential. It is also a primary target for shale gas exploration in China [12,13]. Shale gas enrichment varies greatly with the TOC content of source rocks, which determines the hydrocarbon generation potential, reservoir performance, and gas content [14,15]. Therefore, investigating the mechanism of organic matter enrichment is of great theoretical and practical significance for evaluating the resource potential of shale gas and seeking new sweet spots (favorable exploration targets [2,5,14,15]).

Organic matter enrichment is a complex biogeochemical process [16,17]. Previous studies have proposed three factors affecting this process, including paleoproductivity, paleoredox conditions, and sedimentation rate [17–20]. Paleoproductivity, a controller of the flux of organic carbon to sediments, can be improved by upwelling, terrestrial input, and volcanic activity [18,20,21]. Both reduced bottom water and a high deposition rate are conducive to the preservation of organic matter. The former improves productivity, while the latter preserves organic matter through reducing organic matter exposure to oxygen [19,22,23]. Ibach (1982) proved that the medium deposition rate could contribute greatly to organic matter enrichment. Recent studies suggest that variation in the relative sea level can govern the above three factors and further control organic matter enrichment in shale intervals [19,22–24]. Huang et al. (2020) showed that a highly restricted water environment caused by a sea level drop can increase the organic carbon content in deposits [24].

Currently, the organic matter enrichment mechanism in the upper Yangtze during the early Ludan stage is widely considered to be controlled by the basin topography, where organic matter was enriched in a strong reducing environment with high paleoproductivity and a minor terrestrial supply; as a result, organic-rich shale was primarily distributed at the bottom of the basin or depression [25,26]. Some scholars argue that during the early Rhuddanian stage, organic matter enrichment was primarily driven by upwelling [21,27,28]. This upwelling resulted from a rapid sea level rise caused by geodynamics, such as crustal subsidence or climate change [21,27,28]. This rise caused anoxic and nutrient-rich water from deep basins to flow into shallow shelves, leading to the development of organic-rich shale on slopes [21,27,28]. However, Huang demonstrated that it was controlled by water stratification [24], since biological extinction led to large-scale biological decomposition and oxygen consumption, which formed a strong reducing environment with water stratification, giving rise to organic matter enrichment. Although Guo generally agrees that organic-rich shale was mainly developed during a period of relative sea level rise [12], the primary controllers of organic matter enrichment at the Rhuddanian–Aeronian stage need to be further discussed.

Currently, Wei focuses on the organic enrichment differences between the early Silurian Rhuddanian and Elonian stages and the distribution of high-quality shale gas reservoirs under varying primary controllers [29]. Additionally, Chen posits that shale gas sweet spots mainly form during transgression [30]. Whether shale gas sweet spots can develop during regression, such as in HST and LST, remains a topic for further discussion.

The objectives of this study are to (1) investigate the variations in fundamental factors governing the differential organic matter accumulation model during the Rhuddanian and Aeronian stages and (2) determine whether a shale gas sweet spot with exploration potential was developed during the regressive stage, and pinpoint the successive sweet spot target intervals during the early Rhuddanian rapid transgression.

2. Geological Setting

The Wufeng–Longmaxi sequence within the Sichuan Basin and its periphery in West China originated during the collision of the Cathaysia and Yangtze regions [12–16]. Concurrently, the Upper Yangtze, located between the Leshan and Xuefeng paleouplifts, gave rise to a semi-restricted euxinic basin. Covering an expanse of $2.2 \times 10^4 \text{ km}^2$, the southern Sichuan Basin lies at the juncture of the Tethys–Himalayan and Pacific tectonic zones ([25], Figure 1).

The Luzhou area, situated centrally within southern Sichuan Basin, structurally belongs to the low–steep structural belt in the south of Central Sichuan Paleouplift. This area features a sequence of echelon-oriented narrow anticlines and broad, gentle synclines that extend from north to south [23]. Wufeng Formation is characterized by its lower shale sections containing graptolite belts and the upper Guanyinqiao Member, which has a significant limestone concentration. Longmaxi Formation can be delineated into subsets LM1–9, with LM1–5 deposited during the Rhuddanian stage and LM6–9 during the Aeronian (Figure 2, [15]). After the Middle Ordovician, the Yangtze plate evolved into a foreland basin, positioning Sichuan Basin and its neighboring regions as the elevated posterior portion of this basin [31,32]. Come the Early Silurian, the region underwent continual uplift driven by intensifying southeasterly compression. This dynamic exacerbated sedimentary variation, leading to diminished water depth in central Sichuan [4,22]. During this period, the Luzhou area embodied a deep shelf environment. Subaqueous slopes marked the northwest and northeast, while gravity flow deposits were prominent in the southeast. Central regions featured subaqueous sub-sags, while other areas manifested subaqueous plains [32–35].

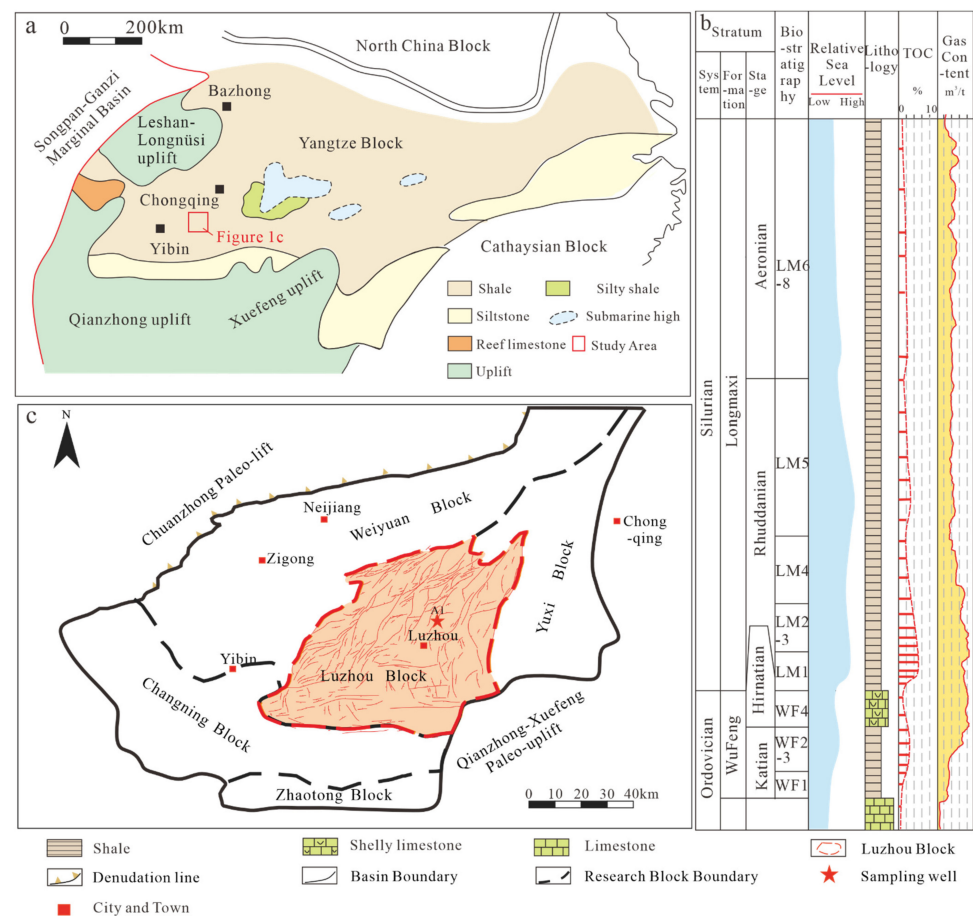


Figure 1. (a) Location of the Upper Yangtze platform during the Late Ordovician, modified from [35]. (b) Simplified stratigraphic units in the study area, modified from [28]. (c) Map showing tectonic sub-units of Southern Sichuan Basin, with the red dashed line indicating the location of Luzhou Block, modified from [36].

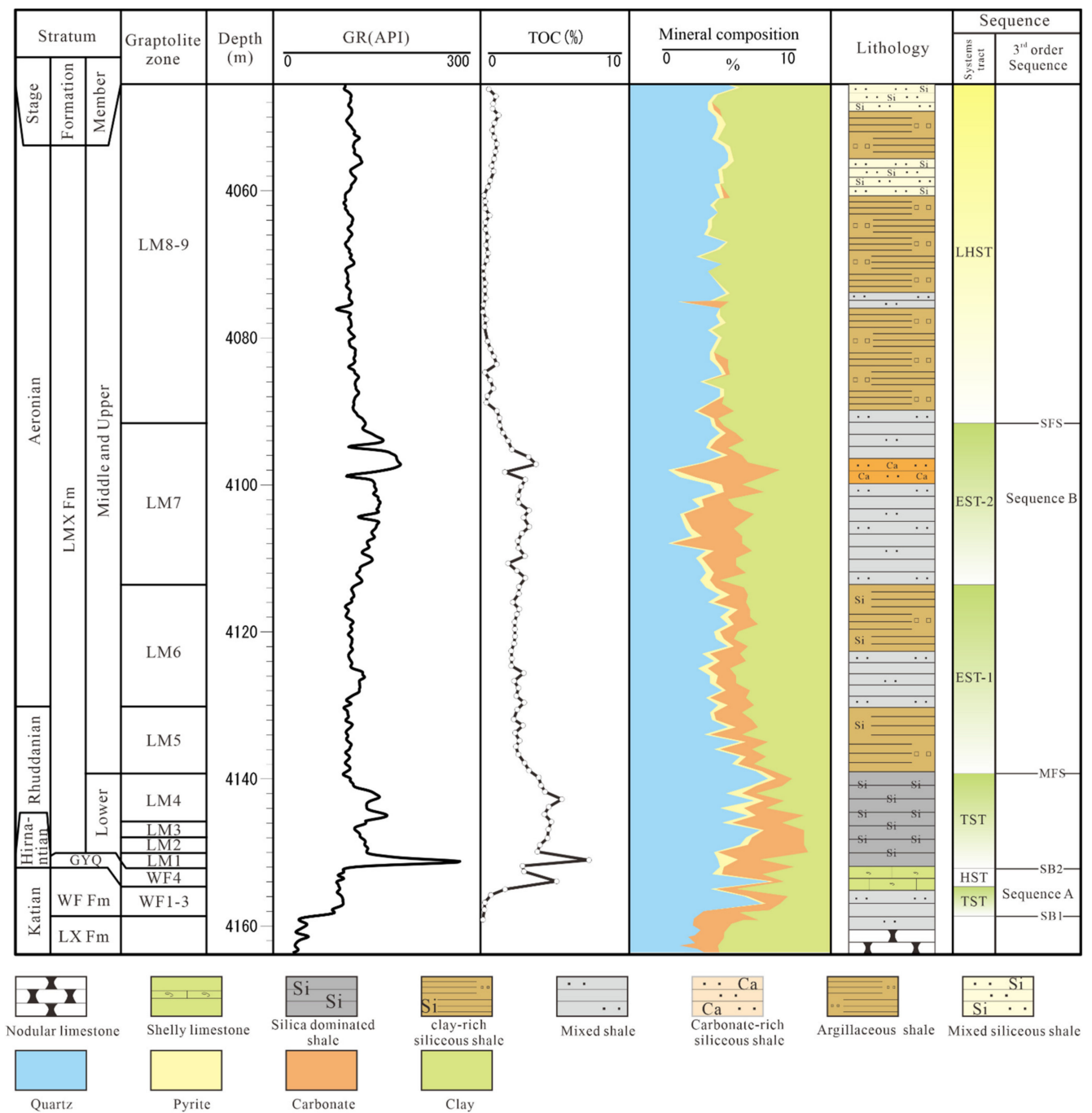


Figure 2. Stratigraphic framework construction and lithofacies classification of the Rhuddanian–Aeronian stage of Well A. HST—highstand system tract; LHST—late highstand system tract; EST—early highstand system tract; TST—transgression system tract; GR—natural gamma ray logging; MFS—maximum flooding surface; SFS—sub-flooding surface; SB—sequence boundary; Mem—Member; LMX Fm—Longmaxi Formation; GYQ—Guanyinqiao Member; WF Fm—Wufeng Formation; LX Fm—Linxiang Formation.

3. Materials and Methods

3.1. Materials

Seventy-four marine shale samples from Well A in the Luzhou area were collected for analyzing TOC content, mineral composition, and major trace elements. Utilizing geochemical analysis of terrestrial inputs, redox conditions, paleoproductivity, and sea level

change, we analyzed organic matter enrichment drivers in Luzhou Area and assessed the Rhuddanian–Aeronian paleoenvironment's impact on shale gas sweet spot development.

3.2. Methods

3.2.1. Major Element Analysis

Shale samples showing no signs of obvious weathering were utilized for analysis. These samples were primarily prepared by crushing them into a 200-mesh powder. Each dried sample, weighing 1.2 g after being oven-dried, was then accurately measured. Prior to analysis, the samples were dissolved in 6 g of solvent ($\text{Li}_2\text{B}_4\text{O}_7$) within a platinum crucible and subsequently spread on a glass sheet to dissolve at a high temperature of 1100 °C. X-ray fluorescence (XRF) was measured using an X-ray fluorescence spectrometer from ThermoFisher, New York. The precision of this method yielded an analysis error of less than 5%. The elements evaluated comprised SiO_2 , Al_2O_3 , Fe_2O_3 , CaO , MgO , K_2O , Na_2O , TiO_2 , P_2O_5 , MnO , MnO_2 , BaO , and SO_3 . All equipment and methodologies were employed at the Research Institute of Petroleum Exploration and Development, Beijing, China.

