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SAMARIUM-NEODYMIUM AND RUBIDIUM-STRONTIUM ISOTOPES OF THE TAMALE AND OTI GROUPS, VOLTAIAN SUPERGROUP, GHANA: IMPLICATIONS FOR PROVENANCE STUDIES

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Abstract

Isotope geochemistry, specifically the Samarium-Neodymium (Sm-Nd) and Rubidium-Strontium (Rb-Sr) systems, has emerged as a robust tool for tracing sediment origins. By comparing the isotopic composition of sediments to potential source regions, geologists can quantify the contributions of different source terranes. This abstract highlight the application of Sm-Nd and Rb-Sr isotopes in provenance studies of the Oti-Pendjari and Tamale-Obosum Group rock formations of the Voltaian Supergroup. Thirty (30) fine sediment samples from the two groups were collected and analyzed for their whole rock elemental compositions using the ICP-MS and XRF. Nd and Sm isotope measurements were conducted using a low-resolution mode on a Nu Plasma II MC-ICPMS (Nu Instruments, Wrexham) located at the Analytical Facility of the University of Johannesburg. Preliminary results indicate distinct isotopic signatures between the Tamale-Obosum Group and the Oti-Pendjari Group. Nd and Sr isotopic analysis of the Tamale and Oti Groups indicates a continental crustal origin for both units. TDM model ages for these groups range from 1.31 to 2.2 Ga, suggesting a dominant Proterozoic crustal source. Specifically, the Tamale-Obosum Group likely sourced from a combination of Birimian and Kwahu-Bombouaka rocks, while the Oti-Pendjari Group likely sourced from a mix of Birimian rocks and rocks from the Pan-African belt.

Keywords
Voltaian Supergroup, Sm-Nd isotopes, Rb-Sr isotopes, provenance, Neoproterozoic, West Africa.

Introduction

Geologists leverage the distinct isotopic signatures of strontium (Sr) and neodymium (Nd) in sediments to trace their origin (provenance) and understand how sediment delivery to basins has changed over time and space (Krom et al., 1999). This technique is powerful because these isotope ratios act like unique fingerprints for different rock types. Additionally, Sr and Nd isotopes can also offer valuable insights into past environmental conditions (Baioumy, 2011; Fu et al., 2012; Weldeab et al., 2003).

The isotopic compositions of Nd and Sr in sediments, specifically the radiogenic variants, have been established as valuable tools for comprehending Earth surface processes. These isotopes remain unaffected by biological fractionation or thermodynamic processes, which could potentially introduce biases to nutrient-based tracers (e.g., Frank (2002)). The varying Nd and Sr isotope compositions found in continental rocks are dependent on age and rock type, enabling the differentiation between regional and local sources of detrital silicates originating from continents and settling on the ocean floor. Neodymium isotopes also function as tracers for water masses due to the distinctive Nd isotope composition characteristic of different sources.

Traditionally, geologists have used the Sm-Nd isotope system to date rocks within the Earth's crust. This approach helps us understand how these rocks formed (petrogenesis) and how the composition of the mantle has changed over time (DePaolo, 1981; Hamilton et al., 1978; Nelson and DePaolo, 1984; O'nions et al., 1983; O'Neill and Palme, 2008). By analyzing the ratios of specific Sm and Nd isotopes in a rock, we can calculate a "model age". This age reflects the time that has passed since the rock first separated from the mantle (McCulloch and Wasserburg, 1978).

However, sedimentary rocks are different. They form from the accumulation of eroded bits and pieces of older rocks with varying ages. Therefore, the Sm-Nd model age of a sedimentary rock doesn't tell us its actual age (stratigraphic age) but rather the average age of the material in its source region. This information is crucial for understanding the origin (provenance) of the sediment. Recognizing this, researchers now widely use the term "dprovenance ageT" in provenance studies of sediments across various geological regions (Dalland et al., 1995; Goldstein et al., 1984; Knudsen, 2001; McCulloch and Wasserburg, 1978; Mearns, 1989, 1992; Mearns et al., 1989; Nelson and DePaolo, 1988).

