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Chemostratigraphy as a Tool for Determining Depositional Ages of Metamorphosed Carbonate Rocks Prior to Supercontinent Formation

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Abstract | Chemostratigraphy or chemical stratigraphy deals with the correlation of sedimentary strata based on systematic variation of a particular chemical composition with time in the history of the Earth. In particular, this method is widely used in decoding the temporal variations seen in marine sediments that are deposited uninterruptedly in deep sea or in shallow marine carbonate depositional environments. In this review, the application of strontium and carbon isotope based chemostratigraphy in marine carbonate sediments is discussed. Irrespective of post depositional geological events such as metamorphism or tectonic displacements to form Mountain chains during continental collision, chemostratigraphy helps in determining the apparent depositional ages of sedimentary rocks in the Precambrian time, where biostratigraphy is not applicable. Although carbonate rocks are vulnerable to post depositional alterations, a systematic geochemical screening can guide in identifying the best chemically preserved carbonate rocks. A novel method of using Mn/Sr ratios for determining the best estimate of strontium initial ratio for carbonate rocks is presented, on the basis of a worked example of metamorphosed carbonate rocks from the Sør Rondane Mountains in the Dronning Maud Land, East Antarctica. The future potential of chemostratigraphy in decoding the early history of the Earth is also discussed.

Keywords: *Chemostratigraphy, Strontium isotopes, Carbonate rocks, Supercontinent*

1 Introduction

The age of a rock formation is one of the fundamental information that is necessary for any geological studies. Without such information we are unable to understand the processes that occurred through the 4.56 billion year protracted history of Earth. Fortunately, geologists are equipped with the latest technological developments that help determine the age recorded in minerals such as zircons by advanced analytical techniques of SHRIMP/SIMS (Sensitive high Resolution Ion MicroProbe/Secondary Ion Mass Spectrometer), which is a double-focusing secondary ion mass spectrometer instrument (Matsuda, 1974) used for the measurement of isotopes, and LA-ICPMS (Laser ablation-inductively coupled plasma mass spectrometer), which is a technique used for the measurement of elemental and isotope composition. These methods use the isotope ratios of uranium and lead, leading to the estimation of an absolute age of crystallization of a mineral using a concordia diagram (Wetherill, 1956). By employing absolute dating methods on minerals formed during magmatism and high-temperature metamorphism (a process that undergoes inside the Earth whereby minerals change to new ones by solid state transformation), it is possible to understand the most prominent geologic events in the Earth's history, right from a magma ocean

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environment at the initial stage of accretion and differentiation of the Earth, through the formation and stabilization episodes of continental crust, and to the processes relating to the present day global plate tectonic regimes. However, geological (and biological) evolution has much more to deal with than the high-temperature processes such as magmatism and metamorphism, both of which are temporally episodic and spatially restricted. We live on a planet where the surface processes, especially those related to 'liquid water', shaped up the Earth's topography and made environment a habitable one. In this respect, weathering, erosion and sedimentation are processes that need prime attention in an evolving Earth.

2 Startigraphy and Chemostratigraphy

Earth's surface processes leave a day to day record in the sedimentary strata throughout the history, some of which are preserved in the rocks surrounding us. Geologists use them as records for understanding the sequence of events and correlate them across the globe to understand temporal relations between sedimentary strata, termed as stratigraphy. They also compare them to identify specific global events, such as glaciation episodes and global warming events and so on (e.g. Snowball earth hypothesis; (Kirschvink, 1992; Harland, 1964); Cretaceous—Paleogene extinction event (Alvarez et al., 1980).

Estimating the age of an Earth surface/near surface process is a difficult task. In the Phanerozoic eon (between present day and 541 ± 1 million years ago; Cohen et al., 2013), fossil records help us in a great deal to understand the age relations in terms of biological evolution and biostratigraphy (assigning relative ages of rock strata by using the fossil assemblages). Prior to the Cambrian (the era defined by appearance of the first multicellular organisms) and the biologic explosion, i.e, in the Precambrian, (the time span in the Earth's history from origin at 4,567 million years to 541 million years; Cohen et al., 2013), constraining the timing of Earth's surface processes is extremely difficult because of the sparse fossil activity. Decoding the age of sedimentary rock records has been restricted to starta that contain volcanic marker horizons that can be dated using zircon U-Pb geochronology. One of the potential ways to overcome this difficulty is to use the temporal variations in the chemistry of sedimentary strata, especially for minerals precipitated directly from seawater, which is defined as "chemical stratigraphy" or "chemostratigraphy" (Berger and Vincent, 1981). This method is employed to correlate temporal variations in sediment chemistry within a depositional basin as well as basins across the globe (Renard et al., 2008). Especially, stable and radiogenic isotope geochemistry of chemically deposited carbonate rocks are extensively used for reconstructing paleoenvironments, determining the tectonic setting of sedimentary basins, indirect estimation of depositional ages, and establishing regional or global correlations (Veizer et al., 1999; Halverson et al., 2005; Halverson et al., 2007; Halverson et al., 2010).

