

Geochemistry of Rare Earth Elements in Pahang River Sediment, Malaysia

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<p>Received: October 30, 2023 Peer-reviewed: December 2, 2023 Accepted: December 27, 2023</p>	<p>ABSTRACT</p> <p>Rare earth elements (REE) are a set of 17 chemically similar metallic elements including 15 lanthanides, scandium and yttrium. The current status of REE as a global strategic commodity has encouraged the identification of REE ore deposits. This research is carried out to identify the mining feasibility of fluvial sediment REE and to understand the sediment's physical and chemical characteristics and effects on the geochemical behaviour of REE in the longest river of Peninsular Malaysia namely Pahang River. Surface sediment samples were collected along Pahang River (n=44) in approximately 10 km distance intervals. The sediment samples were analyzed using XRF to determine the major oxide content. Meanwhile, REE content in the sediment samples was extracted using the Total Digestion method and analysed using ICP-MS. The results show the average value of ΣREE at surface sediments of the Pahang River is 42.58 ppm and can be considered too low to be economically mined. Each area shows higher fractionation of light REE than heavy REE with negative europium anomalies, suggesting sediments in this area were derived from felsic rocks. The concentration of REE in Pahang River surface sediments was controlled by the porosity and organic matter as showed by the correlation of ΣREE with porosity ($R^2=0.65$) and organic matter content ($R^2=0.71$). In conclusion, this research's findings are generally useful for further REE mineral exploration and fluvial sediment environmental monitoring.</p> <p>Keywords: XRD analysis; Rare Earth Elements; Pahang River, Organic Matter</p>
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Introduction

Located on the east coast of Peninsular Malaysia, the Pahang River (459 km) is the longest and river in Peninsular Malaysia with a main bedrock of granite and limestone; and sedimentary rock e.g. shale, sandstone, respectively [1], it contributes

annual fluvial sediment fluxes of 20.4 Mt discharge into the southern South China Sea [2]. Fluvial sediments as the particles or grains eroded from rocks and soils and then deposited on the river bed, carry traces of the parent earth materials [[3], [4], [5]], including rare earth elements (REEs) which are

a group of chemically similar, naturally occurring elements but present in trace amounts [6].

The current high-tech trend in the development and miniaturization of electronic products, clean energy generators, military systems, communication systems, and satellites which rely on the magnetic, luminescent, and electrochemical properties of REEs has immensely induced the increasing demand for REEs and economically making them a strategic commodity [[7], [8]]. Studying the elemental distribution via geochemistry in Pahang Rivers can help us not only to understand the systems and history of Earth, and inspect environmental conditions but more importantly to explore mineral deposits which are high anomalies of certain commercial elements or minerals over appreciable areas which are profitable to be mined [[9], [10], [11], [12], [13]].

Recent research has reported the potential of granite as an REE source [14], therefore, we expect Pahang River with a major granitic background might have the potential to develop fluvial REE mining. In this research, REEs were traced along the river sediments, as one of the geochemical mapping methods in regional and detailed scales is to trace the pathway of elemental movement and locate the source of elements or minerals. There are limited detailed studies available on the geochemistry and mineralogy of the tropical rivers from the East Coast of Peninsular Malaysia as previous studies were focused on the near- and estuary area [[6], [15]], therefore, geochemical studies focusing on REE from upper to lower courses of tropical river sediment is thus needed to decipher the broad view of the sediment provenance. This study was performed to characterize the chemical composition of Pahang River sediments in terms of major oxides and elements, minerals, and rare earth elements (REE) and evaluate the geochemical behaviour of REE in tropical Pahang Rivers.

Experimental part

Sample Collection

The surface sediments were collected at 0 to 20cm from the river bed along the Pahang River by using an Ekman grab sampler, sealed in labelled transparent polyethylene (PE) bag and stored at 4°C for further processing. The coordinates of the sampling locations were acquired by global positioning system (GPS) Garmin GPSMAP 62s and mapped by geographic information system ArcGIS 10.2, as shown in Figure 1 and Table 1.