Research employing geochemical methods, such as those by Anani et al. (2012), has shed light on the provenance of the Anyaboni sandstones. By utilizing Sm-Nd and other techniques, these studies identified a mixed provenance, with contributions from granitic, gneissic, and sedimentary rocks, largely associated with the Birimian Supergroup and additional input from Pan-African rocks. Further exploration, as documented in the study Provenance of the Anyaboni Formation of the Voltaian Supergroup in Ghana, has enriched the understanding of the Anyaboni Formation's source materials. Moreover, broader studies on the Voltaian Supergroup, including its shales and siltstones within the Kwahu and Oti Groups, have highlighted felsic sources (e.g., granites and granodiorites), recycled sediments, and andesitic input

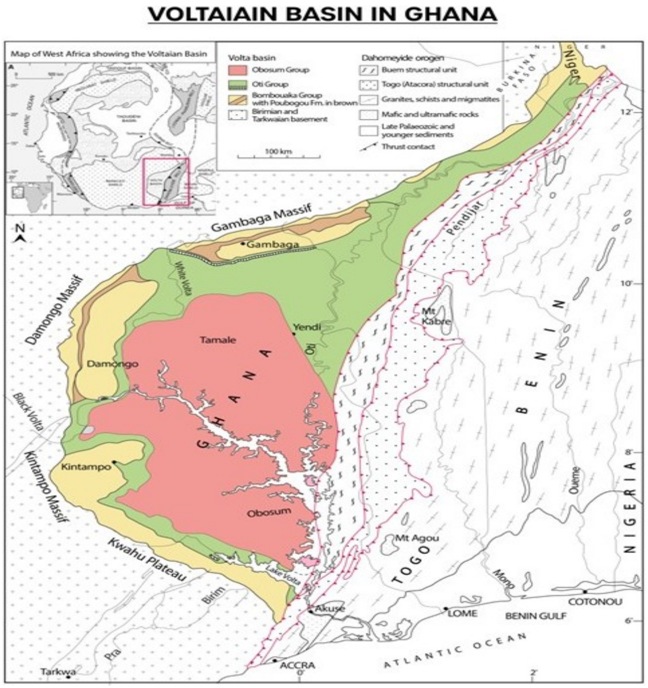

Figure 1. Geological sketch map of West Africa showing the Voltaian Basin (after Carney et al. (2010)).

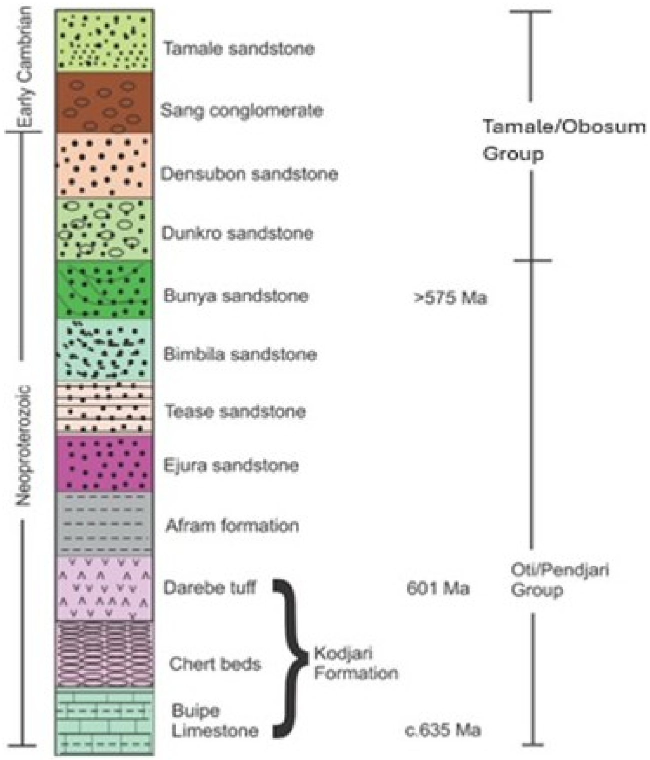

Figure 2. Stratigraphy of the Tamale-Obosum and Oti-Pendjari Groups of the Voltaian Supergroup, Ghana, based on field observations. Ages deduced from Carney et al. (2010)

