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**Elemental geochemistry and Nd isotope constraints on the provenance of
the basal siliciclastic succession of the middle Paleoproterozoic
Francevillian Group, Gabon**

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Abstract

Whole-rock elemental geochemistry and Nd isotope composition of siliciclastic rocks from the basal part of the unmetamorphosed middle Paleoproterozoic Francevillian Group, Gabon (FA Formation and FB₁ Member) were examined to provide insights into their provenance and its crustal evolution. Chemical weathering index diagrams suggest moderate degree of weathering in the provenance and secondary K-addition during illitization. Trace element geochemical proxies including the REEs provide evidence for sediment derivation from felsic-dominated upper continental crust (UCC) similar to Archean trondhjemite-tonalite-granodiorite (TTG) with minor contribution of mafic components. The low ¹⁴³Nd/¹⁴⁴Nd and negative εNd values (-10.83 to -5.76) provide further evidence for old and evolved upper continental crust components in detrital sources. The depleted mantle model ages for the

provenance protolith (2.73-3.05 Ga) are consistent with sediments sourced from the Mesoarchean granitoids of East Gabonian block exposed in the northern and southern parts of Gabon. We infer that the evolved, Mesoarchean felsic TTG-like granitoids were the main source for the sediments of the basal Francevillian Group, while minor mafic inputs from the contemporaneous greenstone belt with the Belinga Group cannot be completely discounted. The predominantly old crustal sources for the sediments suggest crustal addition and recycling of differentiated granitoid plutons instead of formation of juvenile crust during deposition in the Francevillian basin.

Keywords: Elemental geochemistry, Nd isotope, provenance, Mesoarchean, Francevillian Group, Gabon

1. Introduction

The middle Paleoproterozoic Francevillian basin developed in southeastern Gabon and hosting the Francevillian Group, is one of the world's most studied and least deformed middle Paleoproterozoic sedimentary successions. Compared to sedimentary successions of similar age that have been highly deformed and altered, the Francevillian basin sediments were not metamorphosed and have only been affected by diagenetic and/or low-temperature hydrothermal processes (Bros et al., 1992; Gauthier-Lafaye and Weber, 1989, 2003; Mathieu et al., 2001; Bankole et al., 2015, 2016, 2018). Thus, the basin has been extensively studied and is well known for the oldest high-grade, redox-controlled, and sandstone-hosted uranium ore mineralization (Gauthier-Lafaye and Weber, 1989, 2003; Bankole et al., 2016), natural fossil nuclear-fission reactors (Gauthier-Lafaye and Weber, 1989), oldest multicellular macrofossils (El Albani et al., 2019, 2014), and hosts the third largest known manganese-ore reserve in the world (Gauthier-Lafaye and Weber, 2003; Gauthier-Lafaye, 2006).

Although a number of studies have been carried out on the Francevillian Group, little is known about the geochemical characteristics of the sediments and their provenance has not previously been studied in much details. The geochemical composition of siliciclastic rocks is useful in providing information about their provenance, including its age and crustal evolution, surface Earth processes, and tectonic setting of sedimentary basins (McLennan et al., 2003). Thiéblemont et al. (2009) and Weber et al. (2016) used geochemical datasets for the Okondja sub-basin to suggest a change from rift-related deposition of the FA Formation to subduction-related tectonic setting with accompanying alkaline, mafic to ultramafic magmatism at the time when the FB Formation was deposited. However, Bankole et al. (2018) inferred from geochemical data that the interlayered thin K-bentonite beds in the FB_{2b} unit in the Franceville sub-basin were derived from calc-alkaline intermediate to felsic magmatism, which was related to a continental arc in a subduction setting developed during plate convergence. Recently, Ossa Ossa et al. (2020) utilized detrital zircon geochronology to suggest that the uranium-bearing sandstones and conglomerates of the FA and FB formations in the Francevillian basin were mainly sourced from the Mesoarchean granitoids of the East Gabonian block of the northwestern Congo craton.

This study presents new whole-rock elemental and Nd isotope data for the two basal sedimentary units of the Francevillian Group, FA and FB formations, in the Francevillian basin. We screened first for post-depositional overprints and elemental mobility to establish preservation of primary geochemical signals before constraining the intensity of chemical weathering, provenance, and tectonic setting for the sedimentary rocks of the FA and lower FB (FB₁ Member) formations as well as crustal evolution of their provenance.

2. Geological Background

The middle Paleoproterozoic Francevillian Group is an extensive sequence of unmetamorphosed siliciclastic and volcanoclastic strata that unconformably overlie the Archean basement rocks of the northwestern part of the Archean Congo craton in central to southeastern Gabon (Fig. 1). The basin is bounded by the Mesoarchean (3185-2805 Ma) and Neoarchean (2802-2500 Ma) granitoids of the East Gabonian block on the north (North Gabon massif) and south (Chaillu massif) and by the Neoarchean to early Paleoproterozoic (ca. 2515-2435 Ma) migmatites and Paleoproterozoic (ca. 2200-2120 Ma) supracrustal rocks of the Ogooué orogenic belt (West Gabonian block) to the west (Fig. 1; Caen-Vachette et al., 1988; Feybesse et al., 1998; Ledru et al., 1989; Thiéblemont et al., 2009). The Mesoarchean rocks of the East Gabonian block are composed of calc-alkaline and TTG (tonalite-trondhjemite-granodiorite) gneisses and granitoids with minor occurrences of ca. 2820 Ma charnockites and ca. 2920-2750 Ma greenstone belts. The greenstone belts are composed of the stratigraphic unit called Belinga Group, which includes mafic to ultramafic volcanic rocks, BIFs (banded iron formations), conglomerates, and micaschists. The Neoarchean basement rocks of the East and West Gabonian blocks consist of metamorphosed (gneisses and migmatites) and unmetamorphosed (diorites, granites, and syenites) rocks. The Ogooué orogenic belt of the West Gabonian block is bounded by the Ikoy-Ikobé fault to the west and the Ogooué thrust (Lopé High) to the east and was formed during the 2.2-2.0 Ga Eburnean orogeny, which involved collision of the Congo and São Francisco cratons, and several Paleoproterozoic orogenic events in the western and central parts of Africa (Ledru et al., 1989; Feybesse et al., 1998; Weber et al., 2016). It is a fold-and-thrust belt composed of greenschist to amphibolite facies metasediments and metavolcanic rocks with a peak metamorphism at 2120 ± 38 Ma (Feybesse et al., 1998; Weber et al., 2016). Time-equivalent successions on the São Francisco craton include the Monteiro sequence of the Rio Itapicuru

greenstone belt on the western Serrinha block in eastern Bahia, Brazil (Grisolia and Oliveira, 2012; Oliveira et al., 2010), and the Cercadinho-Sabará formations of the Minas Supergroup (Quadrilátero Ferrífero mining district, Minas Gerais, Brazil) (Bekker et al., 2003; Maheshwari et al., 2010; Reis et al., 2002). These two successions were deposited in foreland basins during the ca. 2.08-2.04 Ga collision of the Congo and São Francisco cratons, potentially in the aftermath of the assembly of the West and East Gabonian blocks. Geochemical signatures for these successions are consistent with sediment supply from continental arcs (e.g., Grisolia and Oliveira, 2012; Reis et al., 2002). Speculatively, development of the Francevillian basin could therefore represent a distal response to collision of the Congo and São Francisco cratons.

