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Surficial sedimentary properties of Makran accretionary prism and its application to natural gas hydrate distribution

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The Makran accretionary prism (MAP) resulted from the low-angle, slow-rate subduction of the Arabian Plate beneath the Eurasian Plate. The MAP is an active continental margin, with natural gas hydrate (NGH). In this study, we report an integrated study on surficial sediments, in terms of sediment grain-size, clay minerals and elements. The main results are as follows: (1) the surficial sediments were mainly controlled by hydrodynamic conditions of the Indus River. Along the Indus River channel, it is mainly infilled with silt, while clays are mainly distributed in the accretionary prism. The turbidity records gradually decrease from east to west, along the Indus River channel. (2) Illite and chlorite are the dominate clay minerals, indicating that the surficial sediments were mainly from weathering processes in the Himalaya by the Indus River transport. The very-low contents of kaolinite and smectite indicate the lack of sediments from the Deccan Traps and the *in situ* hydrothermal materials. (3) Elements analysis indicates that elements Al, Mg, K, Fe, and Ti were terrigenous, mainly distributed in the northern MAP, while elements Ca and P were from biogenic sources, mainly distributed in the southern part of the study area. In addition, elements U and Mo distribute generally coupled with bottom simulating reflector (BSR), which can be used as proxies of NGH identification. Therefore, we proposed that the covariation between NGH distribution, BSR identification, and trace elements Mo and U holds the potentials in NGH investigation.

KEYWORDS

sedimentary progress, sediment provenance, clay minerals, natural gas hydrate (NGH), Makran accretionary prism

1 Introduction

The Makran Accretionary Prism (MAP) in the northeastern Arabian Sea resulted from a low-angle (typically < 3°) subduction of the Arabian plate beneath the Eurasian plate (Grando and McClay, 2007). The complex geological structure of this region involves a series of imbricate thrust fractures, including a near east-west thrust fold belt and a deep-sea plain and its inter-transition zone (Gong et al., 2018; Zhang et al., 2003).

The MAP is separated from regions of continental collision in the east by the left-lateral Ornach-Nal fault and in the west by the Minab Fault system. The Jaz Murian and the Hamun-i-Mashkel depressions located between the Makran coastline and the Baluchistan volcanoes are interpreted as forearc basins (Jacob and Quittmeyer, 1979). This large trench-arc distance makes the Makran the widest forearc complex in the world, with an accretionary prism of up to 300km in width (Smith, 2013). The age of the oceanic crust in the study area was considered in the Cretaceous “quiet” zone, because there is no identifiable sea-floor magnetic anomalies between the MAP and Murray Ridge (Whitmarsh, 1979). And heat flow data also suggest an age of between 70–120 Ma (Hutchison et al., 1981; Smith, 2013).

Seismic surveys along the MAP reveal that the sediment thickness at its deformation front is very high, up to 7km (Kopp et al., 2000), with frequent turbidity layers in the north. Before the early Miocene, the Himalayas was the dominant sediment source, and since the Murray Ridge system uplifted, the Indus River was isolated (Gaedicke et al., 2002; Rodriguez et al., 2014). Subsequently, sand and aeolian dusts from the Arabian Peninsula, with riverine materials from the northern arid regions turned to be the main sediment source of the MAP (Kukowski et al., 2001; Mouchot et al., 2010).

Previous studies suggests that the deposition process of the MAP is controlled by several factors: (1) The Indus River in the northeast carries a high flux of terrestrial debris, while the floods carry coarse-grained terrestrial debris as turbidity deposit (Bourget et al., 2011); (2) Winter winds generated by low-pressure cyclones in the Mediterranean region cross the Arabian continent carry a large amount of dust materials into the Arabian Sea (Prins et al., 1999; Clemens and Prell, 2003); (3) Marine organisms in the Arabian Sea and erosional materials from the seafloor also have some contribution (von Rad et al., 1999; Luckge et al., 2001; Fleitmann et al., 2007; Thamban et al., 2007).

Natural gas hydrate (NGH) is an efficient, clean energy with a broad distribution in the world, and is considered as an ideal alternative energy in the future (Milkov, 2000). Global NGH investigations indicated that an NGH system usually locates on the slopes of passive continental margins, often in large contourite deposits, or in an accretionary wedge on subduction margins (Wang et al., 2010). Among them, unique geological units, such as faults, folds, landslides, mud diapirs, and mud volcanoes, are commonly identified (Xie et al., 2013), suggesting the great potential of the MAP as one of the important NGH regions in the world.

Several scientific survey, such as the joint expedition by the R/V Sonne in 1993 and 1997–1998, the R/V Meteor in 2007, and the R/V “Hai Yang Di Zhi Shi Hao” in 2019–2019, found many activities of fluid escape and NGH evidence in the MAP (von Rad et al., 2000; Kukowski et al., 2001; Bohrmann et al., 2008; Römer et al., 2012; Wei et al., 2021; Zhang et al., 2021), particularly the finding of NGH in water depths of 1000m and 2700m by the R/V Meteor in 2007 (Bohrmann et al., 2008). These studies included geological structures (mud diapirs, mud volcanoes, landslides and accretion wedges) (Su et al., 2014), geophysics (bottom simulating reflection, BSR) (Holbrook et al., 1996), mineralogy (carbonate rocks and authigenic minerals such as pyrite) (Zhang et al., 2011), marine halobios (bacterial mats, bivalves, mussels, and tube worms in the cold spring) (Campbell, 2006), and geochemical indicators (anions and cations in sediment pore water, hydrocarbon gases and sulfur isotopes) (Wu et al., 2010). Moreover, Grando and McClay (2007) proposed a “two-floor” model for NGH accumulation: the deep part is mainly controlled by thrust faults, while the shallow part is mainly controlled by normal faults. Smith et al. (2012) and Gong et al. (2016) reported that tectonic activity is the main controlling factor for NGH in the MAP. Zhang et al. (2021) suggested that mud diapir, mud volcanoes, gas chimney structures, and thrust faults are the main pathways for gas migration, playing an important role in controlling the accumulation and distribution of NGH in the MAP.

According to the NGH resource pyramid distribution, more than 90% of the global NGHs are reserved in submarine clayey silt or muddy sediments. Seabed sediments with different characteristics significantly impact the difficulty of natural gas hydrate exploitation. And in particular, clay minerals such as montmorillonite, illite, bentonite, and kaolinite play a significantly role in the storage and exploitation of NGH (Su et al., 2014; Ye et al., 2025). However, regional sedimentary pattern in the MAP and its relationship with NGH distribution are still unclear. To address the linkage between NGH and sedimentary properties, we report a study on surficial sediments collected by the China-Pakistan joint cruise by the R/V “Haiyang Dizhi Shi Hao” during 2018–2019, in terms of sediment grain size, geochemical properties, and clay minerals. Our results reveal the coupling relationship between sedimentary characteristics and NGH distribution.

2 Materials and methods

2.1 Studied samples

During 2018–2019, the R/V “Haiyang Dizhi Shi Hao” executed a systematic sampling of marine sediments in the MAP during the China-Pakistan Joint Cruise, by the Guangzhou Marine Geological Survey, China Geological Survey. A total of 80 sites of surficial sediments were obtained (Figure 1), with a gravity piston corer and a television grab. The collected sediments are mainly brown/grey mud and silt, and two sediment types are identified: fine-grained deposits with turbidite lamination (Shanmugam, 1994; 2002), and grey to brown clay, with biogenic debris and fossils.

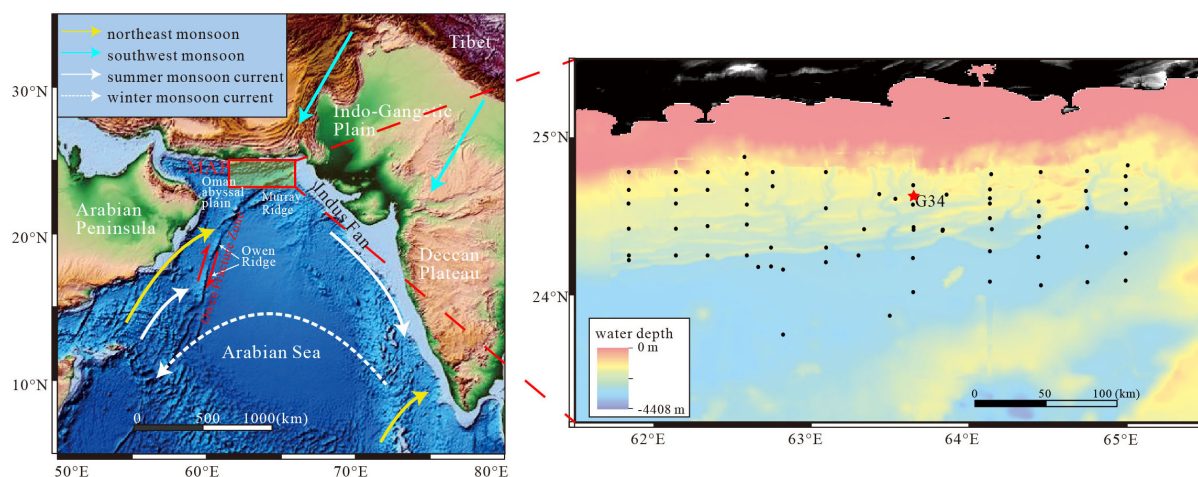


FIGURE 1

Schematic map of the study area, the Arabian Sea monsoon and ocean currents are modified from [Chen et al. \(2019\)](#). Tectonic settings are modified from [Rodríguez et al. \(2014\)](#).