In this review article, first of all I briefly summarize the geochemical proxies that are commonly and efficiently used as chemostratigraphic tools. Thereafter, I introduce a worked example of successful extraction of apparent sedimentary depositional ages from highly metamorphosed and tectonically displaced carbonate rocks form the Sør Rondane Mountains (an isolated inland mountain range in Antarctica located between 22°E and 28°E longitude and 71.5°S and 72.5°S latitude and surrounded by glaciers). This geological entity was formed by the closure of Proterozoic (the time span between 2500 million years and 541 million years; Cohen et al., 2013) ocean and continental collision processes that have obliterated most of the sedimentary imprints. The potential combination of trace and rare earth element geochemistry, stable isotope geochemistry and strontium isotope systematics in identifying the preservation of pre-metamorphic sedimentary isotope signatures is presented here.

3 Strontium Isotope Chemostratigraphy

Marine carbonate rocks deposited directly from sea water are widely used in chemostratigraphy for correlating sequences as well as determining the apparent depositional ages of sedimentary rocks (Trønnes and Sundvoll, 1995; Melezhik et al., 2001; Halverson et al., 2010). Especially, the isotopic composition of strontium is widely used for understanding the chemical composition of paleo-oceans and its evolution through time and space (Veizer et al., 1999; Halverson et al., 2007; Halverson et al., 2010).

Strontium has four naturally occurring stable isotopes (^{84}Sr, ^{86}Sr, ^{87}Sr and ^{88}Sr), of which ^{87}Sr is the daughter nuclide of rubidium (^{87}Rb). The concentration of strontium in naturally occurring rocks vary widely from less than one parts per million in ultramafic rocks (rocks that are common in Earth's mantle and consists almost entirely of ferromagnesian minerals with less than 45% SiO_2 contents) to more than few thousands of parts per million in marine carbonate sedimentary rocks. Seawater acts as the largest reservoir of strontium, by accumulating it from surface runoff throughout the history of the Earth. Strontium in the seawater has a long residence time (~2.4 × 10^6 yr; Jones and Jenkyns, 2001) relative to a short mixing time (~10^5 yr; Kump, 1991; Jacobsen and Kaufman, 1999), and therefore, global ocean is considered to be homogenous with respect to its $^{87}Sr/^{86}Sr$ ratio (Halverson et al., 2007). The $^{87}Sr/^{86}Sr$ ratio of seawater is controlled by the mixing of continental derived strontium, which is radiogenic due to high rubidium content in granitic continental rocks and those from the volcanic sources, especially from the mid-ocean ridge basalts derived from the partial melting (the process though which some minerals in a rock melt initially and accumulate to form a magma) of mantle and are representative of mantle evolution of Rb-Sr isotope system (DePaolo, 1980). Carbonate rocks also act as sources for strontium, where it is recycled. In fact, carbonate minerals, precipitated directly from seawater, contain high concentration of Sr (few hundreds to thousands of ppm on average) and very low contents of Rb (parts per billion levels) resulting in low contribution of radiogenic Sr. Therefore, the $^{87}Sr/^{86}Sr$ ratio of marine carbonate sediments directly reflects contemporaneous sea water composition, irrespective of the age it has formed. This peculiar and systematic geochemical behavior of strontium with time in seawater and its record preserved in carbonate rocks is best suited for chemostratigraphic purpose. Previous studies, as early as in 1970's, have identified the application of strontium isotope evolution in seawater as a potential chronometer for Phanerozoic strata (Faure, 1977). High resolution biostratigraphy and precise age determination in representative geological sections have led to the refinement of the strontium isotope reference curve for the whole Phanerozoic era (Veizer et al., 1999, Figure 1).

