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PALEOREDOX CONDITIONS OF PALEOCENE TO MIOCENE ROCKS OF EASTERN BENIN (DAHOMEY) BASIN, NIGERIA: IMPLICATIONS FOR CHEMOSTRATIGRAPHY

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ABSTRACT

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The major oxides and trace elements, of two wells spanning the Paleocene – Miocene of the Benin Basin, were used to infer the varying paleoredox conditions during deposition of the different rock units and identify the chemostratigraphic boundaries. The major oxides revealed four boundaries in these wells, while trace elements suggest five and three chemostratigraphic boundaries for BH and AJ wells respectively. The information from V/Sc, Ni/V and V/(V+Ni) ratios indicates the prevalence of oxic conditions in the sandy upper 25m of the BH well and the interval between 1046 – 1485m of the AJ well. The redox sensitive trace elements of the shaly middle parts of the BH well suggest dysoxic – anoxic conditions and in most parts of the AJ well.

Contribution/Originality: This study is one of the very few studies which have investigated the variations in oxygenation levels in some Post-Cretaceous sediment of the Benin Basin, Nigeria. The study outlines how the distribution of redox sensitive elements was used to suggest paleoredox conditions and identify chemostratigraphic boundaries.

1. INTRODUCTION

In recent decades, the characterization and fingerprinting of individual sedimentary units based on elemental (major and trace elements) and isotopic geochemistry have emerged to be a potential tool for correlation of widely separated strata where there is paucity of biostratigraphic information; this concept is referred to as chemostratigraphy (Ramkumar, Nagarajan, & Santosh, 2021; Turner, Tréanton, & Slatt, 2016). The method is centered on the principle that the chemical composition of sediments is a genuine recorder of the depositional conditions of the sediments and reflect the changes in the paleoredox conditions.

Several authors have carried out detailed studies on the stratigraphy, tectonic settings, petroleum potential, foraminiferal biostratigraphy and depositional environments of the Nigerian sector of the Benin Basin (Adekeye & Akande, 2007; Okosun, 1990; Olabode, 2006; Oluwajana et al., 2021; Omatsola & Adegoke, 1981). Ajayi et al. (2006) carried out a geochemical study on the Paleogene shales of the Benin Basin and reported an oxygen poor environment for these shales. Akaegbobi and Ogungbesan (2016) investigated the depositional environment of basin using major oxides, trace and rare earth elements. The investigation concluded that a restricted to open shallow marine environments under fluctuating oxidizing to anoxic conditions prevailed in the basin, but the study was restricted to the Paleocene limestones of the basin. Ehinola, Ejeh, and Oderinde (2016) likewise, deduced a probable anoxic condition in quiet, low energy environments and rapid rate of sulphate reduction for the Paleocene limestones. The present study deals with boreholes that cut across the different lithologic units in the Paleocene to Miocene column of the Nigerian sector of the Benin Basin to infer the variations in oxygenation levels downhole the wells.

2. GEOLOGY AND STRATIGRAPHY

The Nigerian sector of the Benin Basin is part of a system of West African Atlantic margin basins, which occupies much of the continental margins of the Gulf of Guinea, extending from southeastern Ghana in the West to the Okitipupa ridge on the western flank of the Niger Delta in Nigeria on the East (Olabode & Mohammed, 2016; Omatsola & Adegoke, 1981). The separation of the Africa and South America plates in the Late Jurassic – Early Cretaceous Period was the precursor to the Benin Basin. This tectonic event led to the opening of South Atlantic Ocean and the development of various faults and fractures; amongst which are the Romanche Fracture Zone (RFZ) and Chain Fracture Zone (CFZ) structures in the South Atlantic Ocean, and other horst and graben features in the basin. The basin is bounded in the west by an offset extension of the RFZ, faults and other tectonic structures associated with the landward extension of the fracture zone. Its eastern limit is similarly marked by the Hinge line, the continental extension of the CFZ, a major fault structure marking the western limit of Niger Delta. The Nigerian sector of the basin is bounded in the north by the Precambrian basement rock and the Gulf of Guinea in the south (Omatsola & Adegoke, 1981).

Early works on the stratigraphy of the Benin Basin detailed successive lithostratigraphic formations spanning the Cretaceous to Recent (Okosun, 1990; Omatsola & Adegoke, 1981). The oldest sediments are from the Abeokuta Group, which are the Ise, Afowo and Araromi Formations. The age of the Ise Formation is Neocomian and it is made up of conglomerates and sandstone facies. The post – Cretaceous sediments are the Ewekoro, Akinbo, Oshosun and Ilaro Formations (Figure 1). The youngest deposits are poorly fossiliferous Continental Sandstone deposits and siltstone.

The study wells however, comprises varying thickness of limestone, conglomerates, sandstone, clay and shale. The upper portions of the BH well is essentially sandstone (Figure 2a), while the middle portions have thick sequences of shale with interbeds of marl, limestone, clay and sandstone. The lowermost part of the well is entirely limestone. The AJ well have an upper section that is composed of shaly-sand, while the lower sections are a monotonous sequence of shales (Figure 2b), with thin interbeds of white – cream – brownish coloured limestone in some parts, marl, clay and sandstone.

3. MATERIALS AND METHODS

Two boreholes, BH well (146m core) and AJ well, were utilized for this study. The samples studied were carefully selected to fall within the age range of study (Paleocene – Miocene). The age of the sediments was confined using marker species of palynomorphs and nannofossils. The upper 137m of the BH well falls within the stratigraphic column of interest, as this depth contains typical Paleogene assemblage of palynomorphs of *Gemmamonoporites sp.*, *Psilatricolporites crassus*, *Verrucatosporites usmensis* and *Constructipollenites ineffectus*, but the lowermost portions is a sort of mixed bag of both Cretaceous and Paleogene palynomorhs. The presence of Paleogene nannofossil marker species of *Neochiatozygus modestus.* at around 1890m (Sample AJ36), in the AJ well was used to confine the depth of study.

Varied geochemical analysis of major and trace element analysis were carried out on a total of 49 samples at the Bureau Veritas Commodities Mineral Laboratory, Canada. 13 of the samples are from the BH well (sample numbers are depths in meters), and 36 ditch -cuttings from the AJ – well at intervals of about 49m (AJ1 – AJ36, 168m – 1890m).