2.2 Methods

Sediment cores are sampled at 20cm interval, and together with the surficial sediments, grain size, clay minerals and elemental contents are carried out in the Guangzhou Marine Geological Survey. Here, we exhibit all surface samples to illustrate sedimentary characteristics, and core sediments (core G34, 63.63659°E, 24.56111°N, 2046m water depth, with a length of 6.32m) were showed to interpret their vertical variabilities. This core was located at the center of our study area, directly above the NGH-rich zone. And also, the core is located in the transition zone between shallow-water and deep-water, receiving both terrigenous and marine sediments. As a result, it provides a comprehensive representation of the sedimentary characteristics of the study area. A total of 2580 sediment samples were collected for grain-size analysis, which were obtained from all sediment cores with an interval of ~20 cm. The samples were placed in an ultrasonic vibrator with $(\text{NaPO}_3)_6$ for several minutes to facilitate dispersion, and measured using a Malvern Mastersizer 2000 grain size analyzer.

805 samples were collected for major and trace elements analysis, with an interval of ~60 cm. Major elements, including Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, and P, were analyzed using the X-ray Fluorescence Spectrometer (XRF; Nippon Science Primus II). The sample handling process for XRF analysis includes (1) placing the 200 mesh sample in a 105 °C oven to dry for 12h, (2) respectively placing 6.0g flux ($\text{Li}_2\text{B}_4\text{O}_7$: LiBO_2 : LiF = 9:2:1) and 0.6g sample, 0.3g oxidant (NH_4NO_3) in a platinum crucible, being melted in a 1150 furnace for 14min. Finally, the crucible was removed and transferred to the refractory brick to cool, and the glass slide was removed for XRF analysis. Precision and accuracy were both better than 5%.

For trace elements contents analysis, the bulk sediments were completely dissolved by HF and HNO_3 solutions. About 50 mg sample was weighed and transferred into a pre-cleaned Teflon beaker followed by the addition of ultra-pure 1ml of HF and 1ml of HNO_3 solution. The beakers were then placed in steel cans and subjected to high

temperature (185 °C) and high pressure. After 36h, the solution was dried on a hotplate. The residues were fully digested using a mixture of concentrated 2ml HNO_3 and 3ml Milli-Q water. Thereafter, the beakers were placed into steel cans at 120 °C for 5h. After cooling, the solution was diluted to 20ml with Milli-Q water. Trace elements were measured via ICP-MS (X Series2, Thermo Fisher Scientific, MA, USA). Precision and accuracy were both better than 5%.

442 clay minerals samples were collected, analyzed by X-ray diffraction (XRD) with an interval of 1m from the cores. The XRD clay mineral study was carried out on the <2 μm fraction, which was separated by conventional Stokes' settling after the removal of carbonate and organic matter by acetic acid (15%) and hydrogen peroxide (10%), respectively. Clay minerals were then identified by XRD using a (Rigaku) D/Max 2500PC 18kW powder diffractometer (SYM125) in the Guangzhou Marine Geological Survey, with an X-ray wavelength $\lambda=1.5418\text{\AA}$ ($\text{CuK}\alpha$), and a scanning rate of $2^\circ(2\theta)/\text{min}$ at $0.02^\circ(2\theta)$. Each sample was measured 3 times under conditions of air-drying, ethylene glycol solvation, and heating at 490 °C at atmospheric pressure. Clay minerals was identified according to the position of the (001) series of basal reflections observed on the XRD diagrams ([Biscaye, 1965](#)).

3 Results

3.1 Sediment grain sizes

Silt is the dominant component in the sediments, with a range of 44.6% to 81.2%, followed by clay content of 16.8-53.4% and sand content up to 6.9%. In a Shepard's nomenclature ([Shepard, 1954](#)), most of the samples in the study area are clayey silt, with silty clay, silty sand, sandy silt, and very little silty sand and sand-silt-clay ([Figure 2A](#)). The grain-size distributions are bimodal, with modal sizes of 5-6 μm and 30-40 μm ([Figure 2B](#)), and only minor differences in the grain-size distributions between different sites, indicating a similar environment

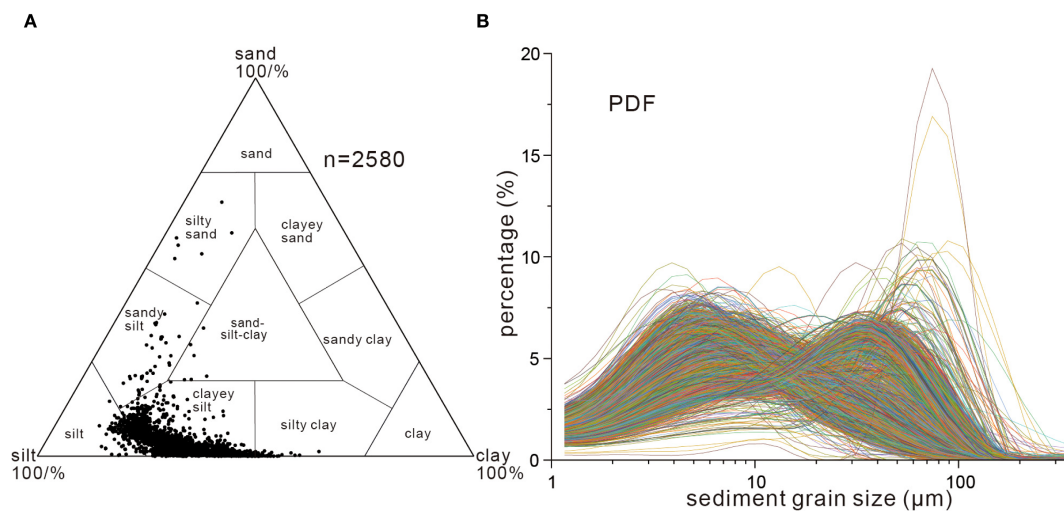


FIGURE 2

Shepard's nomenclature of sediments based on sand-silt-clay (A) and sediment grain-size distribution of samples from all cores (B). PDF, probability density function.

in the MAP (Maynard et al., 1982). For core GC34 (Figure 3), the median size varies from 6.8 Φ -8.42 Φ , with an average value of 7.73 Φ . Sorting varies from 1.24-1.98, with an average value of 1.58. Skewness varies from -0.12-0.17, with an average value of 0.03. Kurtosis varies from 0.88-1.1, with an average value of 0.99. The median size of

sediment samples is generally stable with little variation. Silt (4-63 μm) is the dominant component with an average value of 56.65%, while the clay (<4 μm) component is about ~40%, and the sand is only about 1%.

Ye et al. (2025) demonstrated that Global NGHs are more likely to occur in submarine silty and clay sediments. Taking the NGH sediment

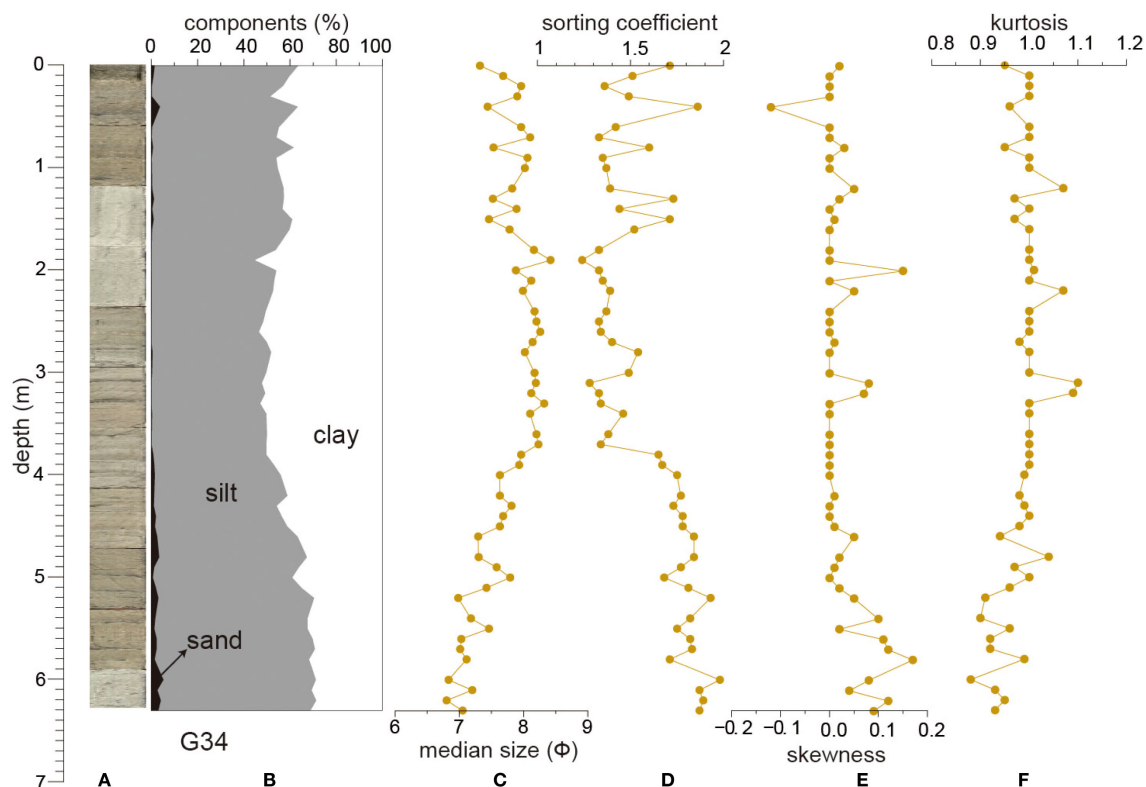


FIGURE 3

Grain-size characteristics of core G34. (A, B) photograph and grain-size composition of core GC34; (C-F) median size, sorting coefficient, skewness and kurtosis of core GC34.