through geochemical analysis. Detrital zircon geochronology has provided constraints on the basin's provenance and stratigraphic correlations, revealing influences from both the West African and Amazonian cratons. Comparative research has also demonstrated distinctive provenance characteristics among the Kwahu-Bombouaka, Oti-Pendjari, and Tamale-Obosum Groups, with the Bombouaka Group being the oldest among them. Although previous research has contributed to our knowledge on the provenance of sections of the Kwahu and Oti Groups, there remains a gap in our knowledge of the provenance of the Oti and Tamale Groups based on their radiogenic isotopic compositions. In this study, we present the first comprehensive radiogenic isotopic analysis of siliciclastic rocks from the Tamale-Obosum and Oti-Pendjari Groups within Ghana's Voltaian Basin. Our goal is to gain a better understanding of the source of these sediments with the aid of Sm-Nd and Rb-Sr isotopes.

Geology of the study area

The Neoproterozoic Voltaian Basin, a smaller counterpart of West Africa's sedimentary cover sequences, occupies the eastern margin of the Man-Leo Shield. This ~5 km-thick basin comprises a diverse suite of sandstones, mudstones, and subordinate limestones as shown in Figure 1 (Affaton, 1990; Affaton et al., 1980; Annan-Yorke and Cudjoe, 1971; Bozhko, 2008; Junner and Hirst, 1946). The Voltaian Supergroup has been traditionally divided into three groups, although variations in nomenclature exist between Francophone and Anglophone authors. The lowermost Kwahu-Bombouaka (or Dapango-Bombouaka) Group, predominantly composed of coarse-grained sandstones, is unconformably overlain by the Oti- Pendjari Group, characterized by shales, greywackes, tilloids, and limestones (Figure 2). The uppermost Tamale-Obosum Group consists primarily of quartzitic and feldspathic sandstones (Carney et al., 2010). The formations constituting the two Groups are represented in the stratigraphic log (Figure 2).

The Voltaian Supergroup's evolution is closely linked to the Pan-African Dahomeyide orogeny. The Kwahu-Bombouaka and Oti-Pendjari Groups are interpreted as continental and passive margin deposits, respectively, associated with the opening of a Pan-African Ocean. The Tamale-Obosum Group, on the other hand, is considered molasse-type deposits formed after collision and uplift during the orogeny (Kalsbeek et al., 2008). Correlations have been drawn between the Voltaian Supergroup and other units in the region. The quartzites of the Togo unit are correlated with the lower parts of the Voltaian succession, and parts of the Buem unit with the middle Voltaian Oti-Pendjari Group. However, the correlation of the upper clastic division of the Voltaian Supergroup remains uncertain.