3.2.2. Trace Element Analysis

Shale samples were used for the analysis of trace element concentrations. These samples, weighing 100 mg each, were initially dried at a temperature of 105 °C. Each sample was dissolved in a solvent composed of HClO_4 (0.5 mL), HF (2.5 mL), HNO_3 (0.5 mL), HNO_3 (1 mL), and H_2O (3 mL). This solution was subsequently diluted for analysis. For the assessment of trace elements, we utilized inductively coupled plasma mass spectrometry (ICP–MS) sourced from AMETEK, Berlin, Germany. The testing protocol incorporated the use of standard samples, including OU–6 (slate), AMH–1 (andesite), and GBPG–1 (plagioclase), to ensure the monitoring and accuracy of measurements. The analysis demonstrated an impressive accuracy rate, reaching up to 99.5% for trace elements. All analyses and equipment were deployed at the Research Institute of Petroleum Exploration and Development, Beijing, China.

3.2.3. X-ray Diffraction Analysis

Sample powders were analyzed for their mineralogical composition. These samples were prepared as per the Chinese oil and gas industry standard (SY/T) 5163-2014 [32]. From this prepared set, X-ray diffraction (XRD) was conducted on all 74 samples, and an additional 30 samples were specifically analyzed for their clay mineral contents, for a total of 74 samples subjected to mineralogical testing. The technical analysis was carried out using the Panalytical X-Pert PRO MPD X-ray diffractometer, operated at a voltage of 50 keV and a current of 800 μA . Diffractograms were recorded over a 2θ range from 5° to 90°. All analytical procedures and equipment operations were conducted at Petroleum Exploration and Development, Beijing, China.

3.2.4. Total Organic Carbon Content Analysis

To evaluate the total organic carbon (TOC) content, samples were initially decarburized by soaking in 4 M HCl at 60 °C for a minimum of 24 h. After this process, the samples were rinsed with distilled water to eliminate impurities and any remaining HCl . Once purified, the samples were dried thoroughly. The measurements were then performed using the LECO CS-400 analyzer from LECO, New York, NY, USA. To ensure analytical precision with an error margin below 0.10%, we repeatedly tested standard samples. All analyses were conducted at the Research Institute of Petroleum Exploration and Development, Beijing, China.

3.2.5. Geochemical Data Presentation

The enrichment factor (EF) is used to assess the degree of elemental enrichment in sediments and is calculated using the formula provided by Munnecke [2]:

$$EF = (E/Al)_{\text{sample}} / (E/Al)_{\text{PAAS}} \quad (1)$$

where E represents the concentration of an element, normalized to the post-Archean Australian shale (PAAS) standard [2,3]. An $EF > 3$ indicates apparent enrichment, $1 < EF < 3$ suggests moderate enrichment, and $EF < 1$ denotes depletion, all in comparison to the PAAS standard [2,3].

The occurrence and content of variable valence elements, e.g., U, Mo, and V, in sediments vary with redox conditions [37–40]. Molybdenum (Mo) is notably enriched under anoxic–euxinic conditions and depleted in dysoxic–oxic environments [40]. It shows moderate enrichment under suboxic conditions ($Mo_{EF} < 10$) and strong enrichment under anoxic to euxinic conditions ($Mo_{EF} > 10$) [18,21,41]. U can occur as dissolved U^{6+} in an oxidizing environment, which can be transformed into insoluble U^{4+} and enriched in sediments in a reducing environment [18,41]. Th is relatively stable in terms of its chemical properties and is not easily migrated, usually being concentrated in oxidizing environments or in weathering-resistant minerals [42,43]. The U/Th ratio has been widely used to identify redox conditions, e.g., $U/Th > 1.25$ indicating anoxic conditions, $U/Th = 0.75–1.25$ representing dysoxic conditions, and $U/Th < 0.75$ indicating oxic conditions [44]. V/Cr and Ni/Co are also used to analyze redox conditions [45]. Cr exists as CrO_4^{2-} under oxic conditions, while it occurs as Cr^{3+} that can be combined with humic acid under anoxic conditions. Ni commonly exists in oxidizing environments as dissolved $NiCO_3$ and is precipitated as sulfide in anoxic environments [46,47]. V is generally combined with organic matter as V^{4+} in reducing environments [48,49]. $V/Cr < 2$ represents oxic conditions, $V/Cr = 2–4.25$ indicates dysoxic conditions, while $V/Cr > 4.25$ means anoxic conditions. $Ni/Co < 5$ suggests oxic conditions, $Ni/Co = 5–7$ means dysoxic conditions, while $Ni/Co > 7$ suggests anoxic conditions [44,50].

The degree of water restriction, which refers to the water exchange between the basin and open water, means an increase in oxygen-carrying currents to the basin floor, supplying oxygen to the basin. Water restriction can greatly change the slope of the Mo-TOC curve [45,51,52]. The Mo/TOC ratios are usually low under strong retention conditions, and are generally high under weak retention conditions because of a sufficient Mo supply in open environments [45,52]. Under anoxic and euxinic water columns, sediments typically have elevated Mo and U contents, but these elements manifest distinct geochemical behaviors [45,52]. The absorption of authigenic U in marine sediments initiates at the Fe (II)–Fe (III) redox boundary, or under suboxic conditions. In contrast, authigenic Mo enrichment requires hydrogen sulfide, implying euxinic conditions [53–56]. The Mn/Fe-oxyhydroxide particulate shuttle enhances the transfer of authigenic Mo to sediments, without influencing authigenic U. The geochemical differences between Mo and U enable the use of their covariation patterns to interpret redox conditions [42,48]. In open-marine systems, Mo and U concentrations rise from suboxic to euxinic conditions, with a concurrent increase in the Mo/U ratio. In restricted basins, Mo and U enrichment depends on the operation of a particulate shuttle. When active, a higher Mo/U ratio occurs due to the stronger enrichment of Mo. Conversely, in conditions of limited Mo resupply, like in the Black Sea, the Mo/U ratio remains lower than in seawater [45,51]. Mn, Cd, Mo, and Co are also usually used to identify upwelling. Intensive upwelling can decrease $Co \times Mn$ significantly, e.g., $Co \times Mn$ in sediments is commonly lower than 0.4 because of upwelling [11].

A higher water nutrition supply will increase the reproductive activities of organisms, which will enhance the ability of organisms to fix carbon through photosynthesis and improve productivity [56]. Stable carbon and nitrogen isotopes, molecular biomarkers, TOC content, and trace element (i.e., Ba, P, Cu, Ni, etc.) abundance have been applied in previous studies to identify variation in paleoproductivity [53,57]. In this study, five

indicators, namely, P, Ba, Cu, Si_{xs}, and Ba_{bio}, were employed to characterize the changes in paleoproductivity. The Si_{ex} is utilized to determine the content of biogenic silica, and the excess SiO₂ content can be computed as follows:

$$\text{Si}_{\text{ex}} = \text{SiO}_{2\text{sample}} - \text{Al}_{\text{sample}}/27 \times 2 \times 60.1 \quad (2)$$

To elucidate the paleoproductivity using the element Ba, it is essential to subtract the terrigenous components. The calculation for Ba_{bio} is as follows [23,33]:

$$\text{Ba}_{\text{bio}} = \text{Ba}_{\text{sample}} - \text{Ti}_{\text{sample}} \times (\text{Ba}/\text{Ti})_{\text{PAAS}} \quad (3)$$

The analyzed shale primarily derives from terrigenous input, with elements such as Al, Zr, and Ti from the continental crust being constrained in marine migration by diagenesis and weathering [58–61]. Elements such as Al, Ti, K, and Zr are high in stability. Al is usually derived from clay minerals in aluminosilicate, while Ti usually exists in stable minerals or heavy minerals, e.g., ilmenite and TiO₂. Zr is usually originated from detrital zircon, which is stable during sediment transportation and deposition [59]. Therefore, they are commonly used to express terrigenous material input [60,61].

The paleoclimate plays an important role in paleoproductivity, marine redox conditions, and terrestrial supply, thus governing element content, mineral composition, and distribution in sediments ([62]). The chemical alteration index (CIA) and index of compositional variability (ICV) are considered as chemical indicators that can describe the weathering of provenance and paleoclimate [63]. The CIA index was proposed by Xu [5].

$$\text{CIA} = [(\text{Al}_2\text{O}_3)/(\text{Al}_2\text{O}_3 + \text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O})] \times 100 \quad (4)$$

Using CIA measurements of modern sediments, the climatic implications are as follows: 50–65 indicates cold and dry conditions (low chemical weathering); 65–80 signifies warm and moist conditions (medium chemical weathering); and 80–100 represents hot and humid conditions (strong chemical weathering) [58–62].

The ICV, index of compositional variability, is defined as

$$\text{ICV} = (\text{Fe}_2\text{O}_3 + \text{K}_2\text{O} + \text{Na}_2\text{O} + \text{CaO} + \text{MgO} + \text{MnO} + \text{TiO}_2)/\text{Al}_2\text{O}_3 \quad (5)$$

It is a common indicator used to distinguish cyclic sediments and original sediments. Specifically, ICV < 1 indicates mature composition and cyclic sediments, while ICV > 1 represents immature composition and original sediments [64]. Wei and Algeo (2020) proposed that Sr/Ba < 0.2 represented fresh water, Sr/Ba = 0.2–0.5 was an indicator of brackish water, and Sr/Ba > 0.5 was seawater [6].

Geochemical proxies, including Sr/Cu, Rb/Sr ratios, and SiO₂ versus (Al₂O₃ + K₂O + Na₂O), have been utilized for paleoclimatic investigations [6,15,24,38], while the Sr/Ba ratio is employed to assess seawater salinity [6,15,24,38]. The SiO₂ vs. Al₂O₃ + K₂O + Na₂O diagram shows that the climate was dry and cold in the TST stage. It became warm and humid in a later stage. The paleoclimate index C quantifies climate preference by relating elements indicative of humid conditions to those of dry environments, with higher values suggesting warm–humid and lower values indicating dry–cold climates [45]. C can be calculated as

$$\text{C} = (\text{Fe} + \text{Mn} + \text{Cr} + \text{V} + \text{Co} + \text{Ni})/(\text{Ca} + \text{Mg} + \text{K} + \text{Na} + \text{Sr} + \text{Ba}) \quad (6)$$

3.2.6. Sequence Boundary Discrimination

The identification of sequence boundaries in this study is primarily based on the 74 samples from Well A and the abrupt transitions observed in eight lithofacies, TOC, GR logging curves, and biostratigraphic changes. Drawing from Lu's standards for the stratigraphic framework partition of Wufeng–Longmaxi Formation in the Fuling area, we

have clarified the geological significance of the main isochronous boundaries during the Rhuddanian–Aeronian period in the Luzhou area (Figures 2–4, [38]).

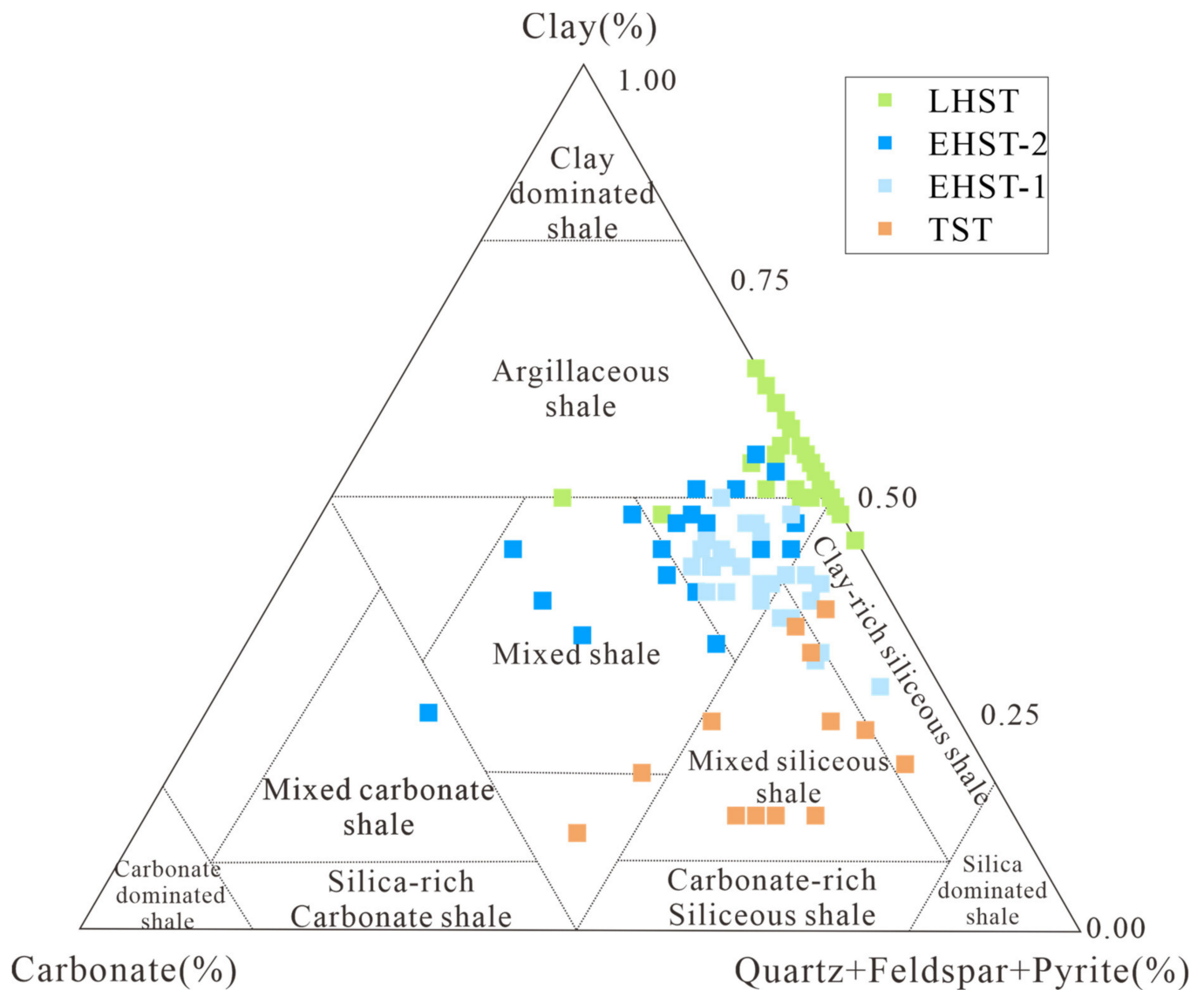


Figure 3. The terminology and classification of the studied Rhuddanian–Aeronian shale. During the TST depositional period, mixed siliceous shale and clay-rich siliceous shale were predominantly developed. This shale lithology discrimination template is modified from [64]. Additionally, in the EHST-1 stage, mixed shale and clay-rich siliceous shale were the main formations. The EHST-2 stage primarily saw the development of mixed shale, while the LHST depositional period was characterized by the predominant formation of argillaceous shale.