samples in the Shenhu area of the South China Sea as an example, its distribution characteristics are between silt and clay. Our sediment samples are predominantly clayey silt, which is aligns with this pattern. A minority of coarse-grained samples were collected from water channels with strong hydrodynamic conditions in the study area.

For sediment cores, the sediment grain size gradually becomes finer from the east to the west (Figure 4), the sand content gradually decreases and the silt and clay fractions increase with the weakening of hydrodynamic conditions from the eastern Indian estuary towards the west; at the same time, several layers with coarser sediment grain size and higher sand fractions were found in the easternmost column samples G67, G61, G56 and G49, which may suggest the enhanced hydrodynamic of the estuary resulting in turbidity events (Burdanowitz et al., 2019).

For spatial distribution, regional sediments are fine, well sorted, positively skewed, and with moderate kurtosis, and become coarser to the south (Figure 4). In the southeast, the sediments become coarser, with a median size of about 5.0Φ - 6.8Φ , bad sorted with the sorting coefficients (σ) up to 2.28, likely attributed to the influences of the Indus River.

3.2 Clay minerals

The sediments are dominated by illite and chlorite, with few kaolinite and smectite (Table 1). The illite content ranges from 40-60%, with an average value of 52.5%; chlorite content ranges from 26-52%, with an average value of 41.2%; kaolinite content ranges

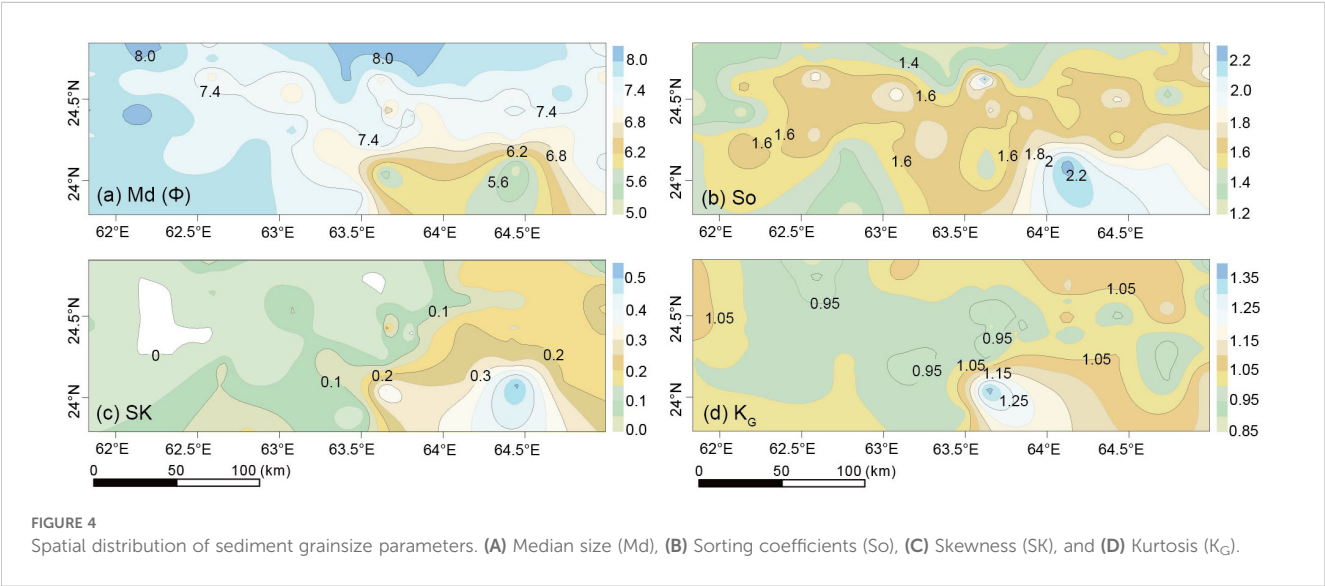


TABLE 1 Contents of clay minerals in the surface sediments and core G34 in the MAP.

Stations	Smectite	Illite	Kaolinite	Chlorite	Samples	Depth (m)	Illite	Chlorite
G01	/	51	/	49	G34-1	0	48	52
G02	/	52	/	48	G34-2	0.2	49	51
G03	/	50	/	50	G34-3	0.4	49	51
G04	/	52	/	48	G34-4	0.6	49	51
G09	/	52	/	48	G34-5	0.8	52	48
G11	/	52	/	48	G34-6	1	50	50
G16	/	50	/	50	G34-7	1.2	51	49
G19	/	53	/	47	G34-8	1.4	52	48
G21	/	53	/	47	G34-9	1.6	51	49
G23	/	49	/	51	G34-10	1.8	50	50
G25	/	48	/	52	G34-11	2	51	49
G27	/	50	/	50	G34-12	2.2	53	47
G34	/	48	/	52	G34-13	2.4	52	48
G35	/	48	/	52	G34-14	2.6	52	48

(Continued)

TABLE 1 Continued

Stations	Smectite	Illite	Kaolinite	Chlorite	Samples	Depth (m)	Illite	Chlorite
G40	/	49	/	51	G34-15	2.8	52	48
G43	/	48	/	52	G34-16	3	53	47
G46	/	51.9	14.1	34.1	G34-17	3.2	52	48
G47	/	55.3	15.8	29.0	G34-18	3.4	55	45
G48	/	54.4	12.9	32.7	G34-19	3.6	53	47
G49	/	53.6	13.3	33.1	G34-20	3.8	53	47
G50	/	54.3	13.2	32.5	G34-21	4	55	45
G51	/	51.6	14.2	34.3	G34-22	4.2	54	46
G52	/	49.9	13.8	36.3	G34-23	4.4	51	49
G53	/	47.4	16.7	35.9	G34-24	4.6	55	45
G54	1.3	50.2	12.5	35.9	G34-25	4.8	54	46
G55	0.8	51.9	13.9	33.4	G34-26	5	56	44
G56	/	50.1	14.2	35.7	G34-27	5.2	56	44
G57	/	47.6	15.0	37.4	G34-28	5.4	55	45
G58	/	46.4	13.5	40.1	G34-29	5.6	53	47
G59	/	50.0	11.6	38.5	G34-30	5.8	55	45
G60	/	49.9	13.7	36.4	G34-31	6	57	43
G61	/	51.7	15.4	32.8	G34-32	6.2	56	44
G62	/	54.6	13.4	32.0				
G63	/	52.5	9.7	37.9				
G64	/	50.1	14.4	35.5				
G65	/	53.3	12.1	34.6				
G66	/	53.3	12.9	33.8				
G67	/	50.4	10.0	39.6				
G68	/	52.6	14.8	32.6				
G69	/	54.3	12.0	33.7				
G70	/	53.8	11.2	35.0				
G76	/	52	/	48				
G77	/	52	/	48				
G80	/	55	/	45				

“/” indicates not detected.

from none to 19.5%, with an average value of 6%; and smectite was the lowest mineral, ranging from 0-3% with an average value of 0.2%. The components of kaolinite and smectite are extremely low in the western part, generally below the detection limit. For core G34, smectite and kaolinite were not detected. Illite is the dominant clay mineral in the sediments, ranging between 48-57% and gradually decreasing from bottom to top. Changes in chlorite is opposite to that of illite.

For spatial distribution (Figure 5), clay minerals are dominated by illite (46.4%-55.3%) and chlorite (29.0%-55.0%). Kaolinite increases in

the east, up to 16.7%, but decreases in the west. Smectite is rare in the whole area, usually less than 1%.