Materials and Methods

A total of thirty (30) sandstones and siltstones from the Tamale-Obosum and Oti-Pendjari Groups of the Voltaian Supergroup were collected for whole rock geochemical analysis (Figure 3). The samples underwent washing, drying, and pulverization before being analyzed at the WIGL of the University of Witwatersrand, Johannesburg. Elemental compositions of Rb, Sr, Nd, and Sm were determined using ICPMS and XRF. The analysis process involved spiking samples with Sm-Nd and Rb-Sr, followed by rigorous chemical digestion. Initial digestion included treatment with concentrated HF and HNO_3, followed by heating on a 110°C hotplate for one week and overnight drying. Additional steps involved treating the samples with concentrated HNO_3, HCl, and H_2O_2, followed by further heating and drying. A final digestion step used 6 M HCl, with extended heating for up to 2 days, ensuring complete digestion. For Rb and Sr separation, 2 mL of BioRad AG50 x8 resin was used. The samples were spiked, centrifuged, and treated with 1.5 M HCl before being conditioned, loaded, and eluted. Following elution, samples were dried and rinsed with concentrated HNO_3 to remove resin residues. LREE separates were prepared by adding 0.3 mL of 0.2 M HCl. For Sm and Nd separation, Eichrom Ln Spec resin was used. The samples underwent similar conditioning, loading, and rinsing steps, followed by separate elution and drying of Sm and Nd fractions. These dried samples were prepared for dilution and analysis. Nd and Sm isotope measurements were conducted using a Nu Plasma II MC-ICPMS at the University of Johannesburg's Analytical Facility. Samples were introduced into a 2% HNO_3 solution using a Glass Expansion MicroMist nebulizer, with plasma settings of 1300W RF power, 13.0 L/min coolant gas flow, and 0.80 L/min auxiliary gas flow. Nd and Sr analyses involved 50 cycles, while Sm and Rb were measured over 20 cycles. Background measurements were taken for each element before the measurements and subtracted. Data reduction used a Microsoft Excel spreadsheet, with mass bias corrections applied according to the exponential law. Nd measurements were corrected using a $^{146}Nd/^{145}Nd$ ratio of 2.0719425, Sm with a $^{152}Sm/^{147}Sm$ ratio of 1.78307, Sr using a $^{88}Sr/^{86}Sr$ ratio of 8.375209, and Rb with a $^{85}Rb/^{87}Rb$ ratio of 2.59376. The JNdi-1 standard was analyzed after every four samples. The analysis of USGS reference material BCR-2 yielded a $^{143}Nd/^{144}Nd$ ratio of 0.512612 ± 04 (1SD, n=4), consistent with the recommended value of 0.512635 ± 29 (GeoRem).

Chondritic Uniform Reservior (CHUR) model ages were calculated from the present-day $^{147}Sm/^{144}Nd$ and $^{143}Nd/^{144}Nd$ of the Tamale-Obosum and Oti-Pendjari Group samples using the equation:

$$T_{\mathrm{CHUR}}^{\mathrm{Nd}} = \frac{1}{\lambda} \ln \left[\frac{\left(\frac{^{143}\mathrm{Nd}}{^{144}\mathrm{Nd}}\right)_A - \left(\frac{^{143}\mathrm{Nd}}{^{144}\mathrm{Nd}}\right)_B}{\left(\frac{^{147}\mathrm{Sm}}{^{144}\mathrm{Nd}}\right)_A - \left(\frac{^{147}\mathrm{Sm}}{^{144}\mathrm{Nd}}\right)_B} + 1 \right] \tag{1}$$

where λ is the decay constant for ^{147}Sm to ^{143}Nd; A – sample today; B – CHUR today

In using $^{143}Nd/^{144}Nd_{CHUR}$ = 0.51263, λ = 6.54E-12, ^{147}Sm /$^{144}Nd_{CHUR}$ = 0.196 from Bouvier et al. (2008). T_{DM}(depleted mantle) model ages were also calculated by substituting the appropriate DM (depleted mantle) values in place of (^{143}Nd/ $^{144}Nd)_{CHUR}$ and ($^{147}Sm/^{144}Nd)_{CHUR}$ in the above equation. Parameters used in calculating TDM are those of Goldstein et al. (1984); (^{143}Nd/ ^{144}Nd)DM today = 0.51316, (^{147}Sm/ ^{144}Nd)DM today = 0.215 and the decay constant l = 6.54 x 10-12yr-1.

