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Geochemical Assessment of River Sediments at the Outlets of Eastern Makran, Pakistan; Implications for Source Area Weathering and Provenance

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Abstract: The river sediments in the eastern Makran were investigated to determine the influence of climate, tectonics, and source rocks based on the geochemistry of sediment delivery to the offshore marine basin. Samples were collected at the outlets of four major drainage basins. The specimens were analyzed by XRF, ICP–MS, and ICP–AES. The geochemical concentrations of major, trace, and rare earth elements show great variation among the four outlets. The Rakhshan Kaur (RK), Shadi Kaur (SK), and Dasht Kaur (DK) represent more sorted and matured detrital sediments than those of the Hingol Kaur (HK). The detrital sediments at all outlets represent mostly humid climates except for the DK which falls at the junction of humid and semi-arid conditions. The studied samples show a low level of source area weathering. The sedimentary provenance of the RK and DK is due to the heterogeneous sources of detritus from Iran in the west and the older Eocene–Holocene sedimentary successions in the east. The sediment carried by SK is probably from the late Oligocene–Holocene succession from central and coastal Makran. The HK represents a heterogeneous provenance from the Kirthar–Sulaiman Belt, Bela Ophiolites, and sedimentary successions of central and coastal Makran. The tectonic environments of all river sediments signify a continental arc rather than an active continental margin.

Keywords: Rakhshan River; Dasht River; Shadi River; Hingol River; eastern Makran; provenance



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1. Introduction

The onshore main rivers of eastern Makran emptying into the Arabian Sea traverse a wide variety of terrain with varying climatic and tectonic forces. The geochemistry of river sediments at basin outlets gives an excellent chance of examining how climate and tectonic forces combine to regulate erosion. If such a sediment archive is to be used, it is necessary to know where the sediment comes from, what its composition is, and how it changes as it travels owing to abrasion and chemical weathering [1]. If the bedrock compositions are diverse enough, the bulk composition of the sediment may provide a means of determining the source of the materials. Many researchers have utilized the geochemistry of bedload and suspended sediments to establish their origin, source area weathering, and the tectonic environment of various river basins [2–9].

The clastic/siliciclastic sediments are the end products of numerous complicated processes such as weathering, transportation, and their relations, and their provenance can be inferred using their geochemical characteristics. The geochemistry of sediments preserves the evidence of these processes from source to sink, after their deposition. Al, Fe, Ti, Sc, Th, Zr, Cr, Ni, Co, and rare earth elements are among the examples of major and minor elements whose distributions are well-suited to distinguishing between different provenances and tectonic settings. These elements tend to be transient in riverine environments and have little mobility throughout sedimentary processes [10]. The geological evolution of a

sedimentary unit or basin province can be decoded from its source region by analyzing the geochemistry of sediments, which in turn exposes information on the nature of source area weathering and provenance, which are governed by climatic and tectonic causes.

Geochemical investigations of the onshore eastern Makran river sediments have been given less priority in conducting such work. Here, we investigate the geochemistry of river sediments collected at the watershed outlets to see how effective major, trace, and rare earth element geochemistry can be in determining provenance. In addition, the sorting, maturity, tectonic environment, climatic influence, and source area weathering of the sediments are also discussed.

2. Geographical and Geological Framework

2.1. Geographical Aspects

Its rough terrains make Balochistan a unique geographical area, which is categorized into five divisions: deserts, plains, lower highlands, upper highlands, and the coast. The east–west coast stretches approximately 760 km from Jiwani near the Iranian border to the Hub River near Karachi. It comprises numerous promontories and peninsulas. The desert areas fall in the Chagai and Kharan districts in the northwest of Balochistan province. The zone covers massive plains in the form of huge sand dunes and black gravel surfaces. The plains contain Makran plain, including Dasht, Bugti plain, and Lasbela plain. The piedmont plain, valley floor, and terrains make up 15% of the area, whereas the greater area is covered by mountains. The lower highlands are commonly found in the southern parts of the province. They range from 600 to 1200 m. The upper highlands include the Kirthar-Sulaiman and Makran ranges at the east and west, respectively. They rise to an altitude of ~1500 to 3700 m [11].

2.1.1. Climate

The climate of the desert zone is extremely arid but extremely hot in summer. The plain areas have mild winters and extremely hot summers. The temperature can rise up to ~52 °C. The climate of the lower highlands varies from place to place. The northern parts are extremely cold in winter, while the winters of the southern parts are mild. Both parts are dry and hot in summer except for the coastal cities. The upper highlands are extremely cold in winter, while summers are warm. The climate of the coast has mild winters and hot summers with high humidity [11].

2.1.2. Rainfall

The annual rainfall ranges from 50 to 500 mm. The northern parts receive the highest rainfall, which ranges from 200 to 500 mm, while the eastern and southern parts receive an average of 25 to 50 mm. The average annual rate of evaporation ranges from 1830 to 1930 mm, which is usually higher than the rate of annual rainfall [12]. The annual rainfall varies from November to March, with a peak in January and February. In addition, glaciation is absent [10]. In [13], the authors stated that the region of the Arabian Sea receives two types of monsoon winds. The humid and warm monsoon winds blow from the southwest in summer, while the drier and weaker winds of semi-arid to arid environments blow from north to north-east in the winter [14]. However, extreme rainfall sometimes leads to extensive flash floods that cause severe erosion and transportation of sediments. These eroded sediments from inner and outer Makran are then transported to the Arabian Sea through the main streams.

2.1.3. Rivers and Streams

Makran is hydrologically distributed into the Rakhshan, Doraski, Hingol, Kech, Nihing, Shadi, Basol, and Dasht rivers. Among them, the Dasht, Shadi, Basol, and Hingol rivers drain directly into the Arabian Sea. The Dasht River is ephemeral, with a catchment area of about 21,000 km², and the rainfall is 110 cm per annum [15]. Two associated tributaries, Kech from Balochistan in the east and Nihing from Iran in the west, join the Dasht

river at Kor-e-Awar in the Nasirabad region. Other tributaries in the Sami area, such as Rakhshan draining from Panjgur, Gishkaur, and Kill Kaur connect the Kech River. The Shadi Kaur (River) is not an ephemeral type, with a total length of about 80 km. The smaller Pidark and Bahri rivers join Shadi Kaur, which finally drains into the Arabian Sea in the Pasni region. The Basol River enters the Arabian Sea about 30–35 km west of Ormara. The tributaries draining from the southern parts of Awaran and Hoshab join the Basol River. On the other hand, the Hingol River is an ephemeral type of river that drains into the Arabian Sea in the Lasbela region, which covers an area of about 35,736 km² [16,17].

2.2. Geological Background

The Makran is bound by two strike-slip faults i.e., Ornach Nal to the east of Makran Accretion Wedge (MAW) in Balochistan, Pakistan (Figure 1) and Menab to the west, in Iran. The region developed at the convergent margin between the Arabian and Eurasian plates throughout the Cenozoic, mainly by recycling of sediments that were eroded from the India–Asia collision belt and from the uplifted older strata [18,19]. Makran is further divided into three regions, i.e., outer Makran comprising the Chagai-Raskoh magmatic arcs region, inner Makran representing Eocene to Miocene Flysch sediments, and coastal Makran representing the post-Miocene Flysch sediments (Figure 1). The oldest parts (Chagai-Raskoh magmatic arcs) are about 400 km inland from the Makran coast. The coastal region contains Plio-Pleistocene sediments [20–22].

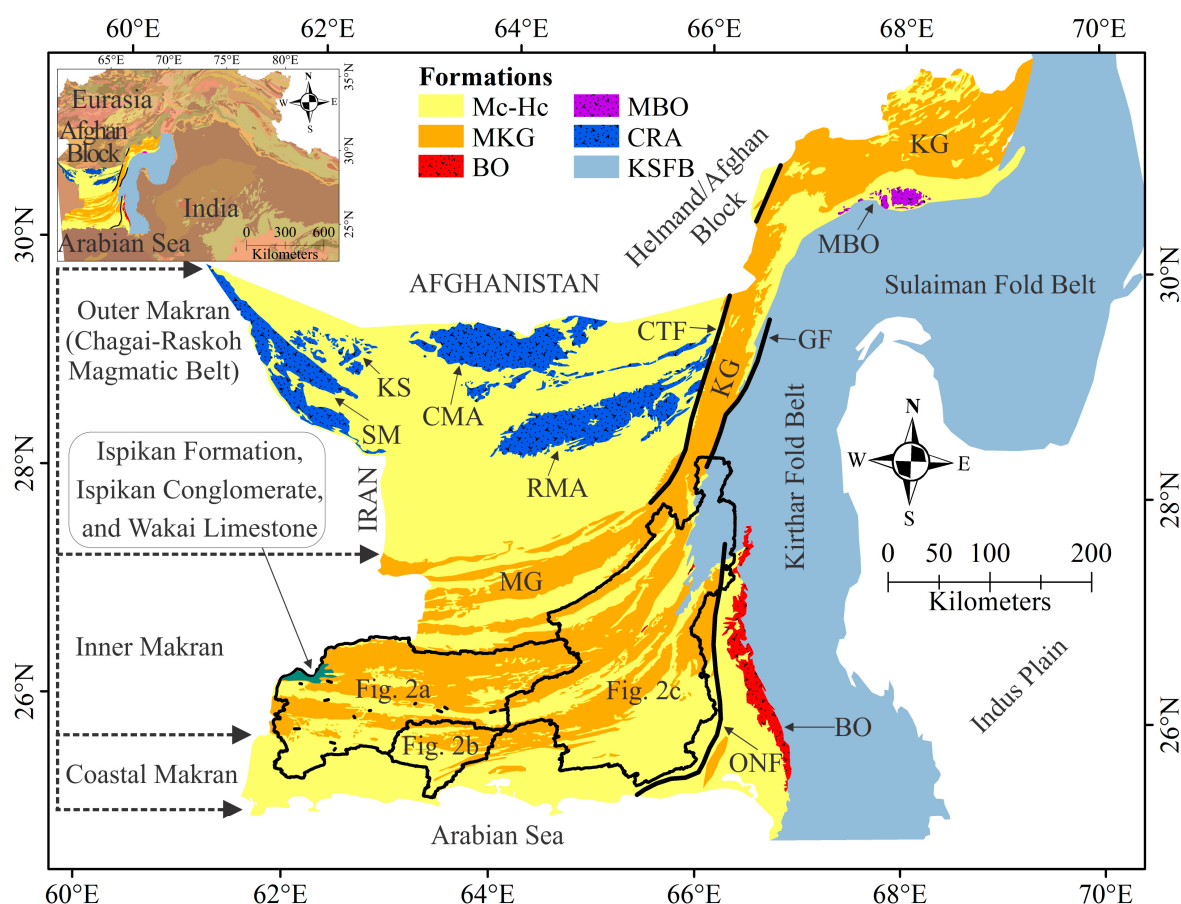


Figure 1. Map showing the geological background of Balochistan. The horizontal arrows refer to the divisions of Makran, for instance, outer, inner, and coastal Makran. MC-Hc: Miocene to Holocene; MKG: Makran-Khojak Group; BO: Bela Ophiolites; MBO: Muslim Bagh Ophiolites; CRA: Chagai-Raskoh arc; KSF: Kirthar-Sulaiman Fold Belt; KS = Koh Sultan; SM = Sandak magmatic arc; CMA = Chagai magmatic arc; RMA: Raskoh magmatic arc; CTF: Chaman Transform Fault; GF: Ghazaband Fault; and ONF: Ornach Nal Fault [20–26].