3.3 Oxides of major elements

The results show that SiO₂ is the dominate composition in the sediments, generally over 50%. From the north to the south (Figure 6), SiO₂ increases from 46% to >55%. Al₂O₃ is the secondary major component, about 10% in the study area. For core G34, CaO shows an

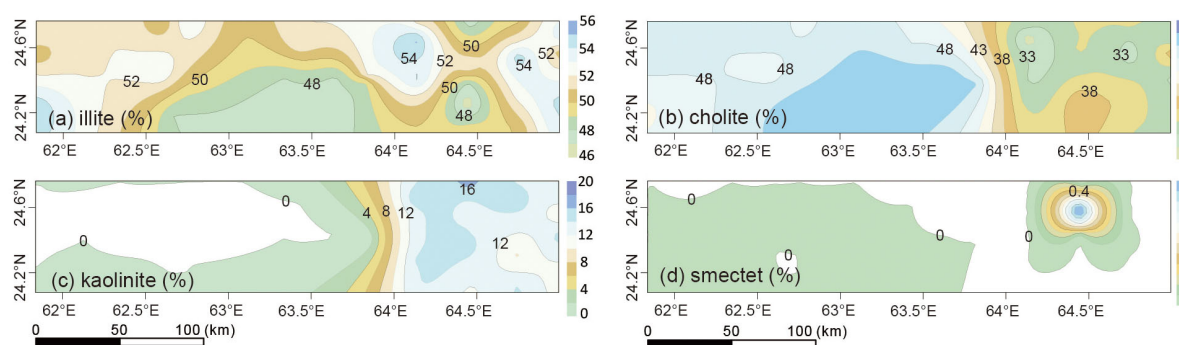


FIGURE 5

Spatial distribution patterns of (A) illite, (B) chlorite, (C) kaolinite, and (D) smectite for surficial sediments from the MAP.

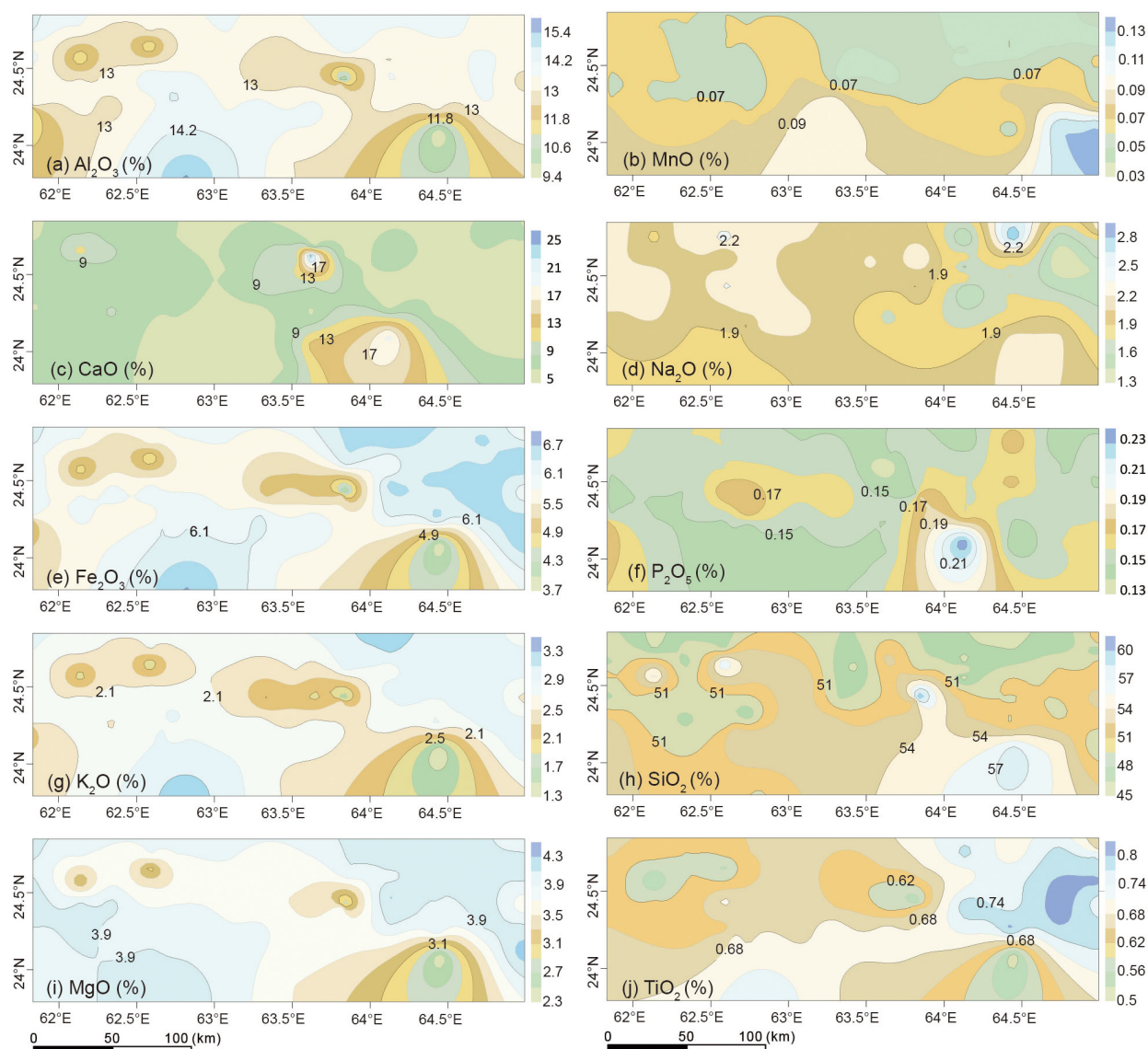


FIGURE 6

Spatial distribution patterns of major elements oxides of sediment samples in the MAP. (a) Al_2O_3 , (b) MnO , (c) CaO , (d) Na_2O , (e) Fe_2O_3 , (f) P_2O_5 , (g) K_2O , (h) SiO_2 , (i) MgO , (j) TiO_2 .

opposite trend to other elements (Figure 7). Based on changes in CaO, two units were identified: in Stage I: below 4.6m, CaO is more than 15%, while within Stage II: above 4.6m, CaO is less than ~10%, likely indicating a decrease in marine biogenic components and an increase in terrigenous debris.

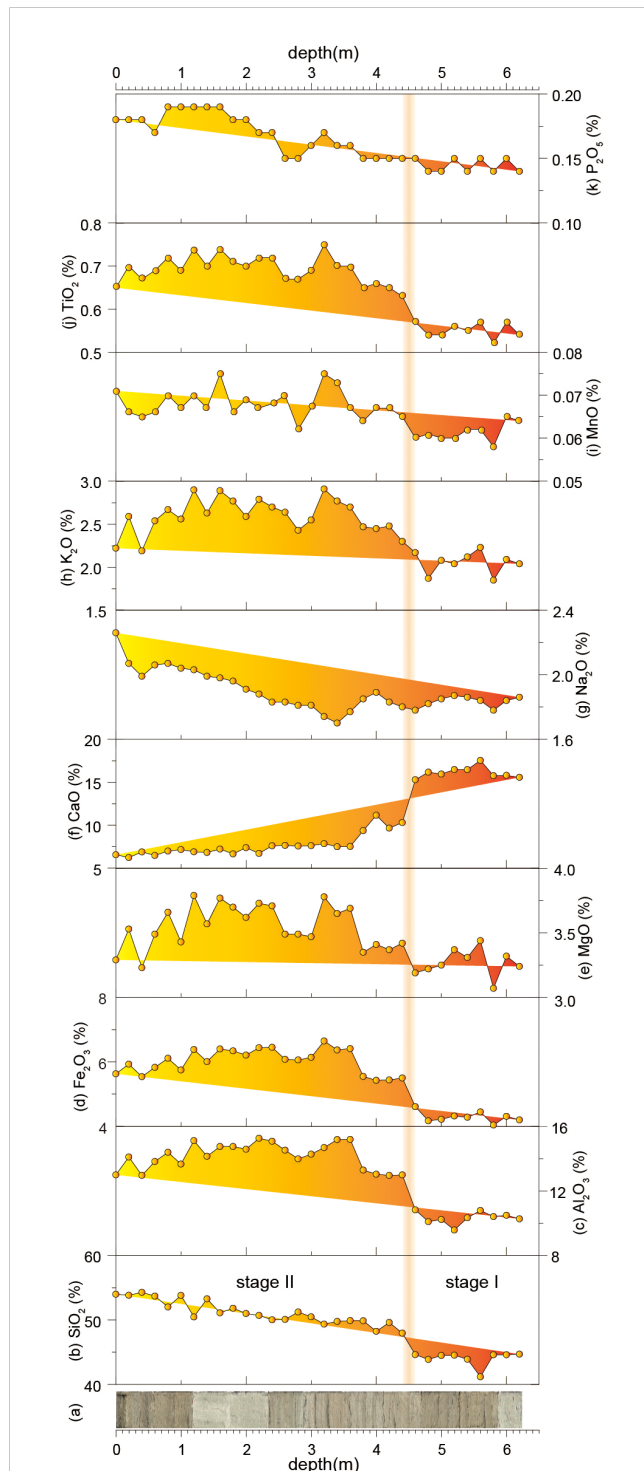


FIGURE 7
Changes in oxides of major elements of site G34. (A) photograph, (B–K) contents of major elements oxides of core G34.