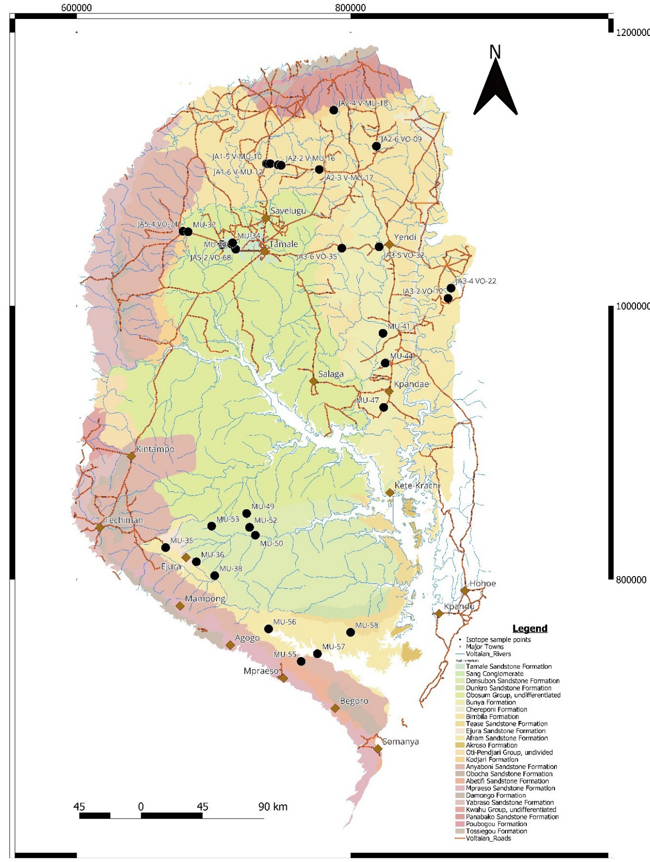

Figure 3. Map of the Voltaian basin in Ghana showing sample location points.

Epsilon Nd (εNd) quantifies the isotopic deviation of neodymium (Nd) in a sample relative to the chondritic uniform reservoir (CHUR). This parameter, derived from the Sm-Nd isotopic system, is a valuable tracer of geological materials' origins and evolutionary histories. Positive εNd values signify mantle-derived, depleted Nd isotopic compositions, while negative values indicate crustal, enriched compositions. The fractionation factor (fSm/Nd), analogous to εNd, measures Sm/Nd enrichment relative to CHUR.

It is often written as fSm/Nd and is calculated from the expression:

$$f = \left[\left(\frac{^{147}\mathrm{Sm}}{^{144}\mathrm{Nd}}\right)_{\mathrm{today}} \Big/ \left(\frac{^{147}\mathrm{Sm}}{^{144}\mathrm{Nd}}\right)_{\mathrm{CHUR}} - 1 \right] \tag{2}$$

A positive f Sm/Nd value, such as the +0.09 adopted for some depleted mantle models, implies a 9% higher Sm/Nd ratio compared to CHUR, resulting in progressively positive εNd values over time.

Table 1. Sm–Nd isotopic compositions of the Tamale-Obosum and Oti-Pendjari Group of rocks