The Francevillian basin, hosting sediments of the Francevillian Group, is bounded by the Ogooué thrust to the west and overlain by the horizontally laying formations of the Mesozoic Congo basin to the east. The Francevillian Group consists of volcano-sedimentary strata that are exposed in Okondja, Lastourville, Franceville, and Booué sub-basins (Fig. 1; Bonhomme et al., 1982; Gauthier-Lafaye and Weber, 1989; Bros et al., 1992; Gauthier-Lafaye, 2006; Prétat et al., 2011). Although the tectonic setting and development of the Francevillian basin remain debated (Feybesse et al., 1998; Thiéblemont et al., 2014; Weber et al., 2016), its geodynamic evolution has been tied to the supercontinent tectonic cycle that started with the break-up of the Archean supercraton Superia and led during the Eburnean orogeny to the formation of the West Central African Belt (WCAB), which subsequently became the core of the Nuna supercontinent. The Francevillian Group has been divided into five lithostratigraphic formations, FA to FE from the base to top, in the Franceville and Okondja sub-basins (Fig. 2; Weber, 1968).

The lower part of the FA Formation, consisting mainly of coarse-grained sandstones and conglomerates with minor intercalations of fine-grained sandstones and mudstones, was

unconformably deposited on the Archean crystalline basement in a braided fluvial environment (Haubensack, 1981; Gauthier-Lafaye and Weber, 1989; Bankole et al., 2015). The upper part of the FA Formation is dominated by fine-grained sandstones and mudstones with interbedded coarse-grained sandstones, and has been interpreted to indicate fluvio-deltaic sedimentation. All the uranium ore deposits are hosted in the upper FA Formation sandstones. The overlying FB Formation is a heterogeneous succession of intertidal and subtidal mudstones and black shales with interbedded fine- to coarse-grained sandstones, breccias, and conglomerates (Azzibrouck-Azziley, 1986; Canfield et al., 2013; El Albani et al., 2010; Gauthier-Lafaye and Weber, 1989, 2003; Ossa Ossa et al., 2013, 2018; Pambo, 2004; Prat et al., 2011; Reynaud et al., 2017; Weber, 1968). The FB Formation has been divided into FB₁ and FB₂ members based on lithological differences and sedimentary structures (Fig. 2b). The transition from FA to FB Formation occurred during marine transgression induced by tectonic subsidence in the foreland basin in front of the forebulge (Ossa Ossa et al., 2020) and associated with the appearance of polygenetic breccias and olistolithes on the present-day southwestern margin of the Franceville and Okondja sub-basins (Gauthier-Lafaye and Weber, 2003, 1989). The FC Formation is composed of shallow-marine to supratidal (sabkha) deposits of massive dolostone and stromatolitic chert with thin beds of black shale (Gauthier-Lafaye and Weber, 2003; Prat et al., 2011; Ossa Ossa et al., 2018). The FD Formation consists of rhyolitic tuffs and epiclastic sandstones interbedded with marine black shales at the top, while the uppermost FE Formation is dominated by cross-bedded, coarse-grained to conglomeratic arkosic sandstones (Gauthier-Lafaye and Weber, 2003; Thiblemont et al., 2014).

The age and the duration of deposition of the Francevillian Group have not been fully resolved, but several attempts have been made to constrain the depositional age of the sedimentary strata. Sm-Nd isotope systematics was used to constrain the age of early

diagenesis of the FB_{1b} unit in the Franceville sub-basin with <0.4 μm and <0.2 μm clay fractions at 2099 ± 115 Ma and 2036 ± 79 Ma, respectively (Bros et al., 1992). The age of late diagenesis of the FB_{1c} unit was constrained with <1.5 μm and <2.0 μm authigenic illites that produced an apparent Rb-Sr isochron date of 1870 ± 50 Ma (Bonhomme et al., 1982). Horie et al. (2005) obtained a precise U-Pb SHRIMP zircon date of 2083 ± 6 Ma for the ignimbrite tuff in the FD Formation for which analytical data was not yet published. Bonhomme et al. (1982) reported a Rb-Sr whole-rock age of 2143 ± 143 Ma for coarse-grained syenites of the N’Goutou alkaline complex that intrudes the FA and FB formations in the Okondja basin (Fig. 1). Recently, Sawaki et al. (2017) published new highly discordant MC-ICP-MS ^{207}Pb - ^{206}Pb zircon age of 2191 ± 12 Ma for the N’Goutou Complex, which, if correct, would place the depositional age of the Francevillian Group to the beginning of the Lomagundi carbon isotope excursion. The FB and FC formations are considered to record the end of the 2.22 to 2.06 Ga Lomagundi carbon isotope excursion based on $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{org}}$ trends, and to be correlative with the Zaonega Formation in the Onega basin of Karelia, Russia (Kump et al., 2011), which is also imprecisely dated (Martin et al., 2015; Ovchinnikova et al., 2007). Elsewhere in Fennoscandia, the end of the Lomagundi carbon isotope excursion is placed between 2.11 and 2.06 Ga (Karhu and Holland, 1996; Martin et al., 2013). Combined, these age constraints place the age of deposition for the Francevillian Group close to 2.11 to 2.06 Ga, and currently provides the best estimate for the depositional age of the Francevillian Group (Canfield et al., 2013).

Detailed discussion of lithostratigraphy, depositional facies, petrography, diagenesis, and mineral chemistry of the basal part of the FA Formation and FB₁ Member in the Franceville sub-basin (this study area) are given in Bankole (2015) and Bankole et al. (2015, 2016). Additional information on the stratigraphy and depositional environment of the

Francevillian Group are provided Haubensack (1981), Gauthier-Lafaye (1986), Gauthier-Lafaye and Weber (1989, 2003) and Reynaud et al., (2017).

3. Samples and Analytical Techniques

Surface outcrops of the FA Formation and FB₁ Member are strongly weathered in the Francevillian basin; therefore, only fresh drill core samples were used for sampling. Samples for this study were collected from eight drill cores from the proximal to distal parts of the Franceville sub-basin (Figs. 2a and 3). Most of the drill cores intersected the transition zone between the upper FA and lower FB formations, except for the BA 2 and GR 15 drill cores where only the FA Formation was encountered. The FA and FB formation sediments as well as their boundary were defined based on distinct lithologies and drill core logs. All the studied samples were carefully cut from drill cores to minimize effect of recent weathering and alteration. We selected 54 samples, including 41 shales and siltstones and 13 sandstones, representing all the major lithological units across the FA Formation and FB₁ Member for geochemical analysis (Fig. 3). All the sandstones are from the FA Formation. The analyzed samples are broadly classified in this study into two main groups: mudstones (shales and siltstones) and sandstones (fine- to coarse-grained sandstones).