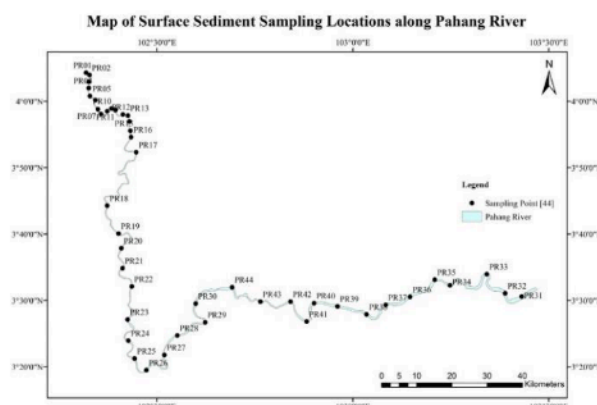


Figure 1 - Map of surface sediment sampling location along the longest river in Peninsular Malaysia namely the Pahang River

Table 1 - Coordinates of surface sediment sampling locations along the Pahang River

Sampling Locations	Latitude	Longitude
PR 01	04° 04' 19.20" N	102° 19' 08.40" E
PR 02	04° 03' 57.59" N	102° 19' 40.80" E
PR 03	04° 02' 56.40" N	102° 19' 33.59" E
PR 04	04° 01' 58.80" N	102° 19' 33.59" E
PR 05	04° 00' 46.79" N	102° 19' 44.39" E
PR 06	04° 00' 10.80" N	102° 20' 34.80" E
PR 07	03° 58' 47.99" N	102° 20' 56.40" E
PR 08	03° 58' 04.79" N	102° 21' 25.19" E
PR 09	03° 58' 30.00" N	102° 22' 22.80" E
PR 10	03° 58' 55.20" N	102° 23' 02.40" E
PR 11	03° 58' 40.80" N	102° 23' 38.40" E
PR 12	03° 58' 01.20" N	102° 24' 46.79" E
PR 13	03° 57' 50.39" N	102° 25' 33.60" E
PR 14	03° 56' 56.39" N	102° 25' 48.00" E
PR 15	03° 55' 33.60" N	102° 25' 55.20" E
PR 16	03° 54' 36.00" N	102° 26' 02.39" E
PR 17	03° 52' 19.19" N	102° 26' 49.20" E
PR 18	03° 44' 16.80" N	102° 22' 22.80" E
PR 19	03° 40' 04.80" N	102° 24' 07.20" E
PR 20	03° 37' 51.59" N	102° 24' 32.40" E
PR 21	03° 34' 51.60" N	102° 24' 43.20" E
PR 22	03° 32' 06.00" N	102° 26' 09.60" E
PR 23	03° 27' 07.19" N	102° 25' 29.99" E
PR 24	03° 23' 56.40" N	102° 25' 37.20" E
PR 25	03° 21' 14.40" N	102° 26' 34.79" E
PR 26	03° 19' 30.00" N	102° 28' 22.79" E
PR 27	03° 21' 46.80" N	102° 31' 08.40" E
PR 28	03° 24' 43.19" N	102° 33' 07.20" E
PR 29	03° 26' 41.99" N	102° 37' 22.80" E
PR 30	03° 29' 31.20" N	102° 35' 56.40" E
PR 31	03° 30' 35.99" N	103° 25' 51.59" E
PR 32	03° 31' 04.79" N	103° 23' 20.39" E
PR 33	03° 33' 57.59" N	103° 20' 31.19" E
PR 34	03° 32' 16.79" N	103° 14' 52.80" E
PR 35	03° 33' 07.20" N	103° 12' 32.40" E
PR 36	03° 30' 32.39" N	103° 08' 45.60" E
PR 37	03° 29' 20.39" N	103° 05' 02.40" E
PR 38	03° 27' 53.99" N	103° 02' 05.99" E
PR 39	03° 29' 05.99" N	102° 57' 39.59" E
PR 40	03° 29' 34.79" N	102° 54' 03.59" E
PR 41	03° 26' 49.20" N	102° 52' 55.20" E
PR 42	03° 29' 49.19" N	102° 50' 27.59" E
PR 43	03° 29' 49.19" N	102° 45' 50.39" E
PR 44	03° 31' 58.79" N	102° 41' 31.19" E

Analysis of Major Oxides and REE in the sediment samples

The sediment sample collected was weighted before and after being heated at 105°C with air ventilation of 50% in the oven until a constant weight was no longer reduced after repetitive heating. The sediment particles with mesh size below 250-µm were selected to be homogenized and pulverized to less than 45µm in a laboratory ball mill machine before sample digestion.

The samples were then analysed for major oxides analysis using Energy Dispersion XRF (EDXRF) which is outfitted with a rhodium (Rh) x-ray tube that operates at a voltage of 40 kV, and a current of 30µA for integration times ranging from 60-600 sec.