However, such information on Precambrian carbonate strata is still sparse, because of the absence of reference geological sections that are precisely dated, as well as the difficulties in preservation of pristine depositional isotope composition for billions of year. Earlier studies have indicated that the strontium isotope evolution has deviated from the mantle evolution curve at around 2.5 billion years (Figure 1-inset, Faure, 1977), where large expanses of continental crust started to contribute to the oceanic strontium reservoir (e.g., Veizer and Compston, 1976; Veizer et al., 1983).

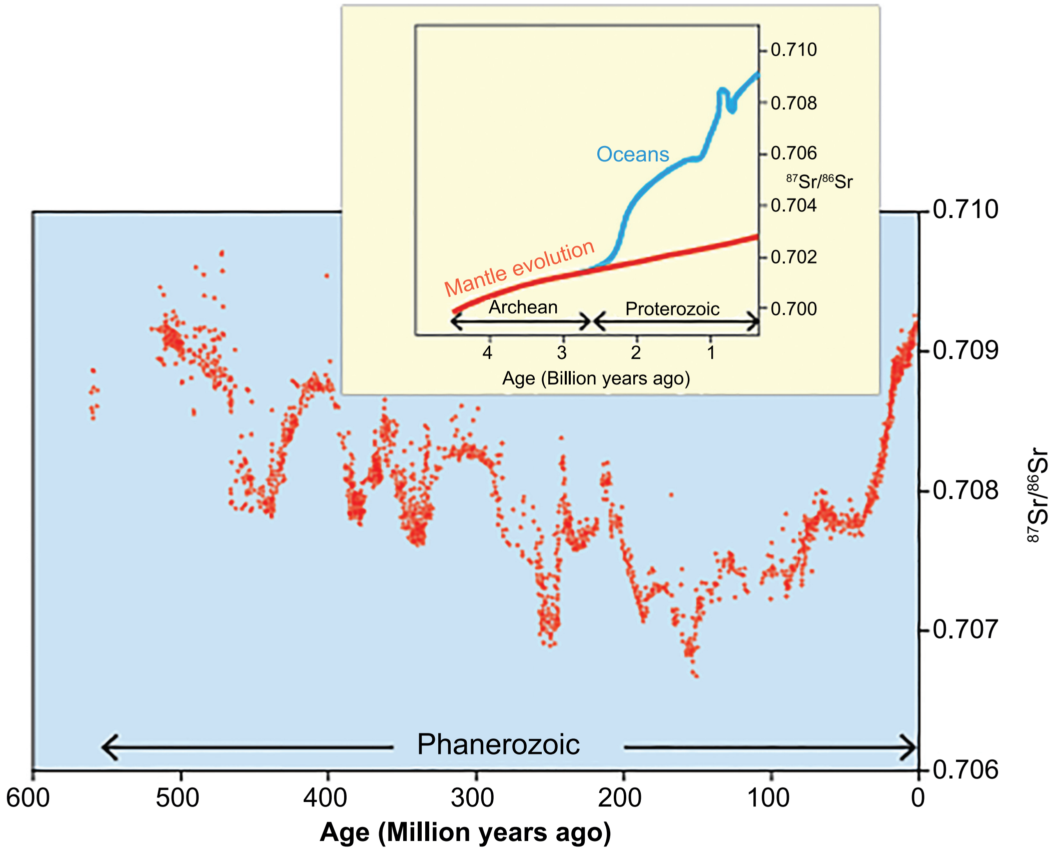

Figure 1: Evolution of strontium isotope ratio with time for Phanerozoic (after Veizer et al., 1999). Inset shows the strontium isotope evolution of Earth's mantle and Oceans (after Faure, 1986).

