

A Study on the Indicative Role of Geochemical Markers Based on Sediment Phase Analysis

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Abstract: Sedimentological research is developing from the basis of qualitative description to the direction of dynamization and quantification. As a bridge between macroscopic depositional environment and microscopic material elements, geochemical signatures will certainly play an increasingly important role in promoting. Geochemical methods can analyse on a microscopic scale, relying on modern analytical techniques. Combined with core and logging data, researchers can gain a more scientific understanding of the sedimentary environment. This research describes the background and current status of geochemistry in the field of sedimentary phase analysis, and focuses on the mechanism of formation of sedimentary geochemical signatures and the specific application of geochemical signatures. The significance of material sources in controlling elemental signatures is first described, and the modification of geochemical indicators by weathering and other sedimentary processes is explained from diagenesis. For the major geochemical signatures, this research introduces the elemental combinations and applications of major elements, trace elements, and organic geochemical signatures. This research emphasizes the application of geochemical markers in sedimentary environments (sea and land phases, redox conditions and paleosalinity) and source analysis. This research is a reference for future scholars to analyze the sedimentary phases by applying geochemical signatures, and it will promote the transformation of sedimentology into a quantitative direction.

Keywords: Geochemical Signatures, Sedimentary Phase Identification, Elemental Ratios, Elemental Content, Depositional Environment.

1. Introduction

The development of sedimentary geochemistry is of great significance for the application of geochemical phase signatures in sedimentology such as the identification of depositional environments. Sedimentology, developed from sedimentary petrology, is a discipline that studies the origin of sediments, the environmental conditions at the time of deposition and their formation processes (sedimentation, diagenesis), and the laws of sediment mineralization. Geochemistry is the study of chemical components and their evolution in geochemical systems. Chemical elements often have the same characteristics in the same depositional environment due to internal factors (their own properties) and external factors (environmental conditions), and show differences when environmental conditions are different. By using the characteristics and patterns of migration, aggregation, and distribution of elements, isotopes, and trace elements, the method of studying sedimentary environments is becoming more and more mature and important. Geochemistry can provide the key for sedimentological research to enter the microscopic scale from the macroscopic scale, which is conducive to the realization of multi-dimensional analysis of sedimentary phases.

The application of geochemistry in discriminating sedimentary environments mainly includes the following aspects: discrimination of marine and terrestrial stratigraphy, discrimination of redox conditions, reduction of paleoclimate and paleosalinity evolution history, paleo bathymetry as well as changes in sea level, and discrimination of stratigraphic exposures. For example, previous studies have found that Sr/Ba values are more suitable for discriminating the paleosalinity of sedimentary waters than trace elements such as B, K, and Ga [1]. Redox-sensitive elements (RSE) are

significant for the discrimination of Eh values in the environment. In oxidizing environments, RSE tends to migrate through seawater as ions, whereas in reducing environments it is stable in the low valence state, and these properties of RSE make it an ideal indicator of the redox state [2]. High salinity depositional environments are often accompanied by the presence of gamma waxanes, whose indices are very indicative of water salinity [3]. The nature of Mn, Ca, Mg, and other elements that can be transported over long distances determines that the MnO/TiO₂ and Mn/Fe ratios can be used as a qualitative indicator to determine the offshore distance (paleodepth) [4]. In addition, geochemical signatures can also be used to determine stratigraphic sequences and to study sediment sources.

The formation mechanism of sediment geochemical signatures and the specific application of them in sedimentary phase analysis will be introduced by the research respectively. Among them, part of the formation mechanism of geochemical signatures will be based on the development process of sedimentation, starting from the source of material to the modification of weathering-transportation. After that, the classification of the main geochemical indicators will be introduced. The research will find that geochemical signatures can accurately differentiate between different sedimentary environments by quantifying key parameters. For the specific application of sedimentary phase analysis, the research will explain and give examples of the main directions of sedimentary environment identification: identification of land and sea phases, determination of redox conditions, and application of geochemical markers in the recovery of paleosalinity. This research will distinguish between the concepts of paleosalinity restoration, which aims at restoring continuous changes in the salinity of sedimentary waters, and land-sea phase identification, which favors the

restoration of macroscopic properties of the environment.