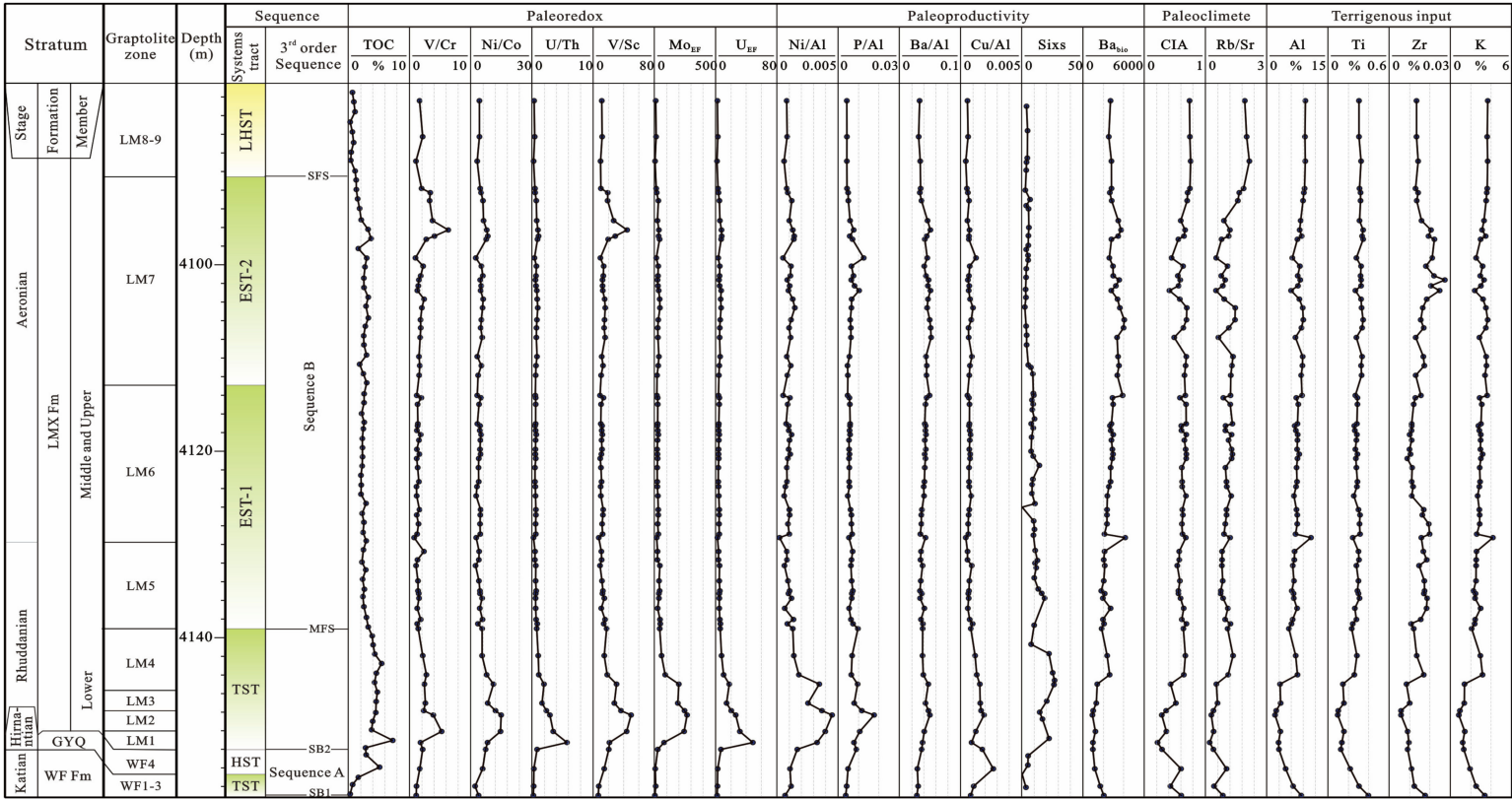


Figure 4. Vertical variation of Well A in geochemical characterization parameters for paleogeographic environments, including paleoredox, paleoproductivity, terrigenous input, and paleoclimate proxies of Rhuddanian–Aeronian shale. HST—highstand system tract; LHST—late highstand system tract; EST—early highstand system tract; TST—transgression system tract; MFS—maximum flooding surface; SFS—sub-flooding surface; SB—sequence boundary; Mem.—Member; LMX. Fm.—Longmaxi Formation; GYQ—Guanyinqiao Member; WF. Fm.—Wufeng Formation.

4. Results

4.1. X-ray Diffraction Analysis

Based on X-ray diffraction analysis, the mineral composition of Longmaxi shale samples appears intricate, as illustrated in Figures 2 and 3. Quartz, clay minerals, and carbonate minerals are the principal constituents. The average quartz content is 42.5% across samples, with biostratigraphic units demonstrating a decreasing trend in the sequence: LM1 (62.1%) > LM2 (58.2%) > LM3 (53.5%) > LM4 (50.3%) > LM5 (47.6%) > LM6 (42.3%) > LM7 (34.0%) > LM8–9 (42.3%) (Figure 2). The shale samples exhibited a clay mineral content range of 12.2–56.4%, averaging 24.3%. When comparing different biostratigraphic units, there was an increasing trend in clay mineral content: LM1 (11.6%) < LM2 (12.5%) < LM3 (16.7%) < LM4 (24.8%) < LM5 (37.0%) < LM6 (43.0%) < LM7 (43.6%) < LM8–9 (54.5%) (Figure 2). Carbonate minerals in the Longmaxi shale primarily consist of calcite and dolomite, collectively contributing 0.2–53.3% of the content. Notably, the average carbonate mineral content across different biostratigraphic units consistently escalates as follows: LM1 (12.5%), LM2 (11.3%), LM3 (12.9%), LM4 (12.5%), LM5 (12.3%), LM6 (11.1%), LM7 (18.7%), LM8–9 (1.3%) (Figure 2).

4.2. Total Organic Carbon Content Analysis

The total organic carbon content (TOC) of the Longmaxi shales varies between 0.6 and 5.1% (Figures 2 and 4), with an average of 2.3% (Figures 2 and 4). Notably, samples within the biostratigraphic units LM1–LM4 exhibit elevated TOC levels, ranging from 3.3 to 5.1% (Figures 2 and 4). Units LM5–LM6 present reduced TOC values, spanning from 2.1 to 3.6%. The TOC in unit LM7 shows a modest increase, lying within the range of 1.7–3.6% (Figures 2 and 4), while that of units LM8–9 undergoes a sharp decline to between 0.6 and 1.8%, with an average content of 0.8% (Figures 2 and 4).

4.3. Major Element Analysis

Major elemental data are presented in Figure 4. Si, Al, and Ca are the primary major elements in the Longmaxi samples. Among these, SiO₂ is the predominant oxide in most shale samples, with its concentration gradually diminishing from units LM1 to LM9, spanning a range of 31.2 to 72.0%. The mean Al₂O₃ contents across units LM1–LM9 are 5.9%, 4.1%, 6.5%, 11.2%, 12.6%, 14.5%, 15.6%, 17.5%, and 18.7%, respectively. The CaO content of LM2–LM6, ranging between 0.3% and 18.1%, is relatively high. Additionally, elements such as Mn, Ba, K, Fe, Na, Mg, P, and Ti maintain relatively low concentrations (0.012–1.23%) in comparison to the primary elements previously mentioned (Figure 4).

4.4. Trace Element Analysis

The trace element results are delineated in Figure 4. Units LM1–4 and LM7 show enriched concentrations of V (avg. 156.8 ppm), Ni (avg. 79.7 ppm), U (avg. 14.9 ppm), and Mo (avg. 37.2 ppm). In contrast, Cr (avg. 82.8 ppm), Co (avg. 12.9 ppm), and Th (avg. 13.3 ppm) exhibit depletions in these same biostratigraphic units (Figure 4). Notably, Cu (avg. 48.4 ppm) is abundant in units LM1–4 (Figure 4). The concentrations of Ti (avg. 0.32 ppm), Zr (avg. 145.7 ppm), and Rb (avg. 164.9 ppm) consistently increase across units LM1–9 (Figure 4). Moreover, a gradual enrichment of Ti, Zr, and Rb is observed throughout units LM1–9 (Figure 4).

4.5. Lithofacies Classification

Eight primary lithofacies were identified in the A1 well from the Rhuddanian–Aeronian shale based on TOC content and mineral components (Figures 2 and 3). The quartz content of siliceous shale is higher than 50%, and pyrite content is lower than 5%, whereas both calcite content and clay mineral content are about 10–20%. The clay-rich siliceous shale is <10 m in thickness, its clay mineral content is up to 40%, and its quartz content is lower than that of siliceous shale (30–50%). It is low in calcite content (5–10%), with a pyrite content of <3%. The quartz content of siliceous shale is higher than 50%, and pyrite content

is lower than 5%, whereas both calcite content and clay mineral content are about 10–20%. The clay-rich siliceous shale is <10 m in thickness, its clay mineral content is up to 40%, and its quartz content is lower than that of siliceous shale (30–50%). It is low in calcite content (5–10%), with a pyrite content of <3%.

Baota Formation is dominated by nodular limestone, which transits upward to the mixed shale and siliceous shale of Wufeng Formation (Figures 2 and 3). The upper part of Wufeng Formation, known as Guanyinqiao Member, has a thickness of 2.8 m and is predominantly composed of marls, with minor inclusions of Hirnantia fauna bioclasts (Figures 2 and 3). This formation is a sedimentary outcome of the Hirnantian lowstand, associated with the global sea level drop due to Hirnantian Glaciation [12–16]. The bottom of Longmaxi Formation is dominated by siliceous shale, and the middle part is interbedded siliceous shale and mixed shale with a thickness of about 10 m, where thin carbonate-rich siliceous shale can be observed. Interbedded argillaceous shale and mixed siliceous shale can be observed at the top of Longmaxi Formation (Figures 2 and 3).

4.6. Stratigraphic Framework

During the Rhuddanian–Aeronian period, the sedimentary fill thickness in the Luzhou area was roughly 90–150 m, primarily composed of the transgressive system tract (TST), early highstand system tract (EHST), and late highstand system tract (LHST). EHST can be further subdivided into two sub-sequences, namely, EHST-1 and EHST-2 (Figure 2). Within the stratigraphy of Wufeng–Longmaxi Formation in the A1 well, two third-order sequence boundaries can be identified: the base of Wufeng Formation (SB1) and the base of Longmaxi Formation (SB2). There are two system tract boundaries, with the maximum flooding surface (MFS) located at the top of LM4 and the sub-flooding surface (SFS) at the top of LM7. Meanwhile, within the EHST, one parasequence boundary is identified (Figures 2 and 4).

The condensed section base sequence boundary (SB1) in Wufeng Formation serves as a significant transgressive process with flooding unconformity, with overlying grey–black mixed shale and underlying nodular limestone from Linxiang Formation (Figure 3). The GR value jumps from 31.8API to 88.0API, accompanied by a sharp increase in TOC (Figure 2). Above SB1, the first third-order sequence (Sequence A) is composed of Wufeng and Guanyinqiao Formation, primarily made up of mixed shale and shelly limestone (Figure 3). GR values range from 38.9 to 102.2API, and TOC ranges from 0.1 to 3.9%. The top boundary of Sequence A in Longmaxi Formation (SB2) also serves as a transgressive process with flooding unconformity, below which is the Hirnantian Guanyinqiao shelly limestone, and above is the LM1 black siliceous shale (Figure 3). The GR curve at this boundary sharply increases from 100.6API to 270.1API, indicating a distinct boundary, and TOC also increases sharply.

The second third-order sequence starts above SB2, with the system tract type being TST (including LM1–LM4), primarily consisting of black siliceous shale characterized by high quartz content (ranging from 42.1% to 69.7%) and high TOC (ranging from 3.3% to 5.1%) (Figures 2–4). The maximum flooding surface at the top of the TST often serves as the boundary between LM4 and LM5, with siliceous shale below and clay-rich siliceous shale above (Figure 3). The GR value at this boundary sharply declines from 114.8API to 100.1API, and TOC also decreases sharply from 3.7% to 2.2% (Figures 2–4). Above the MFS is the early highstand system tract (EHST), including LM5–LM7, mainly composed of argillaceous shale and mixed shale (Figures 2–4). The quartz content in this system tract ranges from 28.2 to 53.6%, and clay minerals from 35.4 to 52.3%, with an average TOC of 2.4%. This study aimed to further identify the shale sweet spots within the EHST succeeding the LM1 black siliceous shale, leading to the identification of the parasequence of the EHST. Within the EHST, a parasequence boundary exists, with mixed shale above and argillaceous shale below (Figures 2–4). Concurrently, both the GR curve and TOC gradually increase, from 1.7% to 3.6% (Figures 2–4). The top of the EHST develops a sub-flooding surface (SFS), often serving as the boundary between LM7 and LM8–9. Below this surface

is grey mixed shale, and above is argillaceous shale (Figures 2–4). This boundary correlates with a GR curve inflection, dropping from 121.2API to 159.3API, and TOC also notably decreases from 1.8% to 0.3% (Figures 2–4). Above the SFS is the LHST, including LM8–9, predominantly consisting of argillaceous shale with significant bioturbation (Figures 2–4). At this stage, the content of clay minerals further increases, ranging from 45.1 to 65.2%; the quartz content ranges from 24.6 to 53.3%; and TOC varies from 0.4 to 1.7% (Figures 2–4).