For REE analysis, about 0.2 g of the pulverized sediment sample was introduced into a 50-ml polytetrafluoroethylene (PTFE) beaker. 68% nitric acid (HNO3), 48% hydrofluoric acid (HF) and 50% hydrogen peroxide (H2O2) with a volumetric ratio of 4:3:2 with a total of 27 ml were poured carefully into the PTFE beaker and swirled to homogenize [[16], [17]]. All REE excluding Pm, were analyzed by Perkin-Elmer Elan DRC-e ICPMS at the laboratory of Universiti Kebangsaan Malaysia.

Results and Discussion

Physical Characteristic and Organic Matter in Pahang River Sediments

Pahang River sediments, as shown in Figure 2, were mainly dominated by relatively clear grains (PR01-PR05, PR08-PR11, PR13-15, PR21, PR28, PR30-PR32, PR35, PR37-39) and other greyish powdery coating grains instead of light brown of Kelantan River surface sediments due to different mineralogical composition and weathering conditions. Dry-bulk density of surface sediments along Pahang River, as shown in Figure 3 had a $\bar{x} \pm s$ of $1.44 \pm 0.45 \text{g/cm}^3$, was least dense at PR42 (0.73g/cm^3) and was densest at PR10 (2.07g/cm^3). Organic matter content (OM) of Pahang surface sediments also had a fluctuated pattern with $\bar{x} \pm s$ of $3.23 \pm 3.19\%$, highest OM at PR40 (10.05%), and lowest OM at PR28 (0.20%) as shown in Figure 3. Pahang River surface sediments OM shows a strong positive correlation with porosity ($R^2=0.87$) suggesting their OM were highly influenced by the porosity as shown in Table 2.

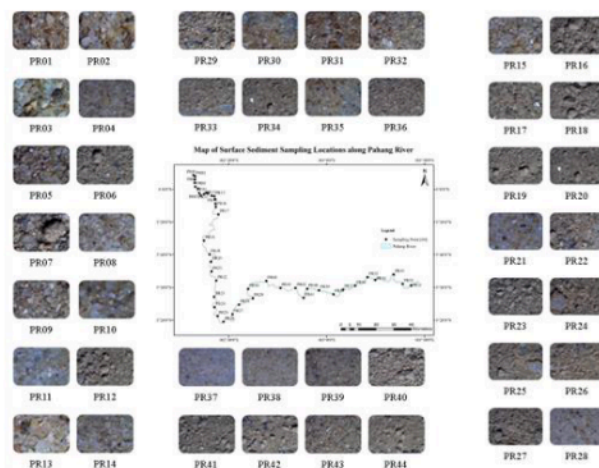


Figure 2 - Pahang River surface sediments under stereoscopic at 5x magnification

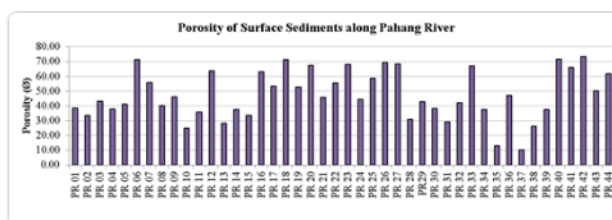


Figure 3 - Porosity of surface sediments along Pahang River

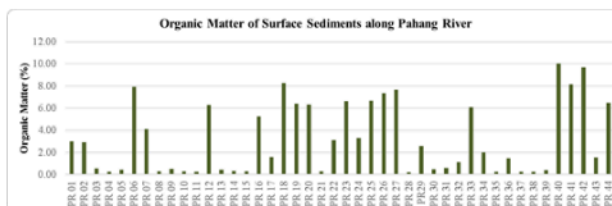


Figure 4 - Organic matter content of surface sediments along the Pahang River

Table 2 - Correlation between physical characteristics of Pahang River surface sediments

Correlation Coefficient, rPR	Organic Matter (%)	Dry-bulk density (g/cm ³)	Porosity (φ)
Organic Matter (%)	1		
Dry-bulk density (g/cm ³)	-0.875	1	
Porosity (φ)	0.875	-1	1