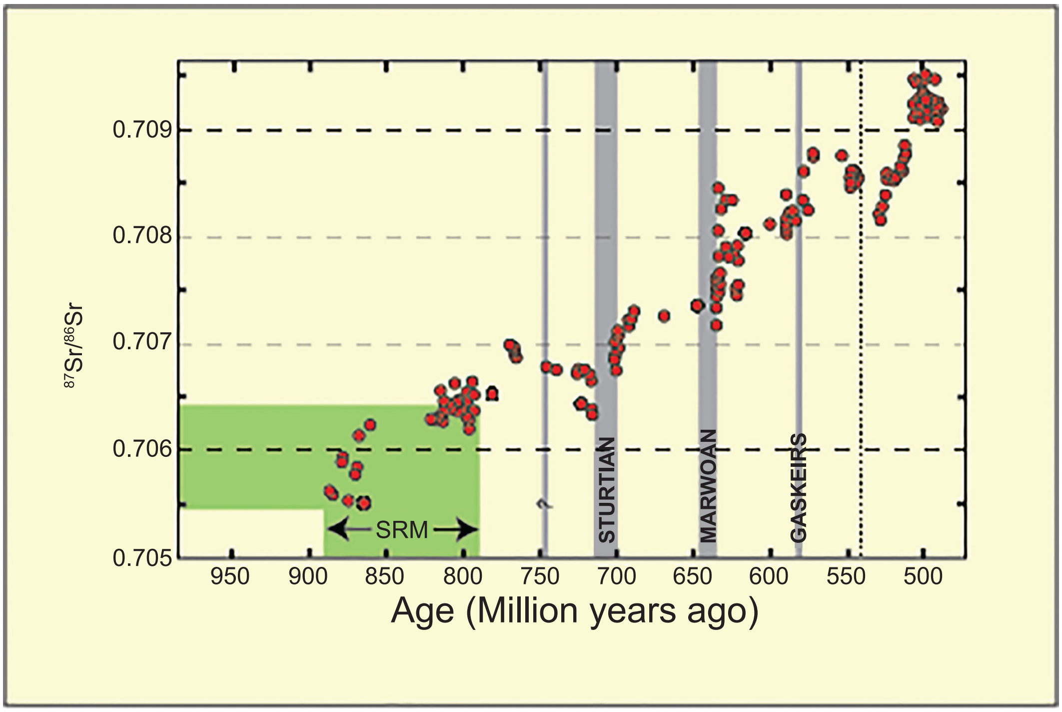

Figure 2: Neoproterozoic chemostratigraphy based on strontium isotope compositions (after Halverson et al., 2010).

A recent compilation of $^{87}Sr/^{86}Sr$ ratios by Halverson and others (2010) from non-metamorphosed carbonate sequences belonging to the Early Neoproterozoic to early-Cambrian strata with precise age information and convincing evidence for the preservation of original geochemical signatures indicate that the strontium isotope ratios have constantly increased with time (Figure 2). Thus, the application of strontium isotope chemostratigraphy has been successfully extended to a larger time span in the Earth's history.

4 Carbon Isotope Chemostratigraphy

The carbon isotopic composition of marine carbonate rocks record the signature of dissolved inorganic pool of carbonate carbon in seawater, except when there are large scale fluctuations in biological productivity. However, it is also highly debated whether carbon isotope ratios of marine carbonates really reflect original seawater composition or not (Bristow and Kennedy, 2008; Knauth and Kennedy, 2009; Frimmel, 2010). Nevertheless, the large volume of reported data indicates that the Neoproterozoic carbonate rocks have positive $\delta^{13}C$ values in general, and large negative anomalies during glacial events (Figure 3) (Knoll et al., 1986; Derry et al., 1989; Kaufman et al., 1991; Des Marais et al., 1992; Shields and Veizer, 2002; Halverson et al., 2005; Halverson et al., 2007; Halverson et al., 2010).

Carbon is a proxy for a variety of environmental factors that controls the biogenic-abiogenic carbon balance in the Earth surface processes (Knoll et al., 1986; Kaufman et al., 1997). Although secular trends in carbon isotope ratios can be applied for chemostratigraphic purposes for shorter time spans (few tens of millions of years) in the Neoproterozoic, recent studies have also indicated decoupling from a simple biologically driven isotopic excursions (Rothman et al., 2003; Knauth and Kennedy, 2009; Derry, 2010). Carbon isotope ratios are relatively resistant to post-depositional alterations, except for isotopic exchange between carbonate carbon and organic carbon or decarbonation reactions during metamorphism (Valley, 2001). Thus, in most cases, carbon isotope ratios of carbonate rocks ($\delta^{13}C_{carb}$) record pristine seawater signals that reflect the secular and temporal variation of global seawater composition, and have been extensively used as a tool not only for paleo-environmental studies but also for identifying excursions that are helpful for chemostratigraphic purpose as time markers.