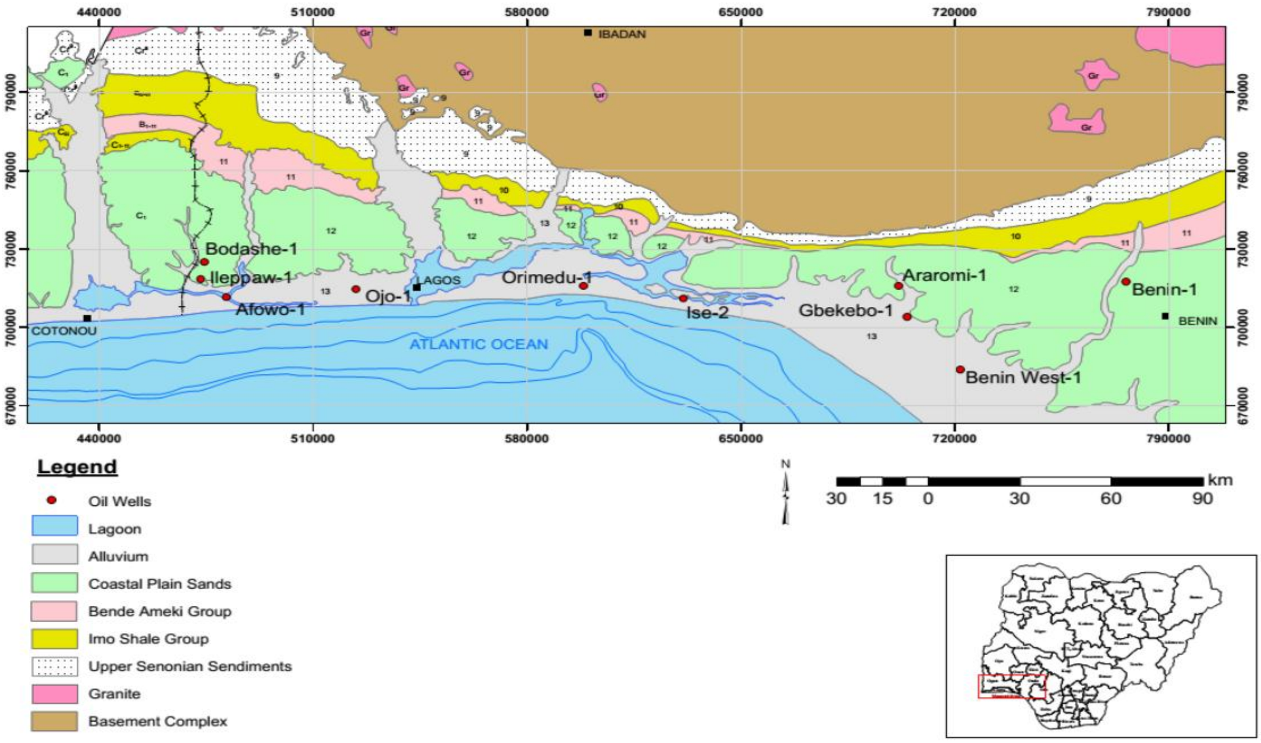

Figure-1. Regional geological map (showing Cretaceous to Quaternary sediments) and well location map of Nigeria's sector of the Benin Basin (Adapted from Ehinola and Oluwajana (2016)).

The multi-acid, Ultra-trace inductively coupled plasma mass spectrometry ICP-MS (MA250) and whole rock Lithium Fusion ICP finish (LF300) methods were used. The MA250 method is capable of dissolving most minerals, which gives near total values for about 59 elements. The method was conducted on 25 samples. A 0.25 g split is heated in HNO_3, $HClO_4$ and HF to fuming and taken to dryness, the residue is then dissolved in HCl. The LF300 (ICP/ICP -MS) analysis employs the fusion technique to completely decompose even the most refractory matrices to measure the concentration for the major oxides and loss on ignition (LOI). This was carried out on all the 49 samples. A split was taken to represent the original sample and were subjected to quality (QA/QC) control checks during the progression of the analytical portion. A detailed explanation on the methods can be obtained from Acme Laboratories Limited, Vancouver, Canada.

The measured concentrations were compared with the Post Archaean Australian Shale (PAAS, Taylor and McLennan (1985)) and also the average Upper Continental Crust concentrations (McLennan, 2001).

4. RESULTS

4.1. Major Element Geochemistry

The SiO_2 content from the upper 33m of the BH core shows enrichment (88.93 - 97.12wt%) relative to PAAS and UCC, but lesser concentrations in the lower parts, with samples BH 83, BH 95 and BH 137 having values below 40.00wt%. Contrastingly, Al_2O_3 and Fe_2O_3 concentrations are lesser in the upper 33m (below 5.00wt%) and a bit higher in the lower parts of the core. A similar trend is observed with MgO, concentrations in the upper parts of the core are low (below 0.30wt%), but higher in the lower portions. CaO content also show some variation, concentration in most of the samples are less than 3% except samples BH71, BH83, BH 95 and BH 137 which recorded 5.02wt%, 17.71wt%, 10.33wt% and 44.64wt% respectively. Na_2O contents are quite low (0.03 - 0.15wt%), so also P_2O_5 (0.02 - 0.32wt%) and MnO concentrations were below minimum detection limit (0.01wt%) in the upper

33m and ranged from 0.02 - 0.20wt% in the lower parts. Small variations of K_2O and TiO_2 were found in the middle portion (33 – 71m) which ranged from 1.10 - 1.19wt% and 1.31 - 1.54wt% respectively, while the other portions recorded concentrations lower than 1%. The recorded LOI varies greatly with lesser percentages in the upper 33m and higher values in the lower parts of the well (16.60 - 36.70wt%).