2. Mechanisms of sediment geochemical characterization

The application of geochemical signatures in the field of source tracing is mainly through the analysis of the geochemical characteristics of the material, combined with its distribution and migration behavior to effectively reveal its source and migration path, where the mechanism is enrichment and diffusion of chemical elements in different media due to physical, chemical, and biological interactions. This research will elaborate on the three aspects of source lithology control, the influence of weathering-transport processes on geochemical indicators, and the classification of major geochemical indicators.

Source supply analysis is of great significance in reconstructing paleogeographic environments and studying sedimentary phases. The characterization of its components usually has a strong correlation with the type of parent rock and even plays a decisive role. The material components in the host rock are usually modified into primitive sediments with different material compositions after chemical weathering. However, during the weathering process of rocks, active elements are easily lost, while others are recalcitrant. Yan et al. proposed the concept of “geochemical genes”, using the stability of inactive elements (e.g., Al_2O_3 , SiO_2 , P, and Ti) to construct gene sequences and verify their heritability in weathering products. This move further proves the control role of physical source lithology on geochemical indicators [5].

The soil formation process uses the products of rock weathering and crushing or Quaternary loose deposits as the parent material. Through physical, chemical, and biological soil-forming processes, the composition, structure, and architecture of the original parent material are changed to a large extent. And the material gradually transformed into a soil that supports biological growth and fertility. The factors in the soil formation process have a profound influence on the chemical composition of the soil profile. For macronutrients, the activity sequence $\text{Ca} > \text{Na} > \text{K} > \text{Mg} > \text{Si} > \text{Al} > \text{Fe}$ is prevalent. For example, in the subtropical monsoon climate of Yunnan, sufficient precipitation and heat make soil formation very strong. As a result, desilication, aluminum-rich iron action, and chemical decomposition are prevalent. Cations and silica are lost and iron and aluminum oxides are relatively enriched, resulting in the formation of Fe-Al soils with typical red soil properties in Yunnan [6].

About the classification of major geochemical indicators, geochemical signatures reveal the ancient sedimentary environment, material source evolution, and rock-forming process through the distribution pattern and combination characteristics of elements (major and trace elements), isotopes and organic components. With the development and progress of the times, the multidimensional classification system centered on trace element ratios (e.g., Sr/Ba and Fe/Mn), isotope systems (e.g., $\delta^{13}\text{C}$ and $^{87}\text{Sr}/^{86}\text{Sr}$), organic geochemistry parameters (e.g., C/N and HI), and rare earth element compositions is maturing and growing. The advantage of geochemical indicators in the study of sedimentary phases lies in the fact that when the lithology does not allow finer discrimination of sedimentary environments (“syncline” problem), geochemical signatures can accurately differentiate between different sedimentary

environments by quantifying key parameters such as palaeosolubility, redox state, sedimentation rate, and source of organic matter, providing a new way of thinking and stronger support for sedimentary phase identification and research. The classification of geochemical indicators is described in detail from the perspectives of major elements, trace elements, and organic geochemistry.

For the major elements (such as Na, Mg, Al, Si, and P), their contents are high and simple to test. They have a wide range of applications in source, weathering, elemental cycling, and geologic response. For example, Si element can be used for source analysis, an indication of the strength of hydrodynamics, as well as redox conditions and chemical component characteristics of sedimentary waters. Different depositional environments at the time of shale formation can be identified by measuring the abundance of Si in the sediments. Since SiO_2 , the oxide of Si, is mainly enriched in quartz, feldspar and other terrestrial detrital minerals, the gradual decrease of SiO_2 content corresponds to the deepening of the paleo-water depth (e.g., the gradual transition from a shallow lakeshore environment to a semi-deep lake or a deep lake). Meanwhile, the trend of Si and other major elements can also indicate the abundance of organic matter in the sedimentary environment, and higher input of land-sourced detrital minerals (higher Si content) implies that organic matter is dispersed, and its abundance will naturally decrease [7]. The oxides of other macronutrients, such as Si, Ca, and Na, can be used to indicate the source and diagenetic environment due to their stable and non-migratory chemical properties.