5 A Case Study of Sør Rondane Mountains, East Antarctica

The Sør Rondane Mountains (SRM), in the Eastern Dronning Maud Land, East Antarctica, form a part of a mountain chain system that developed during the formation of Gondwana supercontinent

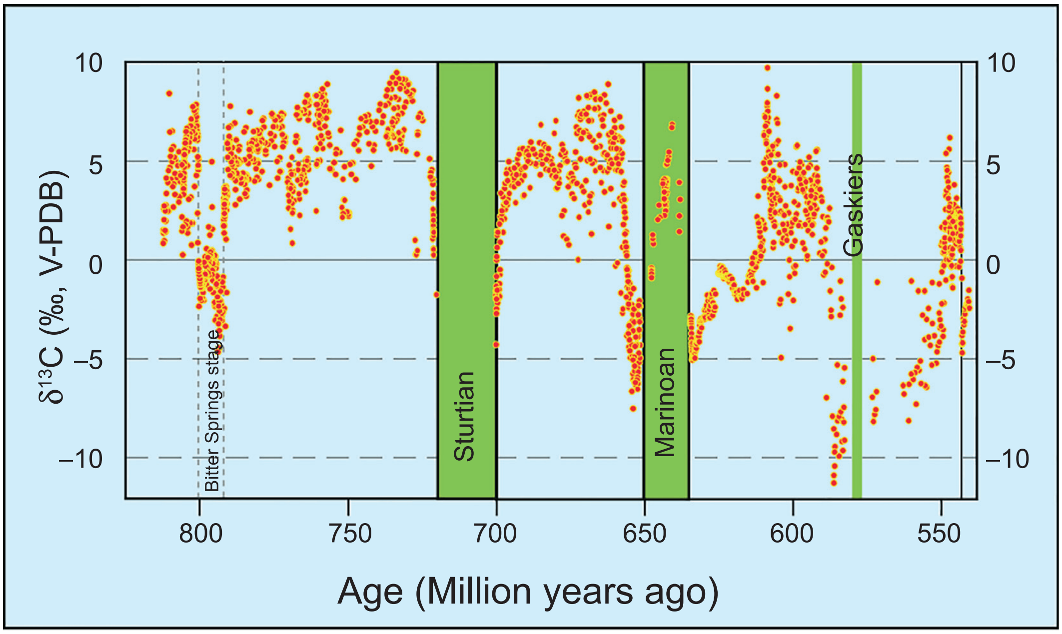

Figure 3: Carbon isotope chemostratigraphic curve for Neoproterozoic (after Halverson et al., 2005).

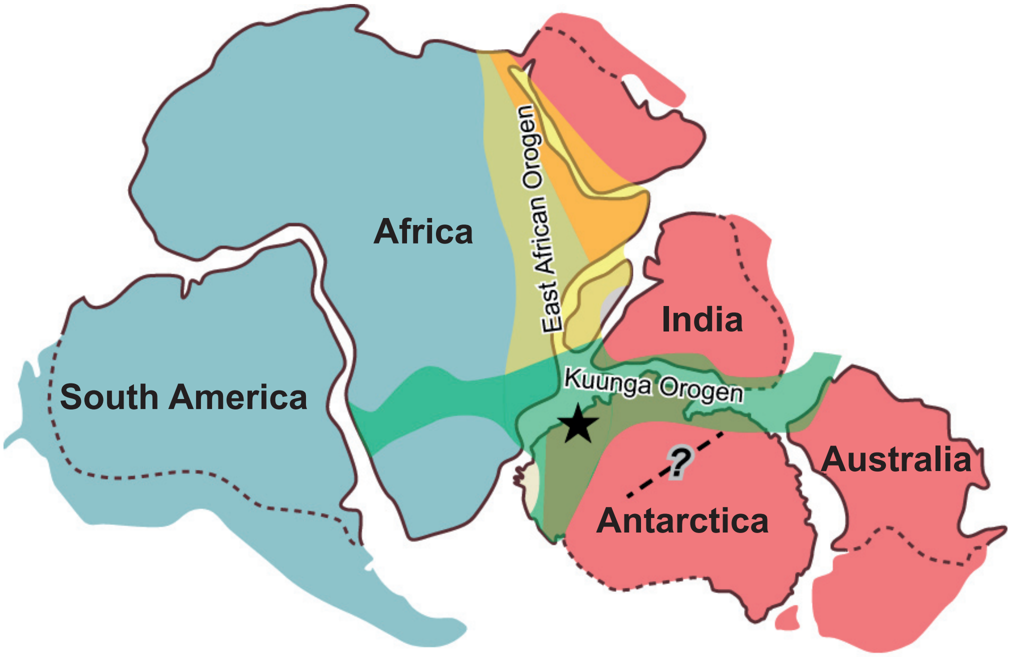

Figure 4: The supercontinent of Gondwana in Early Cambrian period (modified after Meert, 2003). The locality of Sør Rondane Mountains is shows by a start symbol.