5. Discussion

5.1. Environmental Significance of Geochemical Elements

5.1.1. Paleoredox Condition

Organic matter can be enriched and preserved well under an anoxic environment [2–12]. The U/Th ratio follows a decreasing trend from the TST stage at the bottom to the LHST stage at the top (Figure 5a). The U/Th ratio of the TST stage is generally higher than 1.25, indicating an anoxic environment (Figure 5a). The low U/Th ratios of the EHST stage and the LHST stage suggest a dysoxic to oxic environment (Figure 5a). A dysoxic environment can also be identified from the EHST-2 stage (Figure 5a).

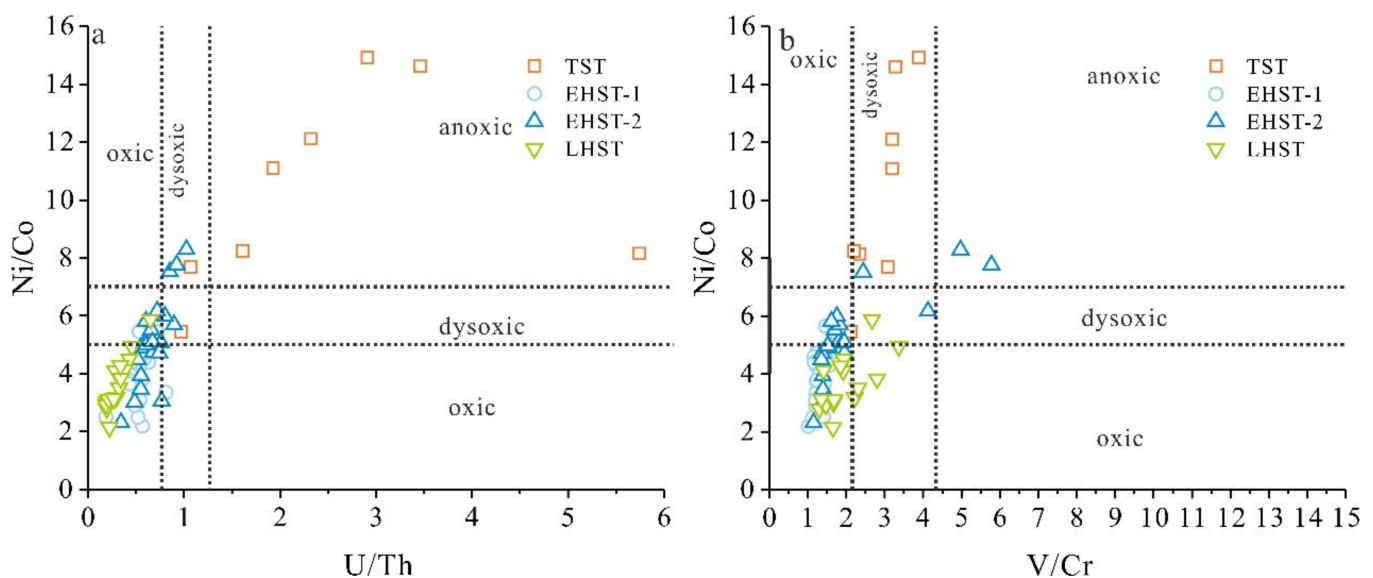


Figure 5. Scatter plots of paleoredox indicators for different system tracts during Rhuddanian–Aeronian transition in Well A. (a) Ni/Co vs. U/Th; (b) Ni/Co vs. V/Cr. In Longmaxi Formation of Well A, values of Ni/Co and V/Cr increase from the TST stage at the base to the LHST stage at the top, while U/Th values decrease from the TST to the LHST stages. This indicates a progressive rise in water oxygen content.

Most V/Cr ratios of the TST stage are between 2 and 4, indicating a dysoxic environment, while the V/Cr ratios from the EHST-1 to the LHST are <2, suggesting enhanced oxidation in later stages (Figure 5b). It is suddenly increased at the contact between EHST-2 and LHST (Figure 5b). The Ni/CO ratio is gradually decreased from the TST to the bottom of the EHST-1, also indicating enhanced oxidation (Figure 5b). The Ni/Co < 5 of the EHST to the LHST indicates an oxic environment. Like the V/Cr, Ni/Co is also increased slightly at the EHST-2, indicating slightly weakened oxidation (Figure 5b). The data points for V/Cr, U/Th, and Ni/Co in the TST stage suggest a predominant hypoxic state, transitioning from anoxic to dysoxic conditions (Figure 5a,b). Within the EHST stage, there is a discernible upward shift from dysoxic to oxic conditions. Notably, a minor increase in the degree of water column reduction is observed in the late EHST stage (Figure 5). Conversely, the LHST represents a wholly oxidized benthic setting (Figure 5).

5.1.2. Degree of Water Restriction

The extent of water restriction influences the trace element availability in basin water, subsequently altering their enrichment in the sediments [32–36]. In the TST, the Mo_{EF}/U_{EF} ratios for most samples were 1–3 times those of typical seawater (Figure 6a). Despite rising enrichment factors, the Mo_{EF}/U_{EF} ratios remained relatively stable, suggestive of a semi-restricted basin (Figure 6a). Conversely, in the EHST and LHST, increasing enrichment factors corresponded to elevated Mo/U ratios in sediments, a hallmark of an open-marine setting (Figure 6a). As depicted in Figure 6b, the samples from the TST typify the characteristics of a semi-restricted marine environment. During the transition from EHST to LHST, a progressive increase in the Mo/TOC ratio suggests an evolving degree of water restriction (Figure 6b).

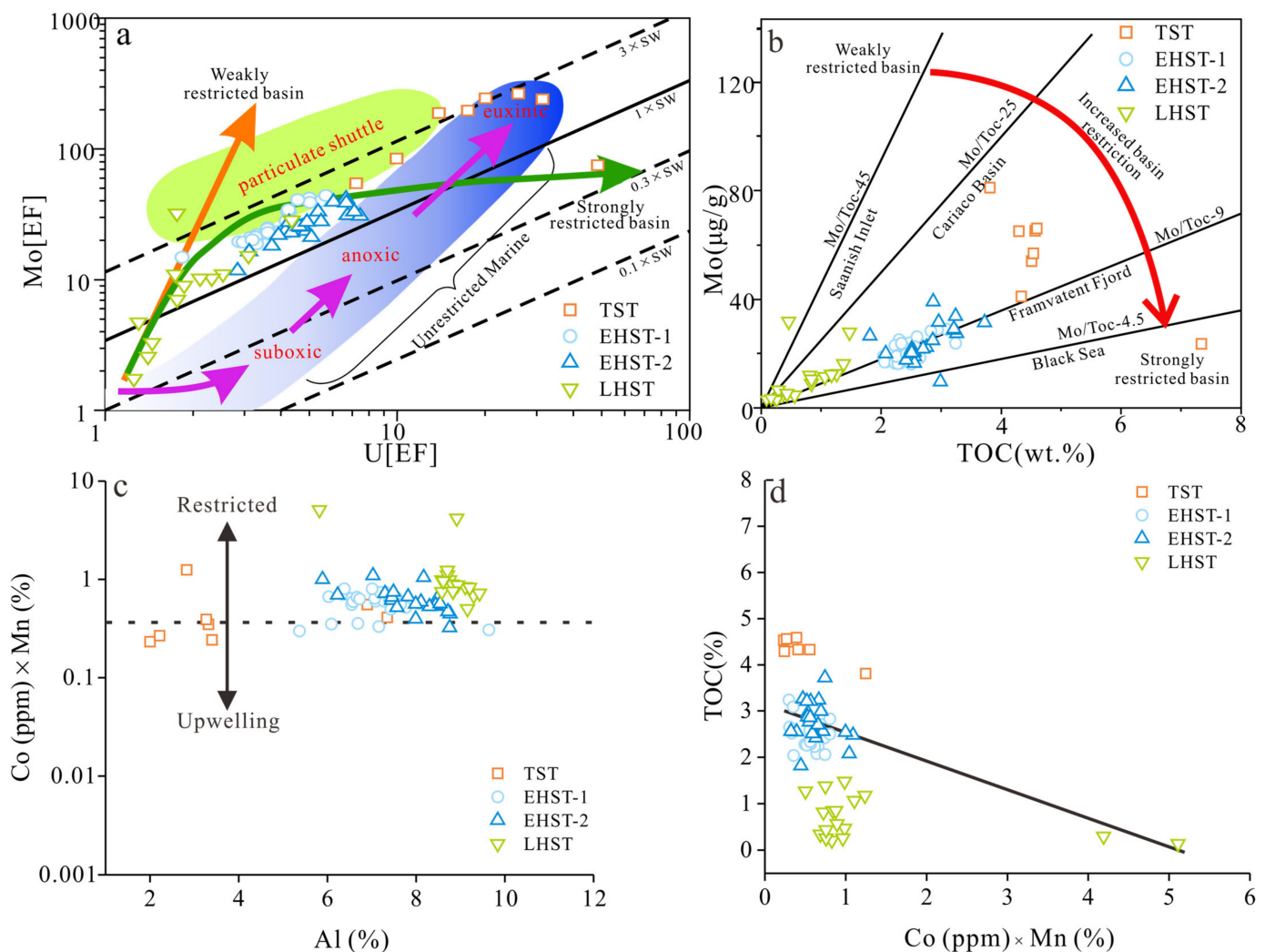


Figure 6. Scatter plots indicating the water mass restriction degree of different system tracts during the Rhuddanian–Aeronian transition in Well A. (a) Mo_{EF} vs. U_{EF} . The orange, green, and purple arrows illustrate the trends of Mo_{EF} and U_{EF} in weakly restricted basin, strongly restricted basin, and unrestricted marine settings, respectively, as the level of water oxidation increases. The dark blue field denotes the “unrestricted marine” trend, while the light green field signifies the “particulate shuttle” trend. This suggests a gradual increase in the degree of restriction from the TST to LHST. (b) Mo - TOC relations in this study and modern anoxic silled-basin environments, indicating that the degree of restriction increases gradually during the TST to LHST. (c) $Co \times Mn$ vs. Al . (d) TOC vs. $Co \times Mn$. (c,d) indicate that the TST was significantly influenced by upwellings, while the EHST and LHST had limited influence from upwellings, suggesting an increase in degree of water restriction.

Following Figure 6c,d, the $\text{Co} \times \text{Mn}$ generally follows an upward trend at the Rhuddanian–Aeronian stage, from 0.6 to 2.3. This suggests that upwelling was developed at the bottom of the TST, while the central part of the TST was in an open environment and related to North Qinling Ocean (Figure 6c,d, [24–26]). During the EHST and LHST stages, the values of $\text{Co} \times \text{Mn}$ exhibited a rising trend, suggesting a continuous increase in the degree of water restriction (Figure 6c,d). Figure 6d illustrates a negative correlation between $\text{Co} \times \text{Mn}$ and TOC. This suggests that during the TST stage, strong upwelling promoted organic matter enrichment, whereas the more restricted water column in the EHST and LHST periods was less conducive to the accumulation of organic material (Figure 6d).

5.1.3. Paleoproductivity

Oceanic productivity is defined by the sea's capacity to synthesize organic compounds through assimilation [23,34,42]. The contents of Si_{ex} , Ba_{bio} , Cu, Ni, and P have significant relevance for marine organisms [23,34,42]. Thus, these elemental concentrations serve as trustworthy metrics for assessing productivity [23,34,42]. Generally, within the TST, elevated values of Si_{ex} , Ni/Al, Cu/Al, and P/Al suggest sustained high marine primary productivity (Figure 4). However, declining trends in Si_{ex} concentrations and the ratios of Ni/Al, Cu/Al, and P/Al imply a gradual reduction in primary productivity, reaching a relatively low primary productivity level during the EHST and LHST periods (EHST-2, Figure 4).

5.1.4. Terrigenous Detrital Inputs

The composition of black shale and the enrichment process of organic matter are primarily influenced by terrigenous detrital [23]. A strong positive correlation between Al and elements like Ti, Zr, K, Ba, and Fe is observed in Figures 4 and 7a–c,f. This suggests that these elements primarily originate from terrigenous fragments. Conversely, either no significant or a mild negative correlation is found among Si, Al, and K (Figure 7d,e), signifying that aluminosilicates are not the main source of Si. During the TST stage, these lower concentrations of Al, Ti, Zr, K, and Fe in the sediment indicate a relatively consistent environment with minimal terrigenous influx (Figure 4). Notably, during the EHST–LHST stage, the concentrations of Al, Ti, Zr, K, and Fe elevate, pointing to an augmented terrigenous input (Figures 4 and 7). However, during the LHST deposition stage, the rate of increase in the Ti concentration lags significantly behind that of K, with Zr even showing a decline as K rises (Figure 4). This trend can likely be attributed to the swift accumulation of fine-grained clay minerals, which dilute their coarse-grained counterparts, leading to inverse trends in Zr and K (Figure 4).

5.1.5. Paleoclimate

As shown by A–CN–K ($\text{Al}_2\text{O}_3\text{--CaO} + \text{Na}_2\text{O--K}_2\text{O}$) (Figure 8a), the average CIA values of the Rhuddanian–Aeronian samples are 73.24, indicating moderate chemical weathering (Figure 8b). Within the TST, EHST-1, EHST-2, and LHST stages, the respective average CIA values are 69.4%, 72.2%, 76.8%, and 79.4%, demonstrating an ascending trend (Figure 8a). Similarly, the ICD displays a corresponding pattern of change; its values for TST, EHST, and LHST in Well A steadily diminish from 4.23 to 1.22 (Figure 8b). These results indicate a gradual paleoclimatic shift from cold and dry to warm and moist during the TST to LHST deposition period (Figure 8).