Major Oxide in Pahang River Sediment

The result of major oxides in sediments from the Pahang River is shown in Table 3. Maturity of Pahang river surface sediments was ranked as the following: PR08> PR24> PR30> PR10> PR21>PR02> PR28> PR13> PR11> PR32> PR15> PR03> PR14> PR29> PR04> PR09> PR01> PR34> PR31> PR05> PR12> PR36> PR35> PR17> PR25> PR37> PR07> PR38> PR16> PR41> PR18> PR33> PR39> PR20> PR19>

PR40> PR42> PR22> PR44> PR27> PR06> PR23> PR43> PR26. By applying the geochemical classification proposed by Herron (1988) [18], among 44 sampling locations along Pahang River, there are 22 subarkose, 7 wacke, 6 arkose, 3 litharenite, 3 quartz arenite and 3 shale as shown in Figure 5.

Table 3 - Major Oxides of Pahang River surface sediments

Location	Major Oxide Percentage (%wt)								
	Al ₂ O ₃	CaO	Fe ₂ O ₃	K ₂ O	MgO	MnO	Na ₂ O	SiO ₂	TiO ₂
PR 01	3.79	0.332	1.68	3.98	0.388	0.202	0.381	88.4	0.136
PR 02	3.4	0.113	0.594	2.83	0.242	0.179	0.265	91.4	0.127
PR 03	3.54	0.16	0.665	3.4	0.269	0.162		91.6	0.165
PR 04	3.94	0.101	0.684	3.98	0.259	0.168	0.102	90.7	
PR 05	4.37	0.102	0.769	4.69	0.265	0.109		89.5	0.1
PR 06	18.1	0.419	4.73	6.49	0.888	0.186		68	0.739
PR 07	9.73	0.25	2.11	5.74	0.377	0.121		81.2	0.302
PR 08	2.95	0.171	0.69	2.38	0.2	0.101		93.5	
PR 09	3.99	0.28	0.721	4.05	0.27	0.18		90.4	0.105
PR 10	3.21	0.186	0.811	2.82	0.28	0.264		92.2	0.146
PR 11	2.83		0.538	2.97	0.19	0.216		93.2	
PR 12	12.8	0.431	4.09	4.31	0.704	0.236		76.5	0.476
PR 13	3.14	0.125	0.812	2.89	0.236	0.105	0.214	92.3	0.102
PR 14	3.42	0.116	1.13	3.75	0.359	0.139		90.8	0.237
PR 15	3.19	0.162	0.594	3.35	0.176	0.212		92.3	
PR 16	14.1	0.469	3.88	5.69	0.846	0.183	0.314	73.6	0.518
PR 17	6.47	0.131	1.41	5.58	0.447	0.117	0.336	85.1	0.225
PR 18	18.2	0.373	4.98	5.83	0.921	0.191	0.12	68.2	0.751
PR 19	15.7	0.295	3.83	6.34	0.705	0.128		72	0.649
PR 20	15.3	0.436	3.89	6.33	0.765	0.17	0.137	72.1	0.612
PR 21	3.3	0.175	1.17	2.83	0.271	0.125		92.1	
PR 22	10.9	0.5	3	7.06	0.759	0.155		76.6	0.469
PR 23	17.2	0.519	5.97	6.54	0.725	0.159		67.4	0.708
PR 24	9.58	0.199	6.66	2.32	1.99	0.123		78.3	0.678
PR 25	14.4	0.277	3.56	5.18	0.643	0.148		74.9	0.538
PR 26	10.7	0.919	9.6	10.2	0.554	0.388	2.32	63.3	1.24
PR 27	16.7	0.527	4.64	6.47	0.807	0.238	0.48	68.8	0.725
PR 28	3.15	0.125	0.683	2.89	0.23	0.22		92.6	0.08
PR29	4.53	0.471	1.54	3.63	0.348	0.153	0.376	87.8	0.265
PR 30	3.1	0.14	0.64	2.78	0.2	0.14		92.9	0.11
PR 31	5.79	0.117	1.18	4.31		0.195		88.2	0.141
PR 32	4.19	0.15	0.79	3.14	0.26	0.16		91.2	0.11
PR 33	13.2	0.474	4.12	6.25	0.85	0.206	0.65	72.9	0.583
PR 34	8.45	0.126	1.54	4.05	0.399	0.104		84.8	0.334
PR 35	1.91	0.387	1.77	5.83		0.187		89.1	0.09
PR 36	7.91	0.162	1.74	5.37	0.413	0.12		83.9	0.243
PR 37	2.02	0.413	1.71	6.19	0.25	0.88		88.4	0.102
PR 38	2.11	0.427	1.61	6.31		0.214		88.4	0.125
PR 39	2.27	0.41	2.05	7.46		0.271		86.5	0.105
PR 40	19.9	0.325	5.22	5.92	0.794	0.208		66.5	0.803
PR 41	18.9	0.241	4.75	5.78	0.783	0.119	0.117	68.2	0.751
PR 42	20	0.354	5.4	6	0.815	0.249		66	0.838
PR 43	5.4	0.532	3.41	10.5		0.224		78.5	0.565
PR 44	15.8	0.253	4.01	6.69	0.692	0.143		71.4	0.63

