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Geochemistry of the Devonian and Permo-Triassic Black Shales in Peninsular Malaysia: Insights into Provenance, Tectonic Setting, and Source Rock Weathering

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Abstract: Selected Malaysian black shale sequences of the Permo-Triassic and Devonian ages that crop out in the Central Belt were investigated to discuss their chemical composition, provenance, tectonic setting, and weathering history. X-ray fluorescence (XRF) analysis shows that the BRSZ Unit 1 black shale has elevated SiO_2, TiO_2, and K_2O contents compared to the Semantan and Gua Musang black shale. In terms of trace elements, the BRSZ Unit 1 black shale has elevated trace element contents compared to the Semantan and Gua Musang black shales. The BRSZ Unit 1 has the highest V, U, Pb, and Mo contents relative to the other two formations. The Mo content is significant in the BRSZ Unit 1 and is thought to be associated with the elevated total organic carbon (TOC) in the BRSZ Unit 1 black shale. Compared to Post-Archean Australian Shale (PAAS), the BRSZ Unit 1 and Gua Musang black shales are low in Fe_2O_3, MnO, CaO, Na_2O, and P_2O_5. The Semantan black shales are deficient in CaO, K_2O, and P_2O_5 and enriched in MnO. The black shales of BRSZ Unit 1 are enriched in V, Cu, Ga, Rb, Mo, Sn, Pb, and U. Except for Pb (mean: 32.3 ppm), the Gua Musang black shales are largely depleted in trace elements. Similar to the Semantan black shales, all trace element concentrations are largely depleted with the exception of Sc (mean: 22.3 ppm), which is slightly higher. Provenance analysis shows that the BRSZ Unit 1, Semantan, and Gua Musang black shales derived from felsic and intermediate parental source rocks. The BRSZ Unit 1 and Gua Musang black shales indicate a collision setting, whereas the Semantan black shales show affinity to a continental arc setting. The CIA values for all the samples analyzed in this study range between 79.4 and 95.8, indicating an intense chemical weathering in warm and wet paleoclimatic conditions. The average CIW values of samples from the BRSZ Unit 1, Semantan, and Gua Musang are 99.4, 94.5, and 98.6, respectively, implying an intense degree of weathering of the source rocks.

Keywords: black shales; provenance; tectonic setting; paleoweathering; paleoclimate

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1. Introduction

The Permo-Triassic and Devonian formations crop out in the central belt of Peninsular Malaysia. These formations are made up of turbidite sequences which are characterized by interbedded sandstone and black shales with some inclusions of volcaniclastic rocks such as tuffs in the Semantan formation. The lithostratigraphic correlation and distribution of Paleozoic and Mesozoic formations in Malaysia is still under investigation and the latest correlations of various formation across the Western and Central Belts are not conclusive and more research is needed to refine the current lithostratigraphy. The Gua Musang, Semantan, and BRSZ Unit 1 were selected because of their proximity to the Bentong-Raub Suture Zone (BRSZ) and the co-existence of clastic and volcaniclastic facies in the Central Belt. According to other authors [1,2], the BRSZ of the Malay Peninsula is the location of

closure for the Paleo-Tethys Ocean basin that serves as the boundary between the Sibumasu Terrane in the west and the Sukhothai Arc in the east. Oceanic radiolarian cherts from the Devonian to the Late Permian are found in the suture, which can be up to 20 km wide [3]. The creation of the Bentong-Raub Suture Zone occurred in the Late Triassic [4]. The description of the selected formations is given below.

BRSZ Unit 1: The BRSZ Unit 1 belongs to the Bentong Group. Reference [5] mapped these rocks in the northwest of Pahang. Reference [6] identified the succession of clastic lithologies of flysch character. In general, the Bentong Group consists of conglomerate at the top, chert in the lower half, and flysch sequences in the upper half. Tuffaceous sediments and fragments of rhyolite were present. The flysch sequences are part of the arenaceous series which contains protoquartzite and greywackes. Lower Devonian graptolites were found in this group. In this study, the term BRSZ Unit 1 was proposed for the flysch-like sequences found along the Bentong-Raub road. Mapping of this unit showed that it consists of interbedded carbonaceous black shale, sandstone, and phyllite and underlain by layers of carbonaceous shales.

Semantan Formation: This formation is the largest cropping out in the Central Belt, which [7] gave the name. It contains a substantial outcrop of rocks of the Middle Triassic age. It is made up of Triassic-aged rhyolitic tuff interspersed with carbonaceous shales. Felsic and intermediate igneous rocks were introduced into this formation. Bivalve species found in shallow water indicate a neritic habitat [8]. Cross or parallel laminations can be seen in tuff beds of the Semantan Formation. Fine tuffs and mud pellet strata coexist with laminated mudstone beds. Reference [9] also reported finding beds of pebbly mudstone. Reference [7] discovered a few species of bivalves too.

Trace fossils such as *Myophoria*, or the shallow-water fauna, and the pelagic fauna, which is represented by the presence of *Daonela, Halobia,* and *Posidonia*, were found in two different fossil assemblages. The Ladinian age of the pelagic sediments is suggested by the presence of *Daonela indica* and *Daonela Pahangensis* [10]. The presence of *Costatoria malayensis, Costatoria chegarperahensis,* and *Neoschizodus ovutus* in the Myphoria fauna indicates an Anisian to Early Ladinian age. The lower part of the Semantan is made up of limestone containing brachiopod fossils, Karavankina sp., indicating a Middle Permian age.

Gua Musang Formation: The Middle Permian to Triassic carbonate facies and the Triassic argillaceous facies make up the Gua Musang Formation. In and around the town of Kuala Lipis in the Central Belt, black shale units of the Triassic age that are a part of the Gua Musang Formation have been reported [11]. The host strata of several gold occurrences that are hosted by sediment and are situated in the eastern portion of the Bentong-Raub Suture Zone also contain these black shales [12,13]. Additionally, [11] reported finding some bivalves, including Costatoria sp., and plant fragments in shale sequences from the Gua Musang Formation. The research in [11] suggested that this formation was deposited in a deep-water marine environment.

Previous studies have been carried out to characterize the provenance, tectonic setting, and paleoclimate in the Western Belt and Sarawak basin in Malaysia [14,15], but there is poor knowledge about the provenance, tectonic setting, and paleoweathering of black shale sequences that crop out in the Central Belt of Peninsular Malaysia.

Clastic rocks are important sources of information that can help identify the type of parental rocks (mafic, felsic, intermediate, and quartzite). The composition of clastic rocks has been used to infer provenance, tectonic setting, and intensity of weathering during their times of formation [16–20].

The purpose of this study is to determine the major and trace element composition of black shales to discuss the provenance, tectonic setting, and intensity of weathering for the BRSZ Unit 1, Semantan, and Gua Musang black shales.

2. Geology and Tectonic Setting

Three north–south trending tectonic belts make up the Malaysian geology (Figure 1). Following [21], reference [22] referred to these belts as the Western Belt, Central Belt, and

Eastern Belt. Figure 1 depicts the spread of the Semantan Formation, Karak Formation, Bentong-Raub Suture Zone, and the accompanying granitoids from the Ilmenite-series to the Magnetite-series. The Langkawi Islands and other formations such as the Kubang Pasu Formation are included in the NW domain, which is a part of the Sibumasu Terrane which is comprised of Sino-Siam, Burma, Malaya, and Sumatra [1]. The term Sino means China, Siam is Thailand, Malaya is Malaysia, and Sumatra is Indonesia.

