

UNDERSTANDING ARC-CONTINENT COLLISION AND CRUSTAL GROWTH: GEOCHEMISTRY OF PHILIPPINE SEDIMENTARY ROCK SEQUENCES

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Abstract

This work synthesizes the results of studies utilizing whole rock sedimentary geochemistry of samples collected from different tectonic settings. Major, trace and rare earth element diagrams effectively discriminate the nature of the source rocks and tectonic setting of the sedimentary rocks. The continental character of Palawan, Buruanga and Mindoro are affirmed by their geochemical components which point to felsic and quartzose recycled source rocks in a continental setting. The Antique samples, when plotted on the various diagrams, are shown to have contributions from both mafic and felsic sources. This indicates their deposition in an overlap basin which marks province linking resulting from the collision of the two blocks – Palawan Microcontinental Block and Philippine Mobile Belt. In stark contrast, the sedimentary rocks from Baguio (Klondyke, Amlang and Cataguingan) have geochemical signatures consistent with their derivation from mafic igneous rocks in an oceanic island arc setting. The variety of diagrams used in this study illustrates their effectiveness in characterizing the provenance and tectonic setting of sedimentary rocks. These results underscore the value of using whole rock sedimentary geochemistry in deciphering the evolution of the Philippine island arc system.

Keywords: Geochemistry of sedimentary rocks, Philippines, Provenance, Tectonic setting

Introduction

The nature and origin of accreted terranes, involving collision and suturing, is an important aspect in understanding the growth and composition of continental crust. The composition of the upper continental crust has been determined primarily by direct sampling of rocks exposed on the surface (e.g. Taylor and McLennan, 1985; Condie, 1993; Gao, 2010). Substantial information, however, has also been gleaned from the determination of the geochemical composition of insoluble and immobile elements in clastic rocks. This leads to an inference of the composition of the rocks at the source region (e.g. Jian et al., 2013; Liu et al., 2013; Roy and Roser, 2013).

The Philippine island arc system is a good laboratory for testing the viability of using geochemical data to extract this information. Fragments of diverse origin which have come together as a result of the collision of a continental fragment with the Philippine Mobile Belt make up the Philippine island arc system. Understanding the geologic evolution of this island arc system had previously been challenging owing to the difficulty in establishing which of these fragments form part of the continental block. Recent geological and geochemical investigations carried out in Central Philippines have led to the identification of various lithologic units which allow for a discrimination of their continental or arc affinity (Yumul et al., 2003; Dimalanta et al., 2009; Gabo et al., 2009; Concepcion et al., 2012).

In much the same way that the geochemical signatures of magmatic rocks provide information on the processes leading to their formation and evolution, the relationship between the geochemical composition of sedimentary rocks and plate tectonics has now been

firmly established. An increasing number of studies which make use of the major, trace and rare earth element data have documented this relationship between the geochemical composition of sedimentary rocks and the nature of their source rocks as well as the tectonic setting (e.g. Dickinson and Suczek, 1979; Basei et al., 2011; Yang et al., 2012; Tao et al., 2013). Moreover, the geochemical results have also been increasingly valuable in providing clues on the degree of weathering at the source area as well as in deciphering the climatic conditions responsible for the degree of chemical weathering (e.g. Goldberg and Humayun, 2010; Mishra and Sen, 2012).

With this end in view, a database consisting of the geochemical signatures of sedimentary rocks was initiated. To date, clastic rocks have been collected from several areas representing different terranes within the Philippine island arc system. These include continental terrane (Palawan and Mindoro), collision zone (Buruanga, Antique) and oceanic arc setting (Baguio) (Figure 1). The whole rock geochemical compositions of these sedimentary rocks are presented in this study.

Terranes of the Philippines

The Philippine island arc system has traditionally been subdivided into two blocks – the Palawan Microcontinental Block and the Philippine Mobile Belt. The oldest lithologic units in the Philippine island arc system spanning from Carboniferous to Eocene are exposed in the Palawan Microcontinental Block. The Philippine Mobile Belt, in contrast, is made up of ophiolitic and island arc rocks, the oldest of which are aged Cretaceous (e.g. Yumul et al., 2009) (Figure 1).

Continental Fragments: Palawan and Mindoro

Located at the westernmost side of the Philippine island arc system is the northeast-southwest-trending Palawan island (Figure 1). Representing a fragment rifted from the southeastern margin of mainland Asia during the opening of the South China Sea, Palawan island exposes some of the oldest lithologic units in the Philippine island arc system. These include Middle to Late Jurassic olistostromes, Permian and Triassic cherts, sandstones and basaltic rocks which are exposed in northern Palawan. An Upper Cretaceous to Eocene sedimentary sequence in thrust contact with an ophiolite complex is observed in central to southern Palawan (Suzuki et al., 2000).