Compared with the major elements, trace elements are usually found in natural systems in amounts less than 0.1% or 1%, and their concepts are relative. Trace elements are categorized based on their different behaviors and modes of distribution in the geochemical system, including rare earth elements (REEs), non-rare earth elements, and rare gas elements. Although the content of trace elements in the earth's crust is relatively low, the status of trace elements should not be underestimated in terms of indicating the depositional environment, tectonics, and mineralization. For example, in the sedimentary environment analysis of the sedimentary rocks of the Yanghugou Formation at the western margin of the Ordos Basin, Lu et al. selected Sr, Ba, U, Th, and V as the evaluation discriminants, and analyzed the sedimentary rocks of the Yanghugou Formation in conjunction with other sedimentary phase indexes. The elemental contents of Li and Sr can indicate the paleo-salinity conditions of the freshwater-mini-brine water, whereas the occurrence of seven samples with high Ni contents in all the mudstone samples can indicate occasional brackish water injection events under overall freshwater-slightly brackish water conditions. Taken together, the mudstone samples from this site are from a transitional phase environment between land and sea. [8]. Ji et al. used Sr and Ba as indicators in analyzing the relationship between trace elements and their ratios and depositional environments in the Baosquanling Formation of the Tangyuan Fault, and the test results showed that the Sr/Ba ratios were low (all less than 0.6), indicative of terrestrial freshwater environments, and the Sr/Cu ratios were high (most of them were less than 10, and some of them were greater than 10), indicative of a warm, wet-dry, and hot climate [9].

Organic geochemical markers are to some extent equivalent to biomarker compounds. After biogenic sediments are modified by the environment and diagenetic

processes, some of the indicators (elemental ratios and compound contents) will respond to the formation process and be left behind, so they can be used to invert the depositional environment and material sources. For example, Yang illustrated the significance of the application of biomarkers by taking tricyclic terpene compounds as an example. Yang pointed out that when the ETR ratio is less than 2.0, there are conditions of freshwater, weak oxygen or anoxia, and a large amount of land-source organic matter input in the depositional environment. And when the ETR ratio is greater than 2.0, the depositional environment is less affected by the input of organic matter from land sources, and the water body is in the condition of high salinity and high reduction. In addition, the different distribution characteristics of tricyclic terpene alkanes correspond to different depositional environments. The changes in carbon number distribution can also be utilized to explore the

maturity level [10]. In order to investigate the depositional environments of crude oils in different areas of the northern Melut Basin, Shi et al. carried out saturated hydrocarbon chromatography and chromatography-mass spectrometry (CHM-MS) analyses of crude oils in the study area to obtain their organic geochemical characteristics. The results show that the gamma waxane index is generally less than 0.1 and the Pr/Ph values are in the range of 0.97-2.31. These indicators can be indicative of a weakly reducing-weakly oxidizing freshwater lake phase formation environment for hydrocarbon source rocks [11]. Compared with other indicators, the advantage of organic geochemical indicators is that when they combine organic matter maturity with inorganic features such as depositional environment and redox conditions, they can realize the multi-dimensional identification of sedimentary phases.