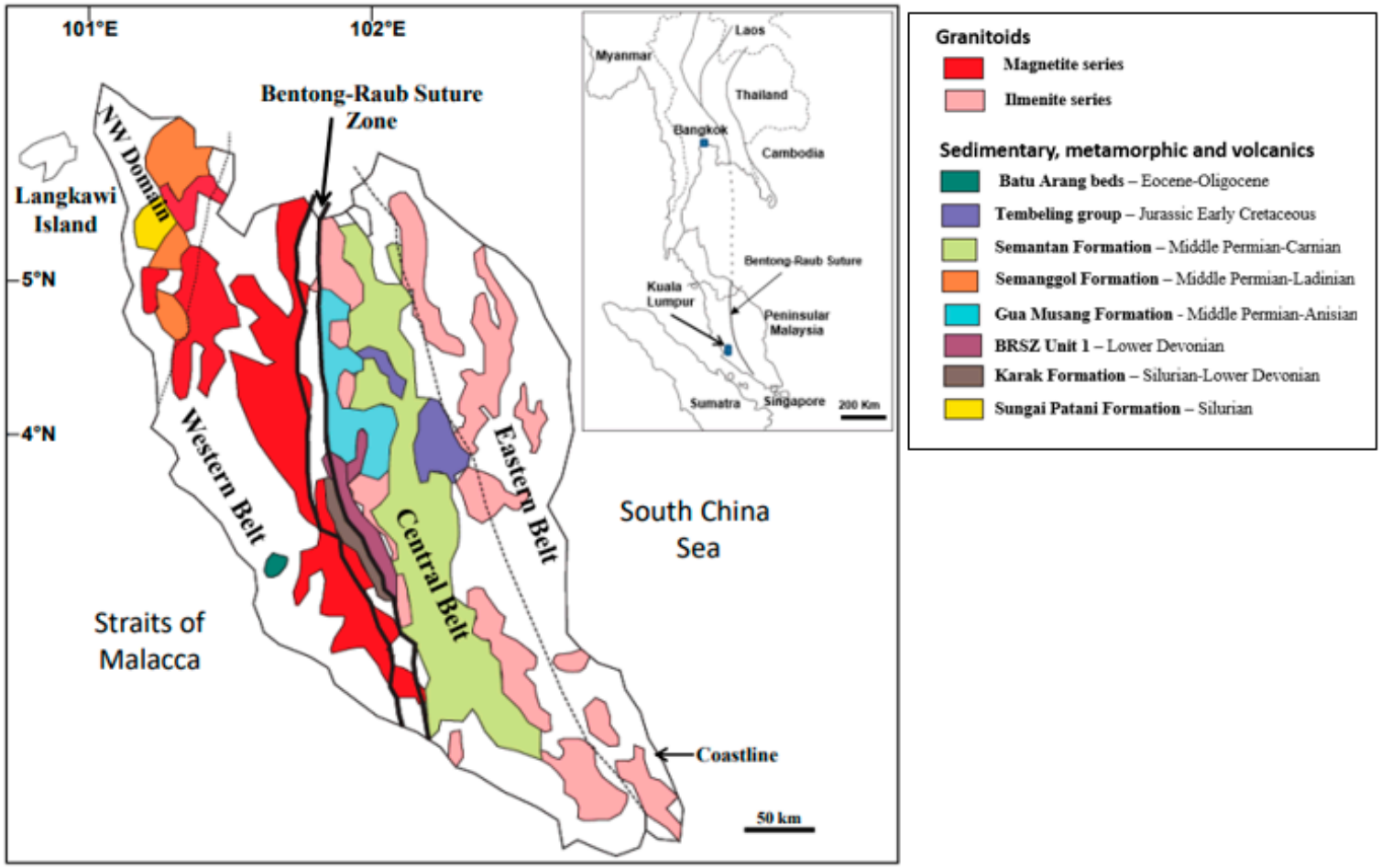

Figure 1. Geological map showing some Paleozoic and Mesozoic formations (modified from [13]). The dotted lines represent boundaries between the tectonic belts. The black solid lines are the extension of the Bentong-Raub Suture Zone. The arrow indicates the Bentong-Raub Suture Zone.

The stratigraphic positions and distribution of the formations from Silurian to Triassic are shown in Figure 2.

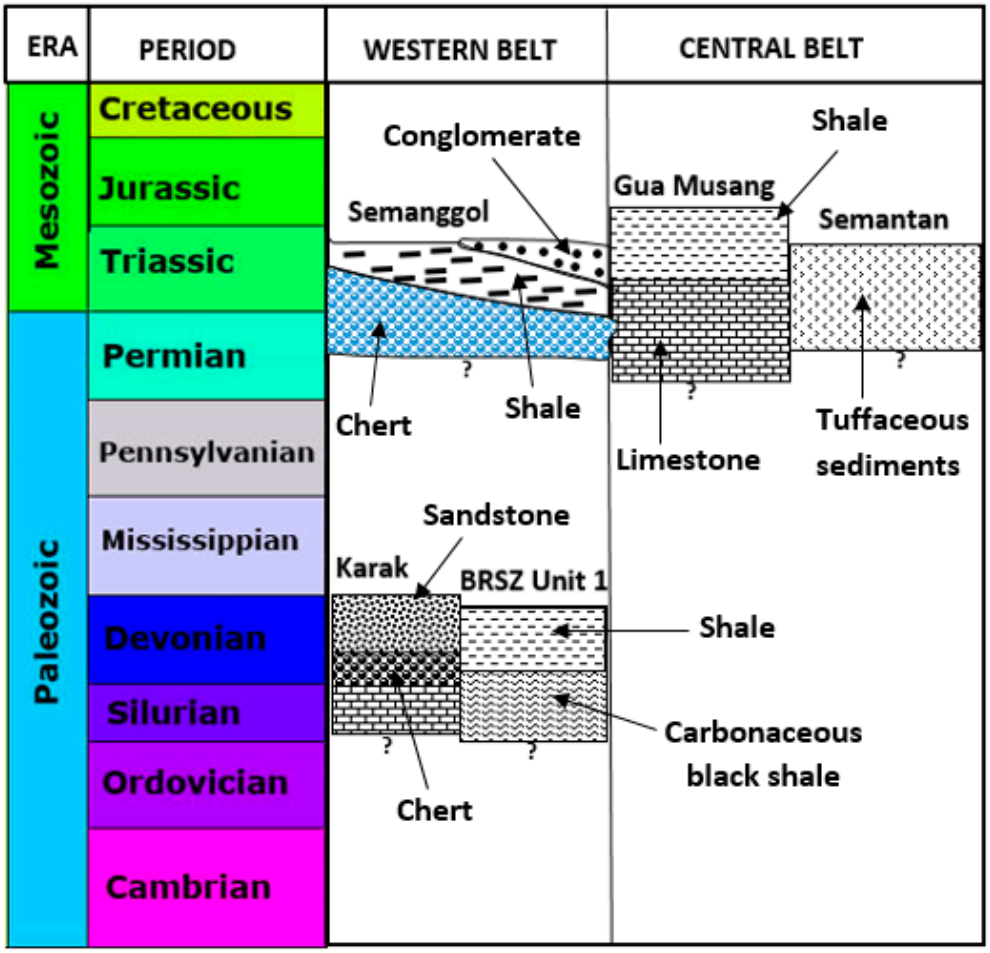

Figure 2. Simplified stratigraphic relationship and distribution of selected Paleozoic and Mesozoic formations in the Western and Central Belts of Peninsular Malaysia (Modified from [23,24]). The question mark represents the unknown underlying geological units.

2.1. The Western Belt

Triassic carbonates, Late Paleozoic platform carbonates, Early Paleozoic continental margin sequences, deep water shale sequences containing Lower Devonian graptolites, and Jurassic–Cretaceous continental deposits are all found in this belt, which is part of the Sibumasu Terrane [25,26]. The countries Siam (Thailand), Burma (Myanmar), Malaya (Malaysia), and Sumatra (Indonesia) form the block called Sibumasu, which was a part of the supercontinent called Gondwanaland.

In the Late Early Permian, the Western Belt developed from the North-West Australian Gondwana boundary [1]. The tin-bearing Main Range Granitoid Province, a sizable plutonic belt that stretches from Malaysia to the southern Peninsula of Thailand and western Thailand, is likewise connected to the Western Belt.

2.2. The Central Belt

East of the Bentong-Raub Suture Zone is where the Central Belt lies. It consists of deep to shallow marine sedimentary rocks and Permo-Triassic metamorphic rocks [11,27]. The Permo-Triassic metamorphic rocks are slate and phyllite.

Limestone was also deposited with intermediate to felsic volcanic and volcaniclastic rocks [5,28–30]. The plutonic rocks in the Central Belt are very similar to those in the Eastern Belt [31]. These are granites of the mid-Triassic age in this area [32,33], the Middle

Triassic ages of granites in this area. The Gua Musang and Semantan formations crop out in this belt.

2.3. The Bentong-Raub Suture Zone (BRSZ)

An important feature in Southeast Asia that crosses Peninsular Malaysia is the Bentong-Raub Suture Zone (Figure 3). According to [34], the suture may have a genetic connection to the orogenic gold deposits in the district. The Malay Peninsula underwent orogenic compression during the Triassic, according to [35]. The Paleo-Tethys Ocean was created or opened in the Devonian [36] when East Malaya and Indochina split off from Gondwana-land [4]. This ocean eventually closed, creating the BRSZ. Some authors [35,37,38] contend that this Triassic collision was the catalyst for the deformation of the Pre-Mesozoic strata and the intrusion of the Mesozoic S-type granitic plutons.