Suzuki et al. (2000) analyzed sandstones from the Upper Cretaceous to Eocene sequence, the Babuyan River Turbidites. This turbiditic sequence comprising of alternating beds of sandstone and mudstone displays parallel laminations, horizontal and cross-laminations, current ripple laminations. These features are believed to represent portions of a Bouma sequence although lacking the graded bedding typically exhibited by a complete turbidite sequence. The medium- to coarse-grained sandstones contain a significant amount of quartz grains (39-46%) and 24-27% acidic volcanic rock fragments.

Lying northeast of Palawan island is Mindoro Island which has also been investigated by previous workers as they tried to establish its geologic affinity (Figure 1). One group argued that it is only the southwestern part of the island which is of continental affinity in contrast with other models suggesting that it is the whole island which is continent-derived (e.g. Taylor and Hayes 1983; Sarewitz and Karig 1986; Concepcion et al., 2012).

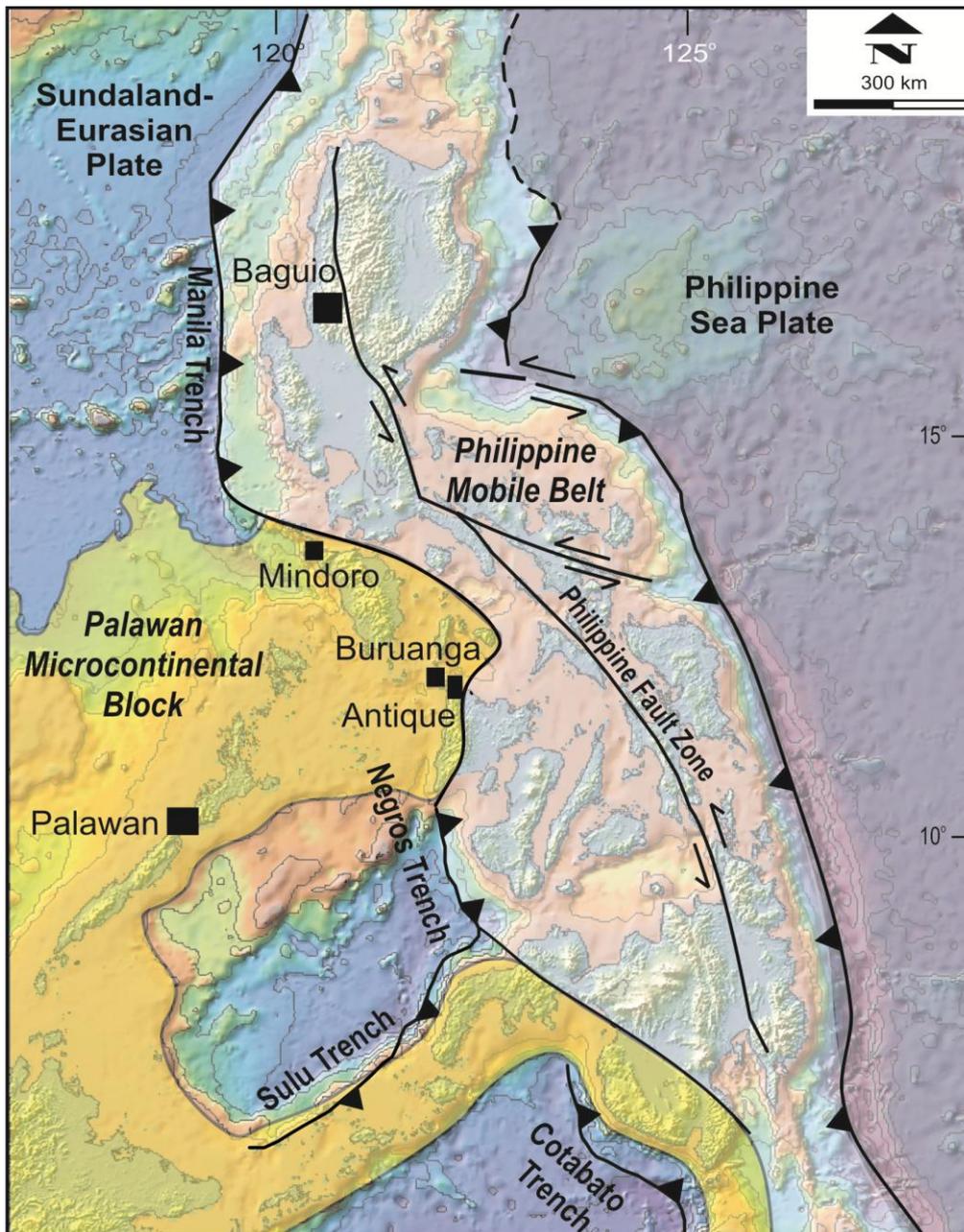


Figure 1. Map showing the tectonic features of the Philippine island arc system formed through the collision between the Philippine Mobile Belt and the Palawan Microcontinental Block. Black squares show the areas where clastic rocks were collected and analyzed for their geochemical compositions.