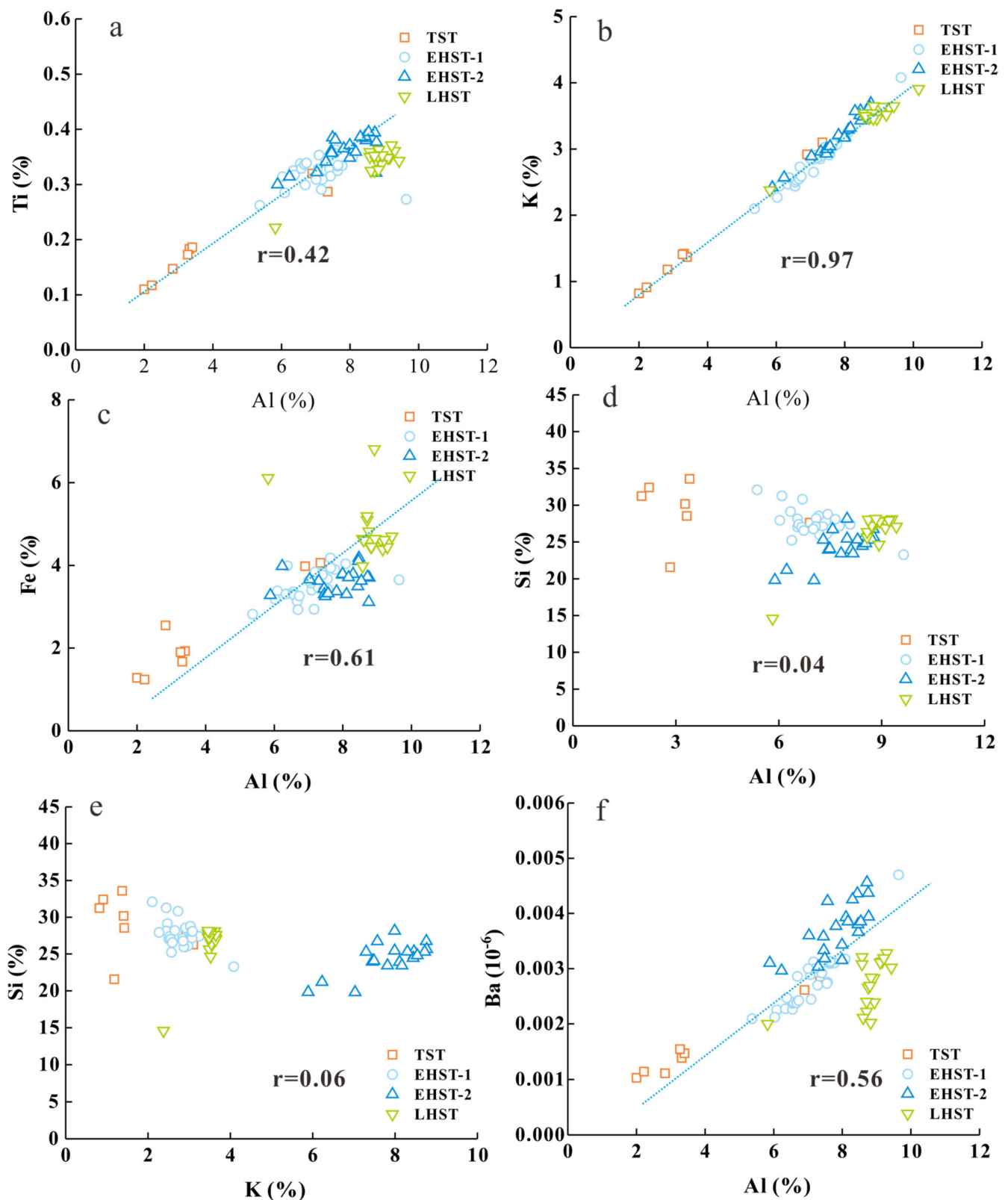


Figure 7. Relationships among Ti, K, Al, Fe, Si, and Ba indicating the degree of terrigenous detrital inputs during the Rhuddanian–Aeronian stage in Well A. (a) Ti vs. Al; (b) K vs. Al; (c) Fe vs. Al; (d) Si vs. Al; (e) Si vs. K; (f) Ba vs. Al. Al shows positive correlations with Ti, K, Ba, and Fe, indicating that these elements primarily originate from terrigenous detritus. The contents of Al, Ti, K, and Zr progressively increase from the TST to EHST and then to LHST, suggesting a continuous increase in terrigenous detrital inputs.

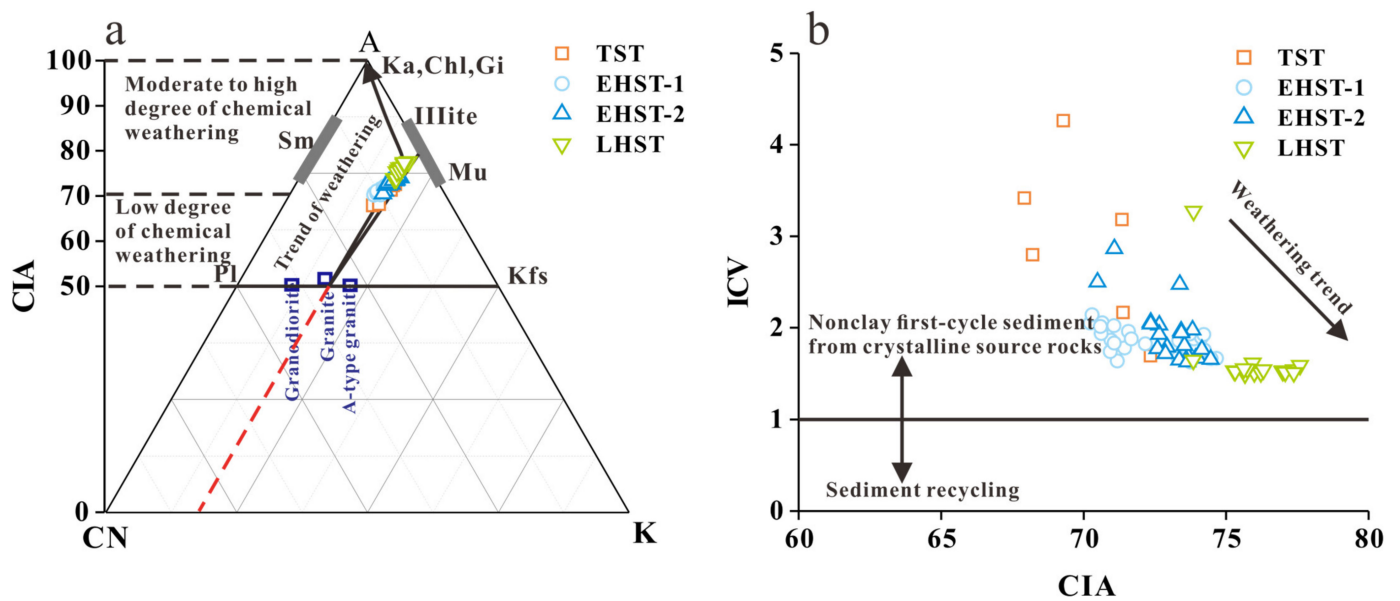


Figure 8. (a) A-CN-K ternary diagrams exhibiting weathering degree (all in mole percentage). (b) Scatter plot of ICV vs. CIA. The CIA and ICV diagrams indicate that during the Rhuddanian–Aeronian stage, the shale was deposited from primary sediments. From the TST to LHST stage, chemical weathering gradually intensified, and following the end of the ice age, the paleoclimate shifted progressively towards warmer and more humid conditions.

The SiO_2 vs. $\text{Al}_2\text{O}_3 + \text{K}_2\text{O} + \text{Na}_2\text{O}$ diagram shows that the climate was dry and cold at the TST stage, and it became warm and humid in later stages (Figure 9a). The cross plot of C vs. Rb/Sr shows a good positive correlation (Figure 9b). The C and Rb/Sr were low in the TST stage, indicating weak weathering intensity and a cold and arid environment (Figure 9b). Weathering was gradually enhanced in the EHST-1, EHST-2, and LHST stages (Figure 9b). The TST, EHST-1, EHST-2, and LHST stages in Longmaxi Formation are characterized by low Sr/Ba ratios (Figure 9c), with average values of 0.14, 0.03, 0.05, and 0.05, respectively. The Sr/Ba is negatively correlated with C (Figure 9d), indicating that a warm and humid paleoclimate with low paleosalinity was crucial to the deposition of the Longmaxi organic-rich shale. The TST sedimentation persisted in tandem with the cooling event triggered by the Hirnantian glaciation [12,23,26]. Indicators, such as CIA, ICV, C, Sr/Ba, and Rb/Sr from the TST depositional stage, suggest that the environment retained its warm to semi-cold climatic conditions (Figures 8 and 9). The minimal chemical weathering in the source area during the TST likely accounted for the reduced terrigenous influx. Following this, during the EHST–LHST transition, the region transitioned to predominantly warm conditions, eventually culminating in a hot climate (Figures 8 and 9).

5.1.6. Provenances

$\text{SiO}_2/\text{Al}_2\text{O}_3$ vs. $\text{K}_2\text{O}/\text{Na}_2\text{O}$, Th/Sc vs. Zr/Sc, La/Th vs. Hf, and Co/Th vs. La/Sc cross plots are used in this study to indicate the provenance of sedimentary rocks (Figure 10a,b, [15,23,32–36]). The Th/Sc vs. Zr/Sc plot shows that samples are concentrated near granite and granodiorite (Figure 10a). As the $\text{SiO}_2/\text{Al}_2\text{O}_3$ vs. $\text{K}_2\text{O}/\text{Na}_2\text{O}$ diagram shows, the $\text{K}_2\text{O}/\text{Na}_2\text{O}$ of the Longmaxi shale from Well A is higher than 1, and the $\text{SiO}_2/\text{Al}_2\text{O}_3$ varies greatly between 3 and 10 (Figure 10b). The La/Th vs. Hf plot shows that samples are located near felsic rocks with volcanic rocks (Figure 10c). The Co/Th–La/Sc, TiO_2 – Al_2O_3 , and TiO_2 –Zr diagrams show that samples are positioned near felsic volcanic rocks (Figure 10d–f). Hence, sediments at the Rhuddanian–Aeronian stage were mainly derived from felsic volcanic rocks, whereas no significant difference occurred in the provenance of the TST, EHST-1, EHST-2, and LHST stages (Figure 10).

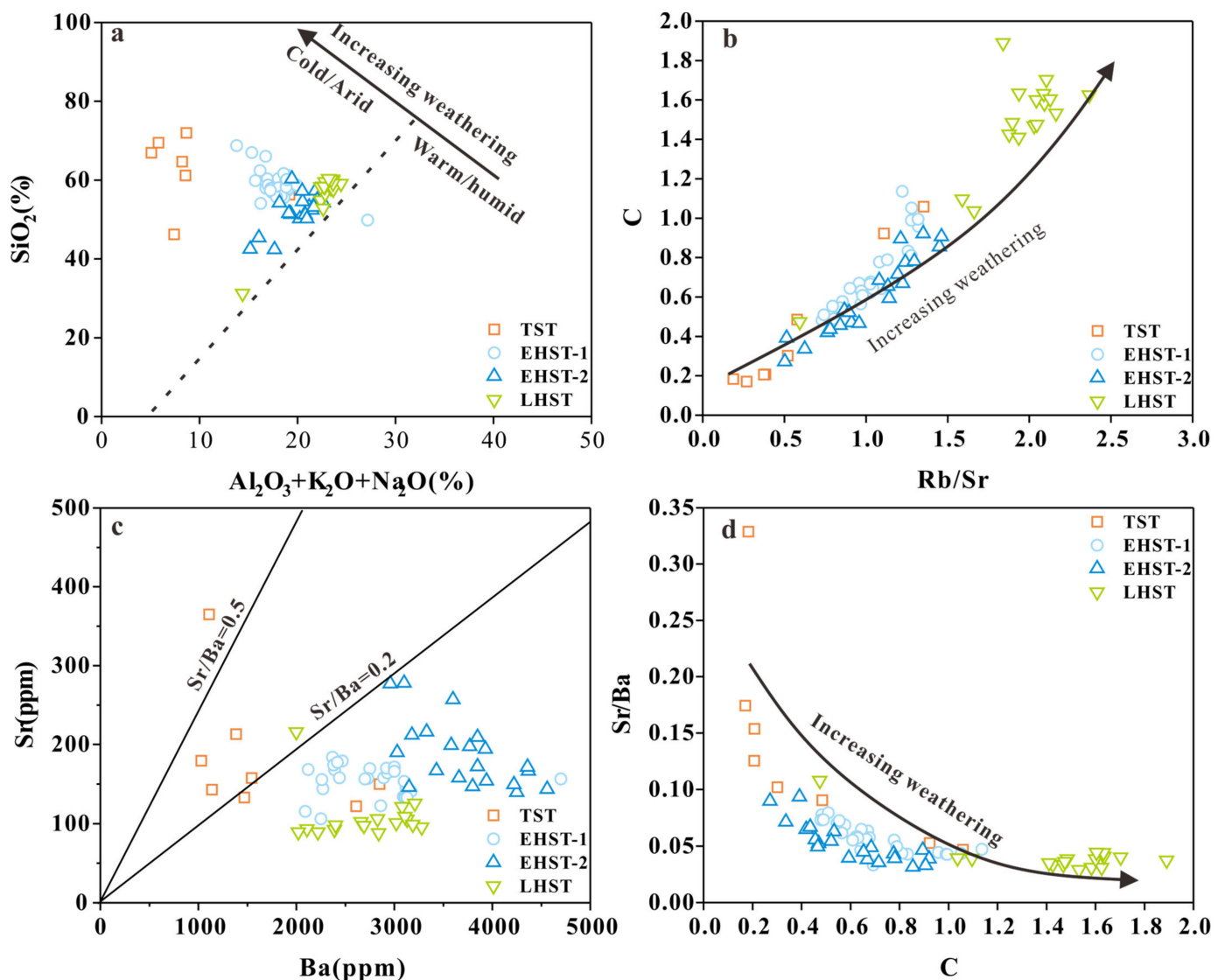


Figure 9. Scatter plots for determining paleoclimate of different system tracts during the Rhuddanian–Aeronian transition in Well A. (a) SiO₂ vs. Al₂O₃ + K₂O + Na₂O; (b) C vs. Rb/Sr; (c) Sr vs. Ba; (d) Sr/Ba vs. C is a paleoclimatic indicator used to determine whether the sedimentary environment is warm–humid or dry–cold. This indicates that during the Rhuddanian–Aeronian stage, the melting of glaciers led to a gradual warming of the climate and a decrease in seawater salinity. The climate was relatively cool and dry during the TST deposition stage. However, as sea levels progressively declined, the climate shifted to a relatively warmer and more humid condition.