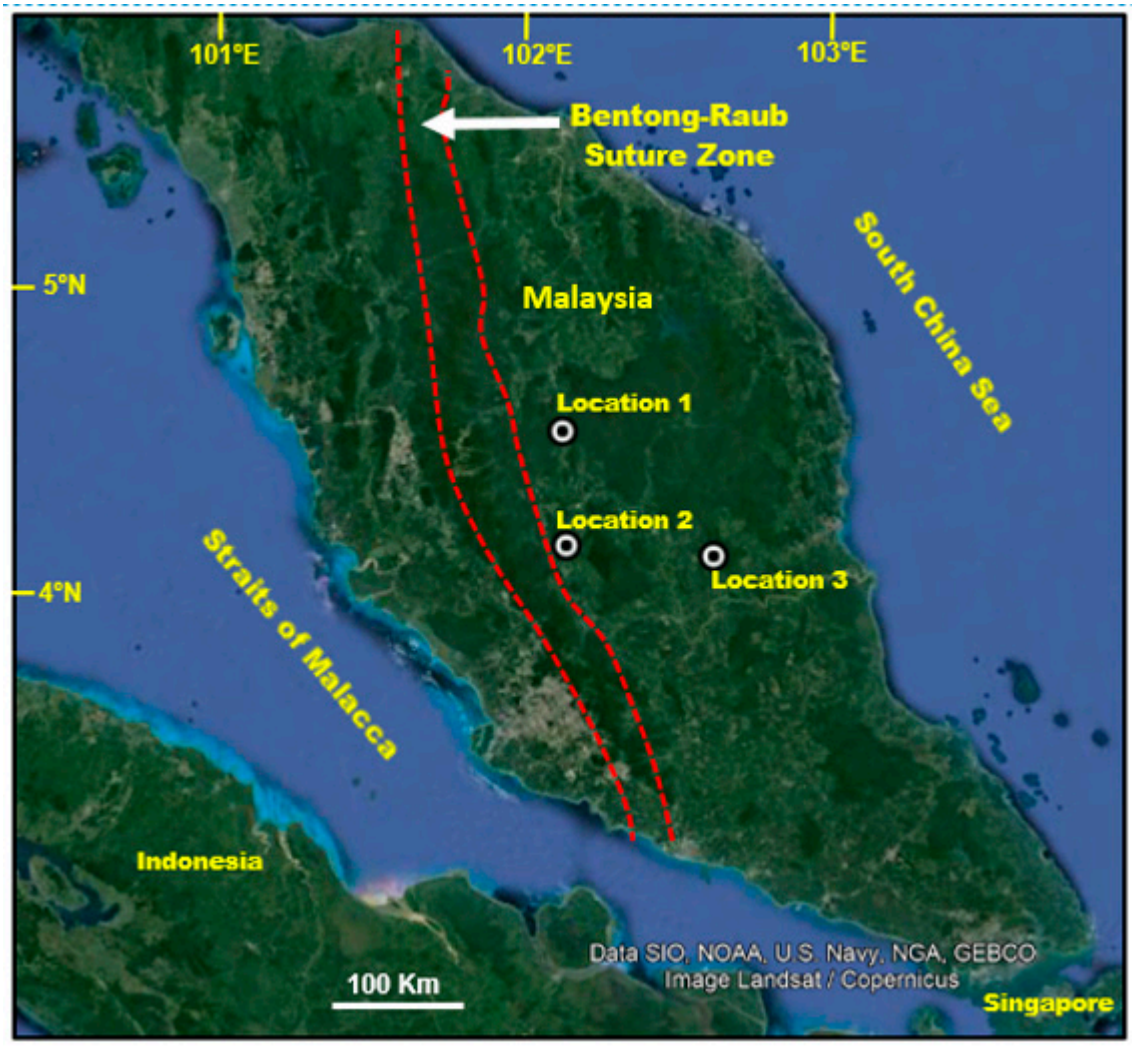

Figure 3. Google Earth map showing the sample locations and the sites where the sedimentary logs were constructed. Sample location 1: Gua Musang; Sample location 2: BRSZ Unit 1; Sample location 3: Semantan. Easting and Northing coordinates of these locations are documented in Table S1 (See the Supplementary Materials). The red lines represent the north-south and east-west extension of the Bentong-Raub Suture Zone.

2.4. The Eastern Belt

This belt, which is a portion of the East Malaya Fold belt, is made up of deformed Late Paleozoic sequences that are irregularly overlain by continental deposits from the Jurassic-Cretaceous period and the Late Permian continental conglomerate. Granitoids

have variable compositions, ranging from diorite-gabbro to hornblende-biotite granite and granodiorite [39].

Samples that were collected with their coordinates are documented in Table S1. Outcrop photographs of selected formations are shown in Figure 4. Lithological logs of the BRSZ Unit 1, Semantan, and Gua Musang formations are presented in Figure 5.

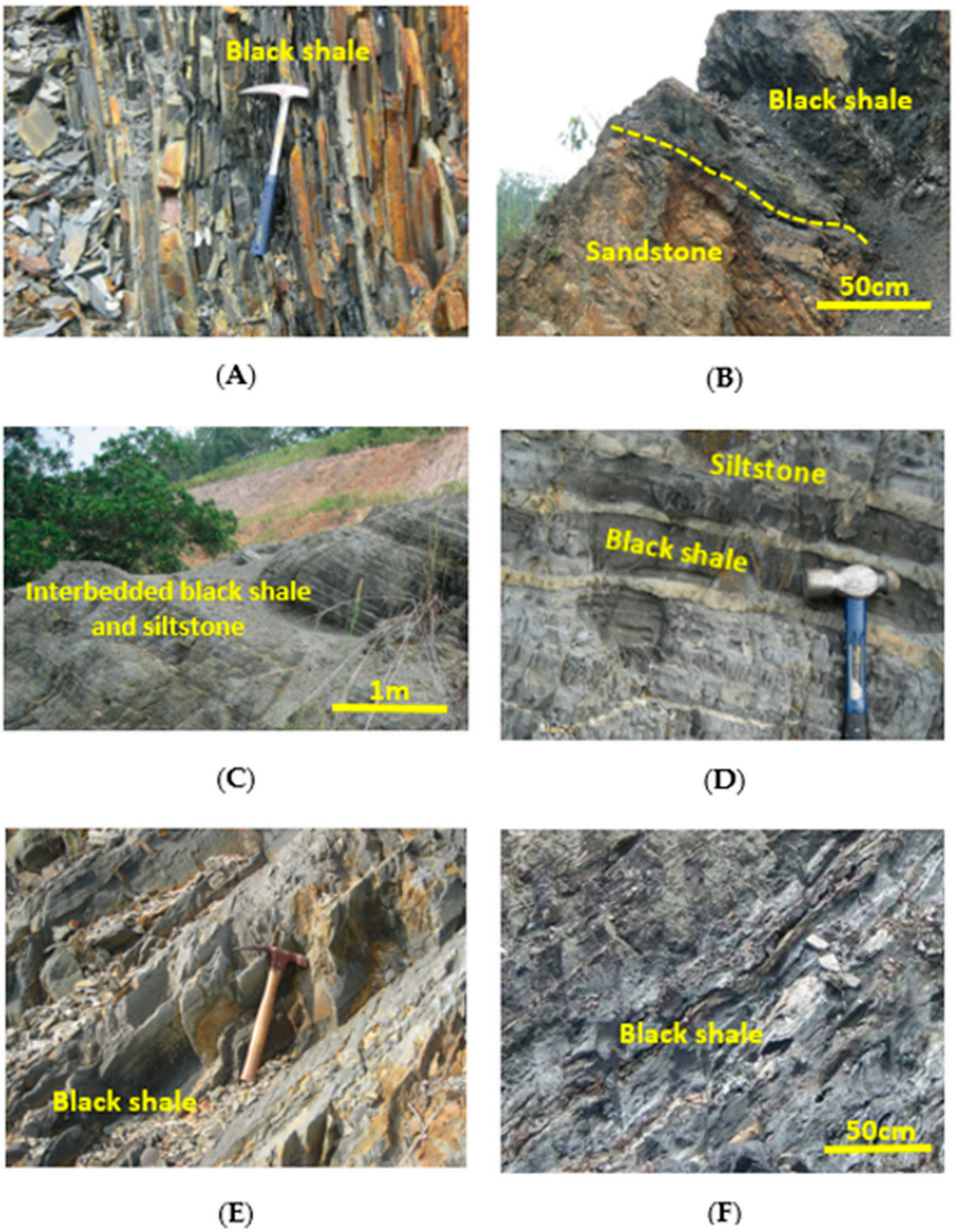

Figure 4. Outcrop photographs of selected formations in Peninsular Malaysia. (**A**) Bedded black shale exposure of the BRSZ Unit 1, close to the town of Bentong, Pahang, Malaysia. (**B**) Geological contact between the sandstone beds overlain by black shales. (**C**) Interbedded black shale and siltstone in the Semantan Formation. (**D**) A close-up of the black shale and siltstone in the Semantan Formation. (**E**) Black shale exposure represents the Gua Musang Formation in the vicinity of Kuala Lips Town, State of Pahang. (**F**) Exposure of black shale close to the town of Gua Musang, State of Pahang.