Northwest Mindoro exposes a Jurassic metamorphic complex overlain by a Late Eocene – Oligocene sedimentary sequence. Thrust onto the older rocks is a Middle Oligocene oceanic lithospheric fragment, the Amnay Ophiolite Complex. This is blanketed by a Plio-Pleistocene sequence of tuffaceous sandstone and siltstone interbeds (Concepcion et al., 2012). In this study, clastic rock samples were collected from the Late Eocene – Oligocene Lasala Formation that is widely exposed in the northwestern tip of Mindoro island. This sedimentary sequence comprises of gray sandstones and gray mudstone interbeds. Minor occurrences of basalt flows, limestones and conglomerates have also been reported. The nannofossil assemblage extracted (i.e. *Sphenolithus* spp., *Coccolithus pelagicus*,

Cyclicargolithus floridanus, *Dictyococcites bisectus/Reticulofenestra bisecta*, and *Reticulofenestra umbilicus*) from the shales and mudstones delimit the age of the Lasala Formation to Late Eocene – earliest Oligocene (Concepcion et al., 2012). The medium-grained sandstones from Northwest Mindoro, similar to the Palawan sandstones, are dominantly made up of quartz grains.

Collision Zone: Buruanga and Antique

The Buruanga Peninsula in Panay island, which lies southeast of the Mindoro island, is believed to represent the leading edge of the continental fragment, the Palawan Microcontinental Block (Figure 1). Exposed here are old lithologic units similar to those found in Palawan island, i.e. Middle to Late Jurassic chert beds, clastic sequences and crystalline limestone. Also exposed in the peninsula are metamorphic rocks (Libertad Metamorphics) which outcrop at the boundary of the chert-clastic-limestone sequence. The limestone unit is cut by a Middle Miocene intrusive, the Patria Diorite. East of the Buruanga Peninsula lies the northern part of the Antique Range where ultramafic rocks associated with the Antique Ophiolite Complex were mapped. This is capped by the Middle Miocene Fragante Formation and the late Middle Miocene Lagdo Formation (e.g. Zamoras et al., 2008; Gabo et al., 2009).

Samples from the sedimentary sequences in the Buruanga Peninsula (Middle to Late Jurassic Saboncogon Formation) as well as clastic units from the Antique Range (Middle Miocene Fragante Formation) are part of the database used in this study. In Buruanga Peninsula, less indurated mudstones are interbedded with fine- to coarse-grained highly indurated sandstones. An examination of the sandstone samples revealed the presence of 36-57% quartz, 30-47% lithic fragments and some feldspar grains (4-12%) (Gabo et al., 2009). This sequence has been assigned a late Middle Jurassic to early Late Jurassic age based on the radiolarians extracted from the mudstones (Zamoras et al., 2008). The sedimentary sequence exposed in the northern Antique Range, the Fragante Formation, was also sampled and made part of this database. This package is made up of pyroclastic flow deposits overlain by conglomerates, sandstones and minor limestones. The sandstones analyzed in this study are fine- to medium-grained and occur as thick beds (30 to 100 cm thick) (Zamoras et al., 2008; Gabo et al., 2009). In stark contrast with the sandstones collected from the Buruanga Peninsula, the Fragante sandstones contain minimal quartz (less than 20%), 4-25% feldspars with a preponderance of volcanic lithic fragments (>50%) (Gabo et al., 2009).

Oceanic Arc Setting: Baguio

The geochemical data for an oceanic island arc setting were collected from sedimentary sequences from the Baguio Mineral District (Figure 1). Based on the geochemical signatures of the igneous rocks exposed in the area, the area was recognized to have evolved from a marginal oceanic basin platform, dominated by basaltic rocks, to an island arc setting characterized mostly by more silicic rocks (e.g. Tam et al., 2005; Yumul et al., 2008).

The Baguio Mineral District is probably one of the well-investigated areas in the Philippines. Its geology as described in various publications is summarized to comprise of a basement represented by the Lower Paleogene Pugo Metavolcanics. This is overlain by Oligocene to Pleistocene sedimentary sequences which were deposited in marine to terrestrial settings. Miocene to Pleistocene island-arc related dioritic to gabbroic plutons to adakitic lavas (e.g. Yumul et al., 2008).

One of the sedimentary sequences sampled for this study is the Middle to Late Miocene Klondyke Formation which consists of massive to thickly bedded polymictic conglomerates. These grade into interbeds of thinly- to thickly-bedded medium- to coarse-grained sandstones

and siltstones. Further west of the Baguio Mineral District, the sequence is dominated by thin beds of shales, siltstones, sandstones and some conglomeratic lenses (e.g. De Leon et al., 1991). These units that make up the Klondyke Formation represent the proximal, midfan and distal facies of a submarine fan deposit (De Leon et al., 1991). Sandstones examined for this study comprise of 10-20% quartz and 25-50% plagioclase grains.

The Amlang Formation, comprising of a thick sequence of rhythmically interbedded sandstones, siltstones, shales with minor conglomerates, is observed to the west of the Baguio Mineral District. Graded bedding, parallel and cross laminations indicate deposition in a deep marine environment (e.g. Lorentz, 1984). This is overlain by the Late Pliocene Cataguintingan Formation which is made up of tuffaceous sandstones, siltstones, shales and conglomerates with minor limestone lenses (e.g. Lorentz, 1984). The sandstone, siltstone and mudstone samples for this study were collected from the Klondyke, Amlang and Cataguintingan Formations.