All the selected 54 samples were powdered in agate mortar, and analyzed for whole-rock major and trace element compositions at the *Service d'Analyse des Roches et des Mineraux* (SARM; CRPG-CNRS), Nancy, France. The whole-rock powder of ~100 mg of each sample was fused with lithium metaborate (LiBO₂) and dissolved in nitric acid for elemental analysis. Major and trace element data were obtained using inductively coupled plasma atomic emission spectrometry (ICP-AES) and inductively coupled plasma mass

spectrometry (ICP-MS), respectively, following the techniques described in Carignan et al. (2001).

Whole-rock Sm-Nd isotope analyses of 30 selected samples were carried out on the 7 collector Finnigan MAT-262 mass spectrometer at the GeOHeLis analytical facility (Géosciences Rennes/OSUR, University of Rennes, France). Samples were spiked with a ^{149}Sm - ^{150}Nd mixed solution and dissolved in a 2:1 acid mixture of 23 mol/L HF with 15 mol/L HNO_3 . They were then dried and taken up with 6 mol/L HCl. In each analytical session, the unknowns were analyzed together with the Ames Nd-1 Nd standard, which during the course of this study yielded average $^{143}\text{Nd}/^{144}\text{Nd}$ values of 0.511938 ± 5 (session 1), 0.511939 ± 5 (session 2), and 0.511928 ± 5 (session 3). All data for the unknowns have been adjusted to the long-term $^{143}\text{Nd}/^{144}\text{Nd}$ value of 0.511963 for Ames Nd-1. Instrumental mass fractionation was monitored and corrected using the $^{146}\text{Nd}/^{144}\text{Nd}$ value of 0.7219. Procedural blank analyses yielded 50 pg for Nd and the blank is therefore considered to be negligible. The initial Nd-isotope composition ($\epsilon_{\text{Nd}(t)} = [((^{143}\text{Nd}/^{144}\text{Nd}_{\text{sample}}/^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}})_t - 1) \cdot 10^{-4}]$), depleted mantle model ages ($T_{\text{DM}} = (1/\lambda) \cdot \ln[(^{143}\text{Nd}/^{144}\text{Nd}_{\text{sample}} - ^{143}\text{Nd}/^{144}\text{Nd}_{\text{DM}(0)}) / (^{147}\text{Nd}/^{144}\text{Nd}_{\text{sample}} - ^{147}\text{Nd}/^{144}\text{Nd}_{\text{DM}(0)} + 1) \cdot 10^{-9}]$), and fractional deviation of $^{147}\text{Sm}/^{144}\text{Nd}$ from that of chondritic meteorites ($f^{\text{Sm}/\text{Nd}} = (^{147}\text{Sm}/^{144}\text{Nd})_{\text{sample}} / (^{147}\text{Sm}/^{144}\text{Nd})_{\text{CHUR}} - 1$); McLennan et al., 1993) were calculated assuming present-day $^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}(0)} = 0.512636$ (CHUR: Chondritic Uniform Reservoir), $^{147}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}(0)} = 0.1967$, $^{143}\text{Nd}/^{144}\text{Nd}_{\text{DM}(0)} = 0.51235$, and $^{147}\text{Nd}/^{144}\text{Nd}_{\text{DM}(0)} = 0.217$, and decay constant, $\lambda = 6.54 \times 10^{-12} \text{ y}^{-1}$ (DePaolo and Wasserburg, 1976). The initial $^{143}\text{Nd}/^{144}\text{Nd}$, $\epsilon_{\text{Nd}(t)}$ values, and T_{DM} ages for the basal FA and FB formations sediments were calculated at depositional age of $t = 2.1 \text{ Ga}$.

4. Results

4.1. Major element geochemistry

The major element data for the studied samples are presented in [Table A1 \(see Appendix\)](#). The FA Formation sandstones falls within the arkose, sub-arkose, litharenite, and quartz arenite compositional fields, whereas mudstones from FA Formation and FB₁ Member fall within the shale and wacke fields in the Herron's (1988) geochemical classification diagram for sedimentary rocks ([Fig. 4](#)). The sandstones generally have higher SiO₂/Al₂O₃ ratios and are therefore quartz-rich compared to the mudstone samples, which are clay-rich ([Fig. 4](#)). Higher Fe₂O₃/K₂O ratios for the FB₁ Member mudstones compared to the FA Formation mudstones and sandstones are related to the abundant Fe-rich dolomite and pyrite, and the absence of K-feldspar in the FB Formation (Ngombi-Pemba et al., 2014; Bankole et al., 2015, 2018). The K₂O/Al₂O₃ ratios are significantly lower for the FB₁ Member mudstones (average = 15) compared to the FA Formation mudstones (average = 30) and FA sandstones (average = 34), consistent with absence of K-feldspar in the FB Formation ([Table A1](#)).

Compared to the Late Archean Upper Continental Crust (LAUCC; Condie, 1993), the analyzed samples, especially the FA Formation sandstones, are depleted in most of the major elements ([Fig. A1; see Appendix](#)). All the FA Formation mudstones and few of the sandstone samples are enriched in K₂O, whereas only few samples are enriched in Al₂O₃ and Fe₂O₃ relative to the FB₁ Member mudstones ([Fig. A1b](#)). The anomalously high MnO content in 2 FB₁ Member mudstones ([Fig. A1a](#)) is due to their stratigraphic proximity to the overlying Mn protore hosted in this unit (Gauthier-Lafaye and Weber, 2003). The greatest elemental variability is shown by the FA sandstones ([Fig. A1c](#)) and the slight enrichment in SiO₂ relative to the LAUCC reflects higher abundance of quartz in these samples. Enrichments in Fe₂O₃, MnO, CaO, and MgO in some samples could be attributed to their high carbonate content indicated by their large LOI values ([Table A1, see Appendix](#)).

4.2 Trace and rare earth element geochemistry

The trace and REE data are presented in [Tables A2 and A3 \(see Appendix\)](#). Although the absolute concentrations of the trace elements are variable in the mudstone and sandstone samples, their LAUCC-normalized patterns are rather similar ([Fig. 5](#)). The large-ion lithophile elements (LILE) such as Rb, Ba, and U are moderately enriched in most of the FB₁ Member and FA Formation mudstones ([Figs. 5a and b](#)), but significantly depleted in the FA Formation sandstones ([Fig. 5c](#)). All of the samples are similarly depleted in Sr and Ca, which is likely controlled by substitution of Sr for Ca in plagioclase. Concentrations of high-field strength elements (HFSE) such as Zr, Hf, Y, Th, Ta, and Nb vary considerably regardless of the grain size of the hosting lithology. Excluding few samples, the samples are depleted in Y, Nb, and Ta, whereas Zr and Hf are relatively enriched in some of the FA Formation samples ([Figs. 5a-c](#)) due to preferential enrichment in zircon during transport. All the transition trace elements (TTE: Co, Cr, Sc, Ni, and V) are depleted in the FA Formation sandstones, but display variable enrichments and depletions in the FA Formation and FB₁ Member mudstones.