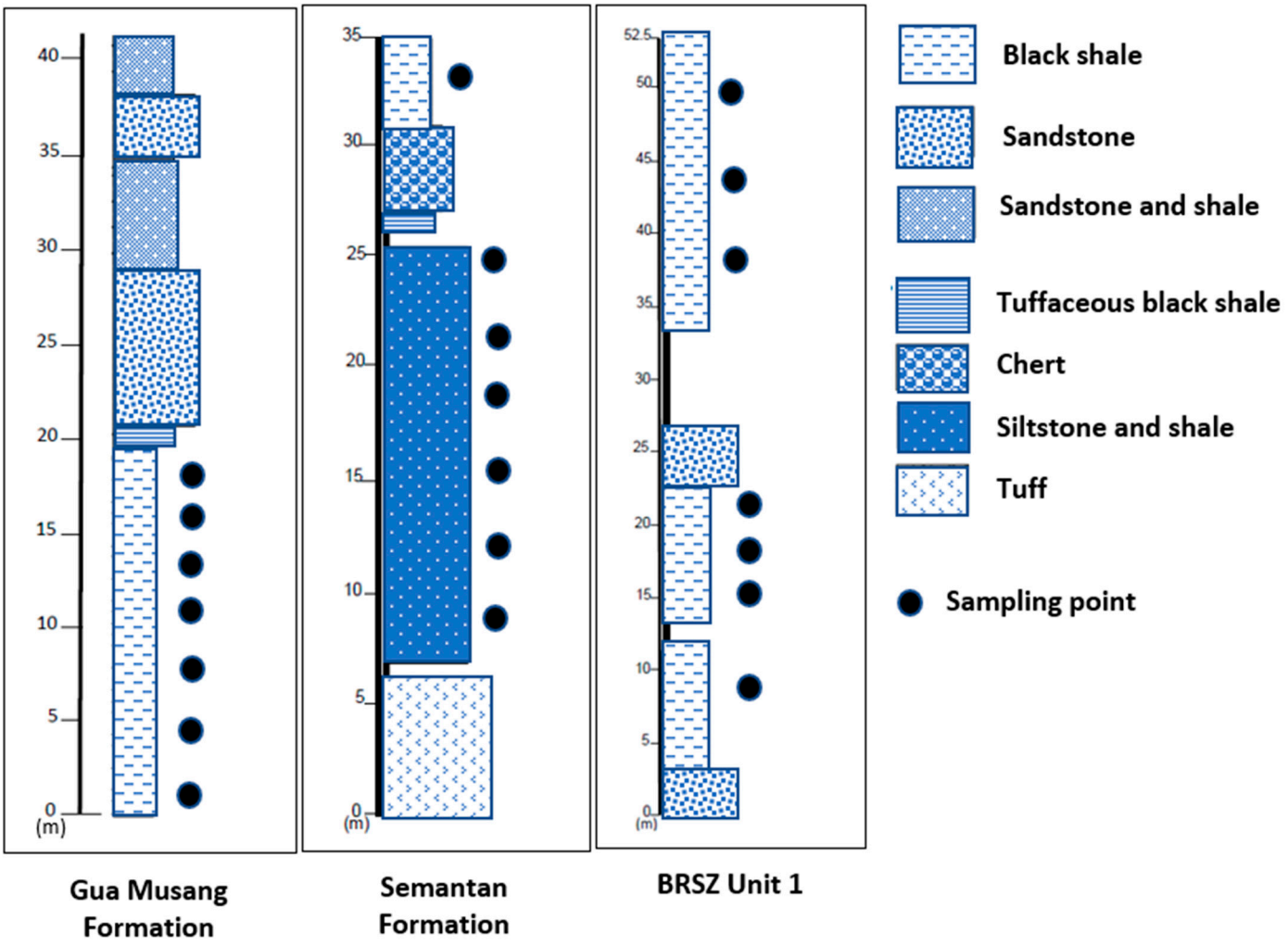

Figure 5. Lithological logs of Gua Musang, Semantan, and BRSZ Unit 1 and sampling positions of the specimen analyzed in this study. X, Y, and Z coordinates are given in Table S1. Vertical scale in meters. Gua Musang Formation is made up of interbedded sandstone and shale. The mapped sequence of the Semantan Formation is composed of tuff at the base overlain by thinly interbedded siltstones and black shales with some chert intercalated in the sequence. BRSZ Unit 1 is basically a flysch-type sequence representing layers of sandstone interbedded with shales.

3. Materials and Methods

Twenty-one black shale samples were analyzed by X-ray fluorescence (XRF) analysis to obtain the whole rock major and trace element composition. Seven black shale samples were analyzed from each unit or formation.

The instrument used for this analysis was PANalytical Axios Advanced X-Ray Spectrometer (Axios^{max}, PANalytical, Almelo, The Netherlands). It is equipped with X-Ray Tubes of 4 KW maximum and an Rh anode end window. The elements that are routinely analyzed are: F, Na, Mg, Al, Si, P, S, K, Ca, Ti, Mn, Fe and trace elements Sc, V, Cr, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Nb, Mo, Ag, Cd, Sn, Sb, Te, I, Ba, La, Ce, Nd, W, Tl, Pb, Bi, Th, and U. The instrument also has the following parts: Crystals: PX-10, LiF 220, PX-1 (for F, Na and Mg), curved PE002, and curved Ge111. The collimators are coarse (0.7 mm), fine (0.3 mm) with high resolution (0.15 mm). The detectors are gas flow proportional counters with P10 gas (10% methane argon), a sealed Xe Duplex, and Scintillation Counter. The sample changer is PANalytical X-Y sample changer with a capacity for 96 fusion discs and 64 pills.

In sample preparation for the major elements, 32mm fusion discs were prepared at 1100 degrees °C in 5%Au/95%Pt crucibles 0.500 g sample, 4.500 g 12–22 Flux (Lithium Tetraborate-Metaborate mix), 0.0606 g $LiNO_3$ for silicates. Platinum/5% gold molds were used for cooling. Sulfide-bearing samples had a different mix with more $LiNO_3$ as an oxidizing agent and the mix was pre-ignited at 700 degrees °C for 10 min. Ore samples and ironstones used 12/22 flux and a higher flux/sample ratio. Dolomites and limestones needed pure lithium tetraborate as a flux. Iodine vapor was used as a releasing agent to remove discs from the mold.

For the trace elements, 32 mm diameter pressed powder pills (10 g, 3.5 tons/cm^{-2}) were used and a sample Binder PVP-MC. Corrections for mass absorption were calculated using PANalytical Super-Q software (version 5, PANalytical, Almelo, The Netherlands) with its Classic calibration model and alpha coefficients. In-house inter-element corrections were also applied. Calibration was done on pure element oxide mixed in pure silica, along with International and Tasmanian reference rocks.

4. Results

4.1. Major Elements

Major element composition results are presented in Table S2 (See the Supplementary Materials). The major element composition in the BRSZ Unit 1 and Gua Musang show a similar trend as opposed to the Semantan Formation that has elevated Fe_2O_3, MgO, CaO, and Na_2O with low K_2O (Figure 6). This figure also shows that the Semantan has significant MnO content compared to the Semantan and Gua Musang black shales. There is no significant variation in composition for SiO_2, TiO_2, and Al_2O_3 for the three formations.

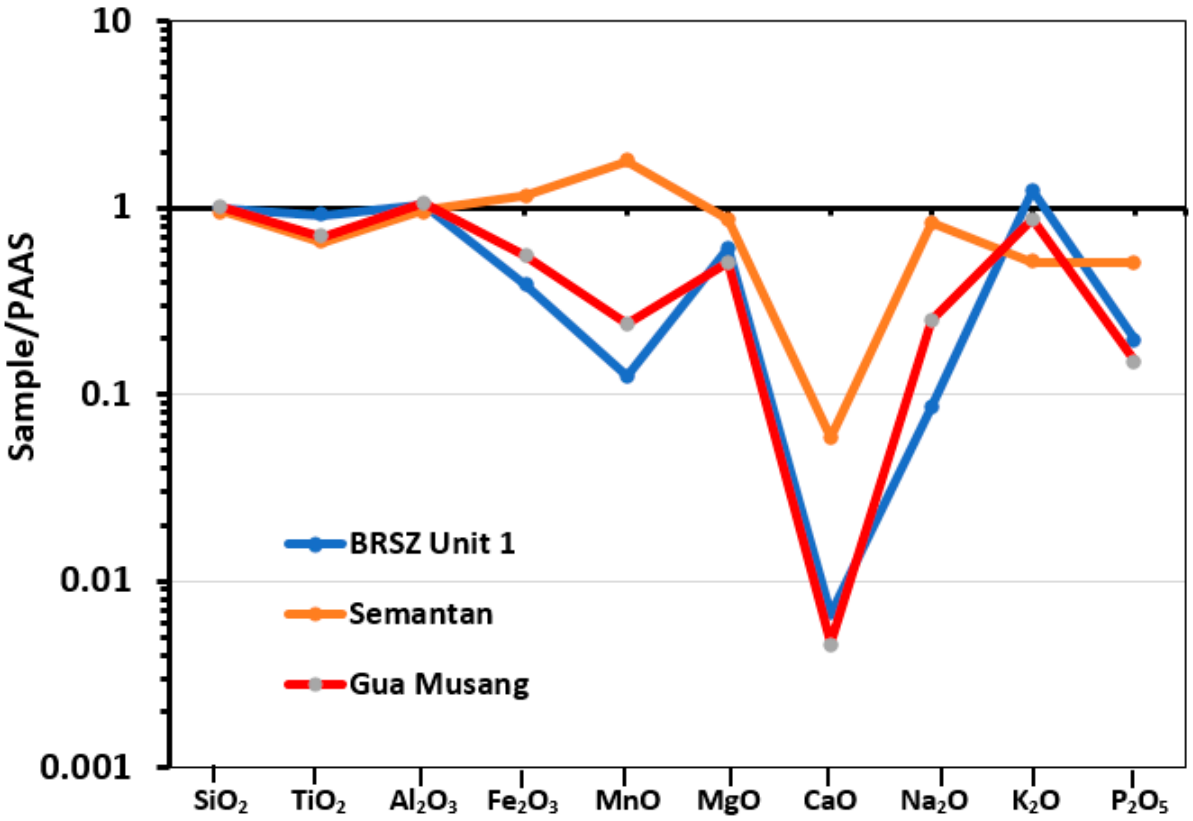

Figure 6. Major element concentrations normalized against PAAS (Post-Archean Australian Shale [16]). Each line on the graph represents an average elemental concentration of the BRSZ Unit 1, Semantan, and Gua Musang black shale samples. Y axis in logarithmic scale.

4.2. Trace Elements

The average trace element values for the BRSZ Unit 1, Semantan, and Gua Musang black shales are given in Table S2. In Figure 7, Mo content is the highest in the BRSZ Unit 1 compared to the Semantan and Gua Musang black shales. The Sr content is depleted in all the three formations. The contents of Pb, U, and V are quite elevated in the BRSZ Unit 1 relative to the other two.