Analytical Methods

Over the past years, we have developed a database comprising of the major, trace and rare earth element geochemical data of clastic rocks from areas representing different tectonic settings in the Philippine island arc system. The geochemical data are from: sandstones from Palawan previously reported by Suzuki et al. (2000); fine- to medium-grained sandstones, siltstones and shales from Northwest Panay (including those reported by Gabo et al., 2009); mudstones, fine- to medium-grained sandstones from Northwest Mindoro (reported by Concepcion et al., 2012); shales, siltstones, fine- to medium-grained sandstones from the Baguio district (including those reported by Tam et al., 2005). The major element compositions were determined using X-Ray Fluorescence spectrometry at the Okayama University and Kyushu University in Japan. Accuracy is within 1%.

Continental vis-à-vis Island Arc Affinity

There have been an increasing number of geochemical studies which rely on major element compositions of sedimentary rocks as indicators of provenance and tectonic setting (e.g. Bhatia, 1983; Roser and Korsch, 1986; Mishra and Sen, 2012; Roy and Roser, 2013).

Some of the most commonly used provenance and tectonic setting discriminants are used in this study to see how well the diagrams will be able to classify the collected samples.

F1 Versus F2 Diagram

Two versions of F1 versus F2 discriminant diagrams based on the concentrations of Al_2O_3 , TiO_2 , Fe_2O_3 , MgO , CaO , Na_2O and K_2O have been widely used in geochemical investigations. Bhatia (1983) offered these equations to obtain F1 and F2: $F1 = 30.638\text{TiO}_2/\text{Al}_2\text{O}_3 - 12.541\text{Fe}_2\text{O}_3/\text{Al}_2\text{O}_3 + 7.329\text{MgO}/\text{Al}_2\text{O}_3 + 12.031\text{Na}_2\text{O}/\text{Al}_2\text{O}_3 + 35.402\text{K}_2\text{O}/\text{Al}_2\text{O}_3 - 6.382$ and $F2 = 56.5\text{TiO}_2/\text{Al}_2\text{O}_3 - 10.879\text{Fe}_2\text{O}_3/\text{Al}_2\text{O}_3 + 30.875\text{MgO}/\text{Al}_2\text{O}_3 - 5.404\text{Na}_2\text{O}/\text{Al}_2\text{O}_3 + 11.112\text{K}_2\text{O}/\text{Al}_2\text{O}_3 - 3.89$. When plotted on the F1-F2 diagram, samples will occupy distinct provenance fields, i.e., quartzose sedimentary provenance, mafic igneous provenance, intermediate igneous provenance and felsic provenance.

In the F2 versus F1 diagram (Bhatia, 1983) (Figure 2), the Palawan samples occupy the felsic igneous provenance field. The Buruanga and Northwest Mindoro samples plot in the quartzose sedimentary provenance, intermediate igneous provenance and felsic igneous provenance fields. The Antique samples straddle both the mafic igneous provenance and intermediate igneous provenance fields. The Baguio (Klondyke, Amlang and Cataguintingan

Formations) samples are consistent with a dominant source coming from mafic igneous rocks.