Distribution of REEs in Pahang River Sediments

Pahang River surface sediments had an average total REE of 42.581 ppm, LREE of 34.531 ppm HREE of 8.050 ppm as shown in Figure 6, and also in Table 4. PR06 with LREE 95.789 ppm and HREE 22.888 ppm contributing to a total REE 118.677 ppm was the highest among other Pahang River surface sediments, also having cerium being the main contributor as shown in Figure 7. The enrichment factor of each REE is also far below the economic value as illustrated in Figure 8. The spatial distribution of total REE was mapped in Figure 9.

Map of Geochemical Classification of Surface Sediments along Pahang River

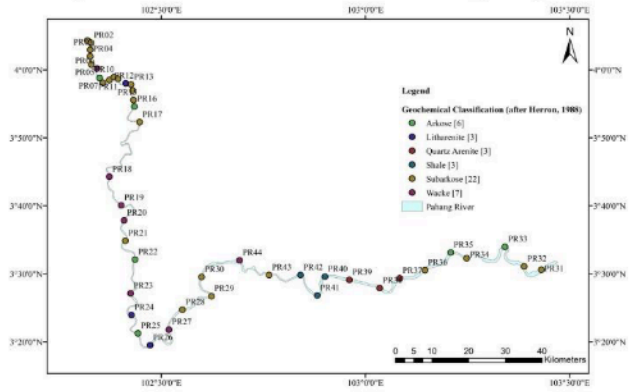


Figure 5 - Map of geochemical classification of Pahang River surface sediments

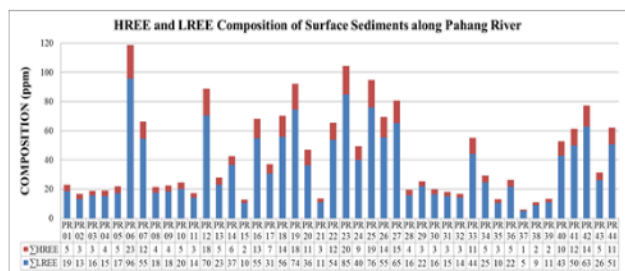


Figure 6 - Total heavy rare earth elements (HREE) and light rare earth elements (LREE) composition (without decimal places) in Pahang River sediments.

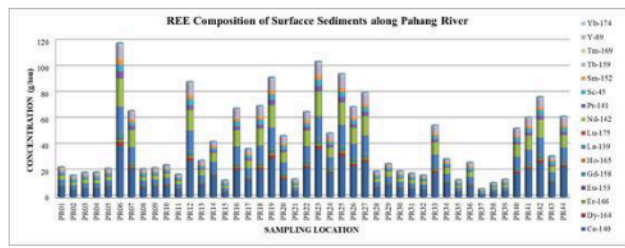


Figure 7 - REE elemental composition in Pahang River surface sediments.