5.2. Main Controlling Factors of Organic Matter Accumulation

The organic matter enrichment is mainly controlled by the following factors [23,65,66]:

- ① organic matter preservation (paleoredox conditions and sedimentation rate) [33,67],
- ② organic matter input (paleoproductivity), and
- ③ organic matter dilution (terrestrial inputs) [23,56,68].

However, it may also be controlled by other factors, e.g., paleoclimate, basin topography, volcanic activities, sea level, tectonic movement, upwelling, submarine current, and hydrothermal activities [21,24,35,69].

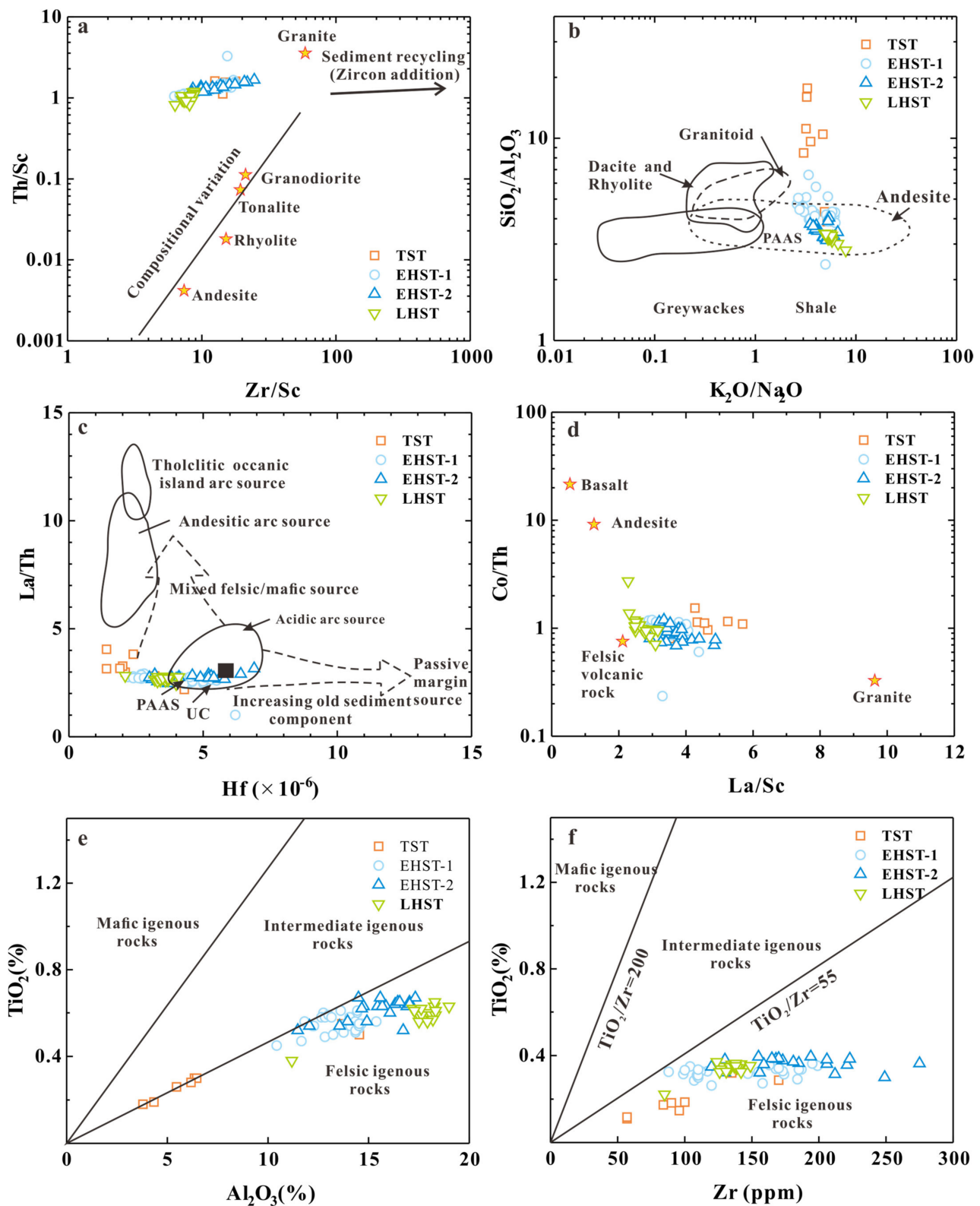


Figure 10. Cross plots for determining provenances of different system tracts during the Rhuddanian–Aeronian transition in Well A. The provenance-indicating diagrams were modified by [15,23,32–36]. (a) Th/Sc vs. Zr/Sc; (b) SiO₂/Al₂O₃ vs. K₂O/Na₂O; (c) La/Th vs. Hf; (d) Co/Th vs. La/Sc; (e) TiO₂ vs. Al₂O₃; (f) TiO₂ vs. Zr. The provenance diagrams indicate that during the Rhuddanian–Aeronian period, there were no significant differences in the provenance of sediments for the TST, EHST-1, EHST-2, and LHST stages. The parent rock type for all these stages was consistently felsic volcanic rock.

Paleoredox indicators (Ni/Co-U/Th, Ni/Co-V/Cr, MoEF-UEF, Mo-TOC, etc.) and paleoproductivity indicators (Ba_{bio} , Cu/Al, P/Al, Si_{ex} and Cu/Al, etc.) show that organic matter enrichment at the Rhuddanian–Aeronian stage was significantly affected by paleoredox conditions and paleoproductivity (Figures 4–6, [19,22,25]). The CIA values during the Hernantian stage are between 52.4% to 62.3%, C values range from 0.1 to 0.2, and ICV values are between 4 and 5. When compared to the CIA, C, and ICV values from the early TST of the Rhuddanian stage in the Luzhou area, it is evident that the values during the TST sedimentary period are significantly higher than those of the Hernantian stage [24,36,42]. Biologically, there is a transition from the Hernantian stage to the early Rhuddanian stage, moving from brachiopods, trilobites, and the Hernantian mollusk fauna to a Graptolite community dominated by *Metabolograptus persculptus* [24,36,42]. This evidence suggests the end of the ice age and a shift towards global warming during the early TST of the Rhuddanian stage. Global warming led to glacier melting and a rising sea level in the TST of the Rhuddanian stage, where deep sea water gave rise to anoxic conditions, positively contributing to the preservation of organic carbon [24,36,42]. The widespread graptolites and radiolarians, as well as high Si_{ex} in the TST stage, indicate high paleoproductivity, leading to the deposition of siliceous shale (Figure 4, [4,6–12,15,36]). The high paleoproductivity could be attributed to volcanic activities and seasonal upwelling at this stage, based on its high value of $Mn \times Co$, which indicates that the Luzhou area was connected to open marine and related to North Qinling Ocean (Figure 6, [4–12]). The upwelling transported the dissolved silica and nutrients present in seawater, promoting the growth and prosperity of surface aquatic organisms [6–12] (Figure 6). As a result, high productivity, combined with the anoxic conditions, played a crucial role in the organic matter accumulation of TST. Moreover, the $Mn \times Co$ values of shale in Tongye-1 Well, located in the shallow shelf and closer to Qianzhong Uplift and Xuefeng Uplift, exhibit higher values compared to shale in the Luzhou area, which is situated farther away from uplifts (Figure 6, [6–14,27,28]). The data indicate a subsequent occurrence of few upwellings, resulting in fewer nutrients available, which, in turn, led to relatively lower productivity in Tongye-1 Well (Figure 6, [6–12,27,28]). Consequently, during the Early Rhuddanian epoch, it appears that robust seasonal upwelling events influenced the entire Upper Yangtze epicontinental sea. In contrast, shale developed at locations farther away from the provenances showed a greater impact from seasonal upwelling during the same period [6–12,18–22]. Moreover, terrestrial input indicators (K-Al and Ti-Al, etc.) suggest that minor terrestrial materials were input at the TST stage, which was increased significantly in the EHST-1, EHST-2, and LHST stages (Figure 7). Hence, the controlling factors for organic matter accumulation of the TST stage were characterized by high paleoproductivity, an anoxic environment, and low terrestrial input.

Minor fluctuations during the sea level fall stage are the primary reason for organic matter enrichment in the late EHST period. Low Mo_{EF} and medium Al content at the EHST stages indicate oxic conditions and medium terrestrial input (Figures 6 and 7). Additionally, the Si_{ex} value decreased, indicating a decline in paleoproductivity (Figure 4). Notably, the reduction degree in the late stage of EHST was enhanced compared with that at the early stage of EHST, indicating the occurrence of upward sea level fluctuation. Compared to the late EHST, the LHST period witnessed a continuous decline in sea level, a consistent increase in terrigenous detrital input, a sustained decrease in paleoproductivity, and predominantly oxidizing water conditions, leading to a relative lack of organic matter accumulation (Figures 4–10). This can be attributed to the intensive compression between Yangtze Block and Cathaysian Block during the LHST stage, which further decreased the relative sea level, amplified oxidizing conditions, and diminished nutrient supply from the open sea (Figures 4–10). The reduced relative sea level exposed uplifts surrounding Sichuan Basin, thereby intensifying weathering. As a result, terrestrial input was increased rapidly to dilute organic carbon content, as confirmed by high Al, K, Fe, Zr, and Ti contents and low TOC content (Figure 7). Furthermore, aluminosilicate can regulate the burial rate of organic matter through adsorbing organic matter, the transport flux of which can affect

organic matter concentration [45,67,70–72]. Hence, redox conditions and terrigenous inputs are important controlling factors for organic matter enrichment in the study area, followed by paleoproductivity. Moreover, the Cd/Mo vs. Co $\times$ Mn diagram is also used to prove the above view by identifying the impact of productivity, preservation, and water restriction on organic matter accumulation (Figure 11). Most samples from the TST stage in the study area are located in the productivity zone, while samples from the late EHST and the LHST stage are primarily positioned in the preservation zone (Figure 11), indicating that organic matter enrichment in the TST stage was mainly controlled by paleoproductivity, and that in the EHST and LHST stages was governed by a dysoxic environment, terrestrial input, and water restriction (Figure 12).

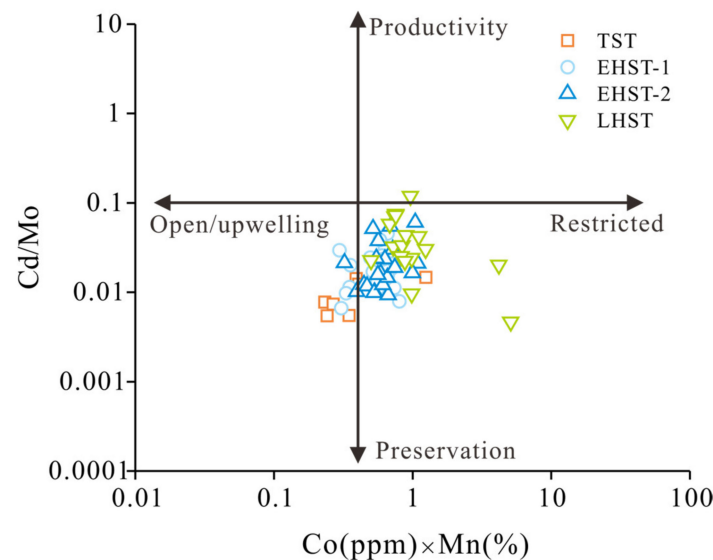


Figure 11. Cross plots of Cd/Mo vs. Co $\times$ Mn exhibiting the controlling factors for the organic matter enrichment of different system tracts during the TST, EHST-1, EHST-2, and LHST depositional periods. The Cd/Mo-Co $\times$ Mn diagrams were modified by [45]. In the Luzhou area, most of the TST samples are located within the productivity region, while the EHST-1, EHST-2, and LHST points are predominantly situated within the preservation area and are influenced by a certain degree of water restriction. This suggests that the organic matter enrichment in the TST stage is primarily controlled by paleoproductivity, whereas the organic matter accumulation in the EHST-1, EHST-2, and LHST units is influenced by a dysoxic environment, a higher content of terrigenous detritus, and the extent of water restriction.

5.3. Paleoenvironmental Evolutionary Model

During the Early Rhuddanian stage, the global temperature rose rapidly; as a result, glacier melting in Gondwana led to rapid transgression, and the sedimentation of TST occurred in the deepwater shelf (Figures 5–9, [71–74]). At that time, the Luzhou area was characterized as an isolated sag, with a sedimentary depocenter located in the southern part of the Luzhou area, where shale thickness reached up to 42 m [4,15,36]. Influenced by the compression from Qianzhong Uplift, Xuefeng Uplift, and Leshan–Longnvsi Uplift from both the north and south, the shale is oriented east–west and gradually thins towards the uplifts in both the northern and southern directions [42–48]. Given the significant increase in sedimentation rate from the Late Ordovician to the Early Silurian [4,15], this suggests that Southern Sichuan Basin underwent intense tectonic subsidence during the early Rhuddanian epoch. At this time, Sichuan Basin was bounded by the Central Sichuan ancient uplift, Kang-Dian ancient land, Qianzhong ancient uplift, and Xuefeng Mountain ancient uplift. The Jiangnan–Xuefeng tectonic belt exerted compressional forces on Sichuan Basin, resulting in flexural subsidence in Southern Sichuan Basin [4,15,36,73]. The sedimentary environment at this period was characterized by a rapid sea level rise, warm and humid climate, low sedimentation rate (0.6–0.8 cm/ky), low terrestrial input, anoxic conditions,

and high paleoproductivity (Figures 5–9). Rapid glacier melting resulted in a sea level at this period that was higher than that at the Wufeng period, connecting water bodies between the middle Yangtze and the Qinling ocean (Figures 8 and 9, [75–77]). Increasing water depth at low latitude led to anoxic water in the upper Yangtze, which was helpful in enhancing the water circulation and anoxic conditions in the bottom water along the continental shelf, and promoted the occurrence of upwelling (Figures 5 and 6, [71–74]). Meanwhile, the continuously rising ocean current brought with it many nutrients (Fe, Si, and P, Figure 2, [71–74]). The improved productivity of the ocean surface accelerated the consumption of dissolved oxygen in the bottom water, further aggravating anoxic conditions in the bottom water (Figure 4, [71–74]). Under this condition, biological remains and silica skeletons were rapidly settled and preserved under anoxic–euxinic environments, widely developing organic-rich siliceous shale (Figure 13).