(Figure 4, Meert, 2003; Satish-Kumar et al., 2013). This region is a part of the Latest Proterozoic to Early Cambrian collision zone in the southern domain of the north-south trending East African-Antarctic Orogen (EAAO; Jacobs et al., 2003) that includes high-grade metamorphic terranes of Mozambique, Madagascar, Sri Lanka and southern India (Figure 4). Recent studies have suggested that the east-west trending "Kuunga" orogeny passes through Eastern Dronning Maud Land and it is highly likely that multiple collision and accretion of smaller continental fragments have resulted in the final amalgamation of Gondwana rather than a single event of East- and West-Gondwana collision (Meert, 2003; Osanai, et al., 2013).

The amalgamation of supercontinent, such as Gondwana, is one of the major tectonic processes that shaped up the present day continents. In simple terms, the amalgamation of a supercontinent include global-scale tectonic processes such as closing of ocean basins and collision of smaller existing continents. The tectonic processes that occurred during the formation of Gondwana supercontinent are comparable to the present day Himalayan orogeny. For example, sediments deposited in the Tethys paleo-ocean that existed between Eurasian continent and Indian continent is now found on top of some of the tallest mountains in the Himalayan Mountain chain. Similarly, carbonate and other sedimentary rocks are found as integral parts of a thick pile of metamorphosed sedimentary sequences at the Sør Rondane Mountains, which form a part of the East-African-Antractic orogeny, suggesting that they were deposited in an extinct paleo-ocean and transformed to metamorphic rocks during continental collision. Tectonic activities relating to the continental collision were active somewhere between 750–530 million years ago, which are recorded in zircons and dated using U-Pb method (Osanai et al., 2013; Adachi et al., 2013; Hokada, et al., 2013; Grantham et al., 2013). This indicates that the sediments that are now transformed to metamorphic rocks should have been deposited prior to it, i.e. earlier than 750 million years ago.

At the Sør Rondane Mountains, most of outcrops contain layers of carbonate rocks that are highly metamorphosed and structurally conformable with the adjoining rock units (Figure 5). Of particular importance is that they are relatively pure, comprising mostly carbonate minerals. Purity of carbonate rocks is related to the amount of impure particulate sediments carried from nearby continents through weathering and incorporated along with the carbonate precipitates.

Otsuji et al. (2013) carried out a comprehensive geochemical study of the metamorphosed carbonate samples collected from the Sør Rondane Mountains to identify the level of alteration during metamorphism and whether pristine depositional geochemical signatures are preserved on not. A rigorous geochemical screening was carried out for more than 300 analyses in 90 samples. Both carbon and oxygen isotope composition of the carbonate rocks were comparable to the average isotopic composition of the Neoproterozoic carbonate rocks reported in literature (Figure 6, Kanuth and Kennedy, 2009). For example, the $\delta^{18}O$ oxygen isotopic composition for Sør Rondane Mountains samples