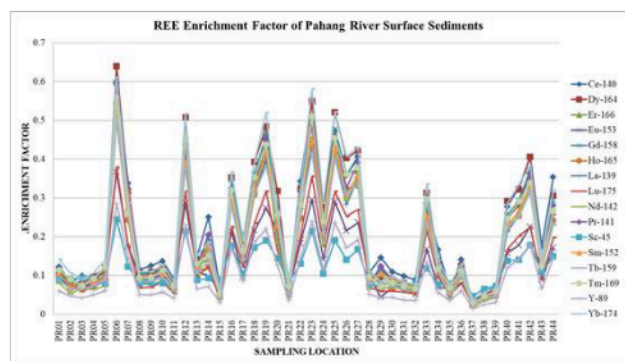


Figure 8 - REE enrichment factor in Pahang River sediments

Table 4 - Total REEs of surface sediments along Pahang River

Location	ΣLREE	ΣHREE	Total REE
PR01	18.527	4.537	23.064
PR02	13.179	3.483	16.662
PR03	15.603	3.218	18.822
PR04	15.186	3.774	18.959
PR05	17.262	4.664	21.926
PR06	95.789	22.888	118.677
PR07	54.641	11.623	66.264
PR08	17.507	4.019	21.526
PR09	18.267	4.078	22.345
PR10	20.073	4.540	24.613
PR11	14.224	3.138	17.362
PR12	70.406	18.264	88.670
PR13	22.864	5.155	28.019
PR14	36.535	6.113	42.648
PR15	10.441	2.283	12.724
PR16	54.835	13.399	68.235
PR17	30.536	6.607	37.143
PR18	55.736	14.441	70.177
PR19	74.311	17.805	92.117
PR20	36.394	10.611	47.005
PR21	10.932	2.748	13.680
PR22	53.841	11.751	65.591
PR23	84.791	19.595	104.386
PR24	39.846	9.363	49.210
PR25	75.970	18.828	94.799
PR26	55.253	14.164	69.417
PR27	65.230	15.349	80.579
PR28	15.749	3.930	19.680
PR29	21.958	3.461	25.419
PR30	16.463	3.450	19.914
PR31	15.127	2.968	18.095
PR32	14.000	2.794	16.794
PR33	44.104	11.074	55.179
PR34	24.546	4.700	29.246
PR35	10.375	2.684	13.059
PR36	21.726	4.715	26.441
PR37	4.922	1.153	6.075
PR38	8.928	1.938	10.866
PR39	11.005	2.394	13.399
PR40	42.858	9.908	52.767
PR41	49.740	11.509	61.249
PR42	62.832	14.393	77.226
PR43	26.182	5.285	31.467
PR44	50.676	11.393	62.069

Map of Total Rare Earth Elements Distribution in Surface Sediments along Pahang River

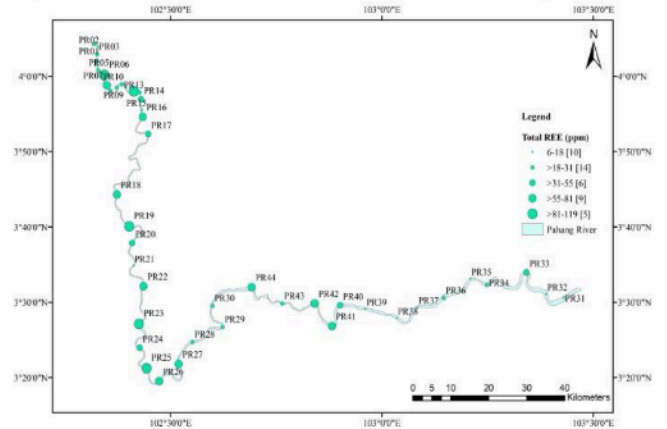


Figure 9 - Maps of total REE in Pahang River sediments.