Moreover, the region shows massive sedimentary beds ranging from Jurassic to Holocene (Figures 1 and 2a–c). The Jurassic formation (AMJ) comprises fossiliferous gray limestone and shale, which is found at the Kirthar Fold Belt (Figure 2c). The Cretaceous Pab Formation (PFC) comprises quartzose sandstones and shales and can be found at the Kirthar Fold Belt (Figure 2c). The Paleocene Gidar Dhor Formation (GDFF) consists of diverse igneous and sedimentary rocks and is found at the Kirthar Fold Belt along with Paleocene undifferentiated rocks (URP) (Figure 2c). The ophiolitic mélanges (BOMP) of Paleocene are found in the Bela Ophiolite (Figure 2c). Many isolated outcrops of Wakai Limestone (WLE) can be found along the Pakistani and Iranian borders in the western Siah Range. This Eocene formation primarily comprises grey reefoid limestones (Figure 2a). The Eocene Nisai Formation (NFE) comprises argillaceous and reefoid gray limestone, shale, and sandstone interbeds in the Kirthar Fold Belt along with Eocene undifferentiated rocks (KPE) (Figure 2c). The Oligocene Nal Limestone (NLO) to the north of the Bela Ophiolite and Nari Formation (NFO) at the Kirthar Fold Belt comprises massive reefoid gray to light brown limestones, shale, and sandstone interbed (Figure 2c). The Oligocene Murgha Faqirzai Formation (MFFO) at the Khojak Group (KG; Figure 1), comprises shale with sandstones, and minor interbeds of shelly limestones (Figure 2c). The Late Oligocene to Early Miocene Panjgur formation (PFOM) is widespread in central Makran and along the coast [20–22,27]. The authors of [18,28] found that the younger succession represents slope and shelf sediments (shallow marine) deposited on top of the wedge, whereas the older succession was deposited on the ocean floor (deep marine) of the precursor of the Arabian Gulf and subsequently tectonically absorbed into the accretionary wedge. The PFOM is composed of greenish micaceous sandstones and turbidites that originated on the ocean floor (Figures 1 and 2a–c). The Neogene Hinglag Formation (HFN) consists of sandstone, shale, and shelly limestone and is found in coastal Makran (Figure 2c). As described by [18] and supported by [29], along the coastal range, the Parkini Formation (PFM) is widely distributed and extensively exposed along the Turbat–Pasni route. With very fine-grained thin sandstone strata, it is frequently composed of light-grey, fine-grained, and less cemented mudstones dating to the Miocene. The sediments of PFM were deposited on an outer shelf region with an upper slope (Figure 2a,b). Along the coast of Makran, the early Pliocene Chatti Formation (CFP) is found. Siltstones and fine-grained sandstones make up the Formation. A middle–outer shelf habitat is indicated by the prevalence of nanofossils in this formation (Figure 2a,b). Along the west coast of Makran, the Gwadar Formation (GFP) can be found. It resembles conglomerate, sandstone, and sandy clay. This formation is late Pliocene in age (Figure 2a). The Bostan Formation (BFP) in the Khojak Group, Kech Conglomerate (KCP) in central Makran, and Haro Conglomerate (HCP) in coastal Makran correspond to the same Pleistocene age and rock properties. These formations are of lacustrine and fluvial origin with less consolidated sandstones, conglomerate, and shale (Figures 1 and 2a–c). In the eastern part of the coastal area, the Holocene Extrusive Mud (EMDH) is found (Figure 2c). In addition, the older alluvial deposits (SRDH) and unconsolidated surface deposits (RDH) are found throughout the region (Figures 1 and 2a–c) [27].

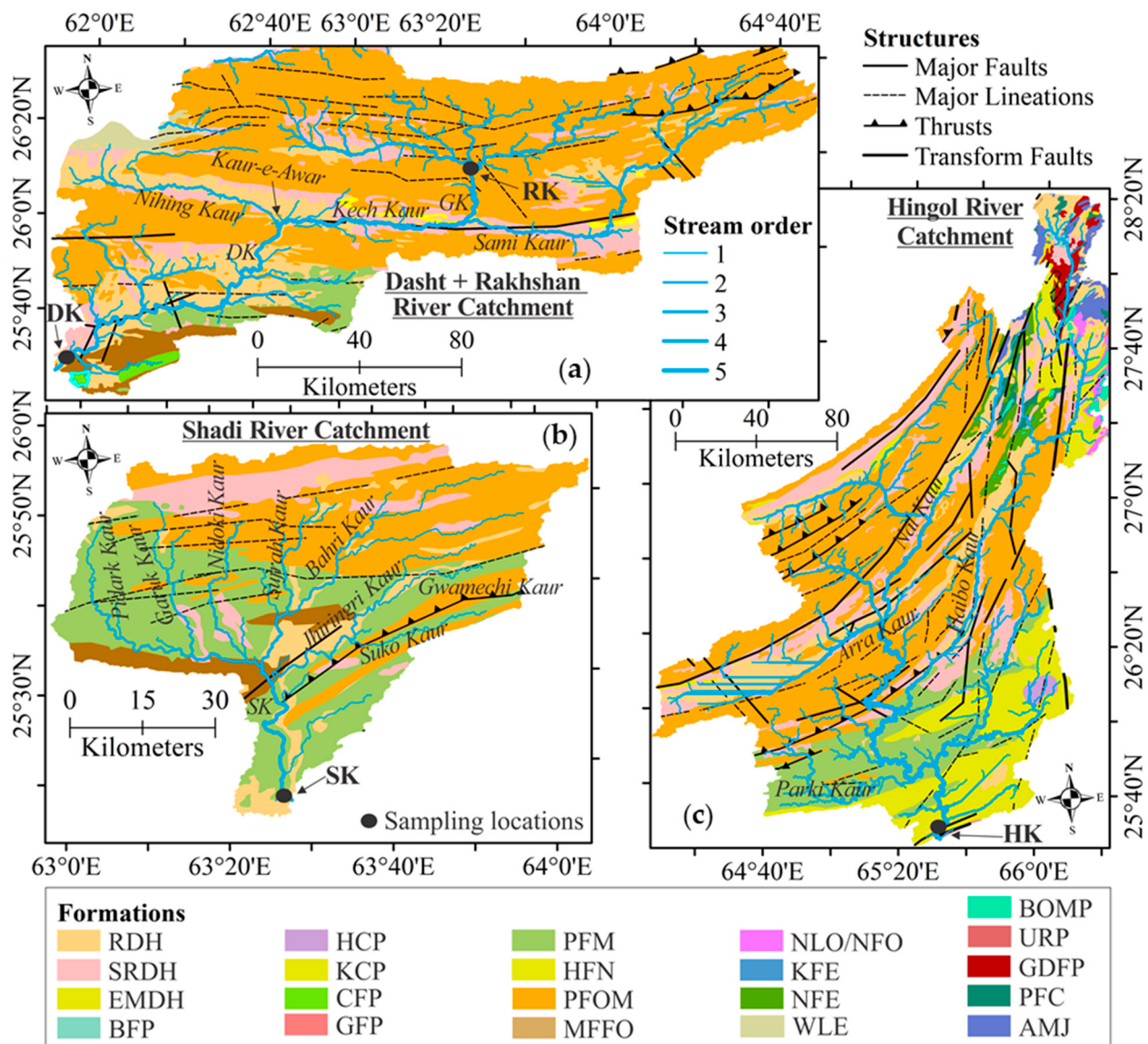


Figure 2. The catchment-based geological maps of eastern Makran in Pakistan. (a) The geological map of the Dasht River catchment including the Rakhshan River catchment, (b) the Shadi River catchment, and (c) the Hingol River catchment. Note that suffix “K” represents Kaur, which is a local word in the Balochi language meaning river. Where, RDH: recent deposits; SRDH: sub-recent deposits; EMDH: extrusive mud deposits; BFP: Bostan Formation; HCP: Haro Conglomerate; KCP: Kech Conglomerate; CFP: Chatti Formation; GFP: Gwadar Formation; PFM: Parkini Formation; HFN: Hinglag Formation; PFOM: Panjgur Formation; MFFO: Murgha Faqirzai Formation; NLO/NFO: Nal Limestone and Nari Formation; KFE: Kirthar Formation; NFE: Nisai Formation; WLE: Wakai Limestone; BOMP: Bela Ophiolitic Mélange; URP: undifferentiated rocks; GDFFP: Gidar Dhor Formation; PFC: Pab Formation; AMJ: Anjira Member [20–26].

3. Sampling and Analytical Techniques

3.1. Data Acquisition and Laboratory Techniques

Some necessary tools were used to collect the sediment samples during the fieldwork. These include a GPS (Garmin eTrex 10 handheld), a shovel, a pan to carry the sediments, a measuring tape, sampling bags, and stationery such as pens, markers, notebooks, etc. Four major outlets were selected for sampling and field observations. These include the Rakhshan River in Panjgur, the Dasht River in Jiwani, the Shadi River in Pasni, and the Hingol River in Lasbela (Figures 1 and 2). Four samples (2 km each) from four outlets

were collected for whole rock geochemical examinations. The samples were collected perpendicularly to the flow direction about 5 m between samples in a row at downstream of the four rivers. This sampling procedure was applied to gain the spatial heterogeneity of river sediments. The fine-grained sediment samples from DK were collected from surface sediment of ~1-m water depth. The dry coarser sediment samples were collected from RK and SK, and coarser wet sediment samples were collected from HK; i.e., the grain size in increasing order is DK < RK < SK < HK. The boulders/coarse materials were removed without sieving procedures and the sediment samples were air-dried prior to the laboratory examinations.