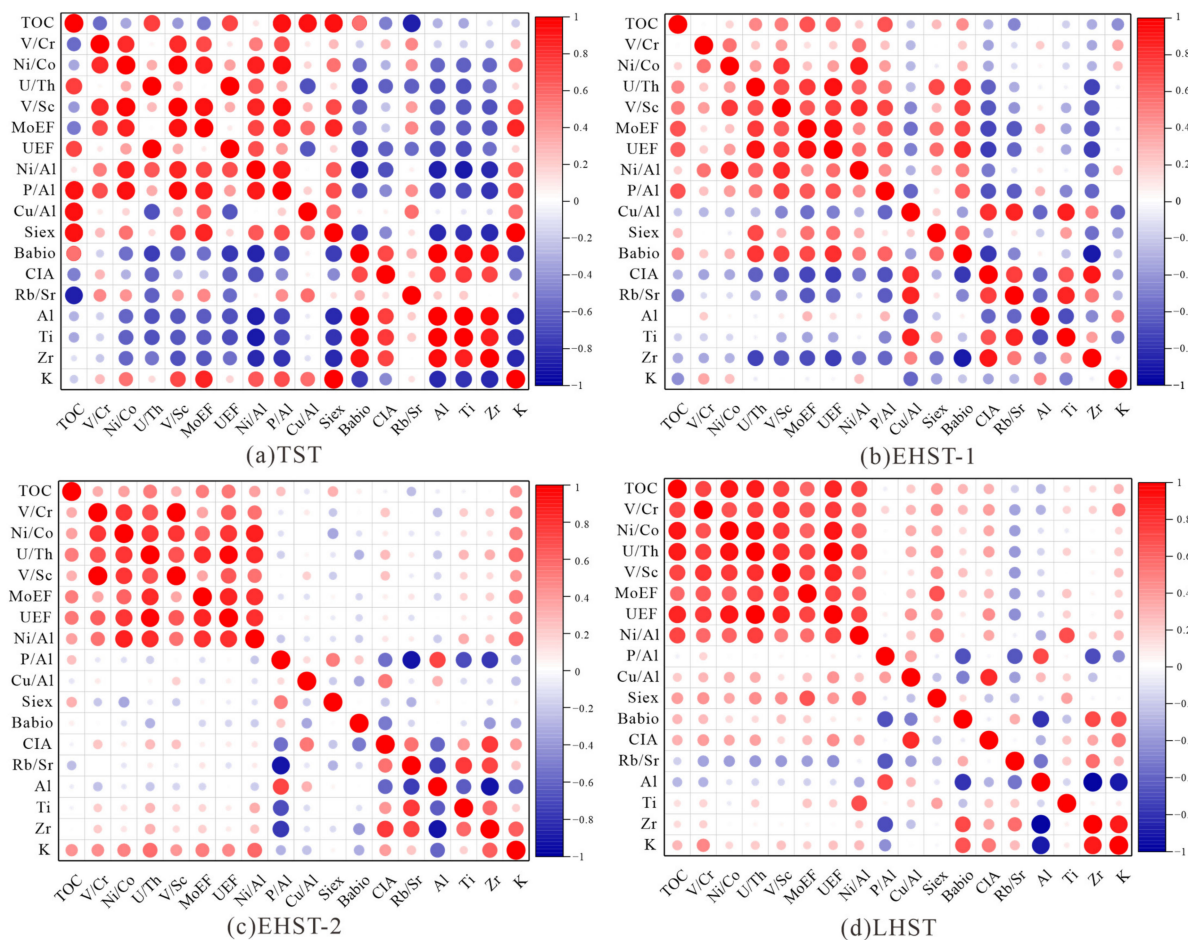


Figure 12. Heatmap for analyzing the correlation among geochemical characterization parameters for paleogeographic environments and TOC of different system tracts during the Rhuddanian–Aeronian transition. (a) The correlation among geochemical parameters and TOC in TST; (b) The correlation among geochemical parameters and TOC in the EHST-1 system; (c) The correlation among geochemical parameters and TOC in the EHST-2 system; (d) The correlation among geochemical parameters and TOC in the LHST system. The heatmap indicates that during the TST period, the correlation between TOC and paleoproductivity indicators (P/Al, Cu/Al, Si_{ex}, Babio) is the strongest, with a considerable correlation with redox indicators (U/Th, U_{EF}). From the EHST-1 to LHST periods, the correlation between TOC and redox indicators becomes increasingly pronounced (V/Cr, Ni/Co, U/Th, V/Sc, MoEF, U_{EF}). This suggests that the organic matter enrichment in the TST unit is more heavily influenced by paleoproductivity, while the organic matter accumulation in the EHST-1, EHST-2, and LHST units is more strongly controlled by a dysoxic environment.

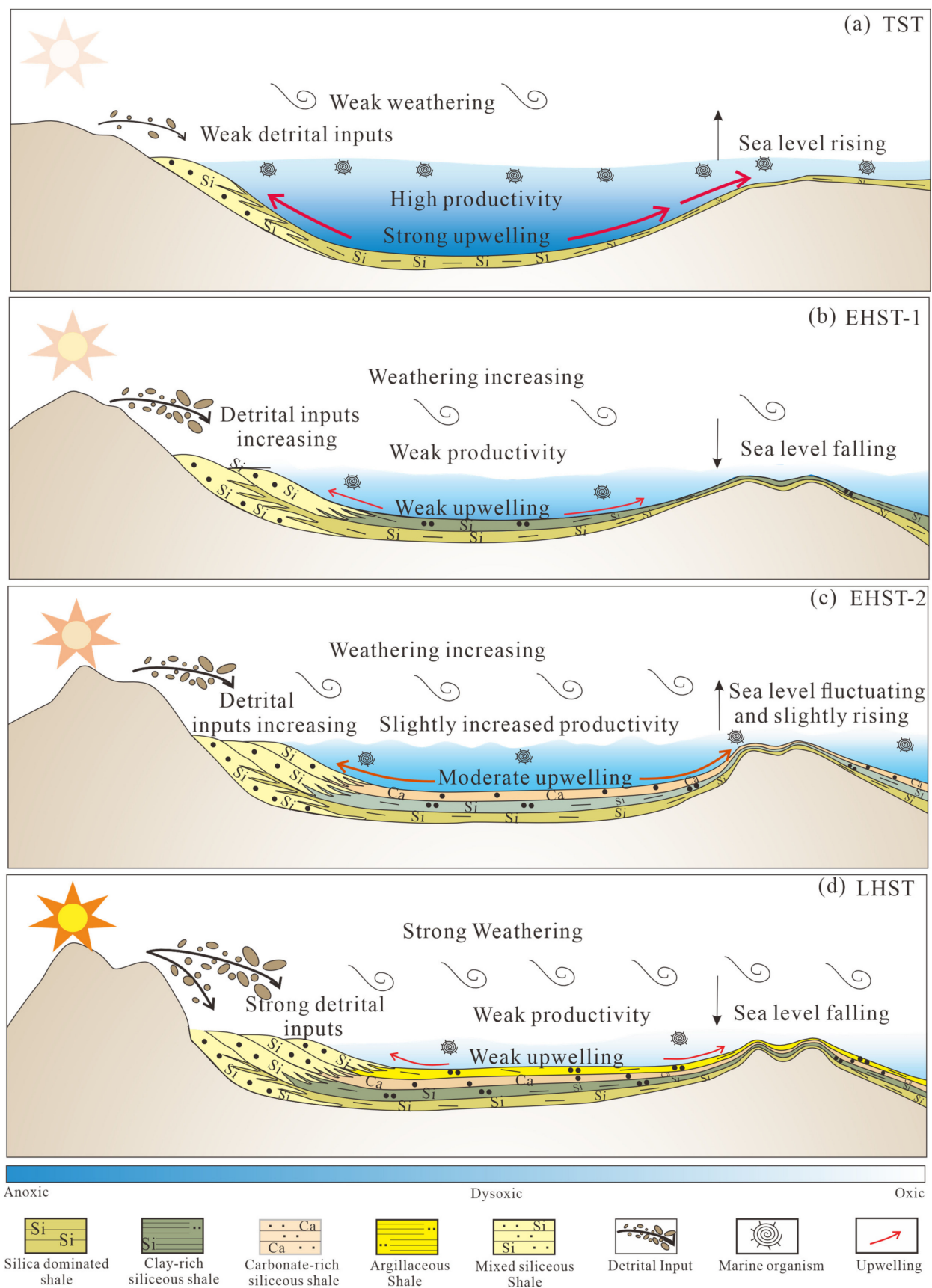


Figure 13. The paleoenvironmental evolutionary model for different system tracts during the Rhuddanian to Aeronian stage. **(a)** Early Rhuddanian stage: Gondwana glacier melting and rapid marine

transgressions. The TST stage showed rising sea levels, warmer climates, low terrigenous inputs, anoxic–dysoxic conditions, and increased paleoproductivity, favoring organic matter accumulation. (b) With the Kwangsian Orogeny’s progress in the late Rhuddanian, the Sichuan Basin’s adjacent areas uplifted, causing a prolonged regional sea level drop. Early EHST witnessed increased water column oxygen due to surface–bottom water mixing, intensified confinement in Luzhou sag, reduced upwelling, declining paleoproductivity, and increased terrigenous input. (c) Late EHST period: Sea levels exhibited minor fluctuations, the oxygen content in the water column slightly decreased, and there was a subtle variation in terrigenous detritus supply. These conditions favored a modest enrichment of organic matter. (d) LHST stage: Influenced by the Guangxi movement, areas like Chengdu and Xuefeng uplifted further, enhancing sedimentation rates. The rapidly declining sea level increased the water column’s oxygen and drastically reduced paleoproductivity.

During the late Rhuddanian to early Aeronian, ancient uplifts surrounding the Luzhou sag continued to rise, caused by Kwangsian Orogeny [78–84], and due to the barrier of the peripheral ancient uplifts, the separation between the Luzhou depression and North Qinling Ocean became more pronounced as a result of the continuous pushing of the Cathaysian block (Figures 2, 4 and 6, [23,63]). The depocenter was essentially retained, with the maximum shale thickness still situated in Southern Luzhou, reaching up to 52 m [53–57]. After the sea level reached its maximum flooding, it began to decrease gradually [32,37]. During this period, the Luzhou depression, in a calm and low-energy environment, experienced an enhanced terrigenous calcium and magnesium input, with a sedimentation rate of approximately 1.2 cm/ky [38]. In areas with strong bottom currents, scouring by these currents can also lead to a relative enrichment of coarse-grained minerals [28,32,42–44]. The shale deposited in the EHST stage displayed distinct bottom-current sedimentary traits, predominantly featuring interbedded argillaceous shale and clay-rich siliceous shale (Figure 4). Horizontal and lenticular bedding were prominent, with a higher clay mineral and sand content compared to the TST deposition stage (Figure 4), indicating a significant influence of the bottom current during the EHST. Moreover, the gray mixed shale of EHST was laid down in a semi-restricted setting with significant terrestrial influx, typically exhibiting a progradational basin fill (Figures 5–8). Normal regression was caused due to a rate of regional tectonic uplift higher than the rate of sea level rise, decreasing the distance between the provenance and the catchment area. The oxygen content in the water column was increased because of fully mixed oxygen-containing surface water and anoxic bottom water [71–73]. Lower organisms grew slowly under a stable ecosystem with fewer nutrients from volcanic activities and upwellings, where primary productivity fell to a medium value (Figures 4 and 5). Increasing terrestrial input gradually enhanced detrital mineral content (clastic quartz and clay), which consisted of primary minerals in shale. Due to the slight decrease in redox environment indicators, such as Mo_{EF} , U_{EF} , Ni/Co , and V/Cr , during the late period of EHST, oxygen content in the water column was decreased slightly, resulting in a dysoxic condition. This suggests a minor upward fluctuation in sea level at that time (Figures 4–6). Compared with the early EHST stage, the water restriction at this period was basically not changed, the paleoproductivity was slightly higher, the terrigenous debris supply fluctuated slightly and increased slowly, and the climate was warm and wet with enhanced weathering (Figures 4 and 6–9).