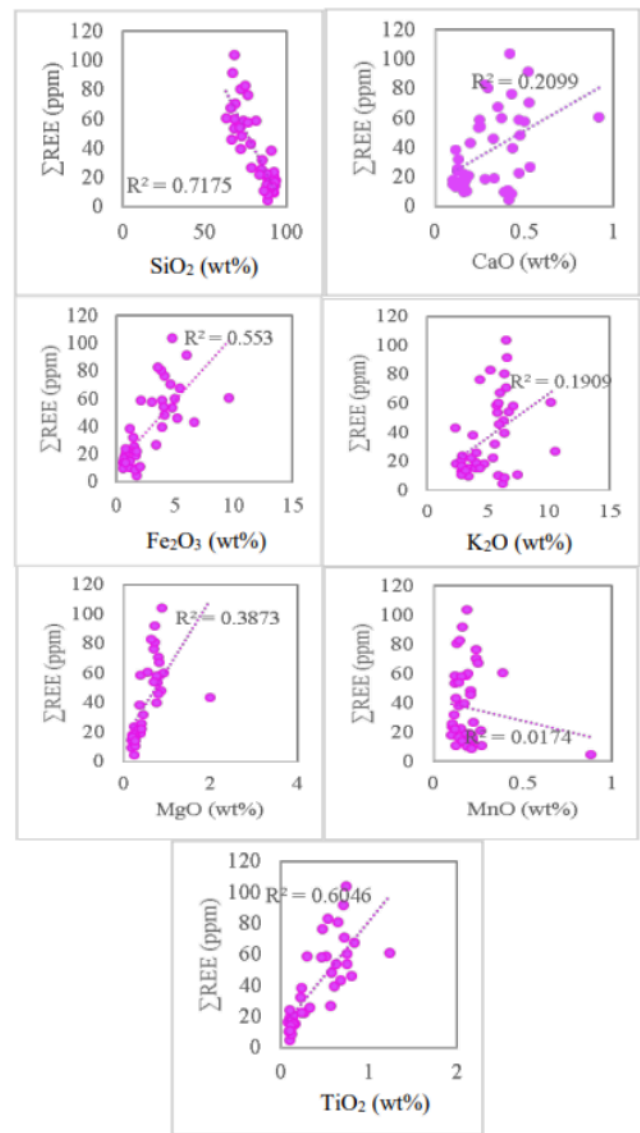


Figure 10 - Relationship of Pahang River sediment total REE and major oxides

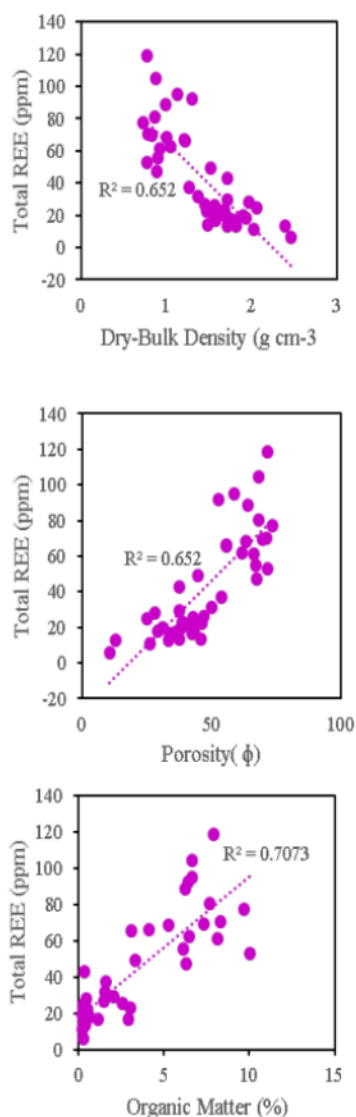


Figure 11 - Correlation of Pahang River sediment total REE and physical characteristics.

Geochemical behaviour of REE in Pahang River sediment

The correlation of total REE in Pahang River sediments was made with oxides as shown in Figure 10. The strongest and weakest regression of surface sediment Σ REE was with Al_2O_3 ($R^2=0.749$) and Na_2O ($r=0.0039$) respectively. Aluminium minerals in Pahang River surface sediments were thus inferred to have a profound effect on the occurrence of REE. Total REE in Pahang River surface sediment had a

moderate positive correlation with Al_2O_3 ($r=0.867$), Fe_2O_3 ($r=0.744$), and TiO_2 ($r=0.778$), showing REE concentrated in aluminium, iron, and titanium minerals; a moderate negative correlation with SiO_2 ($r=-0.848$) showing dilution effect of silicates in REE concentrations. REE in titanite has been proposed to interpret the growth of titanite [19]. Other than chemical influence, total REE also shows some correlations with physical characteristics: dry-bulk density and porosity ($R^2=0.652$) and organic matter ($R^2=0.7073$) as shown in Figure 11. As bulk density and porosity of sediments are mainly controlled by grain size, shape, packing, and distribution [20], it indicates grain size effect may be a factor controlling REE distribution in Pahang River sediments.

Conclusions

Surface sediments collected along Pahang River had a total REE of 42.581 ppm, LREE of 34.53 ppm and HREE of 8.050 ppm. The rivers' sediments showed too low REE content and thus are not economically feasible to mine. The chondrite normalized REE showed a higher fraction of LREE to HREE in this area. Our results show the geochemical behaviour of REE in fluvial sediments was controlled by porosity and organic matter content. In addition, aluminium minerals in the Pahang River have a profound effect on REE occurrence.