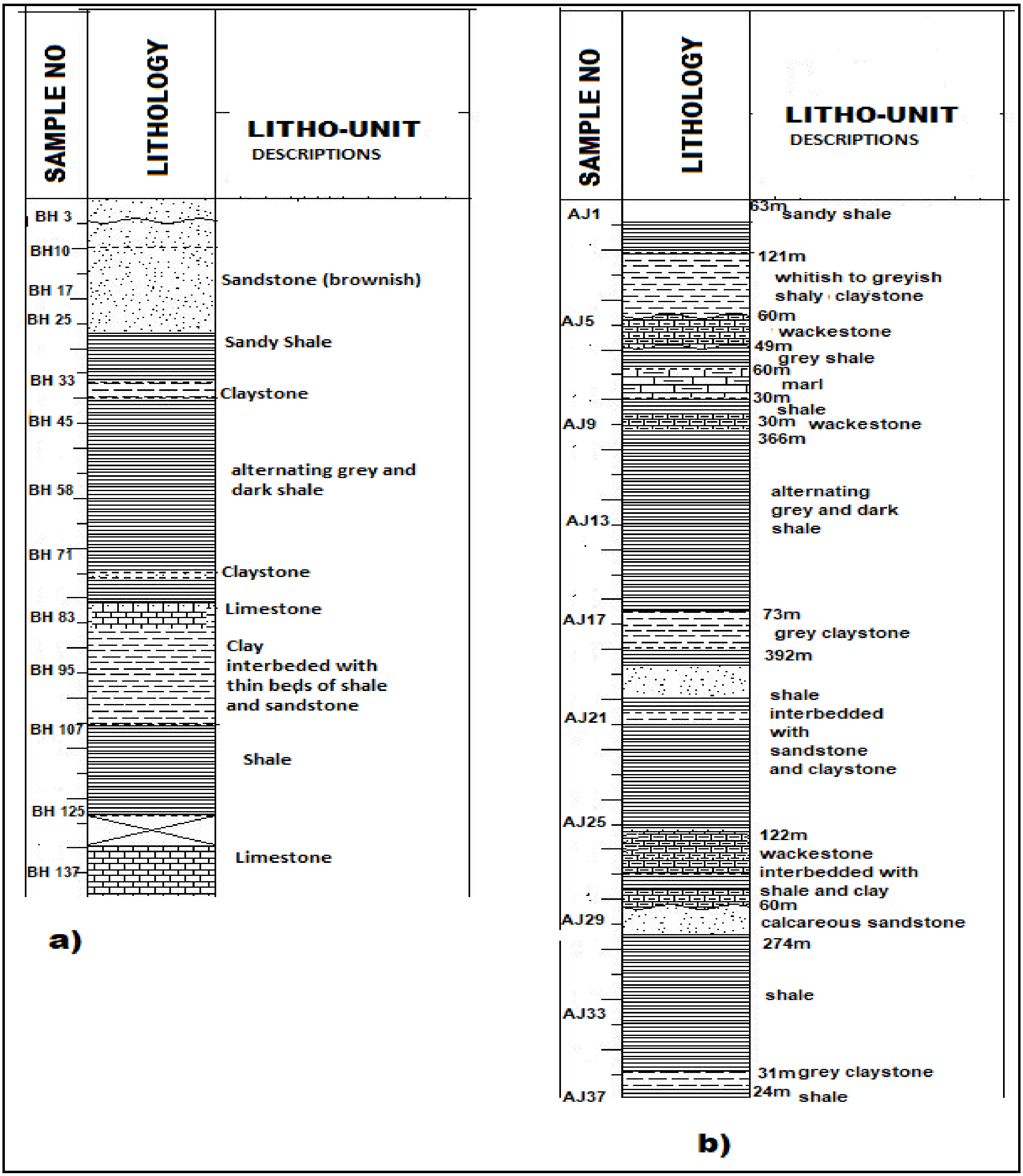

Figure-2. Lithostratigraphy of the Study Wells. **a)** The BH Well has a Basal Limestone Belonging to the Ewekoro Formation and the Upper Parts are Sandstone of the Ilaro Formation. **b)** The AJ Well is Monotonously Shale with Interbeds of Sandstone, Clay and Limestone (wackestone).

The SiO_2 concentrations of 33 samples from the AJ well ranged from 42.88wt% to 77.97wt% (Average 58.17wt%) while samples AJ5, AJ7 and AJ9 recorded lower values (21.74wt%, 22.86wt% and 26.40wt% respectively). The samples with the higher SiO_2 and CaO concentrations recorded lower Al_2O_3 concentrations (below 10wt%; samples, AJ5, AJ7, AJ9, AJ10, AJ17, AJ19, AJ20, AJ21, AJ22, AJ25, AJ27, AJ28, AJ29) while the other 23 samples recorded values above 10wt%. The elemental concentration of iron (in Fe_2O_3 form) is from 1.48 - 10.17wt% and the samples listed above with lesser Al_2O_3 concentrations also recorded lower iron oxide values. MgO concentrations varies from 0.38 - 8.51wt% (average of 2.57wt%, indicating slight enrichment). The CaO concentrations are quite significant and ranged from 1.42 - 31.57wt%. The concentrations of the other major oxides are quite low, TiO_2 (0.13 - 0.98wt%, average 0.58wt%), P_2O_5 (0.07 - 0.39wt%, average 0.06wt%) and MnO shows depletion, 0.03 - 0.20wt% (average 0.06wt%). The LOI ranged from 5.10 - 31.50wt%.

Fluctuations in the major oxide composition of the samples may be explained by the observed variation in mineralogy of the samples. High SiO_2/Al_2O_3 ratios of the sandstones are due to enrichment in quartz relative to other minerals; the low SiO_2/Al_2O_3 ratios of the BH shales are due to enriched Al-rich clay minerals relative to quartz. Also, the covariation in SiO_2 and Al_2O_3 in some of the samples suggests that Al- and Si-poor minerals (like opaque minerals) and calcite are causing varied dilution of SiO_2 and Al_2O_3 (Cullers, 2000). The high LOI values coincides with the samples with the highest CaO contents. The Fe-rich sediments are attributed to shallow and high energy conditions (Nagendra, Nagarajan, Bakkiaraj, & Armstrong-Altrin, 2011).

An attempt at the determining chemical boundaries using major oxides revealed 4 different strata boundaries at 33m, 71m, 83m and 125m for the BH well (Figure 3a). The AJ well also has 4 geochemical boundaries, identified from notable excursions of the major oxide geochemical signatures. The boundaries occur at 607m, 997m, 1192m and 1533m (Figure 3b). The geochemical boundaries are closely related to lithologic changes. Sand (quartz, SiO_2), clay (Al_2O_3) and calcareous (CaO) materials dilution allowed for varied chemical compositions.

4.2. Trace Elements Geochemistry

The measured concentrations of the LILE elements Ba and Rb of the BH core show depletion relative to the PAAS and UCC, with average values of 200.46ppm and 27.48ppm respectively. But the strontium (Sr) content is fairly enriched in the topmost sample and in the lower parts of the core. The measured values for Cr, V, Sc, Co and Ni varied widely with the middle parts showing enrichment, while the upper 25m have values below PAAS and UCC. HFSE elements Zr, Th, Y, Nb and U behaved in an analogous trend to the ferromagnesian trace elements, with lesser values in the upper 25m and higher values in the middle – lower parts of the well. The U and Y concentrations have averages of 2.18ppm and 16.22ppm respectively, with the topmost sample and the lower parts recording the highest values. The concentrations of thorium (Th) ranged from 1.4 – 15.9ppm and the average concentration of Zr is 105.48ppm. Nb shows slight enrichment with an average of 17.47ppm.