While the REE have variable abundances, the chondrite-normalized REE patterns of all the samples are remarkably similar among themselves and to the LAUCC ([Figs. 6a-c](#)). The total REE abundances (ΣREE) are higher in the mudstones than in most of the sandstones ([Table A3](#)), likely related to grain size and quartz dilution. The samples show significant light-REE (LREE) enrichment ($\text{La}_\text{N}/\text{Yb}_\text{N}$), flat to slightly depleted heavy-REE (HREE), negative to negligible Eu anomalies, and insignificant Ce anomalies. Very few samples have $\text{La}_\text{N}/\text{Yb}_\text{N}$ values smaller than the average chondrite value of 12.91 (McDonough and Sun, 1995). The FA Formation sandstones have variable and more pronounced negative Eu anomalies ($\text{Eu}/\text{Eu}^* = 0.30\text{-}0.99$; average = 0.68) and generally higher $\text{La}_\text{N}/\text{Yb}_\text{N}$ (7.20-92.63) values than the FA Formation mudstones ($\text{Eu}/\text{Eu}^* = 0.69\text{-}0.95$; average = 0.81; $\text{La}_\text{N}/\text{Yb}_\text{N} =$

12.18-54.27) and FB₁ Member mudstones ($\text{Eu}/\text{Eu}^* = 0.72\text{-}0.95$; average = 0.85; $\text{La}_\text{N}/\text{Yb}_\text{N} = 6.76\text{-}44.8$).

4.3 Sm-Nd isotope geochemistry

The whole-rock Sm-Nd isotope data of the sandstones and mudstones is remarkably similar and thus does not show any significant fractionation with respect to grain size (Table 1). The initial $^{143}\text{Nd}/^{144}\text{Nd}$ ratios for all the samples range from 0.5094 to 0.5096 and $^{147}\text{Sm}/^{144}\text{Nd}$ ratios from 0.078 to 0.111, except for two outliers with considerably higher values of 0.165 (BA 11_13.9) and 0.192 (GR_243.1) (Table 1; Figure A2). With the exception of the two outlier samples, all the samples have overlapping negative $\epsilon_{\text{Nd}}(t)$ values from -10.83 to -5.76, T_{DM} ages of 2.73 to 3.05 Ga, and $f^{\text{Sm}/\text{Nd}}$ from -0.60 to -0.43 (Table 1). However, the two outliers with high $^{147}\text{Sm}/^{144}\text{Nd}$ ratios have unrealistically large T_{DM} dates of 5.72 and 6.03 Ga and $f^{\text{Sm}/\text{Nd}}$ of -0.16 and -0.02, suggesting post-depositional disturbance of the isotope system, and are not considered further.

5. Discussion

The chemical and mineralogical composition of siliciclastic sedimentary rocks, especially fine-grained rocks, are mainly controlled by the nature of their source rocks, and they have been widely used in constraining the provenance, sedimentary processes, and tectonic setting of sedimentary basins (Bhatia and Crook, 1986; Cullers, 2000, 1994; Large et al., 2018; McLennan et al., 1993, 1990; Roser and Korsch, 1986). Most of the major elements (e.g., Ca, Fe, Na, K, and Mg) have high solubility in water and easily fractionate during sedimentary processes, making them unreliable as provenance proxies. Therefore, provenance analysis typically relies on elements such as REE and certain HFSE such as Al, Ti, Th, Sc, Y, and Zr because of their relative immobility during sedimentary processes and short residence

time in seawater (e.g., Taylor and McLennan, 1985; and references therein). However, sedimentary processes such as grain sorting, chemical weathering, sedimentation, diagenesis, and low-temperature alteration can result in significant changes to mineralogical and geochemical composition compared to that of their original source (e.g., McLennan et al., 1993, 2003; and references therein). In addition, enrichment in heavy minerals such as zircon, monazite, and apatite during sedimentary transport can exert strong influence on trace element and REE contents as they are preferentially concentrated in these minerals (e.g., McLennan et al., 1993; Sugitani et al., 2006). These processes make it difficult to determine whether the geochemical signatures preserved in siliciclastic sediments truly reflect the composition of the provenance or were imprinted by sedimentary processes. It is therefore necessary to first evaluate the influence of sedimentary processes on geochemical composition to resolve primary signals from secondary overprints before using them in provenance analysis.

5.1 Sediment maturity, sorting, and mineral control

Concentrations of major element oxides and their ratios have been used to classify sedimentary rocks and the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio has been used to evaluate their maturity (e.g., Bhatia and Crook, 1986). Sediment maturity increases with increasing $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios reflecting decrease in feldspar and clay content. The moderate to high $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios of the FA Formation sandstones (Fig. 4) are typical of chemically immature to mature sandstones, suggesting derivation of these sandstones from a range of proximal to distal source areas. Furthermore, Th/Sc and Zr/Sc ratios provide insight into sediment recycling and heavy mineral accumulation as both Th and Zr are incompatible elements, and are typically stronger enriched in felsic rocks than the more compatible Sc (McLennan et al., 1993, 2003). The average Th/Sc ratio is lower in the FB₁ Member mudstones (0.52-1.39; average = 0.86) than FA Formation mudstones (0.98-3.36; average = 1.88) and sandstones (2.16-5.64; average =

3.85), indicating more evolved source for the FA Formation sediments compared to the FB₁ Member. Most of the FB₁ Member and FA Formation mudstones cluster close to the Late Archean Upper Continental Crust (LAUCC), and provenance-dependent compositional variation trend on the Th/Sc vs Zr/Sc diagram (Fig. 7), with limited spread along the zircon addition trend, ruling out major role of hydraulic sorting and heavy mineral fractionation on their chemical composition, and probably reflect chemical composition of their source rocks. In contrast, the FA Formation sandstones and few FA Formation mudstones have elevated Zr/Sc ratios, indicating significant zircon addition due to reworking and heavy-mineral concentration (McLennan et al., 1993).