Formation	Nd (ppm)	Sm (ppm)	$^{147}Sm/^{144}Nd$	$^{143}Nd/^{144}Nd$	Error (2s)	εNd	TCHUR	TDM	fSm/Nd
Bunya	14.31	2.77	0.11698	0.51227	0.000005	-6.98	0.69	1.41	-0.4559
Bunya	312.18	50.28	0.09735	0.51217	0.000007	-8.96	0.71	1.28	-0.5472
Bunya	38.38	7.24	0.11409	0.51220	0.000006	-8.29	0.79	1.44	-0.4694
Bunya	19.87	3.90	0.11849	0.51188	0.000005	-14.70	1.48	2.02	-0.4489
Bunya	198.20	33.70	0.10277	0.51210	0.000007	-9.33	0.87	1.44	-0.5220
Bunya	24.03	4.52	0.11363	0.51215	0.000006	-10.38	0.89	1.51	-0.4715
Bimbila	57.60	10.24	0.10752	0.51217	0.000009	-8.00	0.79	1.41	-0.4999
Bimbila	29.16	5.53	0.11472	0.51219	0.000007	-9.74	0.83	1.50	-0.4664
Bimbila	38.08	6.85	0.10881	0.51216	0.000006	-9.58	0.83	1.44	-0.4939
Bimbila	21.12	4.00	0.11445	0.51173	0.000007	-8.01	1.68	2.20	-0.4677
Bimbila	5.98	1.09	0.11059	0.51174	0.000006	-7.87	1.58	2.10	-0.4856
Bimbila	31.32	6.06	0.11690	0.51222	0.000004	-9.25	0.79	1.51	-0.4563
Bimbila	28.56	5.55	0.11741	0.51213	0.000006	-17.60	0.97	1.60	-0.4539
Bimbila	35.24	6.52	0.11181	0.51214	0.000006	-17.31	0.89	1.51	-0.4800
Bimbila	32.02	6.10	0.11519	0.51222	0.000005	-15.05	0.78	1.43	-0.4642
Bimbila	43.87	8.06	0.11112	0.51223	0.000005	-12.87	0.73	1.41	-0.4831
Afram	14.61	2.96	0.12259	0.51197	0.000004	-15.49	1.35	1.96	-0.4298
Afram	16.61	2.62	0.09550	0.51186	0.000005	-15.01	1.21	1.66	-0.5558
Tease	2.66	0.49	0.11127	0.51184	0.000005	-8.72	1.43	1.94	-0.4825
Ejura Ss	4.79	0.86	0.10923	0.51186	0.000005	-11.82	1.35	1.87	-0.4919
Kodjari	65.65	8.75	0.08059	0.51218	0.000004	-10.82	0.59	1.11	-0.6252
Kodjari	29.79	5.16	0.10478	0.51202	0.000006	-14.53	1.01	1.61	-0.5127
Sang Cong.	37.56	7.44	0.11971	0.51208	0.000007	-14.92	1.11	1.73	-0.4432
Tamale	13.57	2.53	0.11286	0.51189	0.000005	-15.14	1.41	1.91	-0.4751
Tamale	7.24	1.07	0.08912	0.51187	0.000007	-13.38	1.10	1.60	-0.5855
Tamale	10.43	1.92	0.11142	0.51185	0.000004	-11.39	1.41	1.92	-0.4818
Undivided Obosum	10.74	1.96	0.11028	0.51194	0.000004	-8.94	1.22	1.81	-0.4871
Undivided Obosum	18.48	3.87	0.12645	0.51205	0.000004	-8.61	1.31	1.91	-0.4119
Densubon	21.61	3.89	0.10881	0.51209	0.000003	-9.61	0.94	1.53	-0.4939
Densubon	52.65	8.38	0.09621	0.51214	0.000004	-19.92	0.75	1.31	-0.5525
Densubon	5.10	0.93	0.10989	0.51161	0.000004	-14.31	1.80	2.24	-0.4889
Densubon	13.46	2.30	0.10328	0.51190	0.000004	-10.49	1.21	1.72	-0.5196

Results and Discussion

Results

The results of the Nd isotopes for the sediments from the Oti-Pendjari and Tamale-Obosum Groups are presented in Table 1. The calculated ε Nd values range from -6.98 to -19.92. The $^{143}Nd/^{144}Nd$ ratios range from 0.5116 to 0.5123 and that of the $^{147}Sm/^{144}Nd$ range between 0.07 to 0.13. The Rb isotopes for both the Tamale-Obosum and Oti-Pendjari Group are also represented in Table 2. The $^{87}Rb/^{86}Sr$ ratios range from 0.29548 to 6.22063 and that of $^{87}Sr/^{86}Sr$ ratios also range from 0.70559 to 0.82003.