In the samples of the AJ well, the LILE element barium (Ba) was anomalously high with a minimum value of 987ppm to values greater than 50,000ppm and the recorded concentrations of Sr is from 125 – 1187ppm (average 342ppm). The ferromagnesian trace elements show some variation Cr (23-162ppm), Ni (21-130ppm), Sc (2-17ppm) and vanadium (V) ranged from 17 -292ppm with an average that is higher than that of the UCC (109.42ppm). The immobile trace elements showed wide variations with Zr content ranging from 75-359ppm (average of 194.81ppm), Th (2.7 – 12.6ppm), Y (7 – 28ppm, average of 17.54ppm) and U has an average of 2.29ppm. Although, there is no clear evidence of fractionation, when compared with UCC. The average Nb concentration was only slightly higher than the UCC.

Generally, the trace element concentrations of the upper parts of the BH core show fractionation and have positive correlations with Al_2O_3. This suggests aluminosilicate source for the immobile HFSE elements and the ferromagnesian elements. There is generally no significant fractionation in the HFSEs of the AJ samples. Zr is slightly positively correlated with Al_2O_3, while Th and Y show strong positive correlations with Al_2O_3, this also suggest aluminosilicate input (Ogbahon & Olujinmi, 2019). Ba is negatively correlated and Rb is weakly correlated with Al_2O_3 which debunks aluminosilicate association for these sediments. The excessive enrichment of Ba, when compared with the PAAS is attributed to high organic contents and paleo-productivity (Ali et al., 2014) as most of the shale samples are dark coloured.

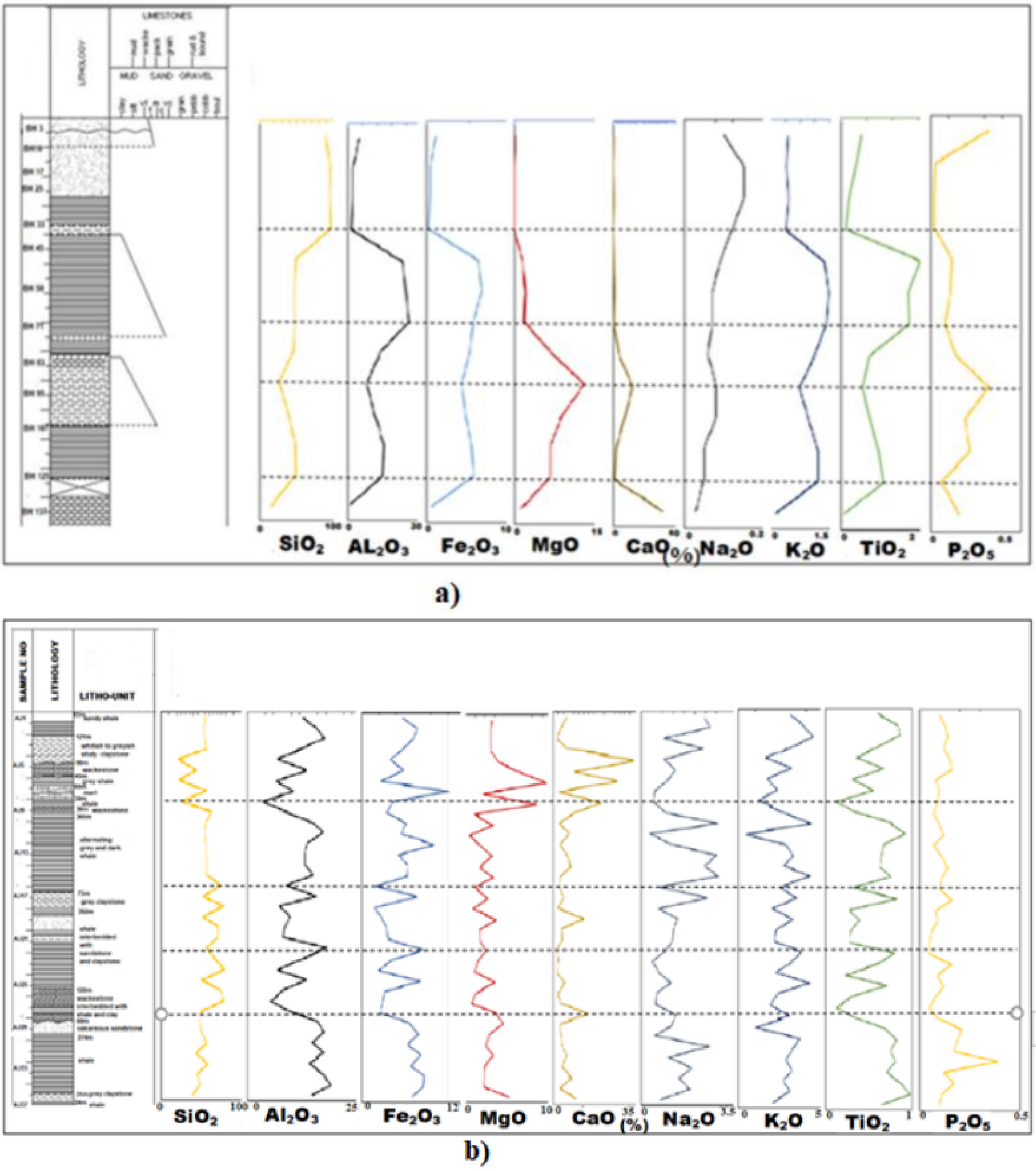

Figure-3. Major Oxide Geochemical Signatures of the Studied Sections **a)** Four Geochemical Boundaries Indicated by the Dotted Lines were Identified in BH well at 33m, 71m, 83m and 125m. **b)** In the AJ Well, Four Boundaries were Established at AJ9, AJ17, AJ21 and AJ28 corresponding to 607m, 997m, 1192m and 1533m respectively.

The plot of the geochemical distributions of the trace elements in the BH well, suggests five chemostratigraphic boundaries at 10m, 25m, 71m, 107m and 125m (Figure 4a). The AJ well displayed noticeable chemical perturbation at around 607m, 1046m and at 1485m (Figure 4b). The distinguishable boundaries are close and similar to the previously identified major oxide chemostratigraphic boundaries.