Trace elements, including REE, in siliciclastic rocks are generally hosted by clay minerals and heavy minerals such as zircon, monazite, and apatite. Zircon is characterized by enrichment in HREE relative to LREE, and this results in a negative correlation between Zr and La_N/Yb_N (Yang et al., 1998). In contrast, monazite and apatite are preferentially enriched in LREE and a negative correlation of Gd_N/Yb_N with Th and Gd_N/Yb_N with P₂O₅, respectively, corresponds to their major contribution to sediment composition (McLennan, 1989). The moderately positive correlation of Al₂O₃ with Σ REE for all samples (Fig. 8a) indeed demonstrates that REE are mostly hosted by aluminosilicates in the mudstones ($R^2 = 0.57$) and, to a lesser degree, in the sandstones ($R^2 = 0.27$), and positive correlation of Σ REE with K₂O (Fig. 8b) suggests illite as the main carrier for REE in the mudstones ($R^2 = 0.59$) and sandstones ($R^2 = 0.73$). Poor correlation between La_N/Yb_N and Zr for the FB₁ Member mudstones ($R^2 = 0.06$), and FA Formation mudstones and sandstones ($R^2 = 0.14$) points to limited influence of zircon as a carrier for REE, except for few samples of the FA Formation (mostly sandstones) with elevated Zr content (Fig. 8c). Additionally, poor correlation between Th and Gd_N/Yb_N (Fig. 8d) and between P₂O₅ and Gd_N/Yb_N (Fig. 8e) in the mudstones and sandstones is inconsistent with phosphate enrichment (e.g., monazite and apatite) and does not

support significant control of these minerals over REE concentration, except for three FA Formation sandstones with high Th content.

5.2 Post-depositional element mobility

Monazite is a common accessory mineral in the FA Formation, and Mathieu et al. (2001) and Cuney and Mathieu (2000) showed that alteration of monazite crystals in the FA Formation coarse-grained sandstones at the Oklo and Bangombe natural reactors by highly saline diagenetic brines resulted in leaching and redistribution of LREEs. Using La as representative of the LREEs and Th as an immobile element, the authors quantified LREEs loss from the FA Formation coarse-grained sandstones due to monazite alteration by comparing their Th/La ratios with the average values for Archean and Post-Archean sandstones and shales. Cuney and Mathieu (2000) found that the whole-rock Th/La ratio increased from 0.27 ± 0.2 in unaltered mudstones of the FA and FB formations to 1.14 ± 0.15 in altered, coarse-grained sandstones of the FA Formation. Using similar approach, we evaluated the post-depositional redistribution and remobilization of LREEs by comparing Th/La ratios of the analyzed samples to the average values of Archean and Post-Archean shales (0.27 to 0.38) and sandstones (0.35 to 0.42) from Condie (1993). We used a Th/La ratio of 0.5 as an alteration baseline, implying that samples with $\text{Th/La} \geq 0.5$ are altered and those having this value below 0.5 are unaltered or minimally altered. The FB₁ Member and FA Formation mudstones have low Th/La ratios similar to those of the Archean and Proterozoic shales, and mostly plot in the unaltered field, indicating insignificant leaching of the LREE from the mudstones, whereas most of the FA Formation sandstones have considerably higher Th/La ratios and mostly plot in the altered field, likely reflecting more porous and permeable nature of the sandstones and remobilization of LREEs (Fig. 8f).

Moreover, post-depositional alteration could potentially impact Sm-Nd isotope

composition because the Nd isotope system is not entirely immune to alteration, especially by processes that mobilize REEs (e.g., Öhlander et al., 2000; Mathieu et al., 2001). For instance, several studies (e.g., McLennan et al., 1993, 2003; Bock et al., 1994; Ehrenberg and Nadeau, 2002) have shown that positive correlation between Sm-Nd ratios and depleted mantle model ages (T_{DM}) is an evidence for post-depositional disturbance of the Sm-Nd isotope system. These studies have shown that an open-system behavior during diagenesis might result in higher whole-rock Sm/Nd ratios and depleted mantle model ages. Excluding two outliers with anomalously high $^{147}\text{Sm}/^{144}\text{Nd}$ ratios (Table 1), there is no apparent correlation between these two parameters for the FA Formation sandstones ($R^2 = 0.04$) and FA Formation and FB₁ Member mudstones ($R^2 = 0.12$), suggesting that the Sm-Nd isotope system of these samples was not disturbed after deposition. Furthermore, parallel chondrite-normalized REE patterns with limited range in sample/chondrite ratio and lack of Ce anomaly displayed by all samples (Fig. 6) support the relative immobility of the REE (cf. McLennan et al., 1993, 2003). Overall, REE in most of the studied samples from the Franceville sub-basin do not appear to be fractionated by sedimentary and post-depositional processes; and thus, geochemical and isotope signatures of the Francevillian Group sediments, especially of the mudstones, were largely preserved on the whole-rock scale. Effect of sorting and heavy mineral enrichment on composition of the coarse-grained sandstones is further discussed below.

5.3 Paleoweathering of the source area

The intensity of weathering in source area can be assessed with the chemical index of alteration (where $\text{CIA} = [(\text{Al}_2\text{O}_3/(\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O})) \times 100]$ in molar ratios and CaO^* is the amount in silicate fraction; Nesbitt and Young, 1982), which measures the degree of feldspar conversion to clays in shales during weathering. CIA values of unweathered to weakly weathered rocks are close to 50 or less, moderately weathered rocks have values

between 60 and 80, and highly weathered rocks have values above 80 (Nesbitt and Young, 1984, 1982; Fedo et al., 1995). We do not have data for carbonate and apatite content, and thus cannot correct for their Ca contents. Hence, we assumed in our calculations that moles of silicate $\text{CaO} = \text{Na}_2\text{O}$ for samples with $\text{CaO} > \text{Na}_2\text{O}$ and accepted measured CaO value if $\text{CaO} \leq \text{Na}_2\text{O}$ (cf. Jian et al., 2013; Roddaz et al., 2006). The analyzed mudstones generally have moderate to high CIA values from 59 to 82 (Table A1), with majority of the samples falling between the range of unweathered igneous rocks (45-55) and typical average shale (70-75), suggesting moderate chemical weathering in the source areas.

The CIA proxy could be compromised by post-depositional addition of K, which is common to aluminous clays, and this is commonly visualized on an Al_2O_3 -($\text{CaO}^* + \text{Na}_2\text{O}$)- K_2O (A-CN-K) ternary plot (Nesbitt and Young, 1984; Fedo et al., 1995). On such diagram, weathering products line up parallel to the A-CN side, while any deviation from this trend towards the A-K side of the diagram indicates significant K addition (Fedo et al., 1995). The FB₁ Member mudstones plot between the TTG and granite weathering trends, while the FA Formation mudstones and sandstones scatter to the right of the weathering trend of the average granite, towards the A-K side of the A-CN-K ternary plot (Fig. 9), which is inconsistent with typical weathering pattern and suggests post-depositional K addition during illitization (Fedo et al., 1995). To overcome the problem of K-addition, we calculated the chemical index of weathering ($\text{CIW} = [(\text{Al}_2\text{O}_3)/(\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O}) \times 100]$; Harnois, 1988) and plagioclase index of alteration ($\text{PIA} = [(\text{Al}_2\text{O}_3 - \text{K}_2\text{O})/(\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} - \text{K}_2\text{O}) \times 100]$; Fedo et al., 1995) to assess plagioclase alteration during chemical weathering in the source area. The PIA and CIW values of unweathered rocks are close to 50 and values close to 100 indicate complete plagioclase breakdown. The PIA and CIW values are high for the FB₁ (80-98) Member and most of the FA Formation mudstones (91-98) and sandstones (92-97), suggesting moderate to intense plagioclase alteration in the source area. Few samples,

however, have values lower than 80 (Table A1). The high PIA and CIW values compared to the CIA values attest that the succession has been strongly illitized after deposition.