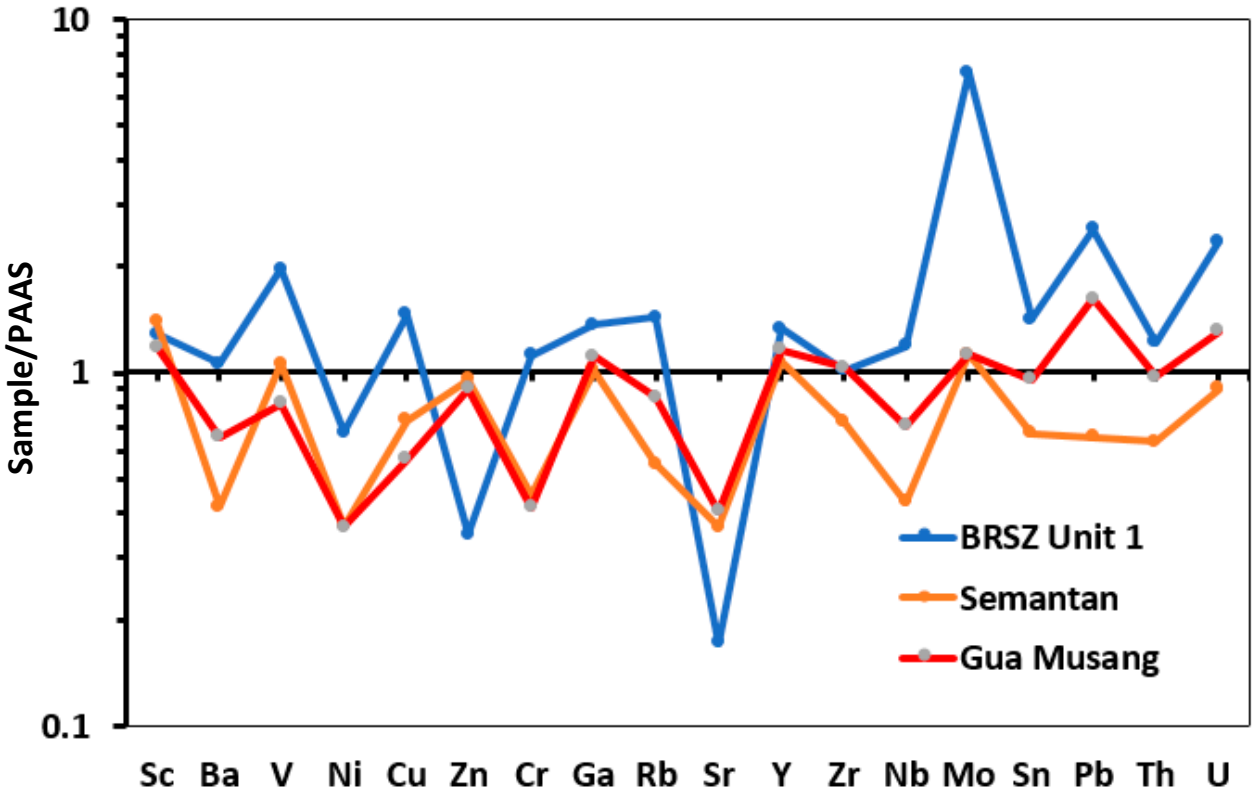

Figure 7. Trace element concentrations normalized against PAAS (Post-Archean Australian Shale [16]. Each line on the graph represents an average elemental concentration of the BRSZ Unit 1, Semantan, and Gua Musang black shale samples.

4.3. Chemical Index of Alteration (CIA) and Chemical Index of Weathering (CIW)

Reference [40] invented the Chemical Index of Alteration (CIA) to objectively assess weathering history recorded in sediments and sedimentary rocks. The CIA values for the black shales from the BRSZ Unit 1 vary from 79 to 82. Black shales from the Semantan Formation have CIA values ranging from 83 to 96. Black shale samples from the Gua Musang Formation have CIA values ranging between 83 and 87 (Table S2). The CIW is a chemical proxy that helps estimate the degree of weathering. It increases as the degree of weathering gets higher. The average CIW values of samples from the BRSZ Unit 1, Semantan, and Gua Musang are 99.4, 94.5, and 98.6 respectively as shown in Table S2.

5. Discussion

5.1. Major and Trace Elements

Compared to Post-Archean Australian Shale (PAAS), BRSZ Unit 1 and Gua Musang black shales are depleted in Fe_2O_3, MnO, CaO, Na_2O, and P_2O_5 compared to PAAS. However, the Semantan black shales are enriched in MnO and depleted in CaO, K_2O, and P_2O_5 relative to PAAS. The black shakes of BRSZ Unit 1 are enriched in V, Cu, Ga, Rb, Mo, Sn, Pb, and U compared to PAAS. The Gua Musang black shales are mostly depleted in trace element concentration except for Pb (mean: 32.3 ppm). Likewise, the Semantan black shales are mostly depleted in trace element concentrations apart from Sc (mean: 22.3 ppm)

which is moderately elevated. Collectively, for all the samples analyzed, the BRSZ Unit 1 black shales are remarkably depleted in Sr (mean: 34.2 ppm) relative to PAAS.

The Zn contents have two maximum concentrations (Table S2), one in the Gua Musang (194 ppm) and another in the Semantan (197 ppm) black shales, which may indicate the presence of sphalerite (ZnS) inclusions in the structure of micron-sized crystals of sedimentary pyrite often found in the black shales in the Central Belt. Considering this, [41] documented that sedimentary pyrites contained in the black shales from the Gua Musang and Semantan Formations are enriched in Zn. For example, the Zn content of Gua Musang sedimentary pyrites reached a maximum of 788 ppm, and that of Semantan reached a maximum of 6990 ppm [41]. This means that when powder samples are prepared for XRF analysis, the enrichment of Zn in sedimentary pyrite can affect the XRF results. Additionally, tiny crystals of euhedral pyrites containing sphalerite (ZnS) inclusions were reported from the Gua Musang black shales [41], which may also have affected the Zn concentration when XRF was carried out. The enrichment of Mo in the BRSZ Unit 1 is explained below.

Reference [41] documented that the BRSZ Unit 1 is enriched in organic matter, with total organic carbon (TOC) content varying from 0.2 to 2.03 wt% relative to the Semantan and Gua Musang, which have TOC contents of 0.28–0.67 wt% and 0.05–0.6 wt%, respectively. Therefore, the BRSZ Unit 1 has the highest TOC content relative to the Semantan and Gua Musang black shales. Reference [42] also indicated that the correlation coefficient between Mo and TOC was R = 0.91 in the BRSZ Unit 1 black shales, implying a strong positive correlation. It shows that Mo was associated with organic matter during sedimentation. The proportion of Mo in the BRSZ Unit black shales is proportional to the amount of TOC in the shales. In simple terms, the higher the TOC, the higher the Mo content.

5.2. Provenance

Low-mobility trace elements including La, Th, Y, Zr, Ti, Co, and Ni are beneficial for distinguishing between different provenances [42–45]. Chemical compositions of shales have been used to infer sediment source areas and their tectonic settings. The composition of the developing sedimentary rocks is significantly influenced by the composition of the source rocks [40].

Due to their poor mobility during weathering, transport, diagenesis, and metamorphism, some elements such as Zr, Th, and Sc are used in this study to determine provenance [46,47]. It is also possible to distinguish between felsic and mafic parent rocks by comparing the ratio of certain immobile elements, such as La and Th (evidence for felsic provenance), to compatible elements, such as Sc and Cr (evidence for mafic provenance) [17,48–50]. Data from this study indicates that samples of Gua Musang Formation mostly plot within the intermediate field together with the BRSZ Unit 1 black shales suggesting an intermediate parental source. The Semantan black shales are mostly plotted within the felsic field implying a felsic source rock (Figure 8).

The Al_2O_3/TiO_2 ratio for mafic rocks ranges from 3 to 8, for intermediate rocks it ranges from 8 to 21, and for felsic igneous rocks it ranges from 21 to 70 [51]. Due to the limited solubility of their oxides and hydroxides in solutions, Al and Ti remain practically stationary throughout the weathering of source rocks [52,53]. Since Al-rich clays, Ti-bearing clays, and mafic minerals exhibit little fractionation during transit, the Al/Ti ratios of the sedimentary rocks have also been taken into consideration for provenance investigations [53]. The plot of Al_2O_3 versus TiO_2 is shown in Figure 9.

To distinguish between different sediment provenances [54] established four domains. Felsic igneous, quartzose sedimentary, intermediate igneous, and mafic igneous are the four fields or sources. According to [55], the Th versus Sc plot of the source rock lithology does not appear to be as reliable as the discriminant diagram based on key elements. The discriminant function diagram of the major elements Al_2O_3, TiO_2, Fe_2O_3, MgO, CaO, Na_2O, and K_2O contents can be plotted using two functions (Figure 10).

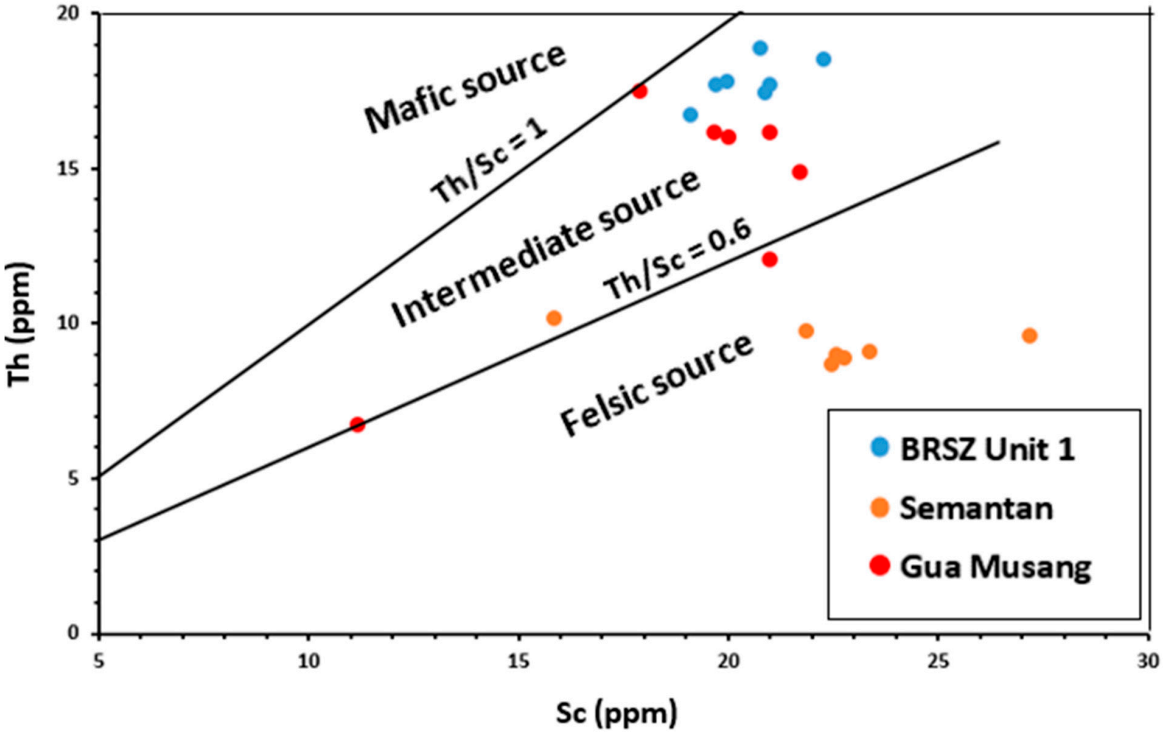

Figure 8. Provenance discrimination diagram Th versus Sc. The BRSZ Unit 1 and Gua Musang plot in the intermediate field, whereas the Semantan samples cluster in the felsic field.