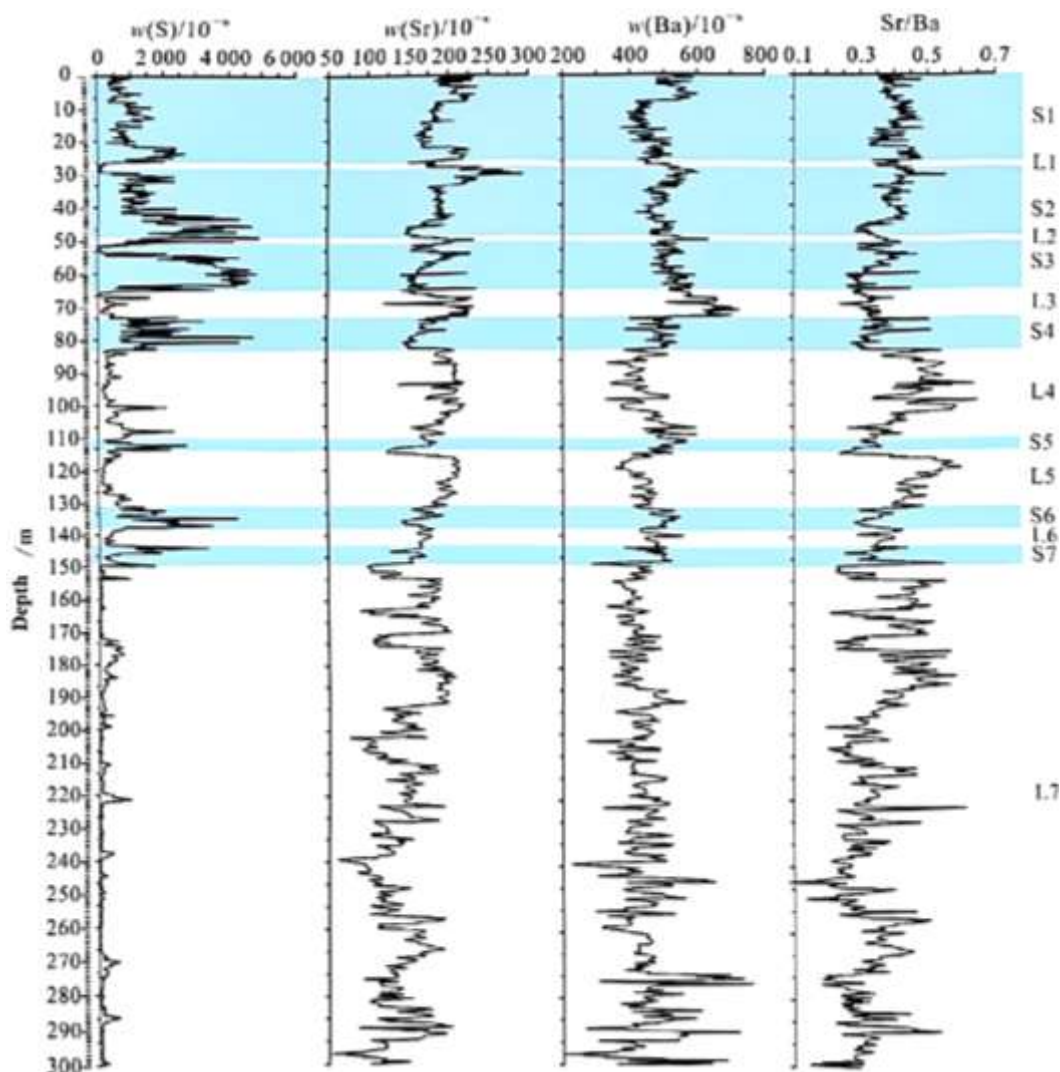


Figure 1. Comparison of distributions of S, Sr, Ba, Sr/Ba in the Core CSDP-1[12]

3. Application of geochemical signatures in sedimentary phase resolution

3.1. Identification of depositional environments

Geochemical signatures are widely used and flourishing in the field of sedimentary environment identification and reconstruction, whose means is to use the geochemical

behavioral differences of elements, isotopes, and organic molecules to explain the physicochemical properties of the paleo-water medium, which provides quantitative and multi-dimensional evidence for the identification of land and sea phases, the restoration of redox environments and the recovery of paleo-salinity conditions. Geochemical signatures for the identification of sedimentary environments are based on a theoretical system that includes elemental geochemical differentiation (caused by differences in the

migratory capacity of elements), isotopic tracing principles (isotope fractionation effects and source area characteristics that record geologic activity), and biomarker responses. The development of modern analytical techniques and the synergistic system of multi-indicators is continuously injecting new vitality into the application of geochemical indicators in the field of sedimentary environment identification, which significantly improves the resolution and credibility of the interpretation of sedimentary phases.

In recent years, research on geochemical signatures in recognizing sea-land phases has focused on the improved application of the Sr/Ba ratio, which in combination with other elements and their ratios (S, B/Ga, TOC/Tn, K/Rb, K/Na, V/Zr and Cu/Zr) can significantly improve the discrimination accuracy of sea-land phase environments. Mei et al. used the mass fractions of S, Sr and Ba elements and the changes of Sr/Ba ratios of different sedimentary units in Hole CSDP-1 in the South Yellow Sea to explore the relationship between seafaring elements and sea-land external geologic forcing with sea-land depositional environments and their changes [12].

As shown in Figure 1, the fluctuation curve of the mass fraction of element S in the stratigraphy (with special attention to the critical point of appearance or disappearance of element S) can provide a basis for judging the sea-land phase. The results show the S element develops in the form of local fluctuation and general increase. This indicates that the stratigraphy of the hole gradually developed from the terrestrial depositional environment of Unit U3 (3.50-1.66 Ma) to the marine depositional environment (since 0.83 Ma). Accompanied by the increase in S elemental mass fraction, the control of sedimentation by rivers relative to the ocean gradually weakened, and the marine environment gradually took the dominant role. However, the significance of the Sr/Ba ratio (terrestrial <0.2, marine >0.3) is not obvious in the hole, and only the ratio can be analyzed to be correlated with the enhancement of marine nature. When recovering the paleoenvironment of the lake basin of the Zhuyi depression, Xie et al. mentioned that the stronger activity of B and Ga in nature makes the two enriched in clay minerals and their ratio can be used as a basis for judging the paleosalinity [13]. When B/Ga is less than 1.5 is the freshwater phase, 5~6 is the nearshore phase, and >7 is the marine phase. As the ratio increases, the contribution of the marine environment to sediment formation increases. Freire et al. used the TOMI index combined with the TOC/TN ratio in quantitatively assessing the contribution of land-based genesis to marine sediment formation in the study area of the Joetsu Basin in the Sea of Japan, and the TOMI index can be judged as marine sedimentation, and when TOC/TN > 20, it is land-sourced sedimentation [14]. It is worth noting that studies have shown that during the ice age, which is a period of low sea level, TOC/TN > 15 corresponds to an increase in input from land-based sources.