5.4 Sediment provenance

5.4.1 Trace elements

The distribution of the least-mobile, incompatible (REEs, HFSE, Th, and Y) and compatible (Sc, V, Ni, Cr, and Co) trace elements have been recognized to reflect the composition of provenance and discriminate felsic from mafic source rocks (e.g., Taylor and McLennan, 1985; Floyd and Leveridge, 1987; Cullers, 1994, 2000; McLennan et al., 1993; Condie, 1993; Large et al., 2018). The incompatible elements are generally enriched in upper continental crust felsic rocks and low in mafic and ultramafic mantle-derived rocks compared to the compatible elements. The variable enrichments and depletions in incompatible and compatible elements in the FA Formation and FB₁ Member mudstones (Fig. 5a-b) suggest a mixed signature of felsic and mafic sources. In contrast, the FA Formation sandstones strongly suggest a predominantly felsic source as they are mostly enriched in incompatible elements and depleted in compatible elements with respect to LAUCC (Fig. 5c). Geochemical discriminant diagrams such as Th/Sc versus Th/Cr (Fig. 10a) and Th/Sc versus La/Sc (Fig. 10b) show that all the samples, except for two FB₁ Member mudstones, have considerable higher Th/Sc, La/Sc, and Th/Cr ratios than the LAUCC, plotting between granite and TTG within the felsic provenance field on Figure 10b. However, the FB₁ Member mudstones have lower Th/Sc, Th/Cr, La/Th, and La/Cr ratios and plot between TTG and mafic source field (Fig. 10a), suggesting stronger mafic input compared to the FA Formation sediments. All samples are characterized by low La/Th ratios and variable Hf content, and predominantly plot within the felsic source and mixed felsic/mafic source fields on the La/Th versus Hf diagram (Fig. 10c). The predominantly felsic composition of the provenance is supported by

the low Cr/V ratios and slightly variable Y/Ni ratios, which might reflect homogenous source or efficient sediment mixing (Fig. 10d; cf. Ghosh et al., 2016).

In addition, distribution patterns of REE and the size of Eu anomaly have also been used as proxies for determining the composition of the source area for siliciclastic sedimentary rocks (e.g., Taylor and McLennan, 1985; Condie, 1993; Cullers, 1994, 2000). The degree of LREE enrichment over HREE ($\text{La}_\text{N}/\text{Yb}_\text{N}$) measures the proportion of felsic to mafic components with LREE enrichment being lower in mafic rocks compared to felsic rocks, and the largest enrichment observed in TTG rocks. The size of Eu anomaly could reflect intracrustal igneous differentiation, separating from the source magma granitic melts and leaving behind residue with feldspar, which is the main host of Eu^{2+} in igneous rocks (Taylor and McLennan, 1985; McLennan et al., 1993; 2003; Gao and Wedepohl, 1995; Cullers, 2000). The seemingly similar chondrite-normalized REE patterns characterized by high $\text{La}_\text{N}/\text{Yb}_\text{N}$ ratios, relatively flat HREE, negative to negligible Eu anomalies, and negative Ta and Nb suggest predominant derivation from granitoids for the FA Formation and FB₁ Member sediments. The distinctly negative Eu anomalies shown by most of the sediments, especially by the FA Formation sandstones, can be attributed to fractionation induced by feldspar-rich residuum during intracrustal fractionation in the provenance (cf. Gao and Wedepohl, 1995), suggesting deposition of differentiated crust-derived sediments in stable cratonic environment comparable to that for LAUCC (Ghosh et al., 2016). On another hand, weakly positive to negligible Ta, Nb, and Eu anomalies in few of the FA Formation and FB₁ Member mudstones are potentially related to their higher feldspar (mostly plagioclase) content (Table A3; cf. Taylor and McLennan, 1985; Bankole et al., 2015), which likely reflects derivation from less differentiated mafic- and mantle-derived source rocks. Together, these features point to heterogeneous provenance dominated by felsic, upper continental crust materials.

5.4.2 Nd isotopes and crustal evolution

The Nd isotope composition can be used as an independent proxy to provide further constraint on the provenance for the Francevillian Group and its crustal residence age. The Sm-Nd isotope system is unique in provenance studies because both the parent (^{147}Sm) and daughter (^{143}Nd) isotopes are REE, and are highly immobile in sediments under most post-depositional processes and during low-temperature metamorphism. According to DePaolo and Wasserburg (1976), DePaolo et al. (1991), and McLennan and Hemming (1992), sedimentary rocks derived from less differentiated, mafic and juvenile sources in active tectonic settings commonly have high $^{143}\text{Nd}/^{144}\text{Nd}$ ratios, positive $\epsilon_{\text{Nd}(t)}$ values, variable and generally high Sm/Nd ratios, and low Th/Sc ratios (≤ 1). In contrast, those derived from old, upper continental crust in passive/cratonic settings are typically characterized by low $^{143}\text{Nd}/^{144}\text{Nd}$ ratios, negative $\epsilon_{\text{Nd}(t)}$ values, low Sm/Nd ratios, and high Th/Sc ratios (≥ 1).

The analyzed FA Formation and FB₁ Member mudstones and sandstones all have almost invariably negative $\epsilon_{\text{Nd}(t)}$ and low $f^{\text{Sm}/\text{Nd}}$ values, uniform Sm/Nd ratios, and significantly higher and nearly uniform Th/Sc ratios (≥ 1), indicating dominance in the provenance of old, differentiated upper crust (Table 1; Figs. 11a-b). The data all scatter around grey TTG granitoids of the East Gabonian block, close to the upper continental crust end-member stretching from the Archean crust to volcanic arc rocks within the LREE-enriched field on the $f^{\text{Sm}/\text{Nd}}$ versus $\epsilon_{\text{Nd}(t)}$ diagram (Fig. 11b). Nd-isotope data for all the samples indicate overwhelming contribution of evolved and differentiated, older TTG-like upper continental crust in a passive margin setting on a stable craton.