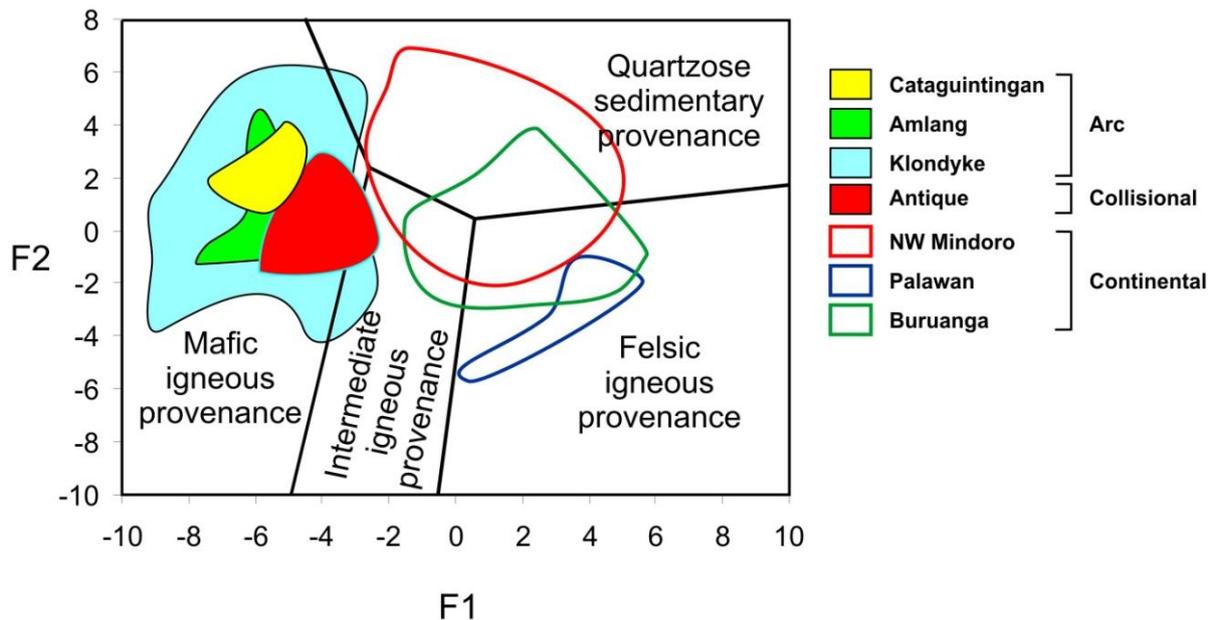


Figure 2. Discriminant diagram to distinguish the nature of the source rocks as proposed by Bhatia (1983). The Baguio (Cataguintingan, Amlang and Klondyke) and Antique clastic rocks plot in the mafic igneous provenance field whereas the Mindoro, Palawan and Buruanga samples occupy the quartzose sedimentary and felsic igneous provenance fields.

In another variation of this diagram, the same major element compositions were used but this time computing F1 and F2 following the equation proposed by Roser and Korsch (1988). The discriminants are computed as: $F1 = 1.773 \cdot TiO_2 + 0.607 \cdot Al_2O_3 + 0.760 \cdot Fe_2O_{3T} - 1.500 \cdot MgO + 0.616 \cdot CaO + 0.509 \cdot Na_2O - 1.224 \cdot K_2O - 9.090$ and $F2 = 0.445 \cdot TiO_2 + 0.070 \cdot Al_2O_3 - 0.250 \cdot Fe_2O_{3T} - 1.142 \cdot MgO + 0.438 \cdot CaO + 1.475 \cdot Na_2O + 1.426 \cdot K_2O - 6.861$.

These parameters allow a distinction among four primary provenance groups: P1 – primarily mafic and lesser intermediate igneous provenance; volcanic lithic are predominantly basaltic-intermediate composition; P2 – primarily intermediate igneous provenance; mostly andesitic volcanic lithic fragments; more evolved volcanic fragments (dacite, rhyolite and trachyte) may be present; P3 – felsic igneous provenance; intermediate-acid volcanic component is fewer; and P4 – quartzose sediments of mature continental derivation; primarily quartz-rich, feldspar- and lithic-poor (e.g. Rahman and Suzuki, 2007; Hossain et al., 2010).

From the resulting F1 and F2 values, the Philippine samples are grouped in distinctly separate fields with the Palawan samples plotting mostly in the P3 field (primary felsic sources) whereas the Buruanga and Northwest Mindoro samples occupy the P3 and P4 (quartzose recycled) fields. In contrast, the Amlang and Cataguintingan samples from Baguio are found in the P1 (primary mafic sources) field. The Klondyke and Antique samples plot across the P1 and P2 fields (Figure 3).

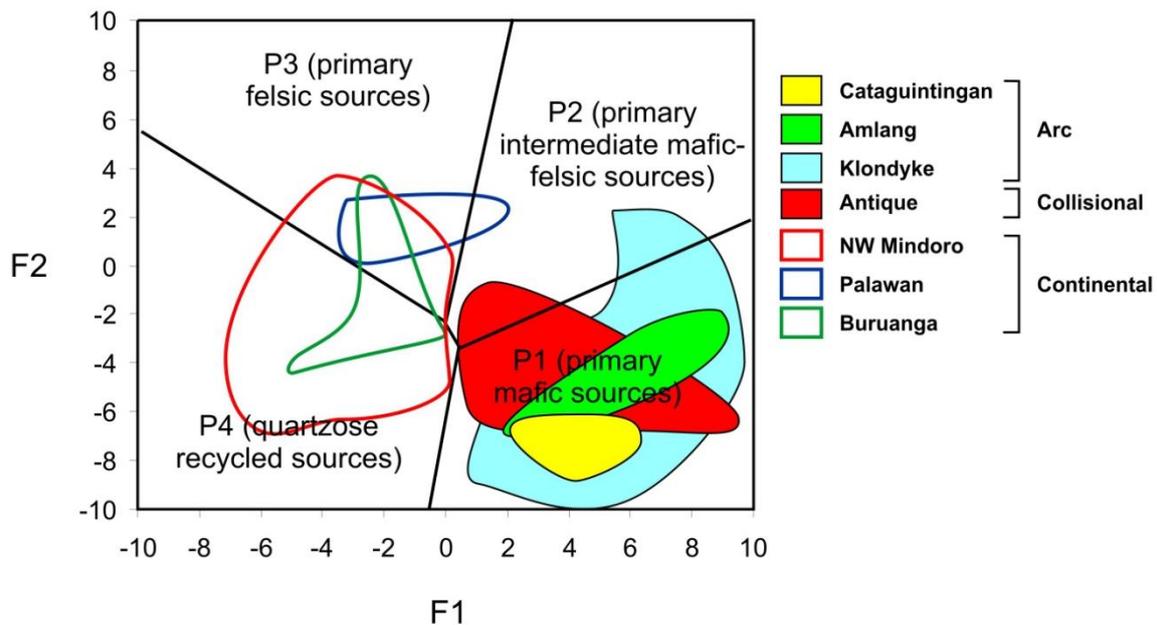


Figure 3. Discriminant diagram to distinguish the nature of the source rocks using the F1-F2 discriminants of Roser and Korsch (1988).