The concentrations of major elements were examined by XRF. The specimens were analyzed using the method of lithium borate fusion. The major oxides were examined by X-ray fluorescence spectroscopy (XRF) following the laboratory's analytical procedures. For the sample preparation, the lithium borate fusion technique was used. First, lithium metaborate (LiBO_2)–lithium tetraborate ($\text{Li}_2\text{B}_4\text{O}_7$) flux containing lithium nitrate (LiNO_3) was mixed with the prepared samples as an oxidizing agent. The mixture was subsequently poured into a platinum crucible. Last, XRF recorded the resulting discs. The inductively coupled plasma–mass spectrometry (ICP–MS) and inductively coupled plasma–atomic emission spectrometry (ICP–AES) methods were applied to measure the trace element concentrations consisting of REEs. The laboratory analytical procedures are as follows: powdered samples (25 mg) were weighed in the first stage and two solution subsamples were subsequently prepared. Perchloric (HClO_4), nitric (HNO_3), and hydrofluoric (HF) acids were digested in a single subsample. The remains were discharged and diluted to volume in dilute hydrochloric acid (HCl). The ICP–MS for ultra-trace level elements and the ICP–AES for trace level elements were used for examinations and then the digestion solution was analyzed. The results were corrected for spectral inter-element interferences. The other subsamples were mixed with the flux of lithium metaborate/lithium tetraborate, blended, and melted at 1025 °C in a furnace. In an acid mixture containing nitric, hydrochloric, and hydrofluoric acids, the resulting melt was cooled and dissolved. To test this approach, ICP–MS was used. In addition, the organic matter content (OMC) of sediment samples is determined using loss on ignition (LOI) analysis. The organic fraction was removed through LOI using a muffle furnace at 1000 °C. The LOI determines OMC by comparing the sample's weight before and after sediment ignition. Before ignition, the sample contains organic matter, but following ignition, only the mineral fraction of the sediment remains. The difference between the sample's weight before and after ignition represents the amount of OMC in the sample [29]. Thus, to test this loss, the concentrations of loss on ignition (LOI) were calculated. The LOI contents are not explained at length in this study but are used to support the reproducibility of data for future research. The geochemical analyses were prepared and analyzed in the Guangdong Provincial Key Laboratory of Geodynamics and Geohazards, School of Earth Sciences and Engineering, Sun Yat-Sen University, China.

3.2. Data Processing

The relationships between elements were explored using correlation and principal component analysis (PCA). Major oxides and trace elements were normalized against upper continental crust (UCC) values [30], and REEs were normalized against chondrite-normalized values [31].

Nesbitt and Young [32,33] offered CIA to calculate the intensity of weathering following the molar proportions, where CIA is solved by Equation (1):

$$\text{CIA} = \frac{\text{Al}_2\text{O}_3}{(\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O})} \times 100 \quad (1)$$

where the weathering intensity scale ≤ 50 refers to unweathered, 50–60 is low weathered, 60–80 is moderately weathered, and ≥ 80 is intense weathered [34].

Cox et al. [35] introduced the index of chemical variability (ICV) for sedimentary maturation. The ICV is calculated to measure the alumina content for the assessment of maturity compared to leading cations. The ICV can be calculated according to Equation (2) as follows:

$$\text{ICV} = \frac{(\text{Fe}_2\text{O}_3 + \text{K}_2\text{O} + \text{Na}_2\text{O} + \text{CaO}^* + \text{MgO} + \text{MnO} + \text{TiO}_2)}{\text{Al}_2\text{O}_3} \quad (2)$$

According to the atomic weights and radii, REEs are typically categorized into LREE or HREE groups [36,37]. There are two distinguishing features of REE concentrations in nature. The concentrations of REEs follow two patterns: (1) La to Lu decrease, and (2) even atomic numbers of REEs are more abundant in their immediate vicinity. As a standard practice, REE contents are normalized to a reference in order to eliminate these two effects and display distribution patterns graphically [38]. The differences in oxidation state and greater fractionation relative to other REEs [39] make Ce and Eu unique among REEs and make them indispensable for redox-state tracing. The methods can be calculated to determine Ce and Eu anomalies by Equations (3) and (4), respectively [36]:

$$\text{Ce} = \frac{\text{Ce}_N}{(\text{La}_N \times \text{Pr}_N)^{0.5}} \quad (3)$$

$$\text{Eu} = \frac{\text{Eu}_N}{(\text{Sm}_N \times \text{Gd}_N)^{0.5}} \quad (4)$$

where the initials represent REEs and “N” subscripts represent values normalized against chondrite values. An anomaly with a value above 1 is considered positive, while an anomaly with a value below 1 is considered negative.

4. Results

4.1. Correlation Matrices of the Major Elements

The correlation matrix and PCA is calculated to examine the range of negative and positive relationship of the studied samples and is shown in Figures 3 and 4. The watershed RK shows a positive correlation of Al_2O_3 , Fe_2O_3 , CaO , Na_2O , and K_2O with SiO_2 , while TiO_2 , MnO , MgO , P_2O_5 , and LOI negatively correlate with SiO_2 (Figure 3 and Table S1). The watershed DK represents the positive correlation of CaO and Na_2O with SiO_2 , while Al_2O_3 , Fe_2O_3 , TiO_2 , MnO , MgO , K_2O , P_2O_5 , and LOI negatively correlate with SiO_2 . The watershed SK denotes a positive correlation of Al_2O_3 , Fe_2O_3 , Na_2O , and K_2O with SiO_2 , whereas TiO_2 , MnO , MgO , CaO , P_2O_5 , and LOI negatively correlate with SiO_2 . The watershed HK signifies a positive correlation of TiO_2 , Al_2O_3 , MgO , Na_2O , K_2O , and P_2O_5 with SiO_2 , while Fe_2O_3 , MnO , CaO , and LOI negatively correlate with SiO_2 . The negative correlation between SiO_2 and LOI represents sediments with low amounts of silicate minerals, for instance, quartz and clay, which have excessive amounts of carbonate and organic materials [40].

SiO_2 vs. CaO shows a positive correlation in the RK and DK but not SK and HK samples, representing the possible influence of detrital carbonates/microfossils in SK and HK sediments, which is further supported by a positive correlation b/w CaO and LOI in SK and HK sediments. The Al_2O_3 vs. SiO_2 negative correlation of river DK is possibly due to quartz dilution and sorting/grain-size effects as fine-grained sediment samples were collected from DK (Figure 3). The MgO , K_2O , Fe_2O_3 , and TiO_2 have a clear positive association, suggesting that these abundances are often regulated in particular size grades by aluminous clay or heavy minerals. The close relationship between P_2O_5 and MnO of rivers RK, DK, and SK indicates the regulation and/or partnership of heavy minerals with phyllosilicate fractions (Figure 4a–c and Tables S1–S6), whereas no such relationship between P_2O_5 and MnO is found in river HK (Figure 4d). In most of the sandy specimens, Na_2O is negatively correlated with SiO_2 , and high concentrations of Na_2O are observed. The sandy samples show an association between CaO and high Na_2O . The

above characteristics suggest that the abundance of CaO and Na₂O is primarily associated with low weathered feldspars. Al₂O₃ is reduced, while in clay-rich samples, its content is fairly enriched, suggesting quartz dilution. However, K₂O, MgO, and Na₂O are markedly depleted (Figure 3), suggesting that during weathering, progressive loss of ferromagnesian minerals occurs [41]. Moreover, the positive correlation between SiO₂ and Al₂O₃, CaO, Na₂O, and K₂O may be controlled by the presence of rock-forming minerals associated with the lithic fragments.

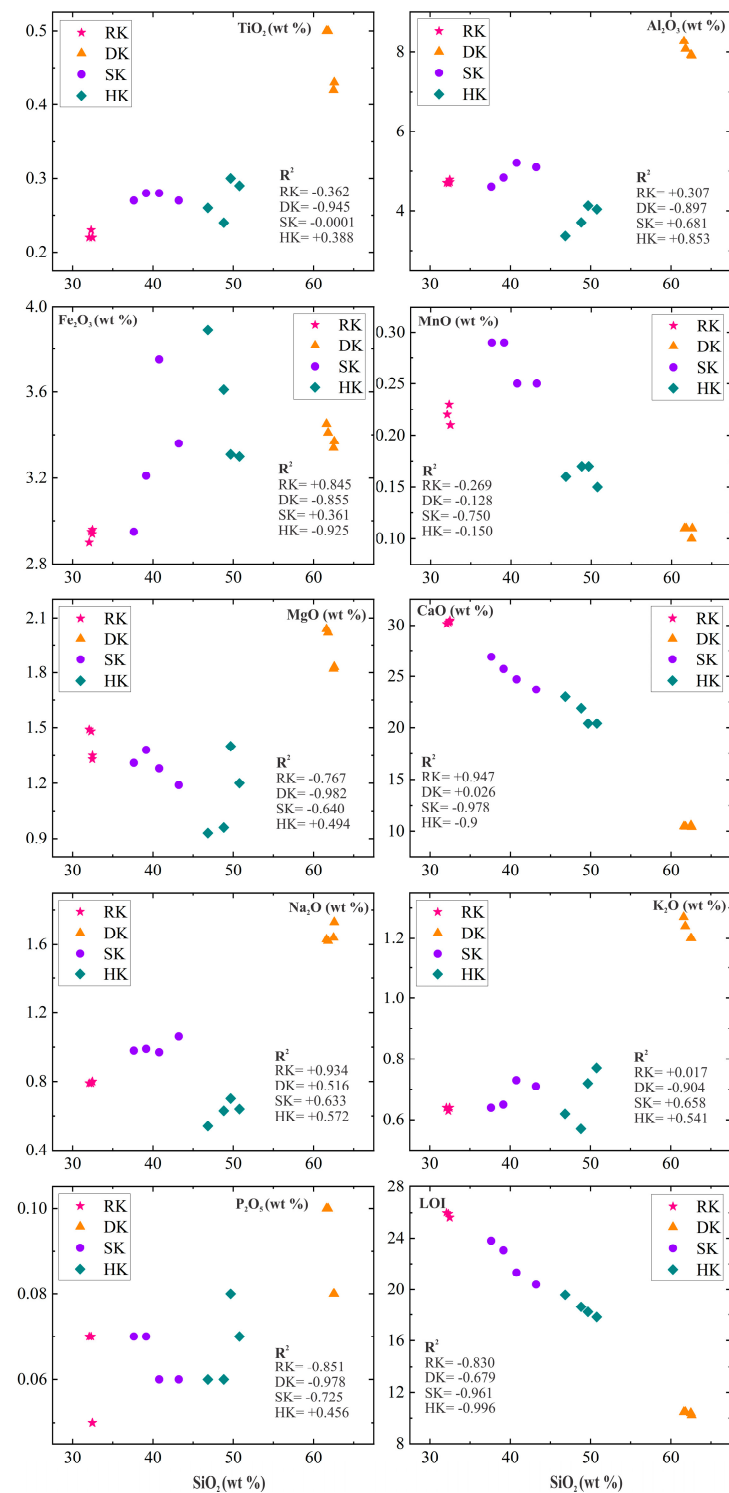


Figure 3. Harker plot showing the correlation of major elements vs. SiO₂.