According to the mixing model of DePaolo et al. (1991), the relative contribution of the Archean crustal (AC) material is given by: $\text{AC} = (1 - \alpha_{\text{VA}}) * 100$, where α_{VA} (contribution of the arc material) = $\beta^* / [(\epsilon_{\text{Nd}}\text{VA} - \epsilon_{\text{Nd}}\text{S}) / (\epsilon_{\text{Nd}}\text{S} - \epsilon_{\text{Nd}}\text{AC}) + \beta^*]$, β^* is the ratio of Nd concentration

in the volcanic arc rocks and Archean crust ($\text{Nd}_{\text{VA}}/\text{Nd}_{\text{AC}}$), $\epsilon_{\text{Nd}}\text{VA}$ is the isotopic composition of the volcanic arc component, $\epsilon_{\text{Nd}}\text{S}$ is the isotopic composition of the analyzed rock, and $\epsilon_{\text{Nd}}\text{AC}$ is the isotopic composition of the Archean crust. We used the average values for the Mesoarchean basement rocks around the Francevillian basin, $\epsilon_{\text{Nd}}\text{AC} = -11.21$ and $\text{Nd}_{\text{AC}} = 37.8$ ppm (cf. Thiéblemont et al., 2009), for the Archean crust end-member, and $\epsilon_{\text{Nd}}\text{VA} = 5$ and $\text{Nd}_{\text{VA}} = 10$ ppm for the volcanic arc end member. The mixing model results do not show any trend with the stratigraphic position, and indicate that the detrital materials for the FA Formation sandstones and mudstones and FB_1 Member mudstones were predominantly derived from the older crystalline basement rocks with negligible to minor volcanic arc input (Table 2). The geochemical provenance model is thus fully consistent with the results of the LA-ICP-MS U-Pb study of detrital zircons from the FA and FB formations, which have exclusively shown Neoarchaeon to Mesoarchaeon dates (Ossa Ossa et al., 2020).

The T_{DM} age range (2.73-3.05 Ga; Table 1) for all the samples is similar and consistent with the U-Pb ages of detrital monazites (Mouélé et al., 2014) and zircons (Bankole et al., 2018; Ossa Ossa et al., 2020) from the underlying crystalline basement rocks, suggesting that the ca. 3.1 to 2.8 Ga granitoids of the East Gabonian block were the principal source for the lower Francevillian Group sediments. The Mesoarchean basement rocks are composed of K-rich granitoids (pink granite; monzogranite) and TTG granitoids with minor occurrences of charnockites and the Belinga greenstone belts in places (Fig. 1). Although the trace element geochemical proxies can separate contribution from felsic vs. mafic rocks in this study, they cannot distinguish between the K-rich granite and TTG suite as the samples fall on the mixing line within the felsic compositional field with basalt, TTG, and granite aligned along it (Figs. 10a-b). Trace element geochemical proxies for the FA Formation and FB_1 Member sedimentary rocks show some similarity; however, it appears that most of the FA Formation sandstones and mudstones have higher content of incompatible elements, more negative Eu

anomalies, and a relatively flat HREE pattern compared to the FB1 Member mudstones. Therefore, it is likely that the FA Formation sedimentary rocks, especially the sandstones, had larger contribution of intracrustally fractionated Mesoarchean K-rich granites than most of the FB₁ Member mudstones, which are predominantly characterized by small negative to negligible Eu anomalies and steeper HREE patterns (larger Gd/Yb ratios) (cf. McLennan et al., 1990). This is in agreement with Ossa Ossa et al. (2020) who used detrital zircon geochronology to argue that the Mesoarchean, K-rich (pink) granitoids were the main source for the uranium-bearing sandstones of the lower FA Formation. However, minor contribution from the Mesoarchean greenstone belts, hosting mafic rocks of the Belinga Group, and charnockites is entirely possible. Lack of juvenile sources for the lower Francevillian Group sediments, and the dominant Mesoarchean age (cf. Bankole et al., 2018; Ossa Ossa et al., 2020) and relatively homogenous chemical composition of the provenance imply crustal addition and effective recycling following magma extraction from the mantle between 2.73 and 3.05 Ga rather than formation of juvenile crust during deposition in the Francevillian basin. Crustal recycling and mineralogical and geochemical fractionation via sedimentary processes point at the existence of thick and stable continental crust during deposition of the Francevillian Group on the Mesoarchean East Gabonian block.

5.5 Tectonic setting during deposition of the lower Francevillian Group

Geochemical discriminant diagrams using certain least-mobile trace elements have been routinely used in resolving tectonic setting for sedimentary basins (e.g., Bhatia and Crook, 1986; Roser and Korsch, 1988; McLennan et al., 1990; McLennan and Hemming, 1992; Cullers, 2000; Armstrong-Altrin et al., 2004; Schneiderhan et al., 2011). Although many of these diagrams were defined for Phanerozoic sedimentary rocks, they have also been applied to characterize tectonic setting of Precambrian sedimentary basins assuming that

plate-tectonic processes were active during this era. However, this approach sometimes results in confusing outcome due to the control of the nature of the source rocks on geochemical composition of sedimentary rocks (McLennan et al., 1993, 1990). On the La-Th-Sc ternary discriminant diagram (Bhatia and Crook, 1986), the FA Formation and FB₁ Member sediments predominantly fall within the passive margin (PM) and active continental margin (ACM) fields (Fig. 12a). However, the FA Formation mudstones straddle the ACM and Continental Island Arc (CIA) fields, while the FB₁ Member mudstones exclusively plot in the CIA field on the Th-Sc-Zr/10 (Fig. 12b) and La/Y versus Sc/Cr discriminant diagrams (Fig. 12c). Although the FA Formation sandstones plot in the PM/ACM and PM fields on the La-Th-Sc and Th-Sc-Zr/10 diagrams, respectively, they extend into the CIA field on the La/Y vs. Sc/Cr diagram (Fig. 12c). Additionally, all the samples are characterized by variable low to high Ti/Zr and La/Sc ratios, and therefore only few samples plot in the ACM, CIA, and PM fields on Figure 12d, while the rest of the samples plots outside the discriminant fields on the Ti/Zr vs. La/Sc diagram. The dominant contribution of older Archean crustal material and lack of juvenile arc source make a CIA tectonic setting unlikely for the FA Formation and FB₁ Member mudstones and sandstones. It seems likely that the CIA signature of sediments might be caused by significant contribution of TTGs, which have similar trace element geochemical signatures to those of CIA granitoids (cf. McLennan et al., 2006). Similarly, high Ti/Zr ratios in most of the samples might reflect the predominantly Archean TTG provenance to the sediments (cf. Martin et al., 2005). Hence, it is challenging to constrain a tectonic setting using trace element composition for the Paleoproterozoic sedimentary basins, which developed on the deeply eroded, Archean upper continental crust with predominantly crystalline rocks in the provenance largely lacking contribution of recycled sedimentary materials.