K₂O/Na₂O Versus SiO₂

Roser and Korsch (1986) also proposed the $K_2O/Na_2O - SiO_2$ systematics as tectonic setting indicators to discriminate between oceanic island arc to active continental margin to passive margin settings (e.g. Mader and Neubauer, 2004; Spaletti et al., 2005; de Araujo et al., 2010). High SiO_2 and K_2O/Na_2O values characterize sediments from passive margin (PM) settings. These include stable continental areas, plate interiors, Atlantic-type continental margins, failed rifts or grabens or depocenters along continental margins. The active continental margin (ACM) includes areas adjacent to active plate boundaries or subduction-related basins, continental collision basins and pull-apart basins associated with strike-slip fault zones (e.g. Roser and Korsch, 1986). Evolved arcs (i.e. ACM, PM) are characterized by high K_2O/Na_2O consistent accompanied by the presence of felsic components. Conversely, sediments derived from oceanic island arcs will likely be enriched in mafic components and will display low SiO_2 and K_2O/Na_2O values (e.g. Batumike et al., 2006; Yang et al., 2012; Verma and Armstrong-Altrin, 2013).

The SiO_2 - K_2O/Na_2O signatures of the Palawan, Buruanga and Northwest Mindoro samples are similar to those displayed by modern sediments deposited at active continental margin/passive margin settings. The Antique and Baguio samples (Klondyke, Amlang and Cataguintingan Formations), which are characterized by low SiO_2 and K_2O/Na_2O values, plot within the ARC field of this diagram (Figure 4).

La/Th Versus Hf

To confirm the results shown by the major element concentrations, some scientists began working with trace and rare earth elements to obtain clues on the provenance and tectonic setting of sedimentary rocks. The rationale behind this is the relatively low mobility of these elements during sedimentary processes. Moreover, these elements are quantitatively incorporated in clastic sedimentary rocks during weathering and transport. Several high field strength elements such as Hf, Th, Zr, Nb and Y have been established to be greater in felsic rocks relative to mafic rocks. In contrast, Co, Sc and Cr occur in greater amounts in mafic

rocks than felsic rocks. These features together with their relatively immobile nature make them suitable indicators of the composition of the parent material (e.g. de Araujo et al., 2010; Jian et al., 2013). There were initially relatively few studies employing trace elements in the geochemical investigation of sedimentary rocks. However, such studies have increased significantly over the past few decades after the work by Bhatia and Crook (1986) showed how valuable trace elements were in distinguishing the source rocks and tectonic setting of sedimentary rocks.

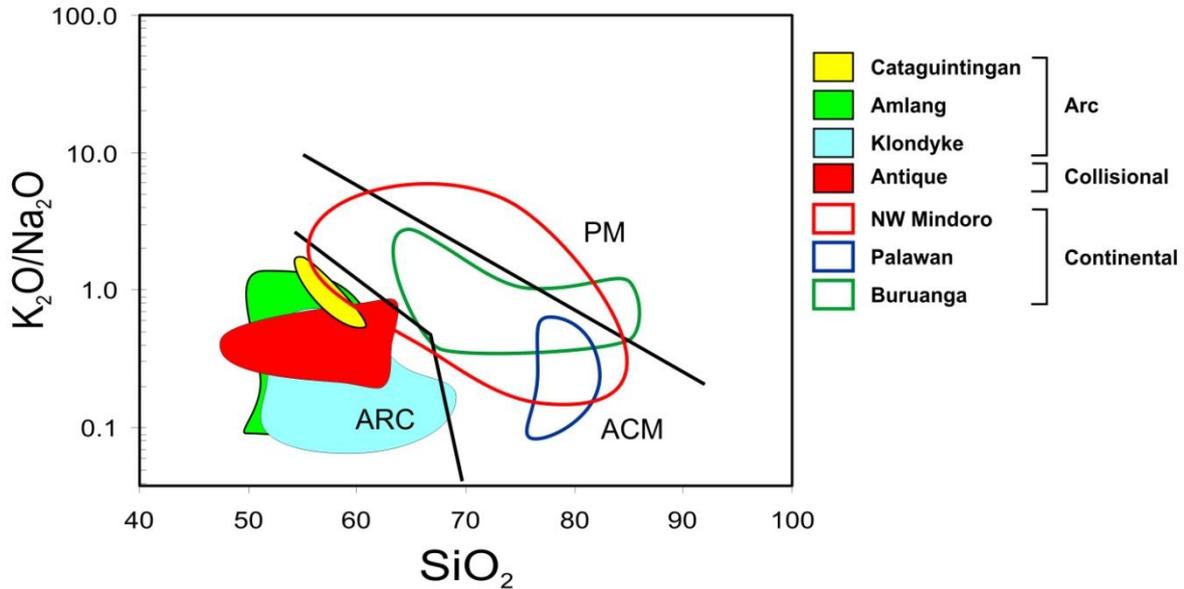


Figure 4. Tectonic setting diagram proposed by Roser and Korsch (1986) to discriminate between oceanic island arc (ARC), active continental margin (ACM) and passive margin (PM) settings.