4.2. Paleoredox Proxies

In marine environments, some trace elements may become authigenically enriched or depleted in the sediments in response to the prevailing oxygenation level during deposition and the various geochemical processes attendant to the environments. The variations in the concentration of these elements and some redox-sensitive chemical elemental ratios serve as proxies to the prevalent paleo-redox conditions in both modern and ancient marine sediments (Madhavaraju et al., 2015). Significant paleoredox geochemical proxies include U, V, Mo, Co, Ni, V/Cr, V/Sc, U/Th, Th/U, V/(V+Ni) and Ni/Co (Ferriday & Montenari, 2016; Jones & Manning, 1994; Madhavaraju et al., 2015). In a reducing marine environment, there is positive excursion of authigenic U principally in the sediment and not in the water column, because uranium is depleted from the water column and it is added to the sediment, while low U contents are ascribed to sediments deposited in oxygenated marine environments (Madhavaraju et al., 2015). The U content in the uppermost sample (BH3) of well BH is higher (8.7ppm) than in the other parts of the well where there was a downward shift to about 1ppm in BH10 – BH25 and to around 3ppm from BH33 downhole (Figure 3a). The values of the U content in the AJ well, is higher in the upper parts of the well, but there was a negative perturbation at AJ18 which extends to AJ27. The lower portions also recorded a positive shift close to 3ppm. Marine sediments deposited under oxic conditions are associated with low concentrations of U, whereas sediments from the anoxic to euxinic environments are high in U content (Jones & Manning, 1994; Madhavaraju et al., 2015).

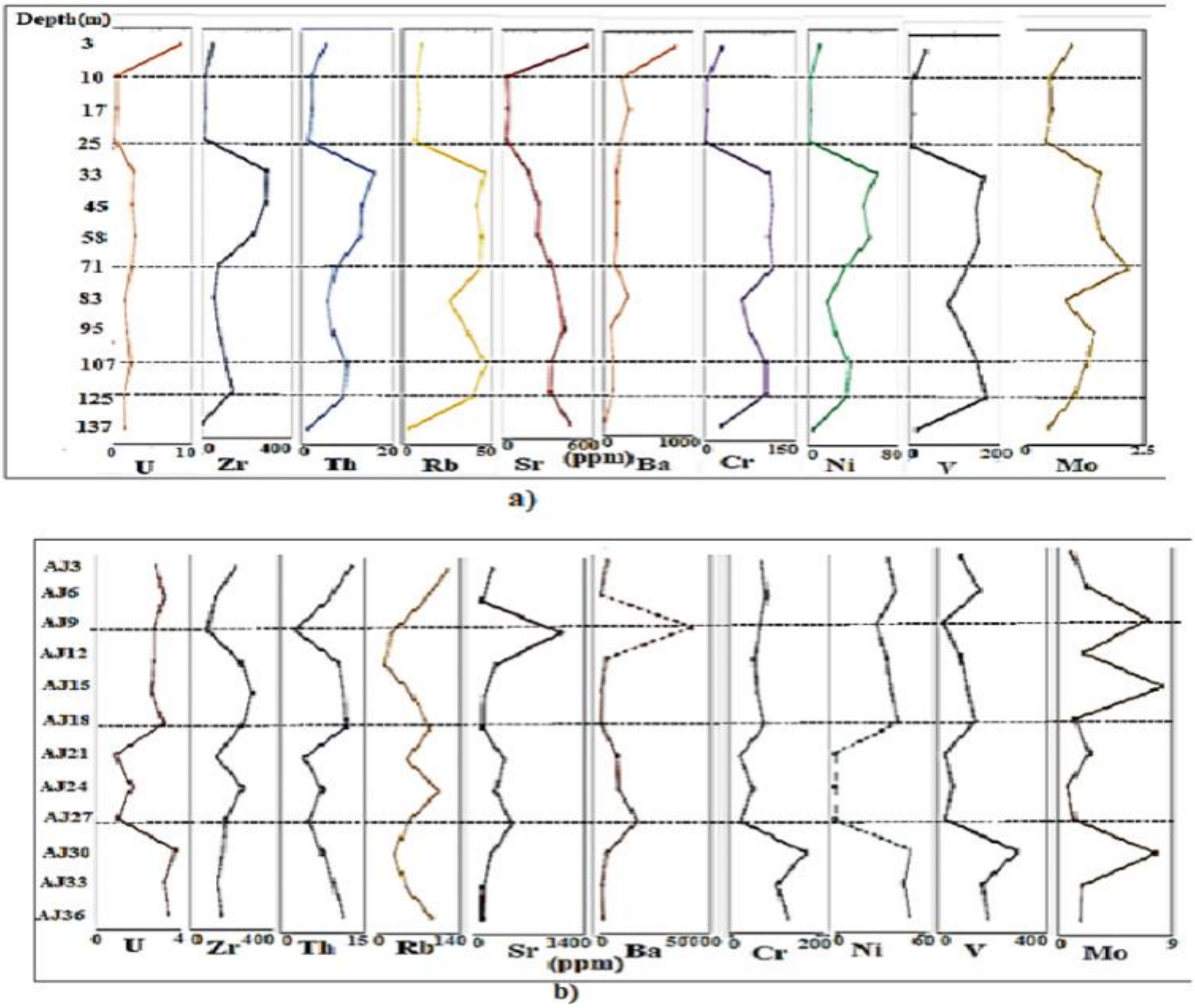

Figure-4. Trace Element Geochemical Signatures of the Studied Sections **a)** Five Geochemical Boundaries Indicated by the Dotted Lines were Identified in BH well at 10m, 25m, 71m, 107m and 125m. **b)** In the AJ Well, Three Boundaries were Identified at AJ9, AJ18 and AJ27 corresponding to 607m, 1046m and 1485m respectively. The Dotted Lines in Ba and Ni are Points Where Concentrations were Below Detection Limits.

The observed concentrations of U in the upper 10 – 25m points to oxic conditions while the lower parts suggest reducing conditions. The upper 1046m and the depths below 1485m of the AJ well point to anoxia, but increased oxygenation levels were observed in the interval between 1046 – 1485m.