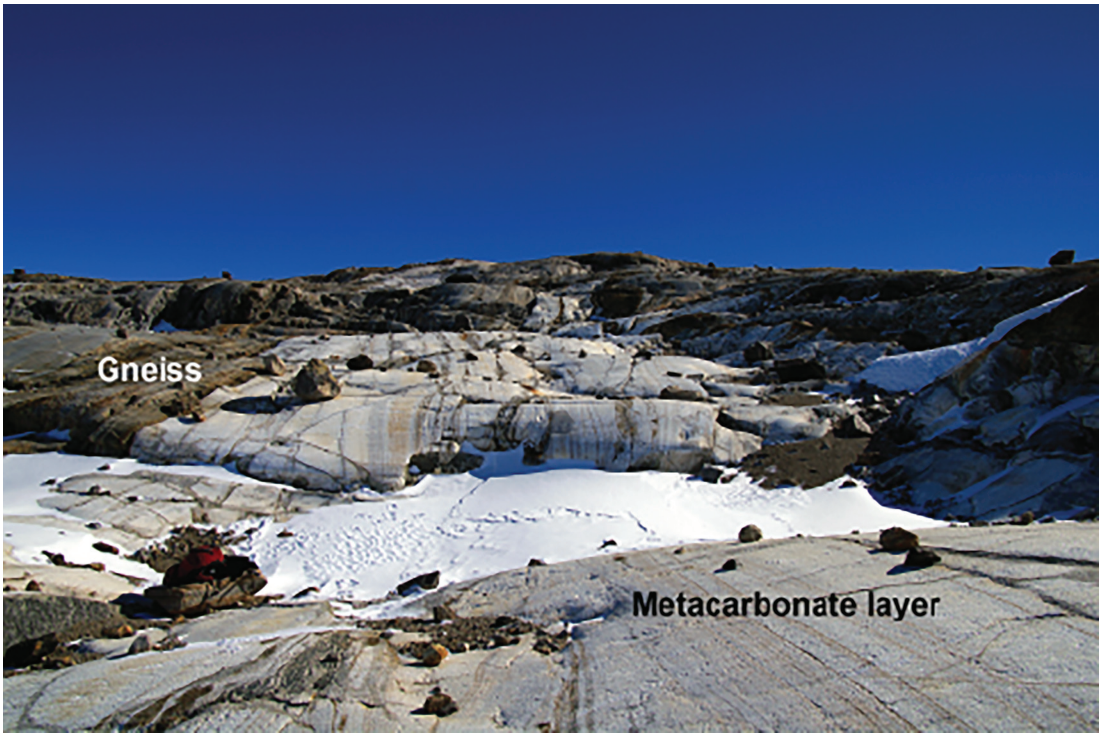

Figure 5: A typical occurrence of a metamorphosed carbonate rock occurrence at the Sør Rondane Mountains, East Antarctica.

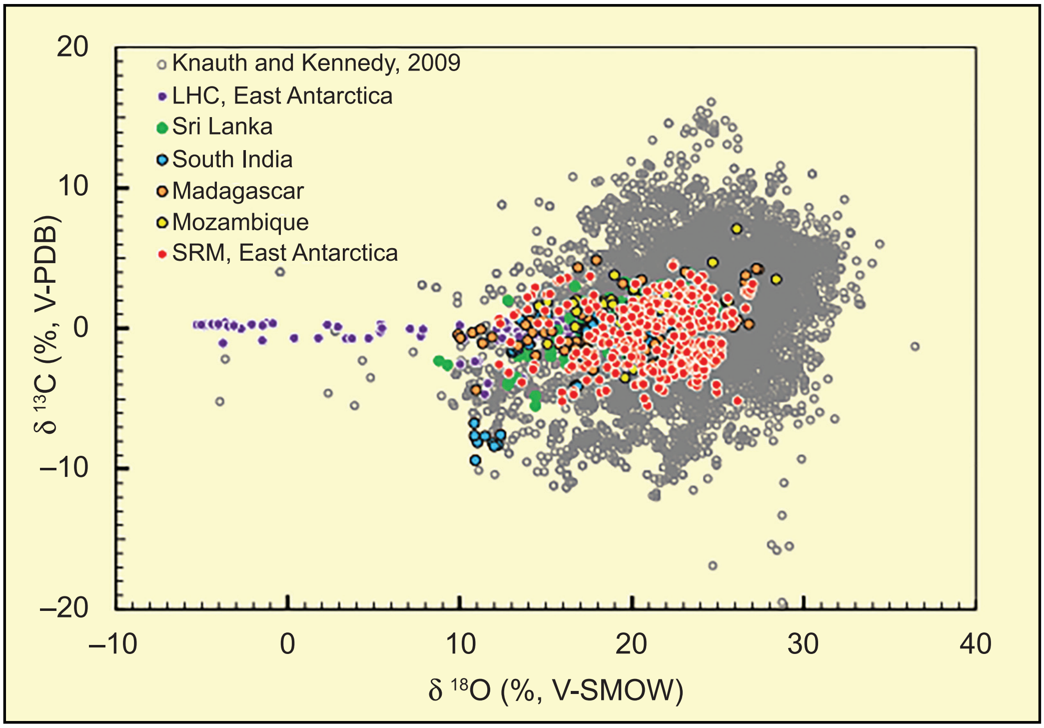

Figure 6: A compilation of carbon and oxygen isotope composition of metacarbonate rocks from East Gondwana (Data source from Satish-Kumar et al., 1998; Satish-Kumar and Wada, 2000; Satish-Kumar et al., 2001; Satish-Kumar et al., 2002; Satish-Kumar et al., 2008; Satish-Kumar et al., 2010, Otsuji et al., 2013). Neoproterozoic carbonate data is after Knauth and Kennedy (2009).

varies from 12.3 to 27.0‰. A conservative cut off value of 20‰ for oxygen isotope ratios were used because carbonate rocks with lower values possibly represent alteration during diagenesis and metamorphism accompanied by hydrothermal fluids, which can easily alter oxygen isotopes (Kanuth and Kennedy, 2009). Selected samples were further screened using trace element composition and rare earth element patterns.