Determining redox conditions in sedimentary waters is another important aspect of geochemical signatures in the field of sedimentary phase interpretation. The enrichment pattern of RSE and ratio analysis are important means to determine the redox conditions of ancient water bodies. The elements and their ratios frequently used in current studies include V/Cr, V/(V+Ni), Ni/Co, U/Th and Ce. The element U is more active and is easily transported by oxidation and leaching. Comparatively, the element Th is more inert and is usually preserved in an adsorbed state, which is not sufficient

for its transport. With insufficient migration ability, it is usually preserved in fine-grained sediments. Due to the different geochemical properties, the U/Th ratio can be used to determine the redox state of sedimentary waters. U/Th < 0.75 can generally indicate oxidizing environments, while U/Th > 1.25 can indicate reducing environments [15]. The chemical activity of element Ce in different redox conditions varies greatly. Under oxidizing conditions, Ce³⁺ is easily oxidized, whereas the high valence Ce⁴⁺ is difficult to dissolve, so there is a deficit of elemental Ce in sedimentary waters, whereas there is an abnormal enrichment of elemental Ce in sediments. When the environment is transformed into a reducing environment, the high-valent Ce⁴⁺ is reconverted into Ce³⁺, which is then released back into the sedimentary water body. The use of rare earth element Ce anomalies to determine the redox conditions of the sedimentary water body has its rationality [16]. In addition, there are still many current judgment indicators for determining redox conditions such as TS/TOC, TOC/P, and Cu/Zn [17].

Paleosalinity restoration and land-sea phase identification are both core tasks in sedimentary environment reconstruction, but there are some differences between them, while at the same time, they are profoundly related. Salinity is defined as the mass fraction of all soluble salts in a medium, and paleosalinity recovery aims to quantitatively or semi-quantitatively recover the salinity value of a sedimentary water body, focusing on the continuous change of salinity. In contrast, land and sea phase identification focuses on determining the macroscopic properties of the sedimentary environment and dividing the sedimentary environment into marine, terrestrial, and transitional phases. Both tasks, share common geochemical mechanisms (e.g., elemental partitioning effects), but paleosalinity recovery differs from land-sea phase identification in terms of technical pathways.

For paleosalinity recovery, the combination of elemental ratios and isotopic indicators, and the correction of test results with regional hydrological characteristics all play an important role in improving the objectivity of analytical results. This research emphasizes the role of isotopic indicators. Due to the correlation between isotope fractionation and salinity, isotopes can well indicate the paleosalinity of sedimentary waters. For example, C isotopes and O isotopes can be used to recover paleosalinity either individually in the range of -5‰ to 15‰ is freshwater, and in the range of -5‰ to 5‰ is seawater [18]. The isotopic composition in the shells of foraminifera in the ocean in different periods can be used to reconstruct the paleosalinity of seawater. An important prerequisite for the application of this method is that researchers must try to extract as much calcite information as possible from the biogenicity of the life-span stages to avoid contamination after deposition and burial [19]. The isotope establishes a quantitative relationship with paleosalinity through the fractionation effect, breaking through the semi-quantitative limitation of the traditional elemental ratio method. Meanwhile, its high sensitivity and wide dynamic range lay the foundation for its wide application [18].