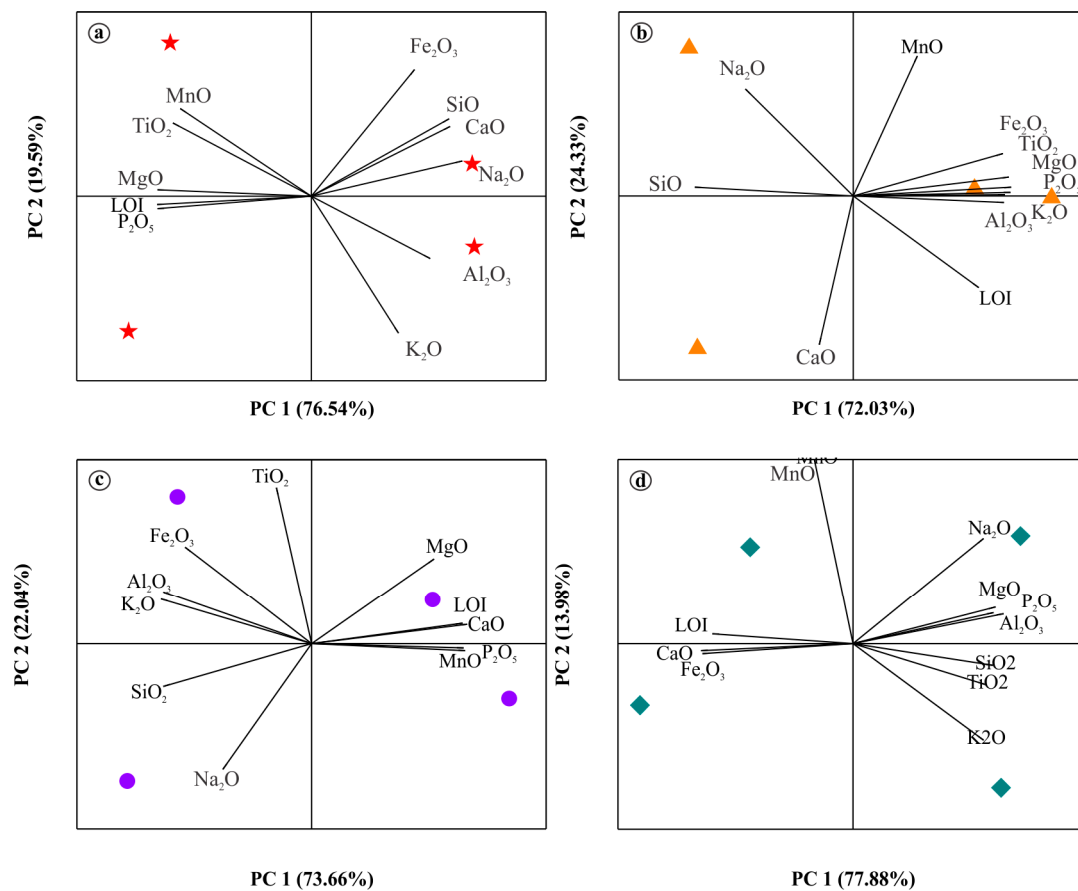


Figure 4. PCA biplots showing the correlation matrices of (a) RK river (b) DK river (c) SK river, and (d) HK river.

4.2. Major Elements

The major elemental concentrations are listed in (Table 1) with the UCC values of [30]. The major concentrations show variations from river to river. The mean SiO_2 concentrations are 32.32%, 62.12%, 40.19%, and 49.03% and the mean Al_2O_3 concentrations are 4.32%, 8.05%, 4.49%, and 3.81%, of river sediments RK, DK, SK, and HK, respectively. This reflects an increase in the ratios of $\text{SiO}_2/\text{Al}_2\text{O}_3$ (6.83, 7.72, 8.14, and 12.91 of river sediments RK, DK, SK, and HK, respectively) as compared to UCC (4.34). TiO_2 contents are significantly depleted as compared to UCC. The contents of total Fe_2O_3 show depletion. The concentrations of CaO are very high, with a mean of 30.34% for RK, 10.46% for DK, 25.24% for SK, and 21.43 % for HK. The concentrations of MgO , MnO , K_2O , Na_2O , and P_2O_5 show great depletion. The enrichment and depletion of the major oxides normalized to UCC are shown in Figure 5a.

Table 1. The examined major oxide concentrations and the calculated parameters of sorting, source area weathering, and provenance of the river sediments of the eastern Makran outlets in Pakistan.

	SiO ₂	Al ₂ O ₃	TiO ₂	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	LOI	S/A	K/Rb	A/T	CIA	ICV
RK1	32.3	4.70	0.23	2.95	0.23	1.48	30.3	0.79	0.63	0.07	25.9	6.87	0.02	20.43	58.8	7.79
RK2	32.5	4.73	0.22	2.96	0.21	1.35	30.5	0.80	0.64	0.05	25.6	6.86	0.02	21.50	58.7	7.75
RK3	32.1	4.71	0.22	2.90	0.22	1.49	30.2	0.79	0.64	0.07	26.0	6.80	0.02	21.41	58.9	7.73
RK4	32.4	4.79	0.22	2.94	0.21	1.33	30.4	0.80	0.64	0.05	25.6	6.77	0.02	21.77	59.0	7.63
Mean	32.3	4.73	0.22	2.94	0.22	1.41	30.3	0.80	0.64	0.06	25.8	6.83	0.02	21.28	58.9	7.73
DK1	61.8	8.08	0.50	3.41	0.11	2.02	10.4	1.62	1.24	0.10	10.5	7.65	0.02	16.16	54.8	2.39
DK2	62.5	7.93	0.42	3.34	0.10	1.82	10.6	1.64	1.20	0.08	10.4	7.88	0.02	18.88	54.2	2.40
DK3	61.6	8.27	0.50	3.45	0.11	2.04	10.5	1.63	1.27	0.10	10.5	7.45	0.02	16.54	55.1	2.35
DK4	62.6	7.91	0.43	3.37	0.11	1.83	10.4	1.73	1.20	0.08	10.2	7.91	0.02	18.40	53.1	2.41
Mean	62.1	8.05	0.46	3.39	0.11	1.93	10.5	1.66	1.23	0.09	10.4	7.72	0.02	17.49	54.3	2.39
SK1	37.6	4.61	0.27	2.95	0.29	1.31	26.9	0.98	0.64	0.07	23.8	8.17	0.02	17.07	58.1	6.61
SK2	43.2	5.10	0.27	3.36	0.25	1.19	23.7	1.06	0.71	0.06	20.4	8.47	0.02	18.89	57.9	8.72
SK3	39.2	4.84	0.28	3.21	0.29	1.38	25.7	0.99	0.65	0.07	23.0	8.09	0.02	17.29	55.0	6.71
SK4	40.8	5.21	0.28	3.75	0.25	1.28	24.7	0.97	0.73	0.06	21.3	7.83	0.02	18.61	56.7	6.13
Mean	40.2	4.94	0.28	3.32	0.27	1.29	25.2	1.00	0.68	0.07	22.1	8.14	0.02	17.96	56.9	7.04
HK1	49.7	4.13	0.30	3.31	0.17	1.40	20.4	0.70	0.72	0.08	18.2	12.0	0.02	13.77	57.2	6.53
HK2	48.8	3.71	0.24	3.61	0.17	0.96	21.9	0.63	0.57	0.06	18.6	13.2	0.02	15.46	58.0	7.57
HK3	50.8	4.04	0.29	3.30	0.15	1.20	20.4	0.64	0.77	0.07	17.8	12.6	0.02	13.93	58.1	6.61
HK4	46.8	3.37	0.26	3.89	0.16	0.93	23.0	0.54	0.62	0.06	19.6	13.9	0.03	12.96	57.9	8.72
Mean	49.0	3.81	0.27	3.53	0.16	1.12	21.4	0.63	0.67	0.07	18.6	12.9	0.02	14.03	57.8	7.36
UCC	66.0	15.20	0.68	5.03	0.08	2.20	4.20	3.90	3.40	0.15		4.34	0.03	22.35		

The concentrations are in wt %. CIA is calculated using molar ratio. S represents SiO₂; A represents Al₂O₃; K represents K₂O; and T represents TiO₂.

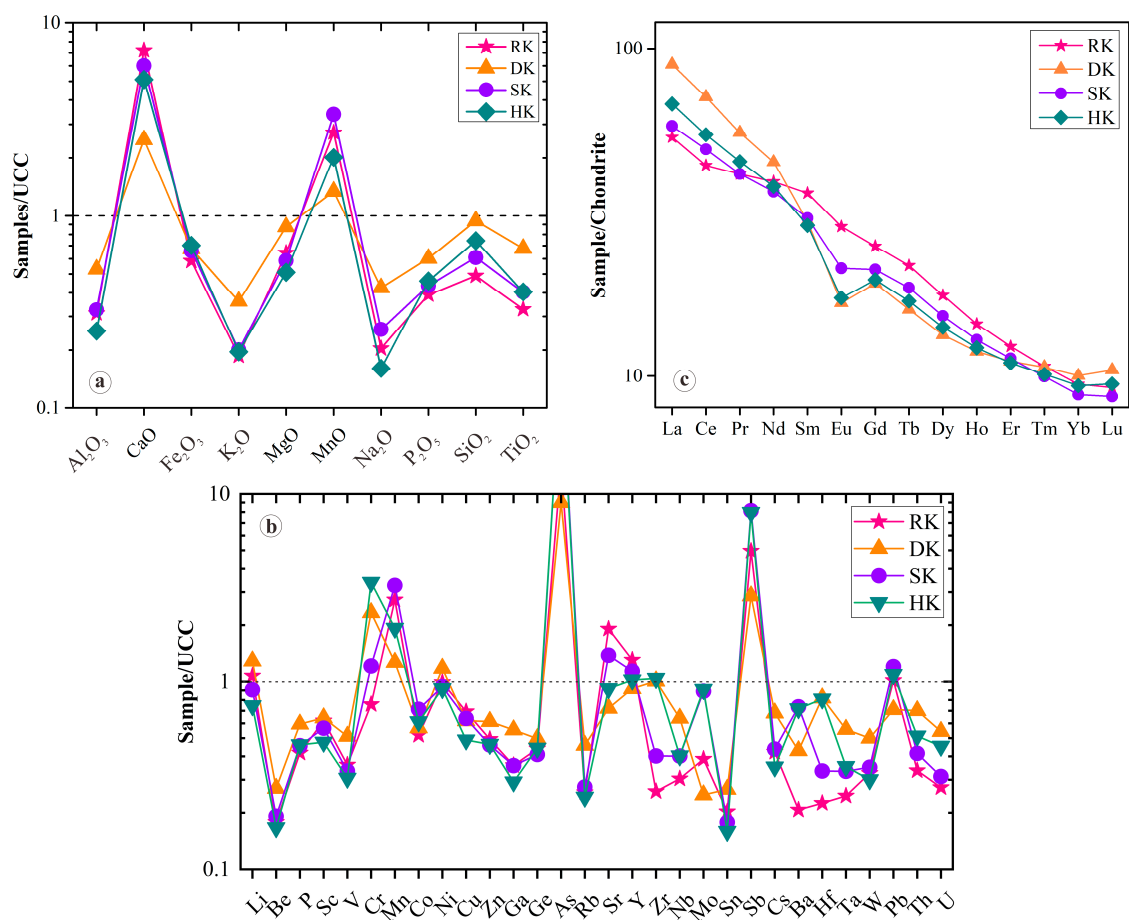


Figure 5. Spider diagrams illustrating the mean normalized patterns of the sediments at the outlets of the eastern Makran in Pakistan: (a) the mean of major oxides normalized against UCC [30,31]; (b) the mean of trace elements normalized against UCC [30]; and (c) the mean of rare earth elements normalized against chondrite [31].