Positive excursions of vanadium (V) indicate oxic environments, as vanadium is relatively insoluble in oxic marine environment, which readily bounds to the sediments by surface adsorption processes and therefore accords a high concentration of V in the sediments (Madhavaraju et al., 2015). The V content in the upper 33m of well BH recorded lesser values (6-37ppm) than in the lower depths with higher concentrations (88 – 171ppm) except for sample BH 137, which also has a low V content (18ppm). The AJ samples showed some variations in V content, with intervals below AJ27 having high V content (162 -292ppm, Figure 4b). In contrast to redox deductions from uranium concentrations, the V contents in BH well suggest anoxia in the upper parts, and oxic conditions prevailed in the depths below 25m. The observed variations in the V content in certain intervals of the AJ well is due to variations in oxygenation conditions (oxic to suboxic/anoxic).

Enrichment in concentrations of Molybdenum (Mo) is indicative of marine deposition under anoxic conditions, while low concentrations have a lot to do with increased oxygenation. The concentration of Mo in the BH core samples are low in the upper 25m and in sample BH137. The lower intervals recorded higher Mo concentrations. The AJ well samples show relatively consistent concentrations in Mo content (0.77 -2.55ppm), while samples AJ9, AJ15 and AJ30 have the highest values 7.11ppm, 8.34ppm and 7.71ppm respectively. Molybdenum is one of the most reliable diagnostic elements for sediment deposition under reducing conditions (Madhavaraju et al., 2015). The lower shaly portions of the BH well suggest prevalence of reducing conditions, and likewise, a drop in oxygenation levels were deduced for certain depths in the AJ well where Mo contents were high.

Geochemical ratios, U/Th and Th/U are good paleoredox proxies in marine environments, which relies on the variability of U concentrations in detrital fractions under changing oxygenation conditions, while Th remains unperturbed by redox fluctuations (Fertl, 1979; Jones & Manning, 1994). A high U/Th ratio (>1.25) suggests an anoxic environment, >0.75 – ≤ 1.25 is dysoxic, low values (<0.75) are associated with oxic environments, whereas a Th/U value of ≤ 2 indicates anoxia (Ferriday & Montenari, 2016; Madhavaraju et al., 2015). The U/Th ratios of BH samples are within the oxic range (0.15 – 0.31, Figure 5), except sample BH3 and BH 137 which were above 0.75. The Th/U ratios of these samples also show a similar trend and fall in the oxic field except BH3 and BH137. The U/Th ratios of AJ samples are likewise low (0.18- 0.43) except sample AJ9 whose value is 0.89. The Th/U for the AJ samples are above 2 (Figure 6), therefore oxic conditions of deposition were inferred.

Different ratios also exploit the response of V to oxygenation levels to better understand the dominant paleoredox conditions during deposition in marine sediments. A V/Cr ratio ≤ 2 indicates oxic conditions, >2 to ≤ 4.25 dysoxic and > 4.25 represents anoxic conditions. Contrastingly, low Ni/V ratios are interpreted as reducing conditions, while high Ni/V indicate oxic conditions. A V/Sc ratio of ≥ 9.1 suggests anoxia whereas lesser values indicate elevated oxygenation levels. The V/(V+Ni) is also a diagnostic redox ratio, where values ≤0.46 are interpreted as oxic, while > 0.84 represents euxinic conditions. The values between 0.46 and 0.84 are dysoxic to anoxic (Jones & Manning, 1994; Rimmer, 2004). The V/Cr ratios for BH samples were < 2, except BH25, that is within the dysoxic field (Figure 5d). However, the V/Sc, Ni/V, V/(V+Ni), all indicate dysoxic to anoxic conditions of deposition (Figure 5e,5f and 5g). The V/Cr ratios of the sediments from AJ well, largely indicated elevated oxygenation levels (Figure 6d), but varied oxygenation levels across the well was indicated by V/Sc ratios (Figure 6e). The V/(V+Ni) and Ni/V ratios reflect a dissimilar paleoredox condition, plotting mostly in the suboxic – anoxic fields (Figure 6f and 6g).

Oxygenation levels for Ni/Co ratios > 5 are suggestive of suboxic/anoxic environments, while Ni/Co ratios < 5 are indications of oxic conditions (Ferriday & Montenari, 2016; Jones & Manning, 1994). All the samples from the BH and and AJ wells have Ni/Co ratios < 5 except samples BH137 and AJ9. In general, the Ni/Co ratios for the studied sections in both wells point to an oxic environment of deposition, aside samples BH137and AJ9.