3.2. Source analysis of material

For stable isotopes, epigenetic geochemistry makes it difficult to change their content. Since the early 1960s, stable isotopes have been used to quantitatively characterize the origin of media. Until today the application of stable isotopes has included the three major rock groups in the lithosphere,

substances in the atmosphere, biosphere, and hydrosphere [20]. The composition of stable isotopes in the samples of the study area can be determined to distinguish the material sources of different geological bodies. For example, Zhang analyzed carbon, oxygen, and magnesium isotopes in rocks from the lower part of the Wufeng Formation-Longmaxi Formation in eastern Sichuan to study the paleoclimate evolution during the Late Ordovician-Early Silurian transition as well as the mechanism of organic matter enrichment [21]. Through the determination of H-O isotope compositions, the water-rock interactions during the formation of hydrothermal deposits can be quantitatively investigated, and then determine the source of mineralizing fluids [20]. Li and Cl isotope compositions also respond well to the elemental migration activities of the basin brines, groundwater, and hydrothermal mineralization. For the evolution of C(O) isotopes in the mineralization system, it is difficult to distinguish the sources due to some overlapping characteristics of carbon isotope compositional features from different sources. If researchers do not make a simple comparison, but further trace the sources on the basis of the isotope system evolution study as well as the reduction of the mineralization environment. The accuracy of C(O) isotopes in providing information on mineralization sources will be greatly improved. Radioisotopes have a monitorable half-life compared to stable isotopes (e.g., ^{239}Pu has a half-life of 24100 years) [22]. They are rare in nature, but relying on their sensitivity and the convenience and non-destructive nature of detection, thus they are able to accurately locate and quantitatively analyze the dynamic changes of substances. The radioisotopes still play an important role in the field of source tracing. Using different radioisotope compositions and ratios, it is possible to date minerals in order to restore the thermal evolutionary history of the source area, identify the amalgamation of multiple sources, and indicate the lithology of the host rock. For example, zircon, as a carrier of the U-Pb system, is readily available, stable, closed, and has a very high closure temperature. These characteristics of zircon allow the zircon U-Pb age system to preserve more realistic and effective information about the source region [23]. The closure temperatures of different isotopic systems such as Rb-Sr, Sm-Nd in zircon, apatite, hornblende, black mica, and other minerals can be used to study the cooling history of orogenic zones, cooling trajectories of endorheic rocks, tectonic movements of orogenic zones, and uplift and denudation relationships, which ultimately involves the change of sedimentary environments after the orogenic movements.

3.3. Prospect

In recent years, the advanced measurement technique, isotopic composition measurement of microcellular in situ radioactive systems, has been rapidly developed by using analytical methods such as laser ablation-multi-receiver inductively coupled plasma mass spectrometry (LA-MC-ICP-MS) and secondary ion mass spectrometry (SIMS), and the results have received wide attention from the international geoscientific community. Its advantages include shortening the analysis period, targeted analysis of different or different parts of the same mineral, and the geological significance of the reaction evolution process. In the future, micro zonation in situ isotope analysis will continue to advance in the fields of improving the transmission rate of stripped samples, analytical sensitivity, and multivariate measurements, which

will continue to promote the advancement of geochemical tracer research.

4. Conclusions

This research discusses the importance of sedimentary geochemistry as a cross-discipline between sedimentology and geochemistry, and points out the reason why geochemical signatures can be used to study sedimentary environments. The various applications of geochemistry can be used for the identification of sedimentary environments. In the part of the formation mechanisms, this research mainly analyzes the decisive role of host rock type on sediment components and the unique advantages of geochemical methods in source tracing, describes the modification of soil composition by weathering-transportation process and the classification of major geochemical markers. It explains the application and advantages of each index in the study of sedimentary phases in detail.

The identification mainly includes the discrimination of sea and land phases, the reduction of redox conditions in sedimentary water, and the recovery of paleosalinity. This research also distinguishes between land and sea phase identification, which focuses on determining the overall properties of the environment, and paleosalinity restoration, which focuses on obtaining specific values. This research emphasizes the important role of isotopic indicators. In terms of physical source analysis, stable isotopes can be used to distinguish the material sources of different geological bodies, while radioisotopes can analyze the precise location of material dynamic changes. For the elemental (ratio) indicators involved in this research, their commonality lies in the fact that differences in depositional environments can lead to large differences in their geochemical behaviors, resulting in recognizable differences in the contents of the same elements (isotopes) or between different elements. The quantitative analysis of sedimentary phases can be realized by choosing appropriate elements and combinations.

The application of geochemical signatures in sedimentary phase analysis faces the problems such as complex elemental geochemical behaviors and high requirements on sample quality. Researchers can further study the formation mechanism and influencing factors, strengthen the cross-fertilization of geochemistry with other disciplines. The use of big data and artificial intelligence will be of great help in the establishment of models and visualization.

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