During the late Aeronian, tectonic compressional activities became significantly pronounced. The peripheral ancient uplifts further elevated, particularly in the southern regions encompassing Qianzhong Uplift, Xuefeng Mountain Uplift, and Leshan–Longnvisi Uplift, caused by Kwangsian movement [4,15,36]. Within this rift basin, the sedimentary thickness belt conspicuously contracted [4,15,36]. At this juncture, the depocenter of the Luzhou area migrated from the south to the northeast, towards the central–eastern parts of the Luzhou area, with a maximum thickness of 60 m. Overall, the shale developed in the sag was oriented from northeast to southwest and thinned further toward the north and south uplifts, with the depression continuing its contraction [4,15,36,42–48]. The LHST

accumulation coincided with a global transgression, yet experienced a regional regression [31]. As the ancient uplift continued to rise and the degree of weathering increased, the supply of terrigenous detrital inputs persistently increased, leading to a continuous input of terrigenous clay and carbonate minerals and an influx of Mg^{2+} , Ca^{2+} , and CO_3^{2-} . These elements from detrital minerals entered the Luzhou area and enabled the deposition of argillaceous shale and mixed shale (Figures 2 and 7–9, [4,15,36,42–48]). Meanwhile, the rapid decline in sea level rapidly increased oxygen content in the water column, further enhancing the oxidation of bottom water (Figures 4–6). Additionally, with the absence of volcanic ash nutrients, the growth rate of lower-level organisms declined [24,32]. This led to a moderate drop in primary productivity and a reduction in Si_{ex} , Ba_{bio} , and P precipitation. Moreover, the influence of bottom currents is even more pronounced in the LHST period (Figure 4). This is evidenced by an increase in fine sandy bands compared to the EHST stage (Figure 4), more developed mixed siliceous shale (Figure 4), and frequent occurrences of low-angle cross-lamination in the shale core [12,24], indicative of sedimentary structures controlled by low-velocity traction currents [12,24]. The influence of the bottom current during the LHST period was prominent and became the dominant factor affecting organic matter enrichment. The concentration of fine-grained sandstone increased, and the impact of bottom currents disrupted the water column conditions, leading to a predominantly dysoxic-to-oxic environment in the bottom waters. This period mainly witnessed the development of low-carbon, dark gray mudstone, and fine sandstone (Figures 4–9 and 13). During this time, argillaceous shale and mixed shale existed in a confined and suboxic environment with significant terrestrial input, typically accompanied by a progradational basin fill. The swift sedimentation rate (>6 m/Ma), combined with unfavorable preservation conditions, led to limited organic matter accumulation ([38], Figures 4–9 and 13).

5.4. Impact of Sedimentary Process for Shale Gas Sweet Spot Formation

The Rhuddanian–Aeronian shale development was directly or indirectly governed by a variety of geological events, e.g., paleostructure, paleoclimate, and paleoenvironment. The mineral composition and TOC variations in the shale can be used as comprehensive records of paleogeography evolution [75,83–85]. TOC, brittle mineral content, and porosity are key parameters of shale gas reservoir evaluation, which can directly characterize shale gas exploration potential. Comparing the key physical parameters and shale geochemical indicators of these four system traces (TST, EHST-1, EHST-2, and LHST) identified two types of shale gas sweet spot in the Rhuddanian–Aeronian shale reservoirs. Type I was developed in the early TST stage, while type II was developed in the EHST-2 stage (Figure 14).

Type I is characterized by high TOC, a high content of brittle minerals, and high porosity. Specifically, the brittle mineral content is about 71.5–87.9%, TOC is distributed between 2.8% and 4.7%, and porosity is 4.4–6.3% (Figure 14). A low bottom-flow rate, low clastic supply, slow deposition rate, anoxic conditions, and high paleoproductivity occurred during the TST deposition, contributing greatly to organic matter enrichment. The warming climate melted the Hernanian ice sheet rapidly, resulting in the development of a large-scale transgression, bringing in strongly anoxic conditions. Besides frequent volcanic activities, upwelling associated with convection between cold water generated by melted ice sheets and water from the equator provided abundant nutrition to further improve productivity [53–55]. Upwelling can significantly promote the productivity and input of organic matter, since it can bring nutrients from deep water to the surface, allowing plankton to flourish [53]. Furthermore, increasing organic matter input can consume oxygen to enhance reducibility, contributing greatly to organic matter preservation and enrichment [54,55]. Meanwhile, the terrestrial input was relatively limited, with low feldspar and clay mineral content. These factors jointly promoted the development of type I shale gas sweet spots [86,87]. Furthermore, the main lithology of type I shale gas sweet spots is silica-dominated shale with a high quartz content.

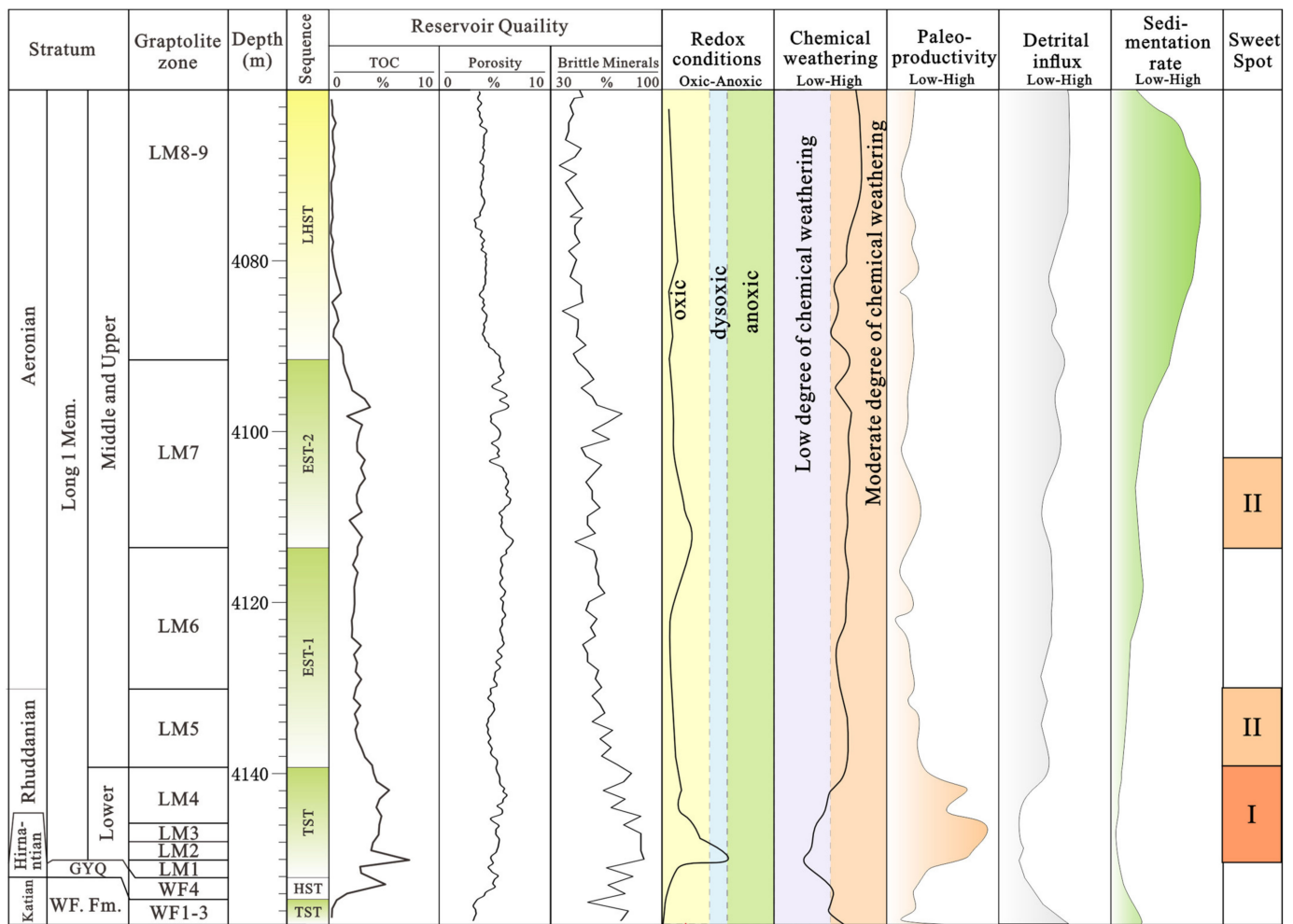


Figure 14. The genetic relationship between the paleoenvironment and the shale gas sweet spot development during the Rhuddanian–Aeronian transition of the Luzhou area. During the Rhuddanian–Aeronian period, two types of shale gas “sweet spots” developed. Type I sweet spots formed within the TST stage, characterized by high TOC, abundant biogenic quartz, and high porosity. Type II shale gas “sweet spots” emerged during the early Aeronian period in the late EHST. Minor, localized sea level fluctuations leading to increased anoxia in the bottom waters were the predominant controlling factor in the formation of type II shale gas “sweet spots”.

The high Siex content and occurrence of massive microcrystalline quartz confirm that the TST shale is dominated by biogenic quartz, with clastic quartz of secondary importance. The clastic quartz grains are characterized by a sub-rounded shape, large size (5–20 μm), poor euhedral properties, and well-developed dissolution pores [24,36,42]. Through our previous work, we found that quartz grains observed from TST shale mostly occurred as microcrystalline quartz (1–5 μm) with good euhedral properties and no fluorescence, which is similar to findings concerning biocrystalline quartz in other studies [24,36,42,47–52]. The biogenic quartz could be sourced from the dissolution and recrystallization of the opal skeleton of siliceous organisms (i.e., radiolarians and sponge spicules), whose reproduction during the TST stage was promoted by high paleoproductivity. Many studies report that biological microcrystalline quartz aggregates can exert a positive impact on pore network development, which can not only provide space for organic pore development, but can also protect intergranular pore networks via a rigid skeleton [16–22,32–34]. Furthermore, these quartz aggregates developing in the early stage have strong compressive strength. Hence, sliding joints are likely to occur between quartz aggregates and organic matter under external force, which can result in the development of effective seepage channels

during fracturing, promoting high and stable shale gas production. Therefore, high TOC and high microcrystalline quartz content in the TST stage were key to the development of type I sweet spots, where pore networks were supported by authigenic microcrystalline quartz, and a large number of organic pores and methane were generated from abundant organic matter [88–90].

Type II sweet spots were mainly developed in mixed shale in the later EHST stage, with an average brittle mineral content of 62.3%, TOC of 2.5–3.9%, and porosity of 5.6–6.1% (Figure 14), all of which were higher compared with the values in the early EHST. A series of geological events, e.g., a slight upward fluctuation of sea level, substantial sinking flux of organic carbon, diminished O₂ saturation caused by escalating global temperatures, and inadequate ocean ventilation during specific periods in South China, enabled the aforementioned circumstances to deplete oxygen and generate severe anoxic–dysoxic conditions, thereby augmenting the preservation of organic matter. During the EHST sedimentary period, as sea levels significantly declined, a minor fluctuation in the late EHST stage slightly intensified the bottom water’s anoxic conditions. This enhanced the preservation of organic matter, which predominantly influenced the development of the type II “sweet spot”. However, it is difficult to identify type II “sweet spots” in the Changning sag and the Weiyuan sag, indicating that the rise in sea level was not obvious in sags near the Luzhou sag. Therefore, sea level fluctuation at this stage only occurred at a small scale, and type II sweet spots developed in limited areas. The relative sea level during the EHST period was lower regionally compared with that at the TST period, where primary productivity was decreased significantly, and oxygen content in the water column was increased. Furthermore, increasing terrestrial input significantly diluted organic matter concentration and greatly increased clay mineral content, resulting in the weak compressive strength of the shale reservoir. Hence, primary pores were difficult to preserve, where intergranular pores and intragranular pores were further blocked during compaction. On the other hand, low organic matter abundance also resulted in a low yield of methane and organic pores. These above elements resulted in the low exploration potential of type II sweet spots compared with that of type I sweet spots, even though the sea level was risen slightly in the later EHST stage. However, the type I sweet spot in the TST stage at the bottom of Longmaxi Formation has been developed on a large scale in recent years. The type II sweet spot at the later EHST can not only serve as a back-up exploration target, but also be used to clarify the exploration potential of subsequent multi-layer integrated development [6,91–93].

6. Conclusions

1. Two third-order sequences (Sq A and Sq B) and four system tracts (TST, EHST-1, EHST-2, and LHST) developed during the Rhuddanian–Aeronian in the Luzhou area, southern Sichuan Basin. Paleoproductivity and redox conditions were primary controllers of organic matter enrichment in the TST stage, followed by terrestrial input. Redox conditions and terrestrial input were the primary controllers in the EHST-1, EHST-2, and LHST stages, with paleoproductivity being of secondary importance.
2. During the Rhuddanian stage, melting Gondwanan glaciers caused a rapid marine transgression. This period was marked by rising sea levels, warm and humid climates, low terrigenous input, anoxic–dysoxic conditions, and increased paleoproductivity, promoting organic matter accumulation. By the late Rhuddanian, the Kwangsiang Orogeny had led to gradual uplift in areas adjacent to Sichuan Basin, causing a regional drop in sea level. In the early EHST, the enhanced mixing of oxygen-rich and anoxic waters increased overall oxygenation. The Luzhou sag’s degree of water restriction grew, weakening upwelling currents, reducing paleoproductivity, and increasing terrigenous inputs. In the late EHST, the sea level fluctuated slightly upwards with increased weathering and a warmer, more humid climate. During the LHST, deposition began, influenced by the Kwangsiang uplift. The Chengdu, Xuefeng, and Qianzhong uplifts further rose, leading to more weathering and the influx of

terrigenous inputs. A decline in sea level enhanced bottom-water oxygenation. The degree of water restriction in the Luzhou sag intensified, alongside a sharp decline in paleoproductivity in an increasingly warm and humid environment.

3. During the Rhuddanian–Aeronian period, two types of shale gas sweet spots emerged. Type I shale sweet spots were found within the TST stage, distinguished by their high TOC (4–7%), significant biogenic quartz content (>30%), and high porosity (4–7%). They were predominantly influenced by factors like low terrigenous detrital inputs, slower sedimentation rates, developed upwellings, anoxic conditions, and elevated paleoproductivity. The authigenic microcrystalline quartz enhanced the reservoir's compressive strength, preserving the primary porosity, while the abundant organic matter gave rise to numerous organic pores and methane during the gas generation stage. Type II shale gas sweet spots originated during the late EHST stage. The reservoir qualities of type II shale gas sweet spots are second only to those of type I shale gas sweet spots, characterized by a TOC distribution of 5–6.5%, porosity ranging from 4 to 6%, and a notably high content of detrital quartz. They were principally controlled by minor, localized sea level fluctuations, causing increased anoxia in the bottom waters. The significant influx of terrigenous clastics strongly diluted the organic content, resulting in a lower degree of organic accumulation and a reduction in the shale reservoir's compressive strength. The discovery of type II sweet spots from the late EHST can serve as backup exploration target, following type I.

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