4.3. Trace Elements

The concentrations of trace elements are shown in Table 2. The enrichment and depletion values normalized against UCC are shown in Figure 5b. A wide variation is observed in trace element concentrations. The large ions lithophile elements (LILE) such as Th, U, Rb, and Ba were relatively depleted in the studied sediments. Sr concentrations were relatively high in the RK and SK samples, while depleted in the DK and HK samples. Cs concentrations were depleted in all river sediments, whereas Pb concentrations were significantly high with the exception of samples from DK. The wide variation in Sr concentrations indicates extreme fractionation of plagioclase probably linked with the source areas [42]. The excessive carbonate, as shown by the negative correlation between SiO_2 and LOI (Figure 3), may have also controlled the Sr content in sediments [43]. The concentrations of Ni and Cr were enriched, while the concentrations of V, Co, and Sc were depleted in the studied samples. Y was enriched in RK, SK, and HK, indicating that the excess was regulated principally by heavy minerals. Relative to UCC, Ba was greatly depleted in the RK and DK samples and relatively depleted in the SK and HK samples. Sr concentrations were greatly depleted in DK and HK, while in SK and RK they were greatly increased, the depletion of Ba and Sr elements indicating feldspar breakdown in chemical weathering.

4.4. Rare Earth Elements

The ΣREE contents were consistently depleted as compared to (UCC: 148) [30] in the studied sediments (Table 3). The higher ratios of $(\text{La}/\text{Yb})_N$ were probably due to a decrease in the HREEs. The $(\text{La}/\text{Sm})_N$ were relatively depleted, as compared to the (UCC: 4.26). The $(\text{Gd}/\text{Yb})_N$ ratios were also depleted. The Eu/Eu^* pattern in the chondrite normalized diagram indicates negative anomalies (Figure 5c). Moreover, the samples from RK show no significant Eu anomaly.

Table 2. The trace elements concentrations in river sediments at the outlets of eastern Makran, Pakistan.

	RK1	RK2	RK3	RK4	Mean	DK1	DK2	DK3	DK4	Mean	SK1	SK2	SK3	SK4	Mean	HK1	HK2	HK3	HK4	Mean	UCC
Li	22.5	20.4	22.4	20.5	21.5	27.2	24.0	28.0	23.8	25.8	18.6	16.4	17.4	20.1	18.1	16.1	15.9	15.5	12.1	14.9	20.0
Sc	8.76	7.90	9.03	8.00	8.42	9.77	7.80	9.72	7.60	8.72	8.15	7.30	7.40	7.89	7.69	7.54	5.10	7.95	5.30	6.47	13.6
V	39.5	37.0	40.6	37.0	38.5	57.9	50.0	58.1	53.0	54.8	35.6	34.0	36.0	37.4	35.8	34.6	31.0	36.2	29.0	32.7	107
Cr	71.7	51.0	76.8	52.0	62.9	280	114	255	125	194	114	82	103	103	101	360	169	401	193	281	83
Co	8.89	8.60	9.11	8.70	8.83	10.2	9.10	10.3	9.00	9.65	11.3	11.9	13.1	12.2	12.1	10.4	10.3	10.8	10.1	10.4	17.0
Ni	44.3	42.5	45.3	42.4	43.6	53.8	49.7	54.6	49.1	51.8	40.1	39.4	43.0	43.0	41.4	41.1	41.9	41.6	37.1	40.4	44.0
Cu	17.2	17.2	17.4	17.5	17.3	15.6	15.3	16.0	15.0	15.5	14.1	17.1	18.5	14.0	15.9	12.4	11.8	13.0	11.7	12.2	25.0
Zn	34.6	35.0	35.5	35.0	35.0	45.0	41.0	45.3	43.0	43.6	31.0	32.0	36.0	31.6	32.7	36.0	30.0	39.6	26.0	32.9	71.0
Ga	6.49	5.58	6.59	5.67	6.08	10.2	8.67	10.3	8.46	9.41	6.36	5.64	5.81	6.46	6.07	5.79	4.35	5.93	3.80	4.97	17.0
Ge	1.31	0.07	1.36	0.07	0.70	1.52	0.08	1.50	0.08	0.80	1.24	0.08	0.07	1.23	0.66	1.32	0.05	1.38	0.08	0.71	1.60
As	-	21.3	-	21.5	21.4	-	13.6	-	13.4	13.5	-	73.4	75.0	-	74.2	-	58.2	-	84.7	71.5	1.50
Rb	28.2	28.8	29.2	28.8	28.8	52.4	50.3	53.2	49.2	51.3	29.5	31.6	32.1	29.8	30.8	28.7	24.5	31.3	24.0	27.1	112
Sr	646	680	664	682	668	247	251	253	260	253	512	456	473	492	483	316	335	311	324	322	350
Y	29.1	27.5	29.9	28.3	28.7	23.6	16.0	25.6	15.7	20.2	27.7	23.2	23.0	26.1	25.0	24.7	16.9	28.9	19.3	22.5	22.0
Zr	73.3	27.7	69.4	27.2	49.4	359	35	336	35	191	134	24.0	25.3	122.0	76.3	359	24.9	379.0	27.6	198	190
Nb	3.75	3.50	3.84	3.50	3.65	8.71	6.60	8.76	6.60	7.67	5.06	4.40	4.70	5.11	4.82	5.54	3.90	6.07	3.80	4.83	12.0
Mo	0.57	0.59	0.58	0.58	0.58	0.37	0.37	0.37	0.38	0.37	0.74	1.17	2.42	1.01	1.34	1.28	1.30	1.44	1.44	1.37	1.50
Ag	-	0.04	-	0.05	0.05	-	0.05	-	0.05	0.05	-	0.06	0.05	-	0.06	-	0.06	-	0.06	0.06	50.0
Cd	0.31	0.24	0.30	0.25	0.28	0.46	0.11	0.46	0.10	0.28	0.27	0.16	0.17	0.27	0.22	0.51	0.14	0.50	0.13	0.32	98.0
In	-	0.03	-	0.03	0.03	-	0.04	-	0.03	0.04	-	0.03	0.03	-	0.03	-	0.02	-	0.02	0.02	50.0
Sn	1.20	1.10	1.16	1.00	1.12	1.61	1.30	1.66	1.30	1.47	0.95	1.00	1.00	0.97	0.98	0.97	0.80	1.03	0.70	0.88	5.50
Sb	0.92	1.11	0.90	1.04	0.99	0.55	0.63	0.54	0.57	0.57	0.80	1.65	2.89	1.17	1.63	1.06	1.49	1.38	2.44	1.59	0.20
Cs	1.89	1.95	1.93	1.99	1.94	3.22	3.05	3.29	2.97	3.13	1.88	2.05	2.17	1.94	2.01	1.68	1.55	1.78	1.45	1.62	4.60
Ba	-	100	142	100	114	267	200	266	210	236	374	360	500	385	405	427	320	422	410	395	550
Hf	1.86	0.80	1.76	0.80	1.31	8.60	1.00	8.31	1.10	4.75	3.33	0.60	0.70	3.12	1.94	8.60	0.60	8.78	0.80	4.70	5.80
Ta	0.25	0.25	0.24	0.25	0.25	0.61	0.50	0.61	0.51	0.56	0.33	0.33	0.34	0.33	0.33	0.38	0.29	0.46	0.28	0.35	1.00
W	-	0.60	-	0.70	0.65	-	1.00	-	1.00	1.00	-	0.70	0.70	-	0.70	-	0.60	-	0.60	0.60	2.00
Tl	-	0.14	-	0.14	0.14	-	0.24	-	0.24	0.24	-	0.17	0.16	-	0.17	-	0.13	-	0.13	0.13	750
Pb	17.7	16.9	17.5	16.9	17.3	12.9	11.4	13.0	11.0	12.1	17.2	21.6	25.0	18.0	20.5	17.1	18.5	20.1	18.5	18.6	17.0
Bi	-	0.22	-	0.24	0.23	-	0.16	-	0.16	0.16	-	0.18	0.17	-	0.18	-	0.10	-	0.11	0.11	127
Th	3.65	3.53	3.68	3.52	3.60	8.34	6.79	8.40	6.48	7.50	4.51	4.40	4.44	4.40	4.44	5.75	4.36	5.96	5.90	5.49	10.0
U	0.84	0.70	0.81	0.70	0.76	1.91	1.10	1.97	1.10	1.52	0.99	0.80	0.70	1.01	0.88	1.58	0.80	1.59	1.10	1.27	2.80
Rb/Sr	0.04	0.04	0.04	0.04	0.04	0.21	0.20	0.21	0.19	0.20	0.06	0.07	0.07	0.06	0.07	0.09	0.07	0.10	0.07	0.08	0.32
Zr/Sc	8.37	3.51	7.69	3.40	5.74	36.8	4.54	34.6	4.66	20.1	16.4	3.29	3.42	15.5	9.65	47.6	4.88	47.7	5.21	26.3	14.0
Th/Sc	0.42	0.45	0.41	0.44	0.43	0.85	0.87	0.86	0.85	0.86	0.55	0.60	0.60	0.56	0.58	0.76	0.85	0.75	1.11	0.87	0.79
Cr/Ni	1.62	1.20	1.70	1.23	1.44	5.20	2.29	4.67	2.55	3.68	2.84	2.08	2.40	2.40	2.43	8.76	4.03	9.64	5.20	6.91	1.89
Y/Ni	0.66	0.65	0.66	0.67	0.66	0.44	0.32	0.47	0.32	0.39	0.69	0.59	0.53	0.61	0.61	0.60	0.40	0.69	0.52	0.55	0.50
Cr/V	1.82	1.38	1.89	1.41	1.63	4.84	2.28	4.39	2.36	3.47	3.20	2.41	2.86	2.75	2.81	10.40	5.45	11.08	6.66	8.40	0.78

The concentrations are in ppm whereas ultra-trace concentrations are in ppb, such as Cd, In, Ti, Bi, Ag, etc.

Table 3. The rare earth element concentration in river sediments from easter Makran (values normalized to chondrite concentration [31]).