Conflicts of interest. On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Малайзиядағы Паханг өзенінің шөгіндісіндегі сирек жер Элементтерінің геохимиясы

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<p>Мақала келді: 30 қазан 2023 Сараптамадан өтті: 2 желтоқсан 2023 Қабылданды: 27 желтоқсан 2023</p>	<p>ТҮЙІНДЕМЕ</p> <p>Сирек жер элементтері (СЖЭ) – 15 лантаноидтарды, скандий және иттрийді қоса алғанда, 17 химиялық ұқсас металдық элементтердің жиынтығы. СЖЭ-ның жаһандық стратегиялық тауар ретіндегі ағымдағы мәртебесі СЖЭ кен орындарын анықтауды ынталандырады. Бұл зерттеу Малайзия түбегінің ең ұзын өзенінде, атап айтқанда Паханг өзенінде шөгінді СЖЭ өндірудің орындылығын анықтау және шөгінділердің физикалық және химиялық сипаттамаларының Малайзия түбегінің ең ұзын өзеніндегі СЖЭ геохимиялық әрекетіне әсерін түсіну үшін жүргізіледі. Жер үсті шөгінділерінің үлгілері Паханг өзенінің бойынан (n=44) шамамен 10 км қашықтықта жиналды. Шөгінді үлгілері негізгі оксид құрамын анықтау үшін РФТ көмегімен талданды. Сонымен қатар, шөгінді үлгілеріндегі ΣREE мәндері Total Digestion әдісімен алынған және ICP-MS көмегімен талданған. Нәтижелер Паханг өзенінің беткі шөгінділеріндегі ΣREE орташа мәндері миллионда 42,58 бөлігі ғана болатынын көрсетеді және оны экономикалық тұрғыдан өндіру үшін тым төмен. Әрбір аймақ теріс еуропий аномалиялары бар ауыр СЖЭ-ге қарағанда жеңіл СЖЭ фракциясының жоғарырақ екендігін көрсетеді, бұл осы аймақтағы шөгінділер қышқыл жыныстардан алынады. Паханг өзенінің беткі шөгінділеріндегі СЖЭ концентрациясы кеуектілікпен және органикалық заттармен анықталды, бұл ΣREE кеуектілігімен (R2=0,65) және органикалық заттардың мөлшерімен (R2=0,71) корреляция арқылы көрсетілген. Қорыта келгенде бұл зерттеудің нәтижелері СЖЭ минералдарын одан әрі барлау және өзен шөгінділеріне экологиялық мониторинг жасау үшін пайдалы.</p>
	<p>Түйін сөздер: рентген құрылымдық талдау; сирек жер элементтері; Паханг өзені, органикалық заттар</p>
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Геохимия редкоземельных элементов в отложениях реки Паханг, Малайзия

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<p>Поступила: 30 октября 2023 Рецензирование: 2 декабря 2023 Принята в печать: 27 декабря 2023</p>	<p>АННОТАЦИЯ Редкоземельные элементы (РЗЭ) представляют собой набор из 17 химически схожих металлических элементов, в том числе 15 лантаноидов, скандия и иттрия. Текущий статус РЗЭ как глобального стратегического товара способствовал выявлению месторождений РЗЭ. Это исследование проводится для определения возможности добычи речных отложений РЗЭ и понимания влияния физических и химических характеристик отложений на геохимическое поведение РЗЭ в самой длинной реке полуострова Малайзия, а именно в реке Паханг. Пробы поверхностных отложений были собраны вдоль реки Паханг (n=44) на расстоянии примерно 10 км. Образцы отложений были проанализированы с помощью РФА для определения содержания основного оксида. Тем временем содержание РЗЭ в пробах отложений было экстрагировано методом полного расщепления и проанализировано с помощью ICP-MS. Результаты показывают, что средние значения ΣREE в поверхностных отложениях реки Паханг составляют 42,58 частей на миллион, и их можно считать слишком низкими для экономичной добычи. На каждом участке наблюдается более высокое фракционирование легких РЗЭ, чем тяжелых РЗЭ, с отрицательными аномалиями европия, что позволяет предположить, что отложения на этом участке произошли из кислых пород. Концентрация РЗЭ в поверхностных отложениях реки Паханг контролировалась пористостью и органическим веществом, о чем свидетельствует корреляция РЗЭ с пористостью ($R^2=0,65$) и содержанием органического вещества ($R^2=0,71$). В заключение можно сказать, что результаты этого исследования в целом полезны для дальнейшей разведки полезных ископаемых РЗЭ и экологического мониторинга речных отложений.</p>
	<p>Ключевые слова: рентгеноструктурный анализ; Редкоземельные элементы; Река Паханг, органическое вещество</p>
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