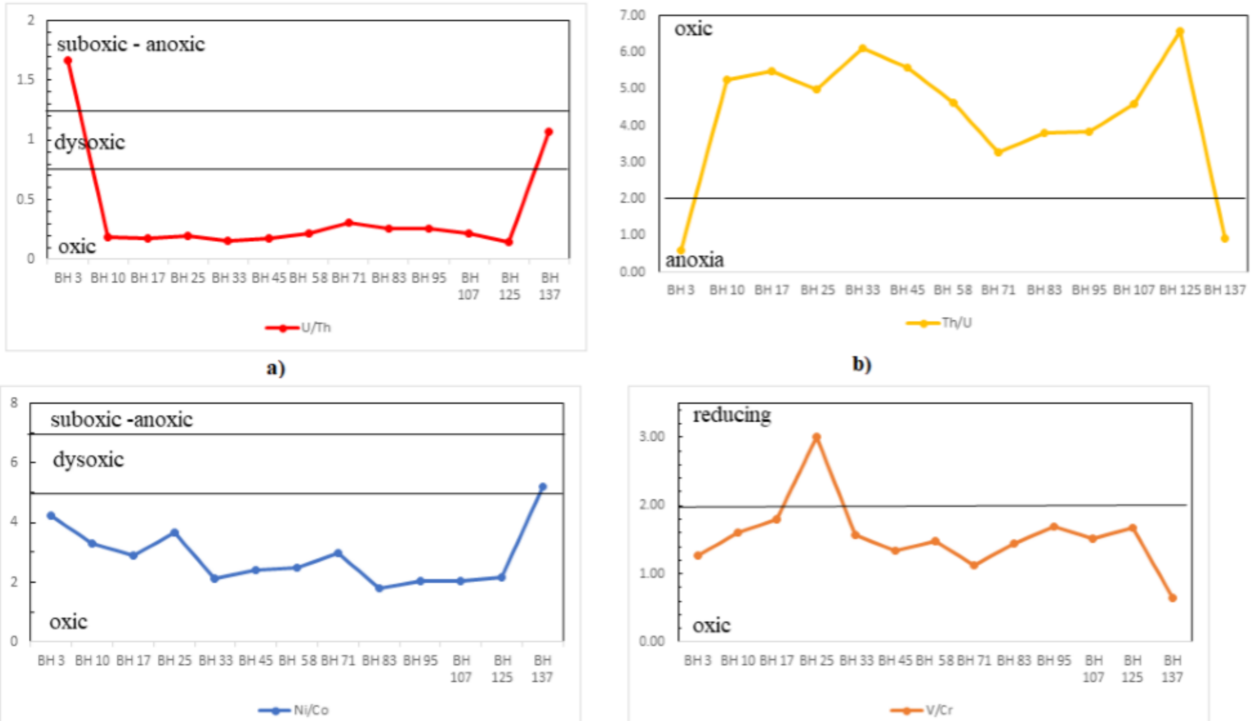

Figure-5. Redox Sensitive Trace Element Ratios in BH well. A high U/Th ratio (>1.25) suggests an anoxic environment, >0.75 – ≤ 1.25 is dysoxic, low values (<0.75) are associated with oxic environments, whereas a Th/U value of ≤ 2 indicates anoxia. A V/Cr ratio ≤ 2 indicates oxic conditions, >2 to ≤ 4.25 dysoxic and > 4.25 represents anoxic conditions. Contrastingly, low Ni/V ratios are interpreted as reducing conditions, while high Ni/V indicate oxic conditions. A V/Sc ratio of ≥ 9.1 suggests anoxia whereas lesser values indicate elevated oxygenation levels. The V/(V+Ni) is also a diagnostic redox ratio, where values ≤0.46 are interpreted as oxic, while > 0.84 represents euxinic conditions. The values between 0.46 and 0.84 are dysoxic to anoxic. **a)** U/Th, **b)** Th/U, **c)** Ni/Co and **d)** V/Cr Ratios Largely Point to Prevailing Oxic Conditions in Most parts of the Well.

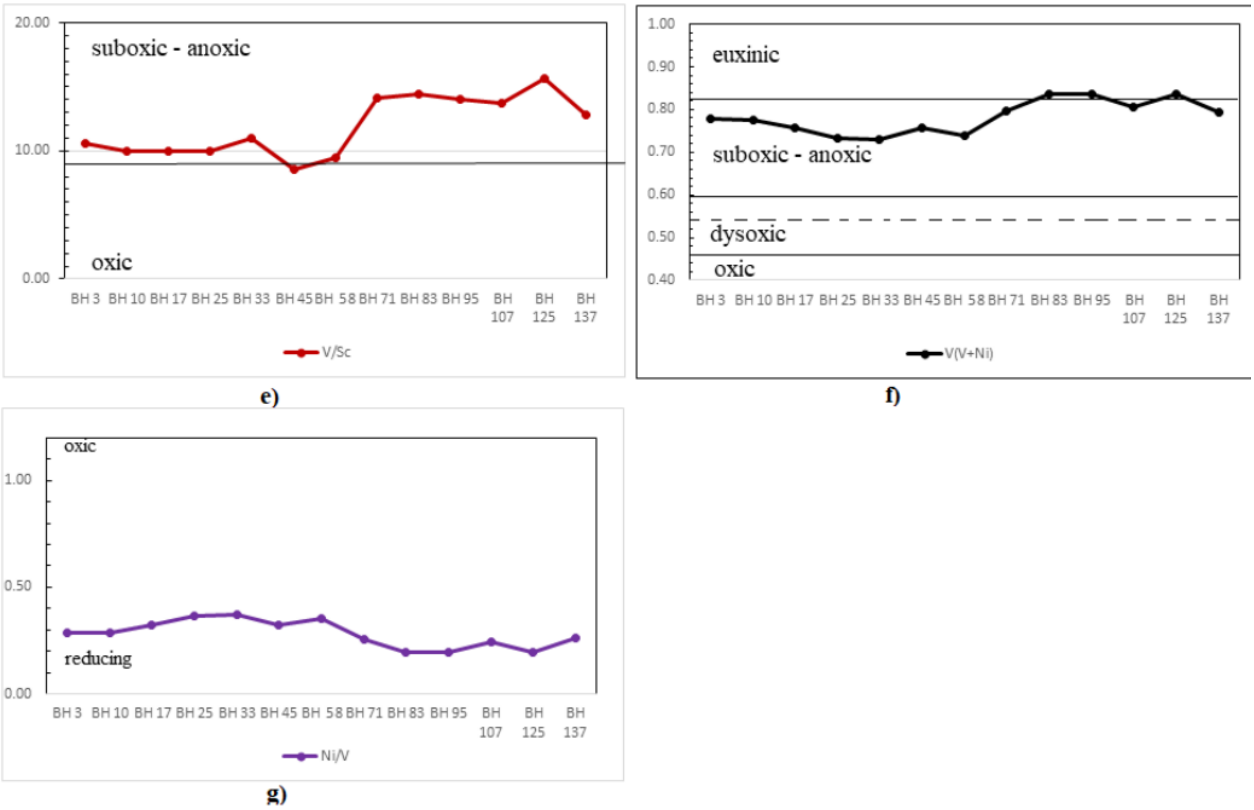

Figure-5(contd.): **e)** V/Sc, **f)** V/(V+Ni) and **g)** Ni/V Ratios Suggests Suboxic – Anoxic Conditions of Deposition.