3.4 Trace elements

Spatial changes in trace elements are relatively stable than major elements (Figure 8). In specific, most of the trace elements, such as Cr, Cu, Mo, Ni, U, V, Zn, Zr, Rb, and REEs, are relatively similar, while element Sr ranges from 163 $\mu\text{g/g}$ (G50) to 1287 $\mu\text{g/g}$ (G52), consistent with that of Ca. In addition, Co, Cu, Ba, Cr, Rb, Ni and Zn contents in the sediments in the west are relatively high, while Sr concentrates in the east. For core G34, changes in trace elements can be similarly divided into two groups (Figure 9): in the stage I, Sr content is $>1000 \mu\text{g/g}$ and decreased sharply in the stage II (above 4.6m). Some trace elements, including Cr, Cu, Ni, V, Zn, and Rb, exhibit an opposite variation, while the others, such as Ba, Mo, U, Zr, and REEs, varies without any patterns.

3.5 Elemental correlations

Correlation analyses on the elements in the surficial sediments of the study area were carried out and the results are shown in Table 2. As shown, Al_2O_3 is positively correlated with MgO, K_2O , TiO_2 , Fe_2O_3 , Co, Cr, Ni, Rb, Ga, V, and REE (Dubinin, 2004), and negatively correlated with CaO and Sr. Al_2O_3 , MgO, K_2O and Fe_2O_3 are the main components from rock weathering, and TiO_2 , Ga, and other elements are mainly derived from terrestrial detritus, which are mainly enriched in fine-grained sediments and endowed in a sorbed state in clay minerals. Hence, Al_2O_3 , MgO, K_2O , Fe_2O_3 and TiO_2 likely represent terrestrial clastic fractions, and CaO and Sr are mainly related with biogenic carbonate fractions, which were less deposited in the MAP and mutually exclusive with other elements (Avinash et al., 2016).

Si in marine sediments consists of terrestrial debris and siliceous bioclastic. The terrigenous clastic source is usually dominated by silicate minerals, such as feldspar, quartz and clay minerals, and the biogenic phase is dominated by siliceous organisms, such as diatoms, sponges and radiolarians. Elemental correlation analysis of the surficial sediments shows that no agreement between SiO_2 and typical terrestrial components, such as Al_2O_3 , Fe_2O_3 , and Ga, was observed, suggesting that Si are mainly from biogenic materials. In addition, a good positive correlation between CaO and Sr was evident, inferring that these two elements are mainly derived from biogenic carbonate fractions.

4 Discussion

4.1 Sediment provenances

Clay minerals in the sediments of the East Arabian Sea are close to weathering and denudation materials from the Himalayas transported by the Indus River (Kolla et al, 1981; Alizai et al., 2012), from the Deccan Plateau transported by the Dabti and Narmada Rivers, and from the southern India (Kessarkar et al., 2003). The Indus River, the world's largest river, has about $250 \times 10^6 \text{t}$ suspended materials to the Arabian Sea for each year. In the

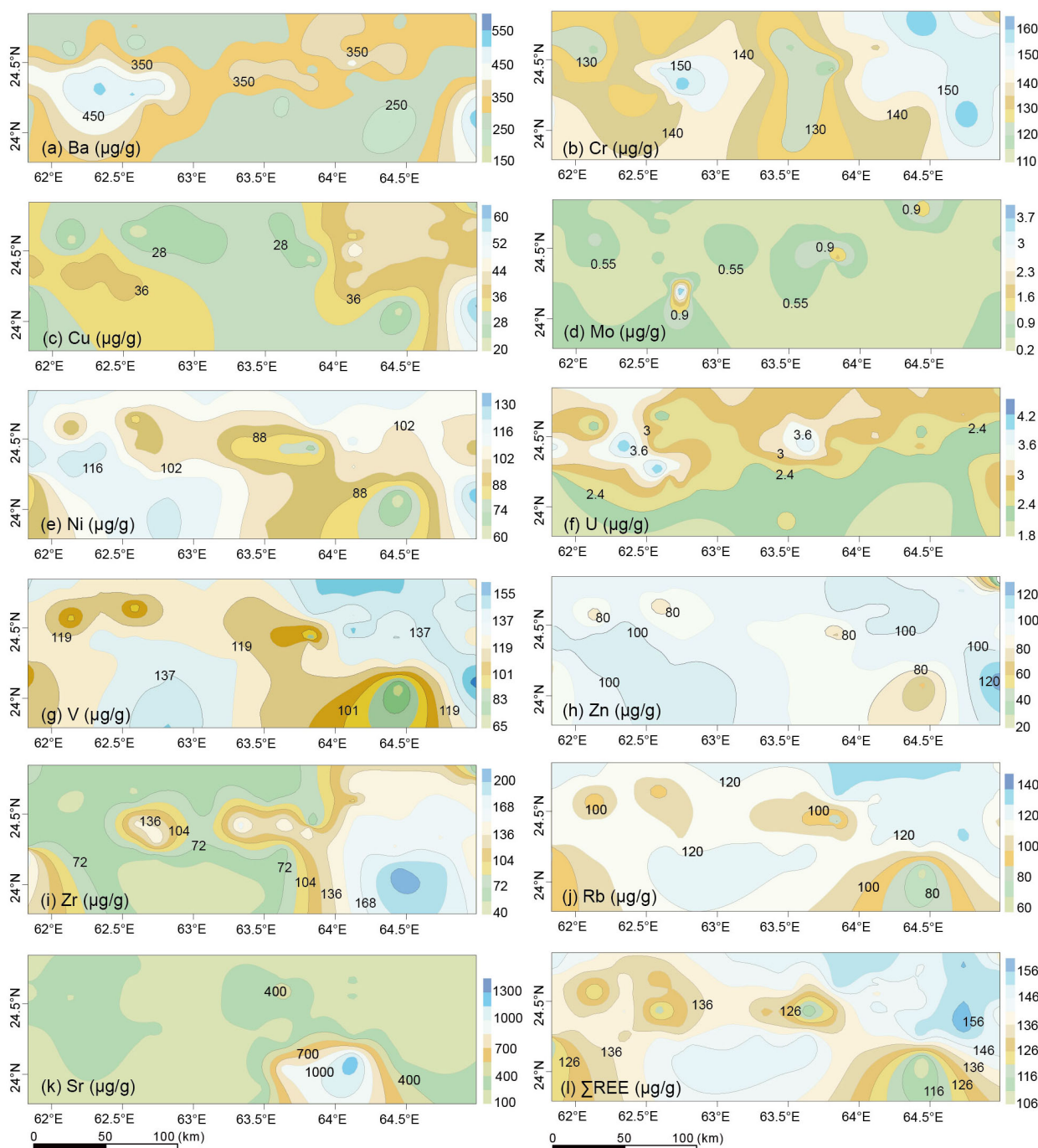


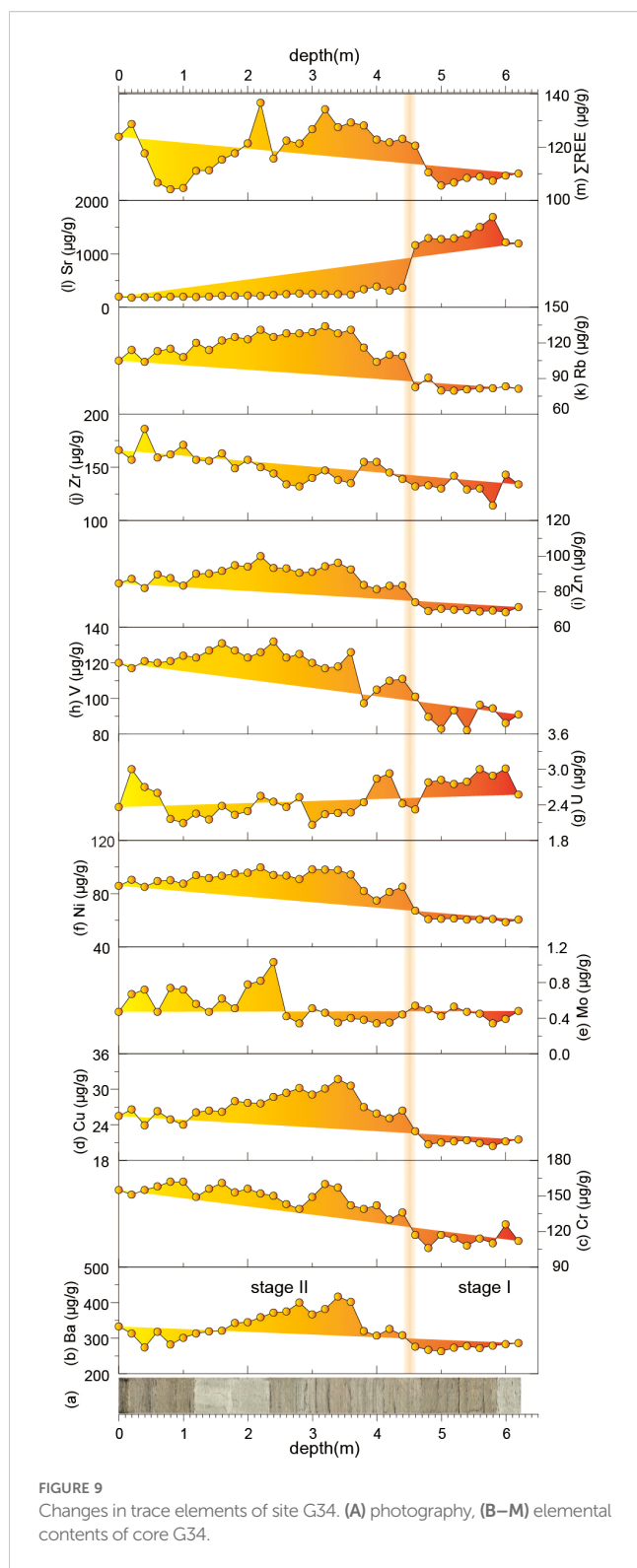
FIGURE 8

Spatial distribution patterns of trace elements of sediment samples from the MAP. (a) Ba, (b) Cr, (c) Cu, (d) Mo, (e) Ni, (f) U, (g) V, (h) Zn, (i) Zr, (j) Rb, (k) Sr, (l) Σ REE.