Discussion

The continental crust exhibits lower Sm/Nd ratios compared to the depleted mantle, resulting in the production of comparatively lower $^{143}Nd/^{144}Nd$ ratios over time (O'Neill and Palme, 2008; Porter and White, 2009; Stepanov et al., 2014). This is because the Sm and Nd elements are resistant to significant fractionation during metamorphism and sedimentation, allowing the Nd isotope compositions to effectively record the isotopic characteristics of their source materials (Sun et al., 2018; Wei et al., 2002). Additionally, the immobility of these elements under hydrothermal conditions ensures that Nd isotope ratios accurately reflect the proportions of source rocks or magmas involved in petrogenetic processes (Jones et al., 2019; Paul et al., 2002).

However, the Sm-Nd system exhibits limited sensitivity to minor amounts of recycled crustal material within the mantle (Condie, 2003; Hofmann et al., 2022). This is because the Nd isotopic composition of the continental crust is typically lower than that of the depleted mantle, and the incorporation of small amounts of recycled crustal material into the mantle does not significantly alter the overall Nd isotopic signature of the mantle (Chu et al., 2013; Guo and Korenaga, 2020).

A plot of $^{143}Nd/^{144}Nd$ against $^{87}Sr/^{86}Sr$ ratios of the Tamale-Obosum and Oti-Pendjari Groups reveals that the formations from both Groups have sediment sources of continental crustal material. (Figure 4a). The Oti-Pendjari Group sediments exhibit middle continental crustal source signatures and align with seawater signatures suggesting the presence of an ocean interaction at the time of formation of the source material. On

Table 2. Rb-Sr isotopic composition and ratios of the Tamale-Obosum and Oti-Pendjari Groups

Formation	Sr (ppm)	Rb (ppm)	$^{87}Rb/^{86}Sr$	$^{87}Sr/^{86}Sr$	Error (2σ)
Bunya	133.33	76.92	1.67027	0.71775	0.000008
Bunya	119.67	55.06	1.33216	0.71895	0.000008
Bunya	143.57	71.32	1.43862	0.71997	0.000009
Bunya	34.04	67.69	5.81497	0.82003	0.000007
Bunya	257.95	69.69	0.78161	0.71066	0.000008
Bunya	219.47	53.94	0.71112	0.71176	0.000006
Bimbila	211.46	45.05	0.61632	0.71050	0.000008
Bimbila	232.93	58.26	0.72377	0.71225	0.000009
Bimbila	188.37	33.38	0.51264	0.71045	0.000006
Bimbila	67.15	81.98	3.54882	0.75973	0.000007
Bimbila	9.60	6.03	1.81584	0.70559	0.000009
Bimbila	195.44	45.71	0.67664	0.71114	0.000007
Bimbila	175.22	54.22	0.89566	0.71413	0.000006
Bimbila	200.86	54.75	0.78875	0.71262	0.000006
Bimbila	204.85	45.93	0.64877	0.71218	0.000010
Bimbila	212.17	45.48	0.62024	0.71061	0.000007
Afram	16.61	1.69	0.29548	0.72038	0.000006
Afram	29.22	62.23	6.22063	0.80851	0.000010
Afram	5.23	1.05	0.57882	0.71598	0.000010
Ejura	14.99	19.05	3.69615	0.76167	0.000006
Kodjari	78.80	19.02	0.69842	0.71169	0.000006
Kodjari	34.62	39.42	3.30565	0.74519	0.000006
Sang Cong.	257.14	47.24	0.53151	0.71042	0.000005
Tamale	30.16	27.31	2.62931	0.74819	0.000006
Tamale	13.67	3.52	0.74614	0.71915	0.000006
Tamale	25.20	39.88	4.60951	0.78025	0.000005
Undivided Obosum	19.33	11.59	1.73822	0.73351	0.000007
Undivided Obosum	153.83	39.01	0.73409	0.71552	0.000007
Densubon	221.56	48.66	0.63542	0.71171	0.000006
Densubon	157.05	59.79	1.10186	0.71338	0.000009
Densubon	7.75	3.96	1.48379	0.74557	0.000013
Densubon	47.79	97.20	5.93660	0.80096	0.000011