	RK1	RK2	RK3	RK4	Mean	DK1	DK2	DK3	DK4	Mean	SK1	SK2	SK3	SK4	Mean	HK1	HK2	HK3	HK4	Mean	UCC
La	12.3	12.9	12.4	13.4	12.8	21.1	21.6	21.8	20.3	21.2	13.1	14.6	15.2	12.3	13.8	16.1	13.9	16.2	18.2	16.1	31.0
Ce	24.6	28.8	24.8	29.5	26.9	42.7	44.9	44.8	42.2	43.7	28.0	32.4	34.0	26.4	30.2	32.7	29.5	33.5	38.3	33.5	63.0
Pr	3.75	4.02	3.79	4.15	3.93	5.21	5.39	5.47	5.04	5.28	3.73	4.11	4.39	3.49	3.93	4.21	3.73	4.45	4.79	4.30	7.10
Nd	17.5	18.8	17.6	19.3	18.3	20.6	21.6	21.5	20.4	21.0	16.4	18.0	18.6	15.3	17.1	17.4	15.6	18.7	19.1	17.7	27.0
Sm	5.35	5.60	5.37	5.81	5.53	4.55	4.53	4.72	4.39	4.55	4.65	4.73	4.90	4.32	4.65	4.29	3.78	4.88	4.72	4.42	4.70
Eu	1.69	1.62	1.70	1.63	1.66	1.03	0.94	1.04	0.88	0.97	1.33	1.21	1.19	1.23	1.24	1.05	0.87	1.16	0.95	1.01	1.00
Gd	5.30	4.98	5.27	4.96	5.13	4.46	3.47	4.66	3.23	3.96	4.73	4.20	4.14	4.36	4.36	4.36	3.24	4.90	3.73	4.06	4.00
Tb	0.87	0.76	0.88	0.75	0.82	0.69	0.49	0.73	0.48	0.60	0.78	0.66	0.63	0.72	0.70	0.70	0.48	0.80	0.56	0.64	0.70
Dy	4.78	4.19	4.81	4.21	4.50	3.92	2.69	4.20	2.73	3.39	4.35	3.55	3.51	4.09	3.88	3.96	2.68	4.55	3.10	3.57	3.90
Ho	0.87	0.76	0.88	0.76	0.82	0.79	0.53	0.86	0.51	0.67	0.82	0.67	0.65	0.78	0.73	0.77	0.52	0.90	0.57	0.69	0.83
Er	2.15	1.89	2.17	1.93	2.04	2.21	1.37	2.42	1.34	1.84	2.11	1.68	1.67	2.02	1.87	2.09	1.31	2.42	1.44	1.82	2.30
Tm	0.30	0.25	0.30	0.25	0.28	0.34	0.19	0.37	0.19	0.27	0.29	0.22	0.22	0.28	0.25	0.31	0.17	0.36	0.19	0.26	0.30
Yb	1.72	1.48	1.72	1.49	1.60	2.16	1.18	2.31	1.16	1.70	1.72	1.24	1.31	1.68	1.49	1.95	1.00	2.22	1.17	1.59	2.00
Lu	0.26	0.21	0.26	0.21	0.24	0.35	0.18	0.36	0.17	0.27	0.26	0.18	0.18	0.26	0.22	0.30	0.14	0.35	0.17	0.24	0.31
ΣREE	81.4	86.3	81.9	88.4	84.5	110	109	115	103	109	82.3	87.5	90.6	77.2	84.4	90.2	76.9	95.4	97.0	89.9	148
Eu/Eu*	0.97	0.94	0.98	0.93	0.96	0.70	0.72	0.68	0.71	0.70	0.87	0.83	0.81	0.87	0.85	0.74	0.76	0.73	0.69	0.73	0.71
Ce/Ce*	0.89	0.98	0.89	0.97	0.93	1.00	1.02	1.01	1.02	1.01	0.98	1.03	1.02	0.99	1.01	0.97	1.00	0.97	1.01	0.99	0.99
(La/Sm) _N	1.48	1.49	1.49	1.49	1.49	2.99	3.08	2.98	2.99	3.01	1.82	1.99	2.00	1.84	1.91	2.42	2.37	2.14	2.49	2.36	4.26
(Gd/Yb) _N	2.55	2.78	2.53	2.75	2.65	1.71	2.43	1.67	2.30	2.03	2.27	2.80	2.61	2.15	2.46	1.85	2.68	1.83	2.64	2.25	1.65
(La/Yb) _N	5.13	6.25	5.17	6.45	5.75	7.01	13.13	6.77	12.55	9.87	5.46	8.45	8.32	5.25	6.87	5.92	9.97	5.23	11.2	8.07	11.1
(La/Lu) _N	5.17	6.58	5.11	6.84	5.93	6.52	12.86	6.45	12.80	9.66	5.40	8.69	9.05	5.13	7.07	5.73	10.6	4.97	11.5	8.20	10.4

The concentrations are in ppm.

5. Discussion

5.1. Sorting and Maturity of Sediments

The authors of [35,44,45] suggested the ICV index to distinguish the clastic mineralogical maturity. The value of the ICV is usually >0.84 for rock-forming minerals, for instance, pyroxene, amphibole, and feldspar, while less than 0.84 is denoted for clay minerals such as muscovites, illites, and kaolinites. In addition, [35,41] contended that the lower values of the ICV indicate recycled sediments and the siliciclastic rocks having greater values represent the immature type and low weathered sources. The calculated mean ICV estimations of the present study (RK = 7.73, DK = 2.39, SK = 7.04, and HK = 7.36) imply immature type, low recycled, and low weathered sources of the studied sediments (Table 1).

Due to the total amount of quartz in the sediment reflecting not only the composition of the sources but also the sorting of the sediment as a result of current activity and the effects of chemical weathering, it appears unlikely that we can use major element concentrations as a provenance proxy unless the sorting process can be accounted for [43]. Because this might fluctuate over time during seasonal flooding and ebbs of the river, as well as throughout the length of the river channel and across the channel with depth, extreme caution would be required. Even in a current river, this would be difficult to do, and it is unlikely to provide any useful results when applied to earlier deposits.

By comparing important ratios against one another, we can analyze the impact of heavy minerals on sediment chemistry. A plot of Th/Al vs. Zr/Al is shown in Figure 6a. This graph (Figure 6a) reveals a positive association, implying that sediments rich in monazite are likewise rich in zircons, as one might anticipate from currents that concentrate these dense minerals. The connection between heavy mineral concentration, as proxied by Th/Al, and relative enrichment in LREE and HREE, as proxied by La/Sm and Tb/Yb, is shown in Figure 6b,c. The LREEs are predominantly found in clay minerals, according to [44], although monazite and allanite are also high in these elements (Figure 6b,c), according to [45]. HREEs, on the other hand, are commonly found in heavy minerals such as xenotime and zircon [45]. The HREE-enriched heavy minerals have a substantial impact on REE compositions, indicating provenance control [46]. In addition, the increasing heavy minerals trend (Figure 6a,d) indicates higher zircon content in the HK river due to granitic detritus.

5.2. Tectonic and Climatic Conditions

The geochemical proxies are globally applied to summarize the tectonic and climatic conditions [10,47–56]. The binary plot of [48] based on SiO_2 vs. $\text{K}_2\text{O}/\text{Na}_2\text{O}$ is largely adopted to scrutinize the nature of tectonic settings. The studied sediments were plotted on the said discrimination diagram, indicating an active continental margin field for all the examined sediments, with some exceptions seen in samples of the RK and SK zones (Figure 7a). In addition, [52] documented a binary plot based on $(\text{La}/\text{Sm})_{\text{N}}$ vs. Nb/La to distinguish the tectonic environments of clastic rocks. The river sediments from the DK zone fell at the junction of the active continental margin and continental arc, whereas the sediments from the RK, SK, and HK zones covered the area of the continental arc (Figure 7b). To sum up all the observations, the tectonic environments of the studied sediments are more possibly continental arc rather than the active continental margin.

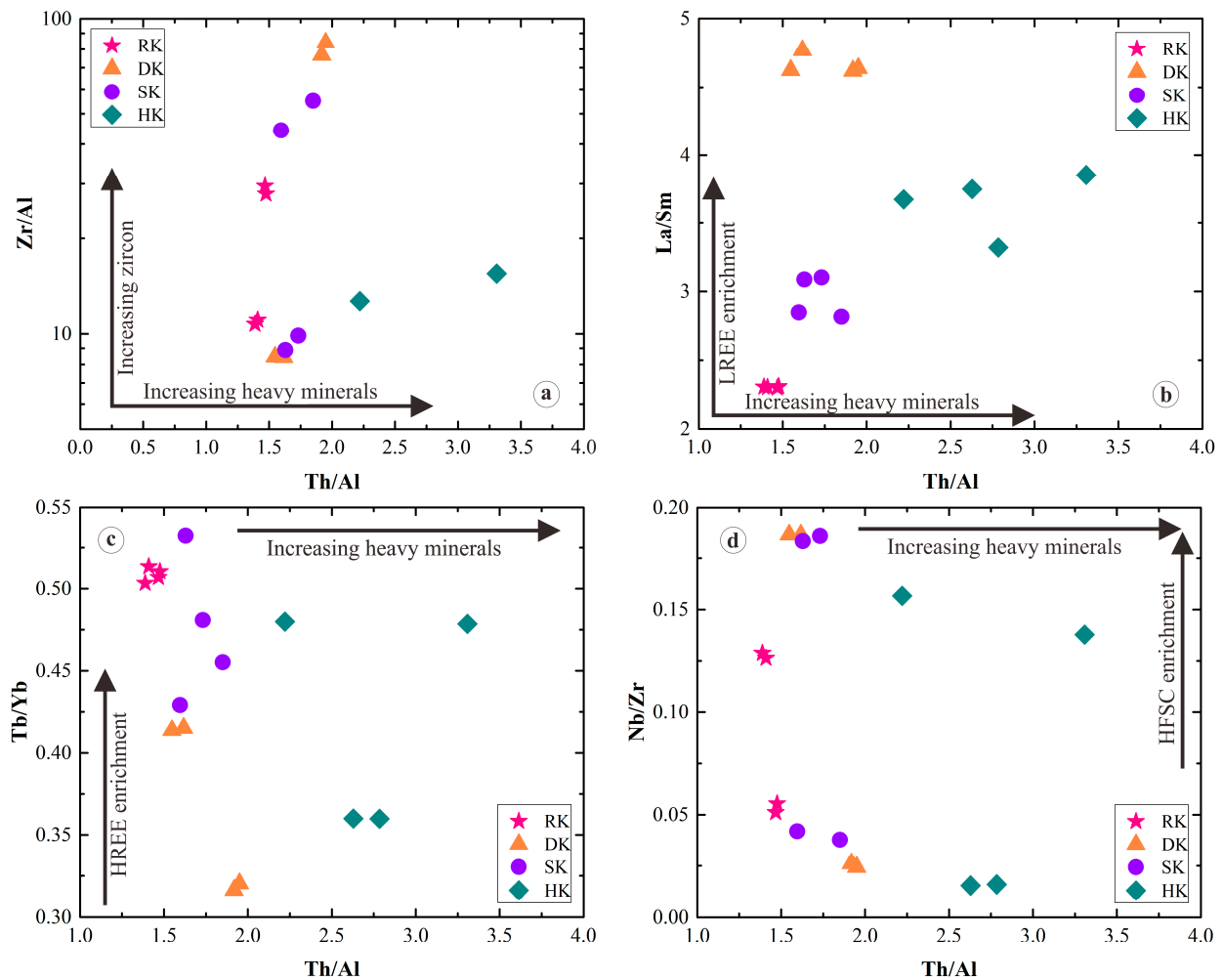


Figure 6. Demonstrating the trace elements variations in the eastern Makran river sediments: (a) Zr/Al vs. Th/Al for increasing zircon compared to the accumulation of heavy minerals; (b) La/Sm vs. Th/Al; (c) Tb/Yb vs. Th/Al; and (d) Nb/Zr vs. Th/Al.