Indus River catchment, Pre-Himalayan Cambrian metamorphic rocks and weathering denudation products from the Himalayas are characterized by crystallinity-rich illite and chlorite (Alizai et al., 2012). The southward coastal currents during the summer facilitate the southward transport of Indus suspensions, with an influence on the distribution of clay minerals in the East Arabian Sea from north to south (Thamban et al., 2002; Govil and Naidu, 2008; Yu et al., 2019). Comparing clay minerals in various sediments (Figure 10), it is inferred that: the surficial sediments in the MAP are mainly from

the Himalayan weathering denudation products imported by the Indus River, while the absence of montmorillonite and kaolinite indicates the MAP deposition less influenced by sedimentary denudation from the Deccan Plateau in the south.

The association between ratios $\text{SiO}_2/\text{Al}_2\text{O}_3$ and $\text{K}_2\text{O}/\text{Na}_2\text{O}$ (Gong et al., 2021) suggests that all surficial and core sediments are deposited from an active continental margin (Figure 11A), agreeing well with the tectonic settings of the MAP. The association between Al_2O_3 , $\text{CaO}+\text{Na}_2\text{O}$, and K_2O , known as an



index of chemical weathering, namely Chemical Index of Alteration (CIA) (Nesbitt and Young, 1982) as Equation 1:

$$CIA = Al_2O_3 / (Al_2O_3 + K_2O + CaO + Na_2O) \times 100; \quad (1)$$

Which indicates that the sediments in the research area are primarily the products of the initial weathering stage (Young and

Nesbitt, 1999; Liu et al., 2012). In addition, these sediments also distribute close to the trend line of granodiorite (Figure 11B), indicating that the parent rocks of these sediments are granodiorite (Xu and Shao, 2018).

Furthermore, inactive trace elements, such as V, Zn, Cr, Ni, and Zr, are primarily controlled by the nature of parent rocks in provenance, and play a crucial role in determining the nature of sediment sources. As shown in Figure 12, the concentrations of these elements in our research area are quite different from those in the surface sediments of the continental shelf off the Indus Delt (Khan et al., 2020), Arabian Sea (Schnetger et al., 2000; Sirocko et al., 2000), average Indus River sediments (Clift et al., 2010) and west coast Indian (Alagarsamy and Zhang, 2010). This discrepancy may suggest that the terrigenous clastic sediments are not the sole provenance in the study area.

Gong et al. (2021) analyzed the trace element compositions of two sediment cores from the MAP, and concluded that the surface sediments possibly contain the Makran-Bela Ophiolite from the northwestern part and Murray Ridge igneous rocks from the southeastern part. The Makran-Bela Ophiolite and Murray Ridge igneous rocks are characterized by an occurrence of basalt and basic ultramafic rocks (Khan and Liu, 2019; Burgath et al., 2002; Ghose et al., 2014). Meanwhile, ultramafic rocks tend to have a high content of Cr, Co, and Ni and basalts tend to have a high content of Cu (Zahid et al., 2015). These trace elements were transported in to the sediments, resulting in elevated concentrations.

Overall, the eastern part of the MAP receives weathering products from the Himalayas carried by the Indus River and wind dust from the Arabian Peninsula, likely with a small amount of volcanic and hydrothermal material from the ocean floors.

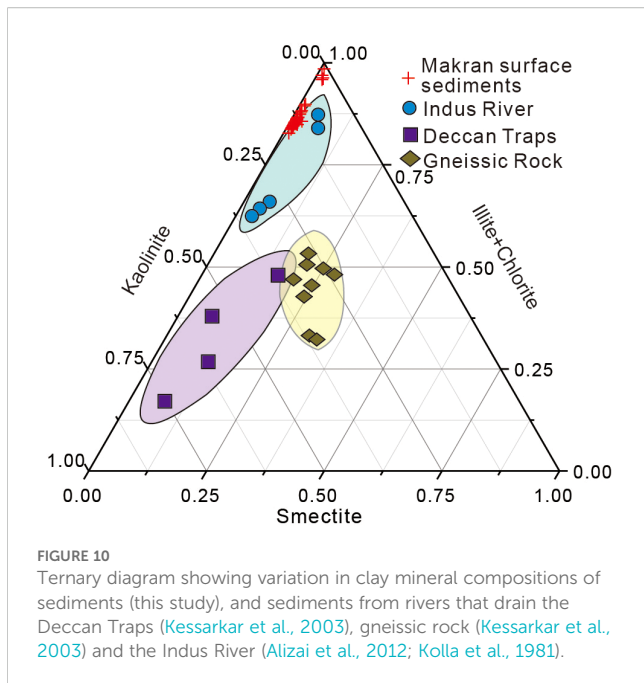
4.2 Relationship between NGH and surficial sediments

NGH are solid substances consisting of methane, ethane, carbon dioxide and other gases and water molecules under high pressure and low temperature (Zhang et al., 2003; Gong et al., 2016, 2018). Classic NGH exploration relies on geophysical methods, typically identifying the bottom simulating reflector (BSR) (Smith et al., 2014). Based on the BSR identification in the MAP (Smith et al., 2014), the sediment samples were classified into three categories: sediment samples over the BSR, and outside the BSR distribution (Figure 13A), respectively, and accordingly, a comparison between trace elements Mo and U similarly exhibits distinct relationships (Figures 13B–D), inferring the coupling between NGH distribution, BSR identification, and trace elements Mo and U. The contents of U and Mo in the sediment samples outside the BSR are generally less than 3 μg/g (Figure 13B), whereas the contents are significantly higher in the sediment samples over the BSR (Figure 13C).

The concentration relationships of redox-sensitive elements such as U and Mo can be used to interpret the marine depositional environment (Figure 14) (Guo et al., 2007; Algeo and

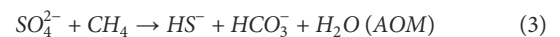
TABLE 2 Earth elements correlation coefficients of surficial sediments from Makran Accretionary Prism.

	SiO ₂	Al ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	TiO ₂	P ₂ O ₅	MnO	Fe ₂ O ₃	Co	Cu	Pb	Ba	Cr	Ni	Zn	Rb	Ga	Sr	V	Zr
Al ₂ O ₃	0.18																					
CaO	-0.71	-0.78																				
MgO	0.02	0.93	-0.69																			
K ₂ O	0.00	0.95	-0.68	0.97																		
Na ₂ O	0.16	0.12	-0.15	0.01	-0.06																	
TiO ₂	0.25	0.81	-0.78	0.83	0.88	-0.30																
P ₂ O ₅	-0.19	-0.24	0.13	-0.11	-0.16	0.07	-0.12															
MnO	0.42	0.19	-0.36	0.09	-0.06	0.18	0.16	0.07														
Fe ₂ O ₃	0.07	0.92	-0.72	0.94	0.97	-0.13	0.92	-0.09	0.16													
Co	0.03	0.74	-0.61	0.72	0.83	-0.07	0.75	-0.04	0.23	0.85												
Cu	-0.25	0.55	-0.32	0.73	0.73	-0.27	0.69	-0.05	0.00	0.74	0.83											
Pb	0.08	0.58	-0.54	0.69	0.72	-0.45	0.86	-0.03	0.13	0.80	0.78	0.80										
Ba	-0.09	0.44	-0.29	0.62	0.52	0.09	0.33	-0.09	0.02	0.48	0.75	0.65	0.48									
Cr	0.33	0.70	-0.79	0.72	0.75	-0.09	0.88	0.02	0.17	0.80	0.72	0.64	0.74	0.40								
Ni	-0.05	0.89	-0.56	0.91	0.87	0.25	0.59	-0.21	0.15	0.80	0.81	0.63	0.47	0.40	0.54							
Zn	-0.08	0.68	-0.44	0.74	0.74	-0.01	0.59	-0.14	0.12	0.70	0.73	0.71	0.58	0.70	0.55	0.75						
Rb	0.01	0.95	-0.67	0.95	0.97	0.03	0.83	-0.18	0.09	0.94	0.73	0.68	0.65	0.60	0.71	0.88	0.72					
Ga	0.11	0.98	-0.72	0.93	0.96	0.05	0.81	-0.24	0.17	0.93	0.76	0.59	0.61	0.50	0.68	0.89	0.69	0.95				
Sr	-0.48	-0.77	0.78	-0.65	-0.67	-0.09	-0.71	0.48	-0.17	-0.64	-0.48	-0.40	-0.45	-0.20	-0.61	-0.57	-0.46	-0.70	-0.71			
V	-0.03	0.91	-0.65	0.95	0.98	-0.04	0.85	-0.13	0.10	0.96	0.86	0.79	-0.45	0.60	-0.61	0.88	0.76	0.96	0.93	-0.65		
Zr	-0.07	0.07	-0.18	0.21	0.25	-0.46	0.52	0.28	-0.06	0.36	0.38	0.56	0.61	0.10	0.59	-0.07	0.17	0.18	0.05	-0.10	0.27	
REE	0.26	0.89	-0.79	0.87	0.91	-0.10	0.92	-0.20	0.14	0.91	0.73	0.58	0.75	0.40	0.77	0.71	0.63	0.91	0.89	-0.74	0.88	0.25