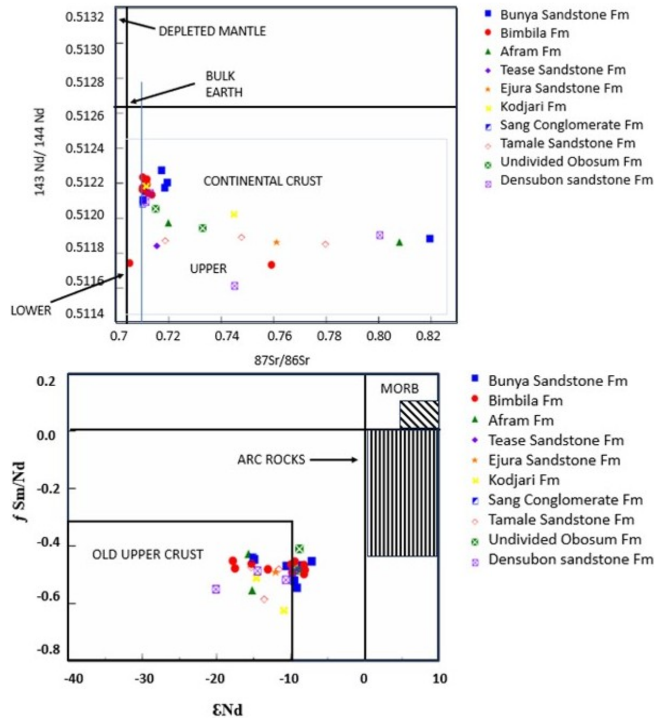

Figure 4. A – Diagrams illustrating a plot of plot of $^{143}Nd/^{144}Nd$ against $^{87}Sr/^{86}Sr$ ratios of the Tamale-Obosum and Oti-Pendjari Groups; B – Diagrams illustrating the Sm-Nd and Rb-Sr isotopic compositions of the Tamale-Obosum and Oti-Pendjari Groups sediments.

the other hand, the Tamale-Obosum samples exhibit higher Sr signatures typical of upper continental crustal material.

Sm-Nd isotopic analyses and model ages for both the Tamale and Oti Groups have been used to assess the source of the sediments. A plot of the fractionation factor fSm/Nd against εNd suggests that both groups originate from an ancient upper crust source (Figure 4b).

Calculated TDM model ages for the Tamale-Obosum Group range from 1.31 - 2.2 Ga and that of the Oti-Pendjari Group ranges from 1.28 – 2.2 Ga. These ages may therefore suggest that both Tamale-Obosum and Oti-Pendjari Groups are possibly derived from a Proterozoic crustal source rock (Figure 2b). With the dominance of Mesoproterozoic and some minor Paleoproterozoic ages, it can be suggested that the possible source of the sediments could be a mix of the Birimian rocks with rocks from the Pan African belt and the older Kwahu-Bombouaka Group. This is supported by research carried out by Anani et al. (2012), who indicated that the source rocks for the sandstones were granitic, gneissic, or sedimentary, suggesting a supply of sediments from the nearby Birimian Supergroup and possibly some contribution from Pan-African rocks. This is further supported by Kalsbeek et al. (2008), who proposed that the sandstones from the Oti-Pendjari and Tamale-Obosum Groups may have been derived from Pan-African arcs, and these sandstones were likely deposited after the closure of the pre-Pan-African ocean.

Conclusion

Nd and Sr isotopic analysis of the Tamale-Obosum and Oti-Pendjari Groups suggest a continental crustal provenance for both groups. TDM model ages for the Tamale-Obosum and Oti-Pendjari Groups range from 1.31 to 2.2 Ga (i.e. prevalence of Mesoproterozoic and minor Paleoproterozoic ages), indicating a dominant Proterozoic crustal source, which we suggest to be likely a combination of Birimian and Kwahu-Bombouaka Group rocks for the Tamale-Obosum Group. For the Oti-Pendjari Group, we suggest provenance source to be a mix of the Birimian rocks and rocks from the Pan African belt.

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