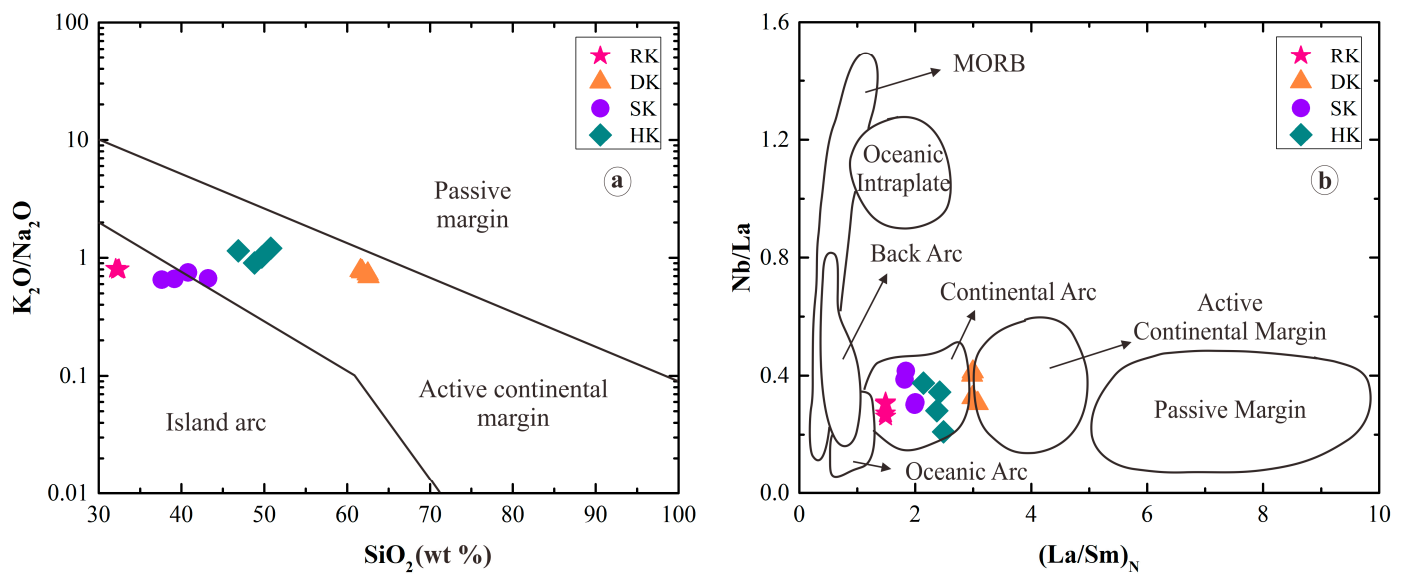


Figure 7. Representative graph of plate tectonic environments: (a) K_2O/Na_2O vs. SiO_2 [48] (b) Nb/La vs. $(La/Sm)_N$ [53].

The authors of [57] proposed $\text{Al}_2\text{O}_3 + \text{K}_2\text{O} + \text{Na}_2\text{O}$ vs. SiO_2 illustration to record the climatic conditions. The river sediments were plotted to test the climatic conditions, indicating that the sediments from the HK zone fall in humid conditions and are somewhat chemically mature due to their provenances, sediments from RK fall at the junction of the humid and arid climate, those from SK fall at the junction of the humid and arid climate, and the DK sediments also fall in the boundary of the semi-arid climate (Figure 8).

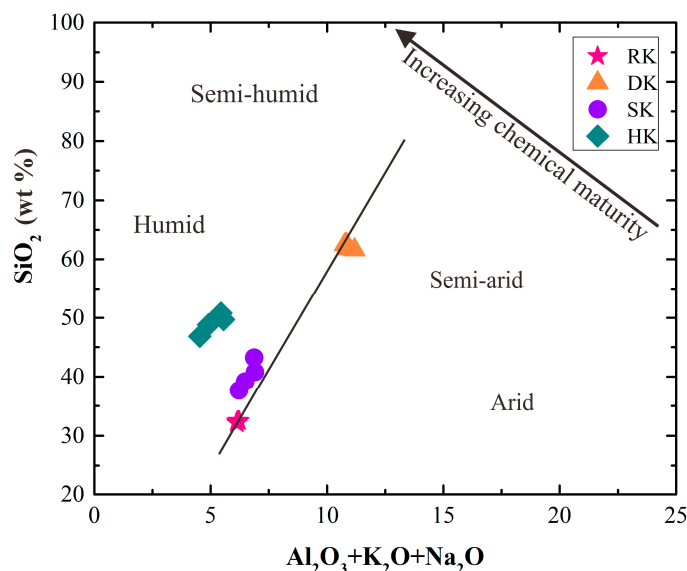


Figure 8. Cross plot showing the determination of the climatic conditions of the studied sediments [57].

5.3. Source Area Weathering

The weathering intensity is largely a consequence of climate variability and rate of tectonic uplift. The CIA drafted by [34] is widely applied to determine the intensities of chemical weathering of clastic rocks. The provenance commonly affects the weathering intensity of various parent materials; thus, the chemical composition of sedimentary rock is controlled by its lithology [34,55,58]. The primary feldspar in association with secondary clay minerals is quantitatively estimated by the CIA index. The altered feldspar ranges from a CIA value of 50 to 100, while the unweathered intensity is usually <50 [32,34]. The mean CIA intensity of the studied river sediments varies between RK (58.87), DK (54.31), SK (56.93), and HK (57.80) with an overall mean of 56.98 (Table 1), indicating a low level of chemical weathering of the parent materials. This can also be visualized by A–CN–K triangular diagram (Figure 9a) to observe the trend of weathering and to evaluate the post-depositional K-metasomatism degree [33,34]. The plotted river sediments display strong depletion in Na_2O and K_2O relative to UCC and track the ideal tendency of weathering (Figure 9a) [55]. The mineralogical compositions and weathering intensities are also determined by the A–CNK–FM graph [59]. The studied river sediments follow the tendency of UCC along with granitic, granodioritic, and andesitic, designating felsic parent materials (Figure 9b). In addition, the ratios of $\text{K}_2\text{O}/\text{Rb}$ and Rb/Sr are commonly considered as proxies for weathering intensity; therefore, the ratio of $\text{Rb}/\text{Sr} > 1$ suggests greater chemical weathering and low intensities inferred by <1 [55,56,60]. All the studied sediments show both $\text{K}_2\text{O}/\text{Rb}$ and Rb/Sr less than 1, indicating low weathering intensities (Tables 1 and 2, respectively). The overall deduced observations suggest a low level of chemical weathering of the river samples.

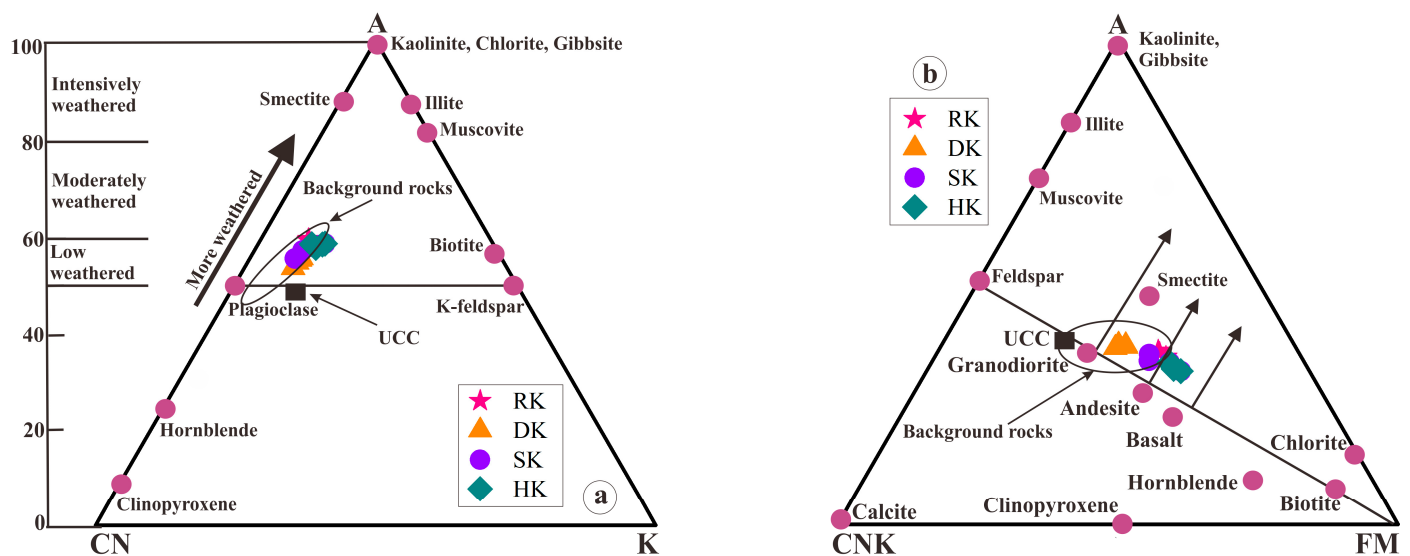


Figure 9. Ternary plots showing the paleo-weathering settings of the studied sediments: (a) the A-CN-K plot [32] and (b) the A-CN-K-FM plot [61].

5.4. Provenance

The compositions of geochemical data such as aluminum and titanium oxides are valuable markers for analyzing the provenance of sedimentary rocks owing to their relatively low mobility in fluvial processes [55,60,62]. The major oxides such as Al₂O₃ and TiO₂ rarely fractionate during diagenesis, weathering, and/or transportation processes [55,63,64]. The ratio of Al₂O₃/TiO₂ is generally applied to distinguish between mafic, intermediate, and felsic provenances of source materials. In [65], the authors documented that the ratios of Al₂O₃/TiO₂ range 3–8 for mafic, 8–20 for intermediate, and 21–70 for felsic sources. The mean ratios of Al₂O₃/TiO₂ from RK (21.25), DK (17.55), SK (18.02), and HK (13.95) (Table 1), indicate that river RK receives materials of heterogenic provenance (mafic + felsic sources) but more felsic inputs, and intermediate parent rocks for DK, SK, and HK (both felsic and mafic equal contributions) (Table 1). Moreover, the low range of chemical weathering (Figure 9a) is also suggestive of marine and Himalayan-derived detritus (unpublished; Khan et al., 2023). The high field strength elements (HFSE) such as Ti, Hf, Zr, Ta, Nb, and Y are incompatible in the mantle phase, while the contents are high in silicate melt. The immobility of these elements helps identify parent rocks. In the crust, these are enriched and representatives of excessive amounts of apatite, zircon, and sphene. The authors of [66] argued that depleted concentrations of these elements are examples of magmatic arc- and subduction-related settings. Similarly, the depletion of these elements in this study followed the argument of [43] that these sediments were linked with magmatic arc-related settings. The transition elements represent compatibility with the mantle phase and are highly concentrated in the solid phase. The higher contents of Ni and Cr represent significant contributions of ultramafic detritus. The negative Eu pattern in the chondrite normalized diagram indicates differentiation of plagioclase and/or crustal magma contamination (Figure 5c). Moreover, the samples from RK show no significant Eu anomaly indicating a mafic/calcareous derived contribution and/or sediment depositing from the nearby bedrocks. In [67] proposed a diagram of Discrimination Function 1 vs. Discrimination Function 2 based on a combination of major elements. The plot was applied to the studied river samples suggesting all the samples fall in intermediate igneous provenance while the samples from DK represent slight differences due to detrital inputs from the Lut Block of Iran (via Nihing River). This further indicates that the sediments are derived from andesitic parent rocks, whereas the acidic plutonic/volcanic rocks may also have contributed (Figure 10a). Further, a combination of trace elements, for instance, Th/Sc vs. La/Sc, is generally illustrated to distinguish the parent arc type of the sediments. The plot

was tested on the studied sediments, indicating a closer relation to the North American shale composite (NASC: $\text{Th}/\text{Sc} = 0.82$ and $\text{La}/\text{Sc} = 2.07$) type of sediments [68] (Figure 10b). In addition, ref. [69] suggested a plot based on trace elements such as Hf vs. La/Th . The studied sediments mostly fall in the field of mixed basic/felsic sources (Figure 10c).