Rowe, 2012; Deng et al., 2017). U is a typical oxygen-loving element, which is reduced to tetravalent in an oxidizing environment and is enriched in sediments as a non-soluble state (Calvert and Pedersen, 1993; Tribouillard et al., 2006). Molybdenum is extremely sensitive to the oxidation-reduction environment of its environment; Mo^{6+} is the most common oxidation valence state and migrates in solution as MoO_4^{2-} ; under reducing conditions, molybdenum is attached to solid-phase sediments as Mo^{4+} or Mo^{5+} (Tribouillard et al., 2006). The U in seafloor cold spring sediments, Mo enrichment can occur. This is mainly because of the presence of large amounts of H_2S in the pore water because of the organoclastic sulfate reduction (OSR,

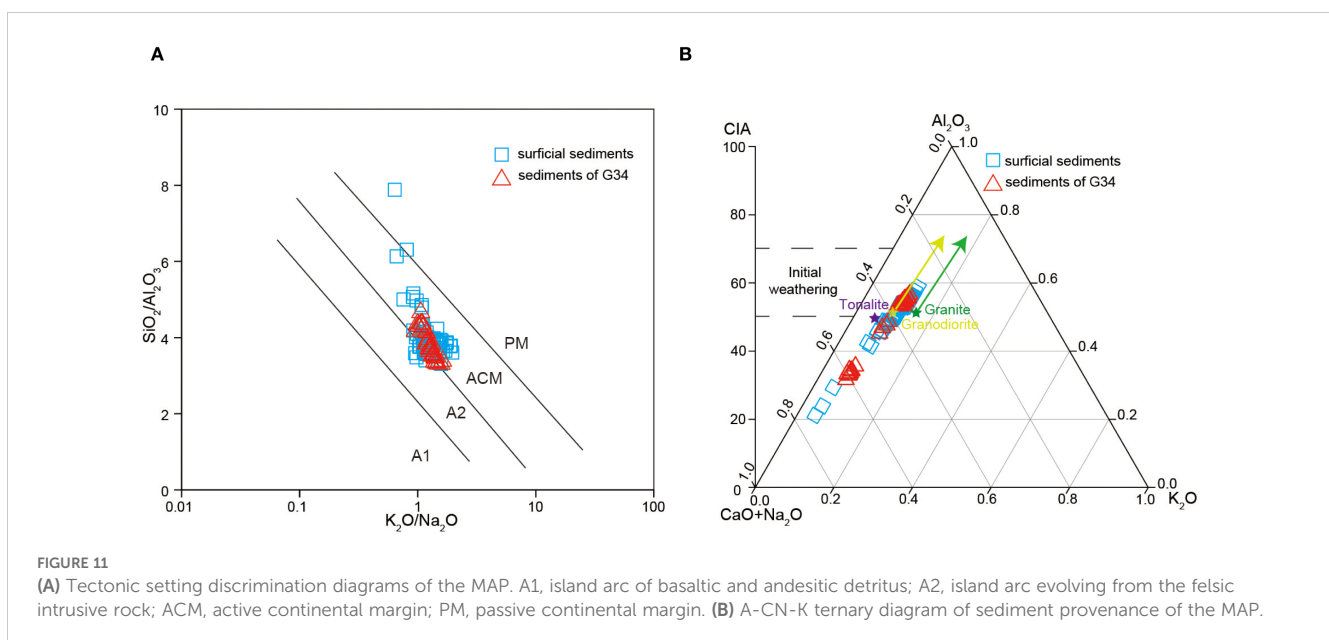
Equation 2), and Anaerobic Oxidation of Methane (AOM, Equation 3) reaction (März et al., 2008; Li et al., 2021), as follows:



Which requires sufficient methane to react with sulfate in a reductive manner, therefore the presence of sulfide zones in the surface sediments and the anomalous concentrations of U and Mo may be due to the high methane fluxes in this area. In areas of no methane seepage, sulfate is not consumed, resulting in a deep sulfate-methane interface (SMI), whereas, in areas of large scale methane seepage, CH_4 fluids may surge upwards into the seawater-sediment interface and massively consume sulfate in the surface sediment pore water, resulting in a shallow SMI in this area. The shallow burial of the SMI interface in the study area results in narrow oxidation and sub-oxidation zones in the sediments and the potential for H_2S from methane-sulfate reactions to transport upwards with the pore water into the surface sediments, resulting in sulfide zones enriched in Mo and U elements (Deng et al., 2017; Li et al., 2021; Gong et al., 2021).

5 Conclusions

Based on analyses of grain size, geochemical properties, and clay minerals of surficial and core sediments collected from the MAP, the linkage between NGH distribution and sedimentary properties were discussed. The main results are as follows. (1) Surficial sediments were mainly controlled by hydrodynamic conditions of the Indus River. Along the Indus River channel, it is mainly infilled



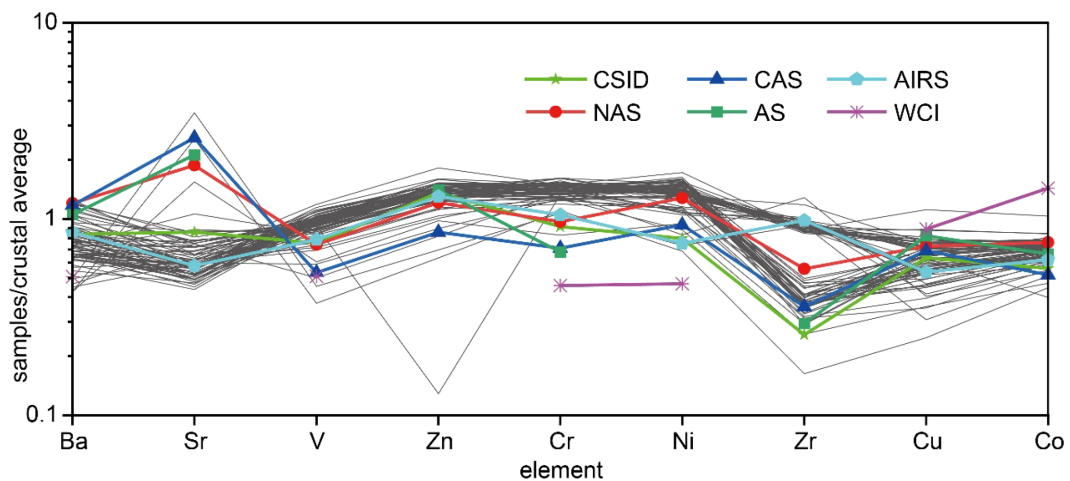


FIGURE 12

Standardized curves of trace elements in the MAP. Crustal average (Taylor, 1964); CSID, surface sediments from the continental shelf off the Indus Delta (Khan et al., 2020); NAS & CAS, North Arabian Sea & Central Arabian Sea (Schnetger et al., 2000); AS, Arabian Sea (Sirocko et al., 2000); AIRS, Average Indus River Sediments (Clift et al., 2010); WCI, west coast India (Alagarsamy and Zhang, 2010).