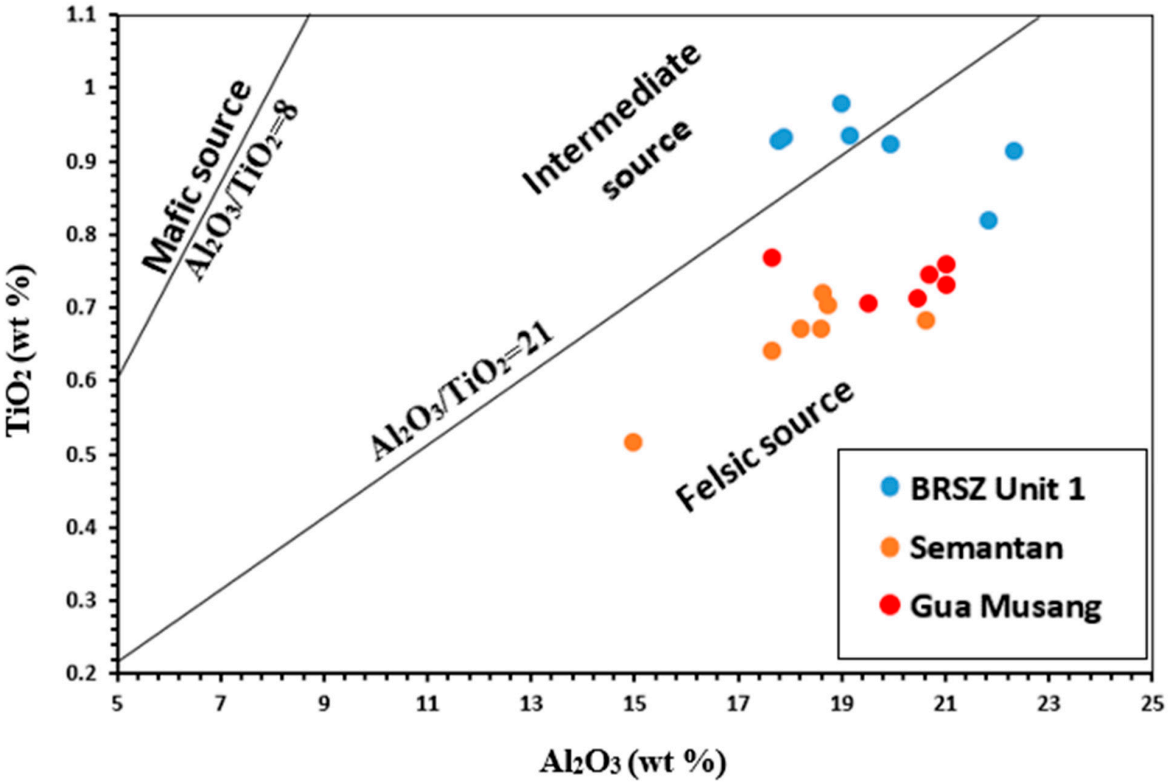

Figure 9. Discrimination diagram of Al_2O_3 versus TiO_2 (Diagram after [49]). The BRSZ Unit 1 samples spread across the intermediate and felsic sources; but the Semantan samples plot spreads in the felsic source field, as does the Gua Musang shifting from intermediate to felsic source.

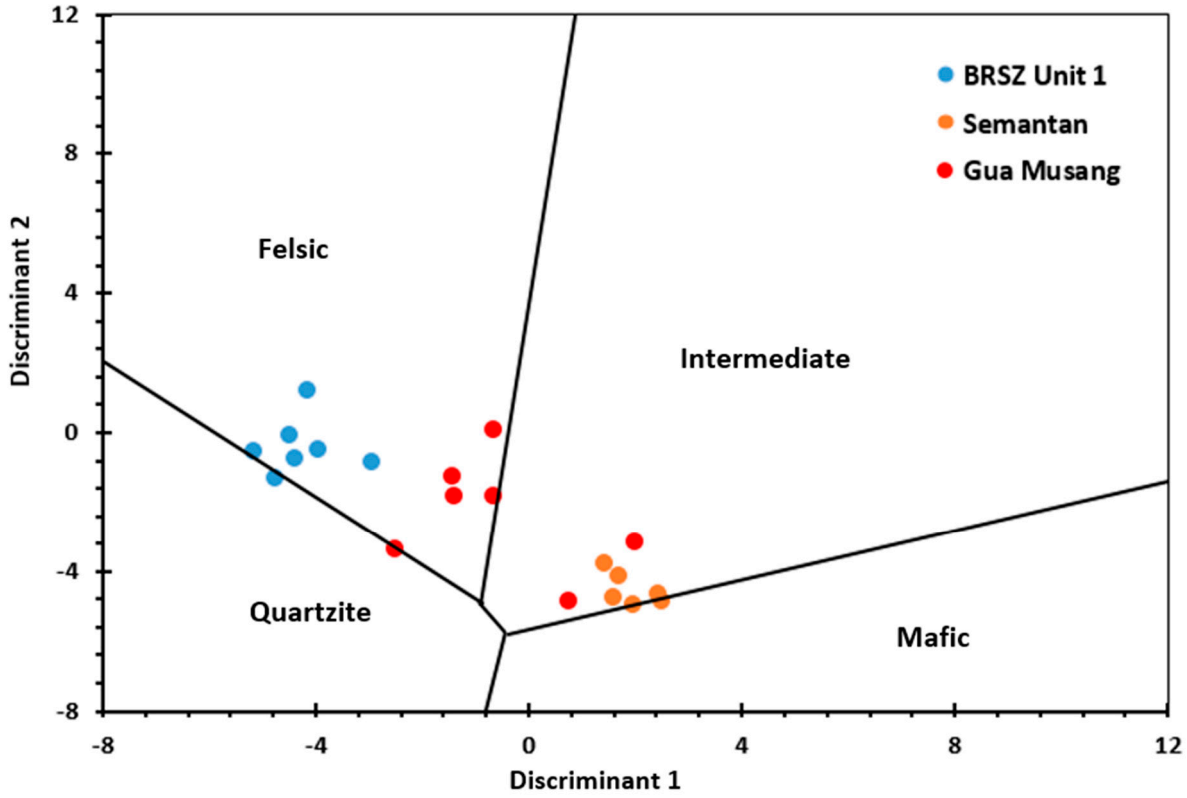

Figure 10. Discriminant diagram showing the main fields of source rocks based on the black shale samples analyzed in this study. The two discriminant functions are the following: Discriminant function 1 = −1.773 TiO_2 + 0.607 Al_2O_3 + 0.76 Fe_2O_3 − 1.5 MgO + 0.616 CaO + 0.509 Na_2O − 1.224 K_2O − 9.09. Discriminant function 2 = 0.445 TiO_2 + 0.07 Al_2O_3 − 0.25 Fe_2O_3 − 1.142 MgO + 0.438 CaO + 1.475 Na_2O + 1.426 K_2O − 6.861. The Semantan samples are in the intermediate field. Gua Musang samples spread across the felsic-intermediate fields.

The evidence shows that black shales of the Semantan Formation may have derived from felsic and intermediate parental rocks. The BRSZ Unit 1 black shale also indicates-felsic parental rocks. The Gua Musang black shales are derived from mostly felsic, and intermediate source rocks. The felsic nature of the parental rocks of the Semantan black shales may be explained by the occurrence of rhyolite quarry (Figure 8) near Lanchang and the existence of sills of intermediate and mafic composition (Figure 9) within the Semantan strata [56]. The shales in the Gua Musang Formation were found interbedded with volcanics. The volcanic rocks are widespread and vary in composition from rhyolitic to andesitic including tuffs, lavas, and agglomerates [5,23,24]. As a result, it may account for the felsic and intermediate source rocks of the Gua Musang black shales (Figures 7–9).

The composition, sorting, and heavy-mineral concentration of sediments are thought to be reflected by the Zr/Sc and Th/Sc ratios [57–59].

5.3. Ratios of Th/Sc and Zr/Sc

Proportions of both incompatible and compatible elements are useful for distinguishing between felsic and mafic source materials, as is the Th/Sc ratio, which is a proxy of igneous chemical differentiation processes because Th and Sc are incompatible and compatible in igneous systems, respectively [58]. In turn, the Zr/Sc ratio is a helpful diagnostic of zircon enrichment since Zr is highly concentrated in zircon while Sc generally retains the provenance signature [48,57].

Previous research has shown that the Puncoviscana metasediments have high and fairly constant Th/Sc ratios (0.86–1.76) that strongly indicate provenance from a relatively evolved igneous source, as well as high and remarkably variable Zr/Sc ratios (17–164) that

indicate zircon enrichment [60]. Compared to the present study, Th/Sc ratio values for the black shales vary from 0.35 to 0.98 indicating highly evolved igneous source. The Zr/Sc ratio values range between 5.94 and 15.35 suggesting less zircon enrichment.