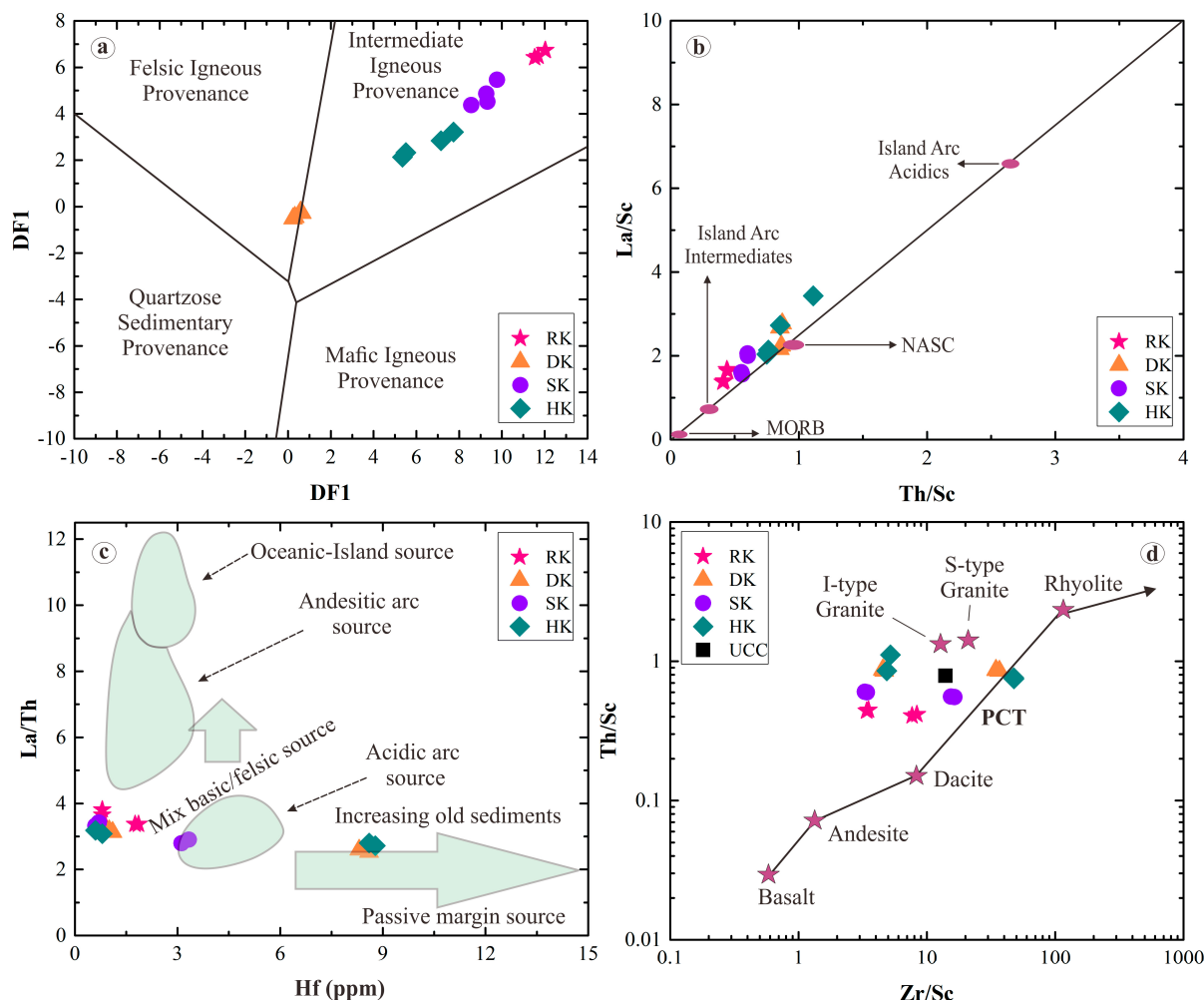


Figure 10. Cross plots illustrating the provenance types of the eastern Makran river sediments: (a) DF1 vs. DF2 [67]; (b) Th/Sc vs. La/Sc; (c) Hf vs. La/Th [69]; and (d) Zr/Sc vs. Th/Sc [68].

Moreover, the ratios of trace elements, for instance, Ti/Zr , Zr/Sc , and Th/Sc , are reliable indicators of felsic and mafic sources [7,60]. The binary plot of Zr/Sc vs. Th/Sc is used to examine the provenance type and also for determining the sorting/recycling effect [60]. The river sediments follow the PCT (primary compositional trend) but are unrelated to I-type granitic or S-type granitic provenances and indicate intermediate characters with slight variations between samples due to grain size variation/sorting input of sedimentary materials (Figure 10d). The scattered distribution of sediments is possibly the provenance from recycled materials of the Makran-Khojak Flysch Basin (Figure 1a; unpublished: Khan, 2023) and also reflects the addition of zircon and loss of Sc (Table 2). The authors of [7] argued that the Th/Sc ratios are good indicators of mafic and felsic source materials. A Th/Sc ratio less than 1 indicates Archean/mafic provenances, whereas greater than 1 suggests granites and approximately 1 for post-Archean source materials. The studied river sediments comprised the mean ratios <1 for individual rivers RK (0.43), DK (0.86), SK (0.58), and HK (0.87), representing significant mafic contributions (Table 2) similar to that of the adjoining Oligocene–Miocene composition (0.79) (unpublished: Khan et al., 2023).

Furthermore, the ferromagnesian concentration of Cr > 150 ppm and Ni > 100 ppm and the relative ratio of Cr/Ni of 1.3 to 1.5 reflects provenance from ultramafics [70]. The Cr/Ni of the river sediments imitated a sufficient input of ultramafic (except for samples from river RK) and their slight increase compared to (UCC: 0.78) is perhaps measured by phyllosilicates (Table 2). In addition, the ferromagnesian minerals in mafic/ultramafic parent rocks are generally high, which results in a decrease in Y/Ni (<0.5) and an increase in Cr/V (>8). The ratio of Cr/V in calc-alkaline, UCC, and Phanerozoic granite is relatively <1 [7,60,68]. The studied river sediments comprised mean Y/Ni of 0.66 (RK), 0.39 (DK), 0.61 (SK), and 0.56 (HK), indicating the fact that river DK receives sources from calc-alkaline volcanic rocks from the Lut Block of Iran (via Nihing river Figure 1) but the rest of river sediments are slightly enriched in Y/Ni. The mean ratios of Cr/V were 1.63 (RK), 3.47 (DK), 2.81 (SK), and 8.40 (HK), signifying higher than that of the proposed ratios; thus, the increased Cr/V ratio indicates an effective influence of mafic/ultramafic.

Ref. [60], the authors suggested that the absence of strong chemical weathering of siliciclastic rocks and the distributions of REEs and europium anomaly describe the felsic and mafic sedimentary source of the materials. The REEs patterns of mafic sediments show lower fractionation, slight or negative europium anomalies, and depleted (La/Yb)_N. On the other hand, the REEs patterns of felsic sediments describe higher fractionation, enriched (La/Yb)_N, and good Eu negative [7]. The REE normalized patterns of all river sediments have good Eu negative anomalies except sediments from river RK which comprised no effective Eu negative anomaly (Figure 4c), demonstrating sedimentary and crystalline source materials for the DK, SK, and HK rivers, and more basaltic/calcareous rock contributions for RK. The Eu pattern of RK was probably due to the absence of unaltered feldspar coming from granitic and gneisses detrital materials present within the older rocks in MAW, and calcareous inputs from the Kirthar Fold Belt and/or adjoining Jurassic limestones (AMJ) (Figures 1 and 2a–c). The mean (La/Yb)_N values of RK (5.75), DK (9.86), SK (6.87), and HK (7.64), indicate significant mafic assistance. In addition, the slight negative Ce anomaly of the RK river indicates arc magmas of the parent rocks [71,72] i.e., Bela Ophiolitic region (Figure 1), and this Ce depletion also represents derivatives of carbonates. The DK watershed comprises several major tributaries such as Nihing, Kech, and Rakhshan. The Kech and the Rakhshan run through the central Makran and carry mostly the sedimentary type of rocks, while Nihing runs from Iran, enters Pakistan, and joins the Dasht river at Kaur-e-Awar. Igneous and metamorphic sediments from the Ispikan-Wakai Formation in Pakistan and the Lut Block in Iran are carried by the Nihing river.

6. Conclusions

The study concluded with the following findings:

1. The ICV values for all studied sediments indicated immature, low recycled, and low source area weathering of the parent rock.
2. The DK along with RK and SK indicated more mature, sorted sediments, and variation in tectonic and climatic environments than HK due to their heterogeneous provenances. The higher zircon content in the HK river only represented by the increasing heavy minerals trend is due to granitic detritus, whereas other river sediments show variations due to their source rocks.
3. The tectonic environments of all river sediments signify a continental arc rather than an active continental margin and humid climatic conditions.
4. The sediments of HK have a provenance of intermediate character due to a mixture of felsic and mafic with or without sedimentary/meta-sedimentary rocks and ultramafic rocks. The heterogeneous sources of the sediments include Himalayan-derived recycled sediments found in the Khojak-Makran flysch basin and the Kirthar-Sulaiman belt. Bela Ophiolitic complexes also supply detrital metamorphic and volcanic lithic to HK.
5. The detrital sediments carried by the SK watershed represent the complex tectonic condition of a continental arc with a humid climatic setting. They also show low source

area weathering conditions. The provenance indicators of SK signify an intermediate igneous character as their main source of sediments is central Makran.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/min13030348/s1>, Table S1: Major elemental individual and mean positive and negative correlations of the Eastern Makran rivers samples, Table S2: Major elemental PCA correlation matrix of RK, Table S3: Major elemental PCA correlation matrix of DK, Table S4: Major elemental PCA correlation matrix of SK, Table S5: Major elemental PCA correlation matrix of HK, Table S6: Major elemental PCA correlation matrix of mean.

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