One such diagram was proposed by Floyd and Leveridge (1987) and relies on the correlation between La/Th and Hf to classify source rock compositions. The degree of recycling in sediments is reflected by Hf whereas La/Th gives an indication of the degree of differentiation of magma. More evolved magmas will be characterized by lower La/Th ratios (e.g. Basei et al., 2011; Drobe et al., 2011; Jian et al., 2013). The samples from Buruanga are characterized by low Hf and La/Th ratios indicating components derived from an acidic arc source with some contributions from mixed felsic/basic source rocks. The Northwest Mindoro samples display low La/Th (<5) and Hf values and cluster near the composition of upper continental crust indicating felsic sources. In contrast, the Baguio samples (Klondyke, Amlang and Cataguintingan Formations) have Hf values <3 but the La/Th ratios spread from 4-14 (Figure 5). These values are consistent with materials sourced from andesitic arc to tholeiitic ocean island sources.

Th – Co – Zr/10

Using the Th, Co and Zr/10 concentrations in clastic rocks, Bhatia and Crook (1986) were able to show distinct fields representing four different tectonic settings. The oceanic island arc (OIA) and continental island arc (CIA) fields in the Th-Co-Zr/10 discrimination diagram correspond to convergent plate margins where orogenic volcanic rocks are formed. They defined the active continental margin (ACM) field to include Andean-type continental margins, characterized by plate convergence and orogenic volcanic rocks, and strike-slip continental margins. Depositional basins related to active continental margins are typified by

thick and elevated continental crust. Rifted continental margins of the Atlantic-type formed along the edges of continents, inactive or extinct convergent plate margins and sedimentary basins on trailing continental margins supplied from collisional orogens all fall within the passive margin (PM) field (e.g. Augustsson and Bahlburg, 2008; Yan et al., 2012).

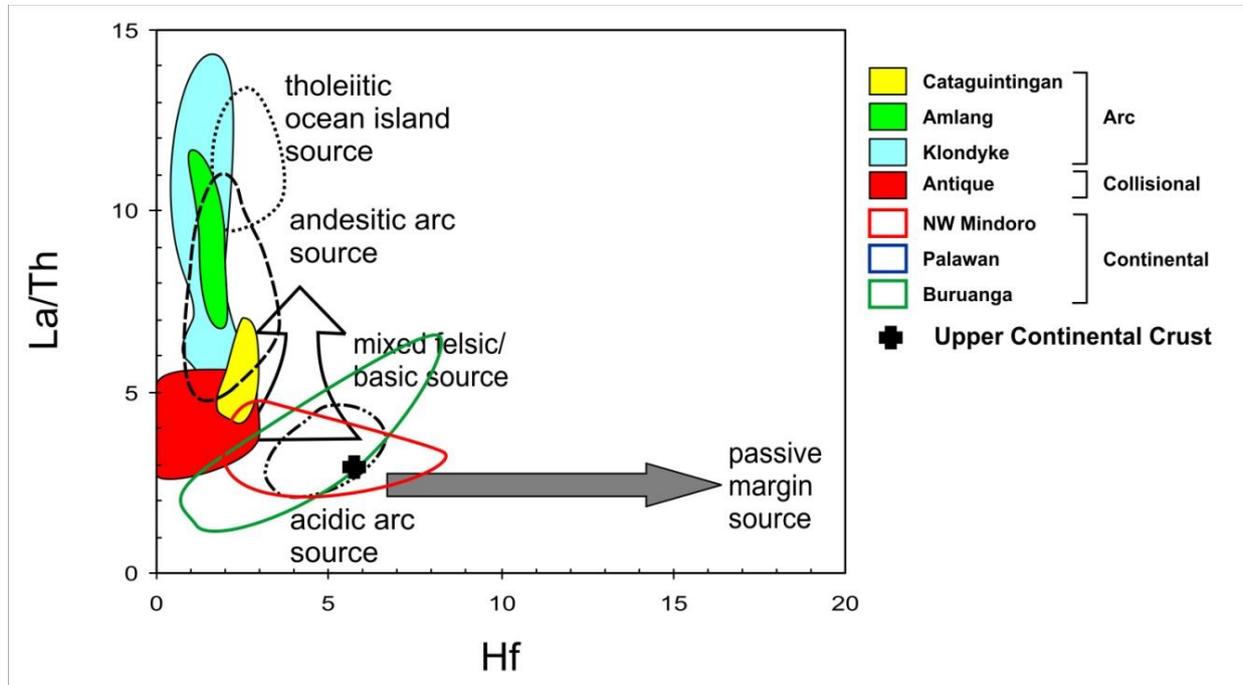


Figure 5. Provenance diagram using La/Th ratios versus Hf (Floyd and Leveridge, 1987).