5.4. Tectonic Setting

The tectonic context of derived sedimentary material may be inferred using the chemistry of sedimentary rocks. The method developed by [61] is one of the most used tectonic setting discrimination techniques. It is predicated on the idea that there are considerable differences across tectonic settings in terms of SiO_2 concentration and K_2O/Na_2O ratio. Many authors [62–64] have employed the K_2O/Na_2O against SiO_2 plot as shown in Figure 11 to discuss tectonic setting. In this study, the BRSZ Unit 1 black shales indicate a passive margin setting of the source area.

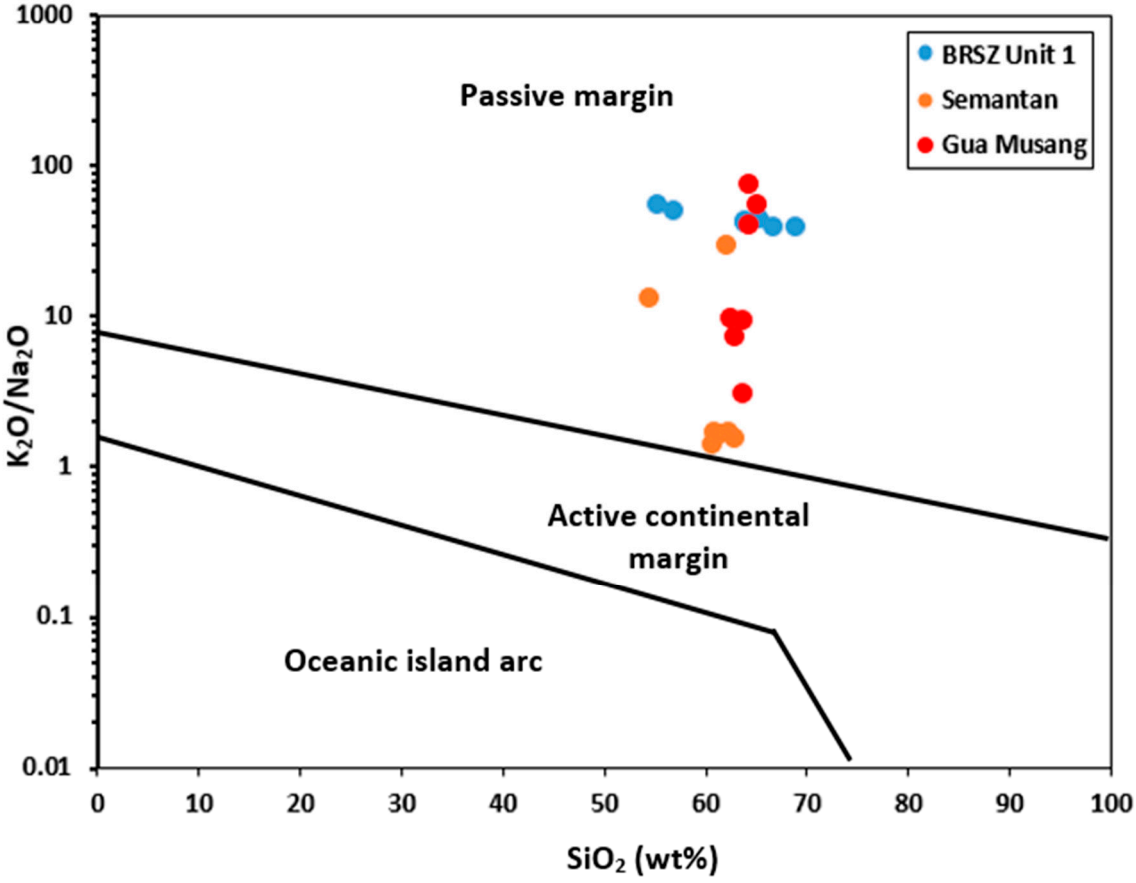

Figure 11. Tectonic setting discrimination plot K_2O/Na_2O versus SiO_2 for the studied black shale samples (Diagram after [62]). All samples plot in the passive margin setting.

The Gua Musang black shale samples also indicate a passive margin setting of the source area. Similarly, the Semantan black shales plot in the passive margin setting.

Low-mobility trace elements represent the tectonic context more accurately during transport and deposition [65–67]. The La–Th–Sc plot adapted from [42] shows that all samples from BRSZ Unit 1 fall within the passive margin and the Gua Musang black shales show a chemical affinity to a passive margin and continental island arc, whereas the Semantan black shales point to a continental island arc (Figure 12). The diagnostics of the continental island arc may be explained by the presence of shallow-deep volcaniclastic facies in the Gua Musang and Semantan areas that have been interpreted to be related to volcanic activities that occurred in the region [11,68].

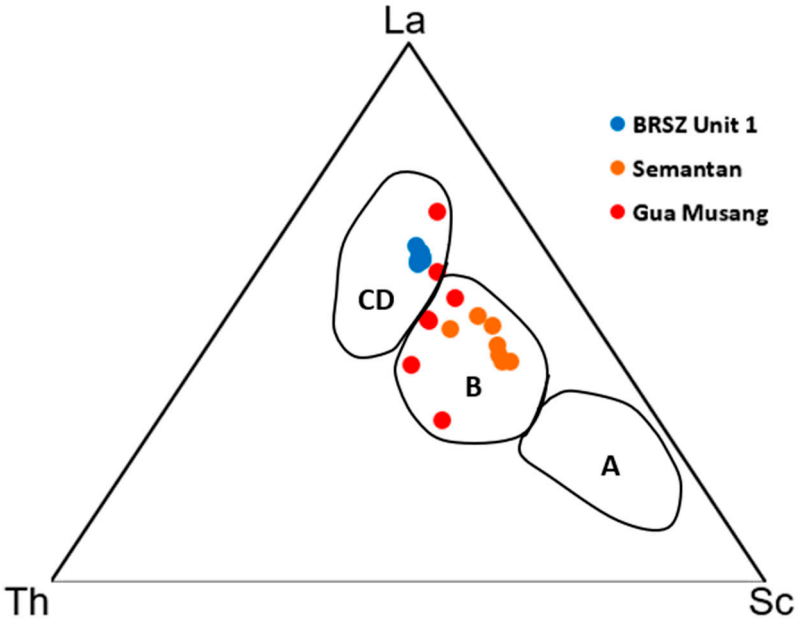

Figure 12. La–Th–Sc discrimination plot (diagram after [43]). Note: A = Oceanic Island arc; B = Continental Island arc; C =Active continental margin; D = Passive margin. The BRSZ Unit 1 plots in the passive and active continental margin. Gua Musang samples plot in the CD and B fields. Semantan samples cluster in the continental island arc setting.

Reference [69] discussed the sedimentology and stratigraphy of the Middle Triassic rocks of the Semantan Formation that crop out near Lanchang, Pahang in Malaysia. In terms of provenance and geographic setting, the research work documented that the fine material consisting of black shales may have derived from fine ash or altered volcanics. The site of deposition of the black shales was on the western or southwestern part of a Triassic volcanic island or island chain.

With regards to water depth, the black shales do not contain evidence of bioturbation and primary structures of shallow water setting but had transported neritic bivalves [69] indicating a deeper marine environment. Compared with literature data, the sedimentological and geochemical evidence points to a continental island arc setting for the Semantan black shales (Figure 12).

Some Gua Musang samples plot within the field of continental island arc may be explained by the fact that a lot of volcaniclastic rocks were reported to be cropping out in the Gua Musang area (Figure 12).

The two discrimination plots shown above (Figures 10 and 11) do not precisely define the tectonic setting of the BRSZ Unit 1, Gua Musang, and Semantan black shales. It is important to mention that these black shale samples were collected in the east of the Bentong-Raub Suture Zone (Figure 1). This suture formed during the collision between the Sibumasu and Indochina tectonic blocks [70]. For this reason, a new discrimination diagram is shown below that is adapted from [71]. These authors proposed a new multi-dimensional diagram for tectonic discrimination of clastic rocks with an emphasis on the fact that no discrimination diagrams were proposed to distinguish between an island or continental arc, continental rift, and collision. Reference [71] also found that the success rate of their diagrams ranged from 84.5% to 93.6%. This study shows that the BRSZ Unit 1 and Gua Musang black shales plot in the collision setting field, whereas five samples of the Semantan black shales plot in the arc setting field and two samples plot in the collision setting field (Figure 13). The Semantan samples that plot in the arc setting field in Figure 13 show tectonic setting similarity with those that plot in the continental island arc in the La–Th–Sc plot (Figure 12).