Conclusions

The whole-rock geochemistry and Nd isotope data for the unmetamorphosed, middle Paleoproterozoic Francevillian Group FA Formation and FB₁ Member sediments demonstrate that their geochemical signatures have not been considerably disturbed by sedimentary and post-depositional processes and are thus suitable for the provenance study. The moderate paleoweathering indices and position of the whole-rock geochemical data on the A-CN-K ternary diagram suggest that the FA Formation sandstones and most of the FA Formation and FB₁ Member mudstones have undergone significant chemical weathering and post-depositional K addition. The geochemical signatures of selected relatively immobile elements, including REE, and their ratios, and Nd isotope systematics indicate geochemically similar sources for the FA Formation and FB₁ Member sediments that were predominantly derived from the Mesoarchean TTGs of the East Gabonian block upper continental crust with only minor, if any, mafic contribution from the Mesoarchean greenstone belts hosting the Belinga Group. We infer that minimal to no contribution of juvenile arc materials to the middle Paleoproterozoic Francevillian Group sediments is consistent with the recently inferred foreland basin setting (Weber et al., 2016; Bankole et al., 2018; Ossa Ossa et al., 2020) developed on the deeply eroded upper continental crust of the Mesoarchean East Gabonian block.

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List of figures

Figure 1. Geological map of Gabon showing distribution of the Francevillian Group and the Franceville sub-basin on the inset (modified from Thiblemont et al., 2009).

Figure 2. A. Geological map of Franceville sub-basin showing location of the drill cores (modified from Aubineau et al., 2018) and (B) Lithostratigraphic column of the Francevillian Group in the Franceville sub-basin, Gabon (modified from Weber, 1968). Sources of geochronologic age constraints: ¹ - Horie et al., 2005; ² - Bonhomme et al., 1982; ³ - Bros et al., 1992; ⁴ - Bouton et al., 2009.

Figure 3. Lithostratigraphy of the studied drill cores in the Franceville sub-basin showing

position of the studied samples. Two additional drill cores (BA 74 and GR 3), located on Figure 2, are not shown on this figure since only partial drill core logs are available for them and only three samples with well-defined stratigraphic positions from these two drill cores were used in our study.

Figure 4. Geochemical classification of FA Formation and FB₁ Member sandstones and mudstones (Herron, 1988).

Figure 5. Trace element spider diagrams for (A) FB₁ Member mudstones, (B) FA Formation mudstones, and (C) FA Formation sandstones normalized to the composition of the LAUCC (Late Archean Upper Continental Crust) from Condie (1993).

Figure 6. Chondrite-normalized Rare Earth Element (REE) patterns for (A) FB₁ Member mudstones, (B) FA Formation mudstones, and (C) FA Formation sandstones, compared with the Late Archean Upper Continental Crust (LAUCC) from Condie (1993). Chondrite values are from McDonough and Sun (1995).

Figure 7. Plot of Th/Sc versus Zr/Sc for the studied Francevillian Group sediments (after McLennan et al., 1993). Data for basalt, TTG (tonalite-trondhjemite-granodiorite), granite, and LAUCC (Late Archean Upper Continental Crust) are from Condie (1993).

Figure 8. Plot of (A) Σ REE versus Al₂O₃, (B) Σ REE versus K₂O, (C) La_N/Yb_N versus Zr, (D) Th versus Gd_N/Yb_N, (E) Gd_N/Yb_N versus P₂O₅, and (F) Th versus La.

Figure 9. Ternary A-CN-K (Al₂O₃-CaO+Na₂O-K₂O) diagram in molar proportions (Nesbitt and Young, 1984) for the lower Francevillian Group sediments. Data for basalt, andesite, TTG (tonalite-trondhjemite-granodiorite), and granite are from Condie (1993). The arrows 1, 2, 3, and 4 indicate possible weathering trends for unmetasomatized basalt, andesite, TTG, and granite, respectively.

Figure 10. Trace element discriminant plots of (A) Th/Cr versus La/Cr (after Bhatia and Crook, 1986), (B) Th/Sc versus La/Sc (after Bhatia and Crook, 1986), (C) La/Th

versus Hf (after Floyd and Leveridge, 1987), and (D) Cr/V versus Y/Ni (after Ghosh et al., 2016) for the studied sediments. Potential sources are plotted for comparison. B: basalt; LAUCC: Late Archean Upper Continental Crust; TTG: tonalite-trondhjemite-granodiorite. Data for basalt, LAUCC, TTG, felsic volcanics, and granite are from Condie (1993).

Figure 11. Plots of (A) $\epsilon_{\text{Nd}(t)}$ versus Th/Sc and (B) $f^{\text{Sm}/\text{Nd}}$ versus $\epsilon_{\text{Nd}(t)}$ (after McLennan et al., 1993), excluding altered samples, BA 11_13.9 and GR_243.1, with anomalous $\epsilon_{\text{Nd}(t)}$ values. Data for the Late Archean Upper Continental Crust (LAUCC), MORB, and Arc Andesite are from Taylor and McLennan (1985), and Sm-Nd isotopic data for the Gabon basement rocks (pink granite and grey TTG) are from Thiéblemont et al. (2009).

Figure 12. Trace elements tectonic discriminant plots for the studied sediments (after Bhatia and Crook, 1986): (A) La-Th-Sc ternary diagram, (B) Th-Sc-Zr/10 ternary diagram, (C) La/Y versus Sc/Cr diagram, and (D) Ti/Zr versus La/Sc diagram. PM: Passive Margin, ACM: Active Continental Margin, CIA: Continental Island Arc, and OIA: Oceanic Island Arc.

List of tables

Table 1. Sm-Nd whole-rock isotope data for the FB₁ Member and FA Formation sediments. Sm-Nd isotopic data for Gabon basement rocks (pink granite and grey TTG) are from Thiéblemont et al. (2009).

Table 2. Modelled fractions of compositionally different Archean crust in the analyzed samples.

Appendix

Figure A1. Major element spider diagrams of (A) FB₁ Member mudstones, (B) FA Formation mudstones, and (C) FA Formation sandstones, normalized to the composition of the Late Archean Upper Continental Crust (LAUCC; from Condie, 1993).

Figure A2. Plot of Sm/Nd ratios versus $^{143}\text{Nd}/^{144}\text{Nd}_{(2.1 \text{ Ga})}$. Sm-Nd isotopic data for Gabon basement rocks (pink granite and grey TTG) are from Thiéblemont et al. (2009).

Table A1. Whole-rock major element geochemical data for the studied sediments. CIA (Chemical Index of Alteration) = $(\text{Al}_2\text{O}_3/(\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O})) \times 100$, where CaO* correspond to CaO content corrected for CaO concentrations in carbonate and apatite.

Table A2. Whole-rock trace element geochemical data for the studied sediments.

Table A3. Whole-rock Rare Earth Element geochemical data for the studied sediments. Eu anomaly, $\text{Eu}/\text{Eu}^* = \text{Eu}_\text{N}/(\text{Sm}_\text{N} \cdot \text{Gd}_\text{N})^{0.5}$ (Taylor and McLennan, 1985). N refers to the chondrite-normalized value.

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Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Figure A1

Figure A2

Highlights

- Sediments have undergone moderate weathering and secondary alteration
- Sediments mainly derived from felsic source with minor mafic components
- Provenance from Mesoarchean East Gabonian block with no juvenile contribution
- Sediments sourced from crustal addition and recycling of differentiated granitoids

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Journal Pre-proofs