In this study, we plotted our data on the Th-Co-Zr/10 discrimination diagram of Bhatia and Crook (1986). The Baguio and Antique clastic rocks are characterized by low Th and Zr values. The higher amounts of Co consistent with their oceanic island arc setting led the samples to plot in or adjacent to the OIA field. The Buruanga and Northwest Mindoro samples have higher Th and Zr concentrations. As a result, the Buruanga samples occupy the CIA field whereas the Northwest Mindoro samples straddle the CIA and PM fields (Figure 6).

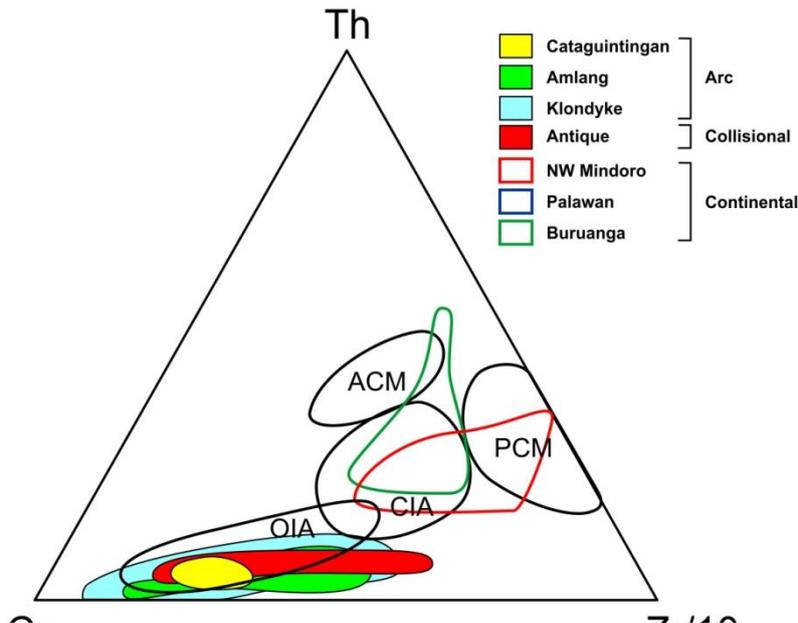


Figure 6. Tectonic setting discriminant diagram using Th-Co-Zr/10 (diagram after Bhatia and Crook, 1986).

Synthesis

The geochemical compositions of clastic rocks collected from several sites representing different tectonic settings were examined. Based on the work done by Suzuki et al. (2000), the well-established continental affinity of Palawan is evident from the whole rock major element compositions displayed by the Palawan clastic rocks. Discriminant diagrams point to the felsic nature of the provenance of the clastic rocks and their deposition in an active continental margin setting. The geochemical data indicate that the Buruanga clastic rocks were sourced from felsic igneous and quartzose recycled rocks in a continental margin setting. The clastic rocks sampled from Northwest Mindoro share similar geochemical signatures as the Palawan and Buruanga clastic units (Concepcion et al., 2012). As a result, this has provided additional proof that Mindoro is part of the continental fragment that broke off from mainland Asia.

Being situated within a collision zone, the clastic rocks from Antique (east of the Buruanga Peninsula) display geochemical signatures which indicate that their components were derived from both mafic and felsic sources. This is consistent with this sedimentary sequence being deposited in the overlap basin that marks the suture zone produced during the collision between the Palawan microcontinental block and the Philippine Mobile Belt (Gabo et al., 2009).

The variety of discriminant diagrams, binary and ternary plots used in this work have clearly characterized the rocks from the Klondyke, Amlang and Cataguintingan Formations to have been derived from mafic sources in an oceanic island arc setting (Dimalanta et al., 2013).

Conclusions

This study shows that the use of geochemical diagrams in determining the provenance and tectonic setting of clastic rocks from the Philippine island arc system is applicable. The whole rock geochemistry of the sedimentary rocks provides an additional tool in the delineation of how the different terranes of the Philippine island arc system evolved. Based

on the geochemistry of the Palawan, Buruanga and Northwest Mindoro sedimentary rock samples, these areas are part of the continental fragment which broke off from mainland Asia. The two source rock signatures displayed by the Antique samples point to their derivation from felsic and mafic igneous rocks which were eroded and deposited into the depositional basin. This provides the time line for province linking in this area situated within a collision-related suture zone. The mafic source and oceanic island arc setting for the Baguio area is clearly manifested by the whole rock geochemical signatures of its sedimentary rocks. The geochemical signatures of sedimentary rocks provided valuable information on the nature and affinity of terranes which became amalgamated and led to the growth of the Philippine island arc system.

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