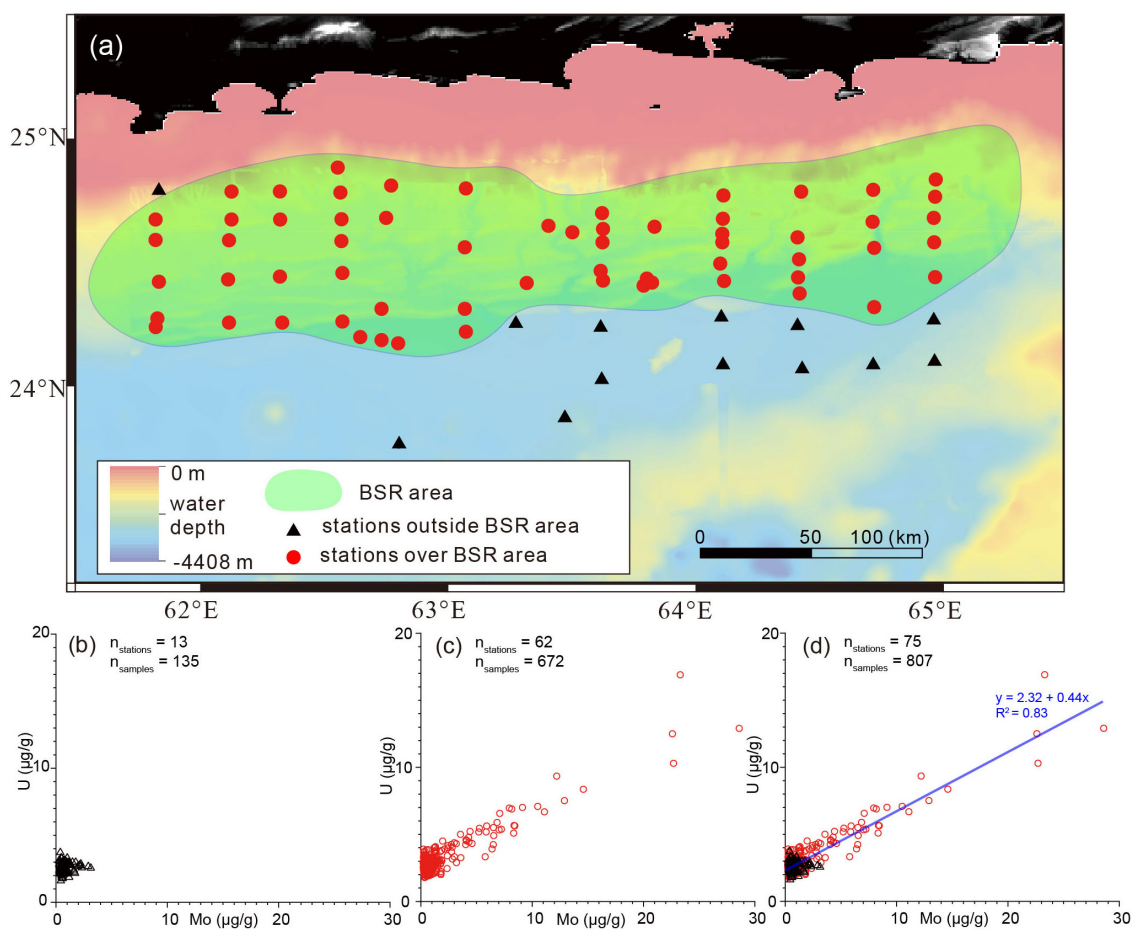


FIGURE 13

Relationship between distribution of BSR and trace elements U and Mo in the Makran accretionary prism. (a) locations of sample stations and BSR area; (b) sample stations outside the BSR area; (c) sample stations over the BSR area; and (d) all samples. The BSR area of the study area was modified from (Smith et al., 2014).

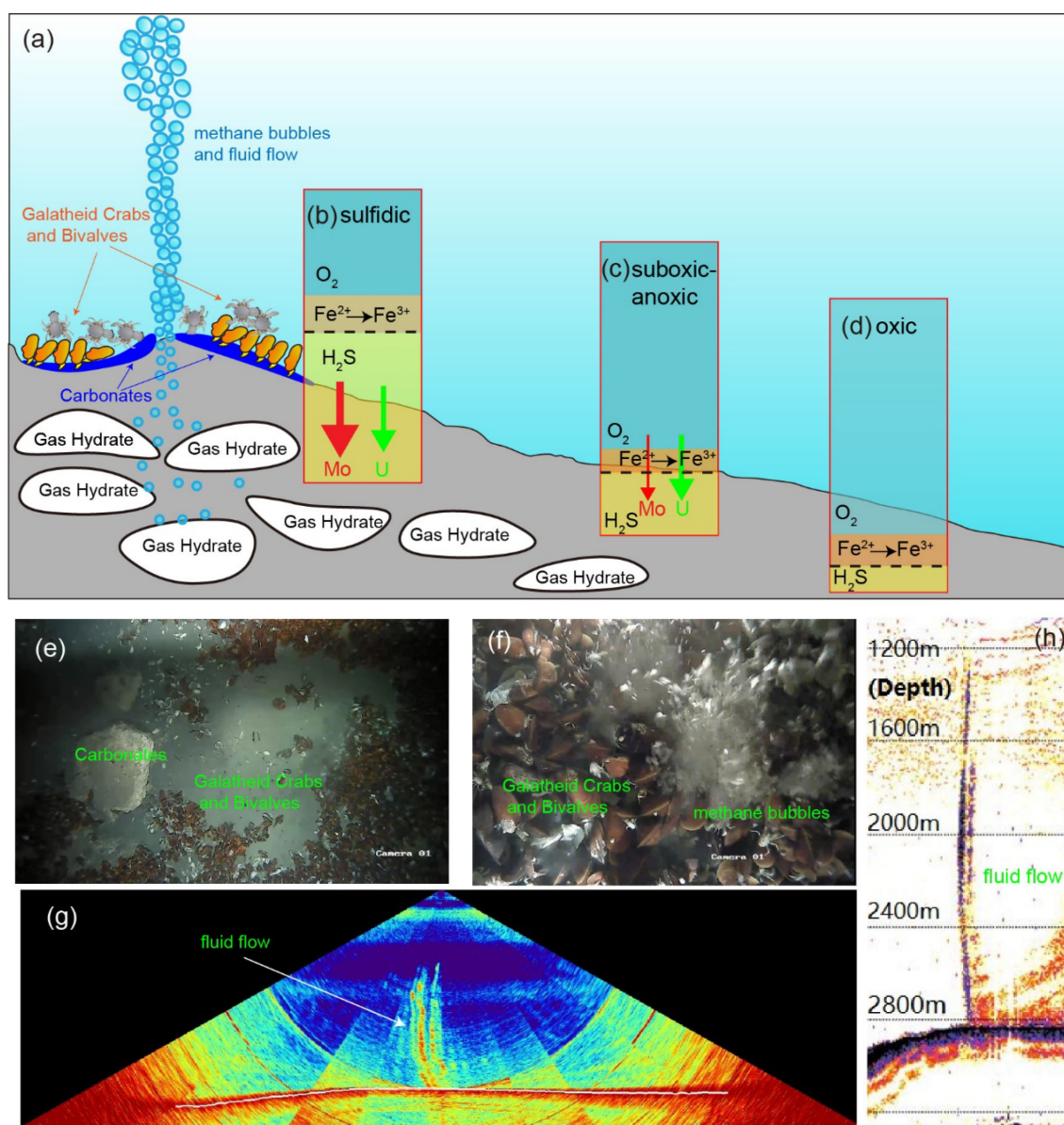


FIGURE 14

(A) Schematic diagram of the gas hydrate cold seep system (modified from Bohrmann and Torres, 2006; Delong, 2000); (B–D) relationships of Mo and U authigenic enrichment to benthic redox variation (modified from Algeo and Tribouillard, 2009); (E, F) biota and carbonates in the MAP cold seep area revealed by underwater imaging; (G, H) detection of cold seep fluid flow using multibeam and sub-bottom profiling aboard the R/V “Hai Yang Di Zhi Shi Hao”.

with silt, while clays are mainly distributed in the accretionary prism. The turbidity records gradually decrease from east to west, along the Indus River channel. (2) Illite and chlorite are the dominate clay minerals, indicating that the surficial sediments were mainly from weathering processes in the Tibet Plateau by the Indus River transport. The very-low contents of kaolinite and smectite indicate the lack of sediments from the Deccan Traps and the *in situ* hydrothermal materials. (3) Elements analysis indicates that elements Al, Mg, K, Fe, and Ti were terrigenous, mainly

distributed in the northern Makran accretionary prism, while elements Ca and P were from biogenic sources, mainly distributed in the southern part of the study area. (4) Based on these results, a close relationship between NGH distribution and sedimentary properties was observed, and we therefore proposed that the covariation between NGH distribution, BSR identification, and trace elements Mo and U holds the potentials in NGH investigation. The anomalies of U and Mo in surface sediments can be used to efficiently conduct reconnaissance for NGH.

Data availability statement

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

Author contributions

HW: Data curation, Formal Analysis, Methodology, Visualization, Writing – original draft. LY: Data curation, Formal Analysis, Investigation, Methodology, Visualization, Writing – review & editing. FW: Data curation, Writing – original draft. YL: Formal Analysis, Writing – original draft. YC: Writing – review & editing. ZZ: Data curation, Writing – original draft. MY: Investigation, Writing – original draft. XD: Formal Analysis, Writing – original draft. YY: Data curation, Writing – original draft. HY: Writing – review & editing. XD: Investigation, Writing – review & editing. GH: Funding acquisition, Writing – review & editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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