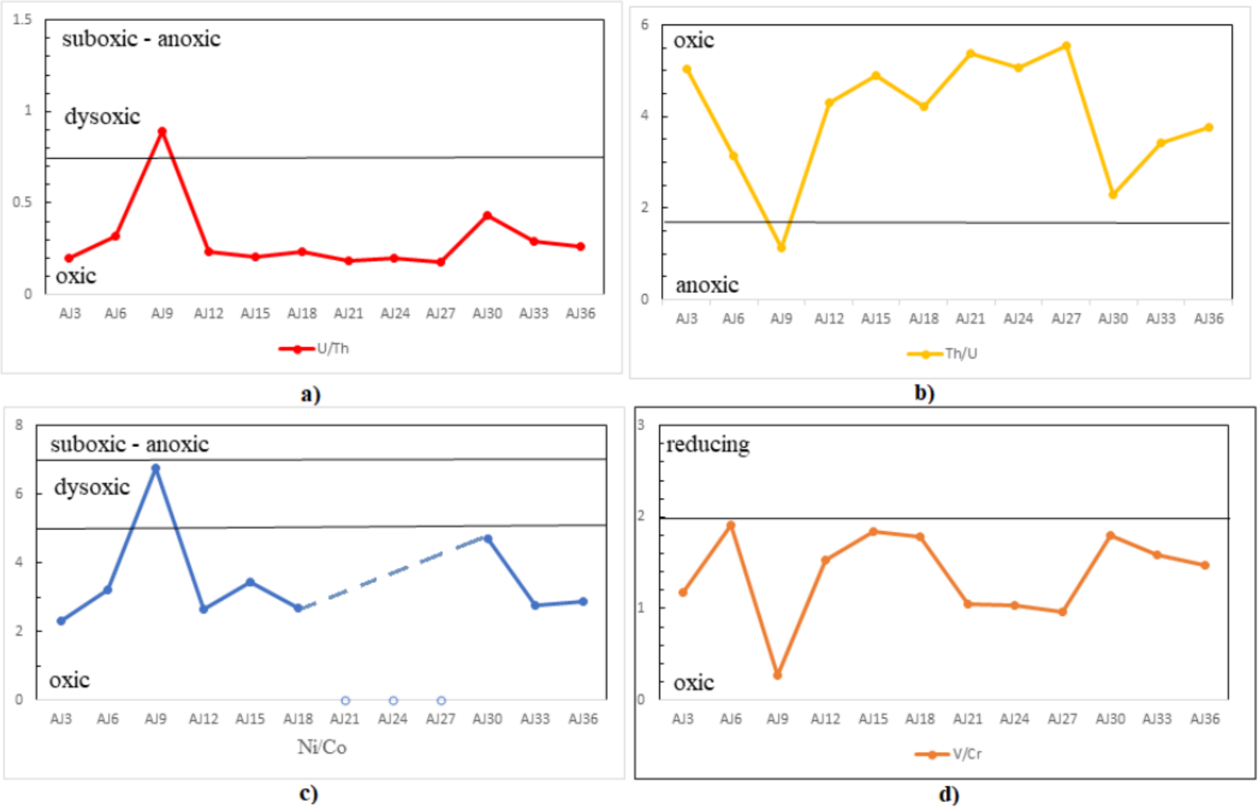

Figure-6. Redox Sensitive Trace Element Ratios in the AJ Well **a)** U/Th, **b)** Th/U, **c)** Ni/Co and **d)** V/Cr Ratios Largely Point to Prevailing Oxic Conditions in Most parts of the Well Except in Sample AJ9.

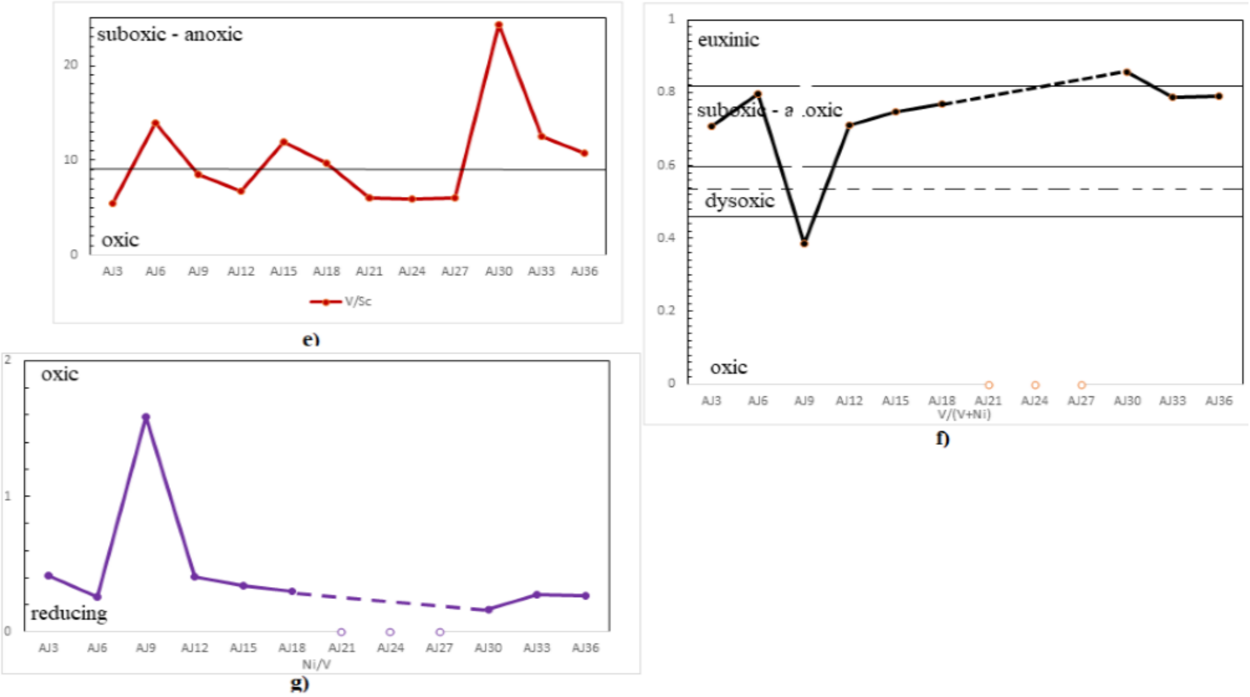

Figure-6. (contd.): e) V/Sc, **f)** V/(V+Ni) and **g)** Ni/V Ratios Suggests Dysoxic – Anoxic Conditions of Deposition.

5. CONCLUSIONS

The plot of the geochemical signatures of major oxides concentrations suggests that, there are at least four chemostratigraphic boundaries in the BH and AJ wells. However, five and three boundaries were identified for the BH and AJ wells respectively, from the trace element geochemistry. These boundaries are related to lithostratigraphic changes. The recorded values of U and Mo in the upper 25m of the BH well suggest prevalence of oxic conditions, but suboxic – anoxic conditions in the lower portions of the core. This inference was supported by V/Sc, Ni/V and V/(V+Ni) ratios, all pointing to extensive dysoxic – anoxic conditions. Conversely, the interpretation from paleoredox proxies, V, U/Th, Th/U, V/Cr indicates dominance of oxic conditions, and this information contradicts the physical observations of most of the shaly strata, which lack bioturbations. Dysoxic – anoxic conditions were inferred for most intervals of the AJ well using trace element proxies of U, V, V/Sc, Ni/V and V/(V+Ni), and oxic conditions the interval between, 1046m – 1485m (AJ21 -AJ27). Although, the interpretation from the Mo, U/Th, Th/U, V/Cr and Ni/Co values differs, as they all point to prevailing oxic conditions. The disparity in the deductions from paleoredox proxies, U/Th, Th/U, V/Cr and Ni/Co may be related to preferential fractionation and post-depositional processes in the environments of deposition.

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