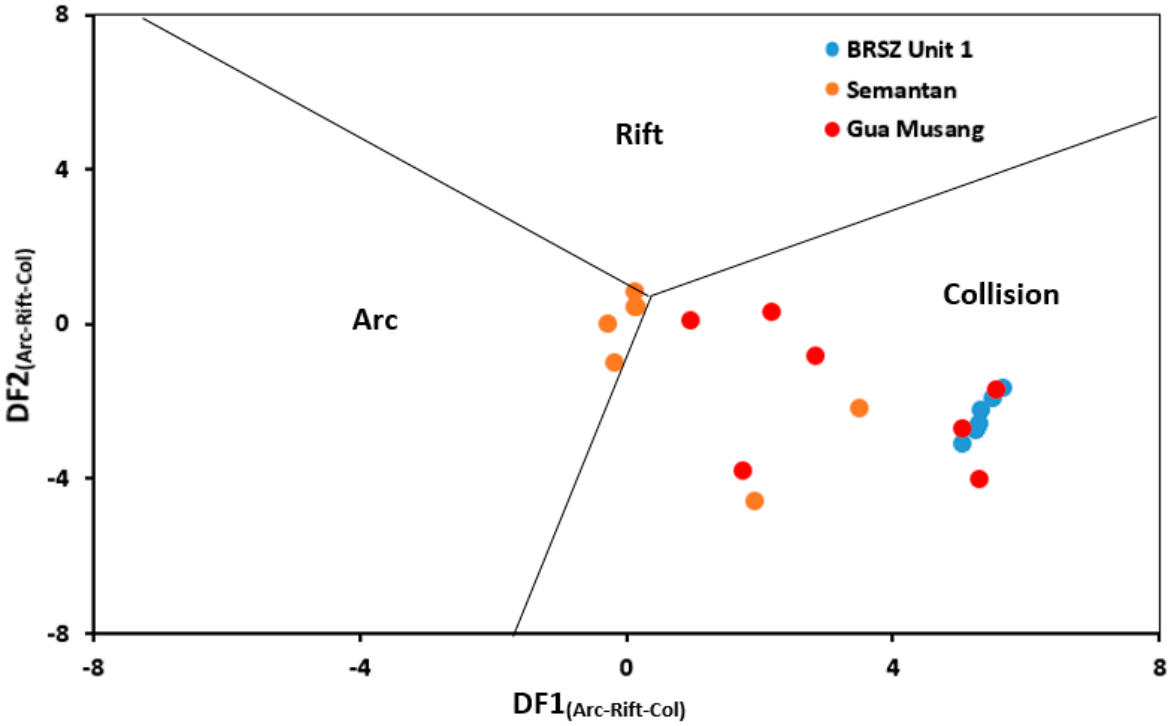

Figure 13. Discrimination-function multi-dimensional diagram for low-silica BRSZ Unit1, Semantan, and Gua Musang black shales highlighting the three tectonic setting fields (arc, continental rift, collision). $DF1_{(Arc\text{-}Rift\text{-}Col)}$ = (0.608 × ln(TiO_2/SiO_2) + (−1.854 × ln(Al_2O_3/ SiO_2) + (0.299 × ln($Fe_2O_3{}^t$/ SiO_2) + (−0.550 x ln(MnO/SiO_2) + (0.120 × ln(MgO/SiO_2) + (0.194 × ln(CaO/SiO_2) + (−1.510 × ln(Na_2O/SiO_2) + (1.941 × ln(K_2O/ SiO_2) + (0.003 × ln(P_2O_5/SiO_2) − 0.294. $DF2_{(Arc\text{-}Rift\text{-}Col)}$ = (0.608 × ln(TiO_2/SiO_2) + (−1.854 × ln(Al_2O_3/SiO_2) + (0.299 × ln($Fe_2O_3{}^t/SiO_2$) + (−0.550 × ln(MnO/SiO_2) + (0.120 × ln(MgO/SiO_2) + (0.194 × ln(CaO/SiO_2) + (−1.510 × ln(Na_2O/SiO_2) + (1.941 × ln(K_2O/SiO_2) + (0.003 × ln(P_2O_5/SiO_2) − 0.294. The DF1 and DF2 were calculated using the low-silica rocks (SiO_2 = 35%–63%) discrimination-function diagram [71].

5.5. Paleoweathering

The degree of weathering can be inferred by using the Chemical Index of Alteration (CIA). CIA is determined by the following formula: CIA = [$Al_2O_3/(Al_2O_3 + CaO^* + Na_2O + K_2O)$] × 100 [40,72].

The values in this equation are in molar proportions and the CaO content is the amount of CaO found in the silicate fraction of the rock sample. CIA values between 50 and 60 indicate a weak chemical weathering, values between 60 and 80 indicate a moderate degree of chemical weathering, and CIA values > 80 indicate intense chemical weathering [73].

CIA values of 50–65 indicate weathering in cold and dry environments. A value of 65–85 points to weathering in warm and humid environments. A value of 85–100 indicates weathering in hot and humid environments. The CIA values for all the samples analyzed in this study range between 79.4 and 95.8 (Figure 14), indicating an intense chemical weathering in warm and wet paleoclimatic conditions.

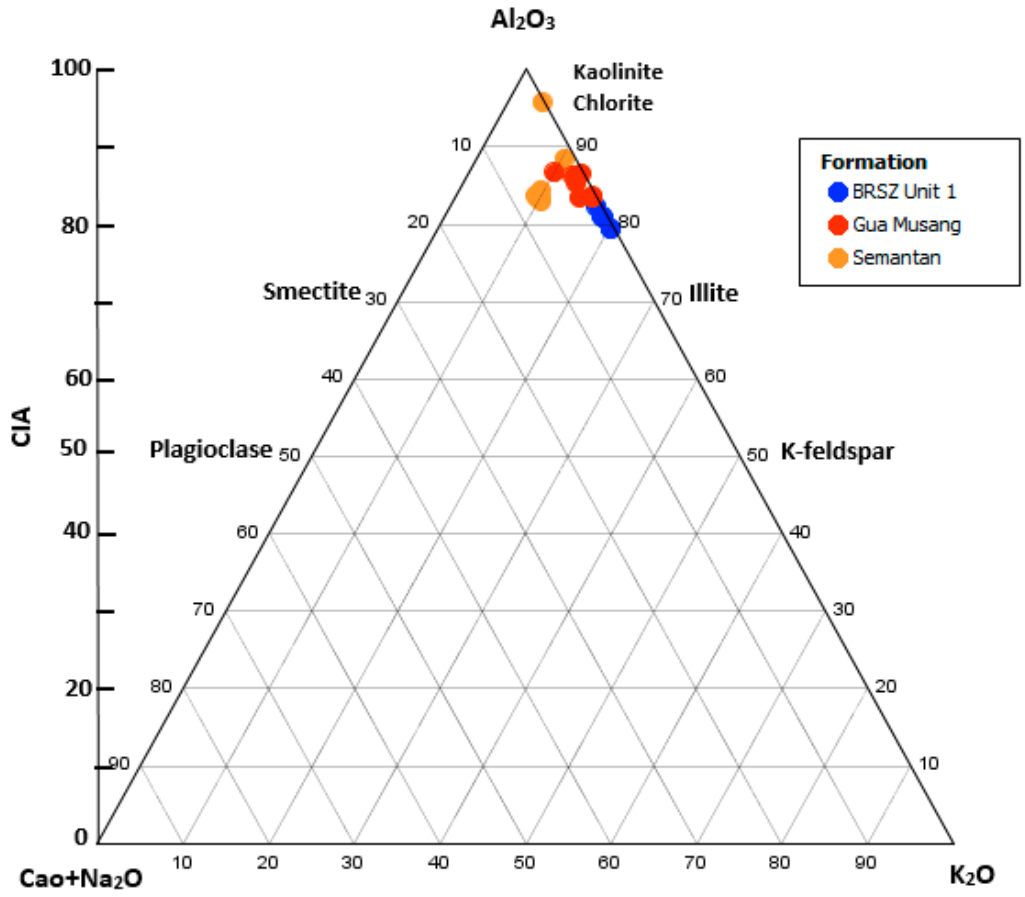

Figure 14. Ternary plot for the estimation of the chemical index of weathering of the black shales on molecular proportions of Al_2O_3, CaO, Na_2O, and K_2O [74].

To counter the effects of potassium alteration, the Chemical Index of Weathering (CIW) has been estimated to further evaluate the degree of weathering [75–78]. The CIW is calculated using the following formula:

$$CIW = [Al_2O_3/(Al_2O_3 + CaO + Na_2O)] \times 100 \text{ (in molecular proportions).} \quad (1)$$

The CIW values of all samples range between 92.3 and 99.8. The average CIW values of samples from the BRSZ Unit 1, Semantan, and Gua Musang are 99.4, 94.5, and 98.6, respectively. All these CIW values indicate an intense degree of weathering of the source rocks.

6. Conclusions

Major element compositions indicate that the BRSZ Unit 1 black shale has elevated SiO_2, TiO_2, and K_2O contents compared to the Semantan and Gua Musang black shale. Trace element compositions show that the BRSZ Unit 1 black shale has elevated trace element contents compared to the Semantan and Gua Musang black shale. The BRSZ Unit 1 black shale has the highest V, U, Pb, and Mo contents relative to the other two formations. The Mo content in the BRSZ Unit 1 is the highest because Mo is associated with the elevated total organic carbon (TOC) in the BRSZ Unit 1 black shale. Compared to PAAS, the Semantan black shales are deficient in CaO, K_2O, and P_2O_5 and enriched in MnO. The BRSZ Unit 1 and the Gua Musang black shales are low in Fe_2O_3, MnO, CaO, Na_2O, and P_2O_5. The black shales of BRSZ Unit 1 are enriched in V, Cu, Ga, Rb, Mo, Sn, Pb, and U. The Gua Musang black shales are largely depleted in trace element content. Similar to the Semantan black shales, all trace element concentrations are largely depleted with the exception of elevated Sc content.

Provenance analysis indicates that the BRSZ Unit 1, Semantan, and Gua Musang black shales were derived from felsic and intermediate parental source rocks. Tectonic discrimination diagrams show that the BRSZ Unit 1 and Gua Musang black shales point to a collisional setting; however, the Semantan black shales show affinity to a continental arc setting. The CIA values for all the samples range between 79.4 and 95.8, indicating intense chemical weathering in warm and wet conditions. The average CIW values of samples from the BRSZ Unit 1, Semantan, and Gua Musang are 99.4, 94.5, and 98.6, respectively, implying an intense degree of weathering of the source rocks.

The Semantan and Gua Musang Formations occupy a wide area in the Central Belt. Therefore, further research is needed to acquire robust data in terms of major and trace element composition, provenance, tectonic setting, and intensity of weathering.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/min13070911/s1, Table S1. Location coordinates of samples collected in the field; Table S2. Major and trace element concentrations.

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