In order to obtain the minimum strontium isotope initial ratio, the last criterion used by Otsuji et al. (2013) was to compare the Mn/Sr ratio of the samples from the same region with its ^{87}Sr/^{86}Sr ratios. Manganese is an element derived from continents, whereas strontium is available in large concentrations in sea water. This means that the Mn/Sr values will be lowest for carbonates, which has been least affected by continent input. As can be seen from Figure 7, the positive slope of the lines indicates that higher Mn/Sr values have higher strontium isotope ratios corresponding to the continental input. Using this relation, Otsuji et al. 2013 estimated the best possible value of strontium initial ratio (Figure 7).

Based on the representative Sr isotope chemostratigraphic curve for Neoproterozoic by Halverson et al. (2010), the strontium isotope ratios between 0.70566 and 0.70630 of the least altered samples reflect apparent depositional ages between c.880–790 million years ago (Figure 2). Whether the strontium isotope curve of Halverson et al. (2010) represents a global representative curve is yet to be confirmed, however, the apparent age of deposition estimated is consistent with the geological and geochronological information obtained from the region. Thus, irrespective of the high-grade metamorphism and tectonic displacement of the carbonate sediments, apparent depositional ages were estimated, and such information provides valuable clues for our understanding of the tectonic evolution of Sør Rondane Mountains, in particular, and the East African Antarctic orogeny in a global scale.

As discussed earlier carbon isotope chemostratigraphy has some inherent problems. Although the secular variations of δ^{13}C values of unaltered carbonates are often used to estimate apparent depositional ages (e.g. Melezhik et al., 2008; Halverson et al., 2010; Johnston et al., 2012), in the case of metamorphic rocks, carbonate can exchange its carbon isotopes with organic matter during metamorphism.

Existing data suggest that the carbon isotopic composition of carbonate sediments between ca.

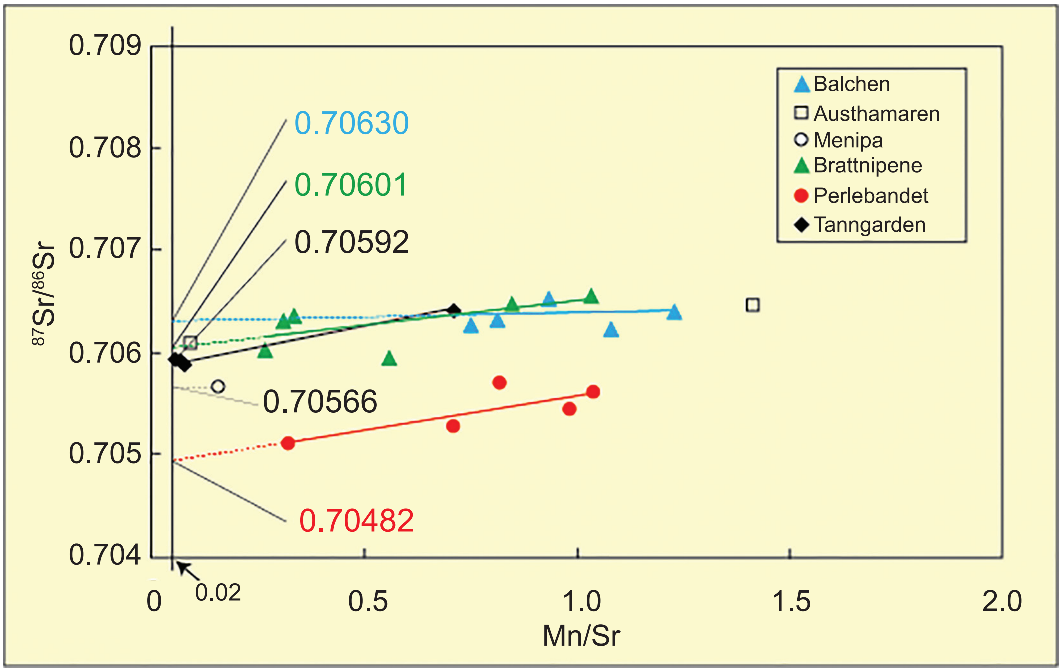

Figure 7: Estimation of strontium initial ratio of carbonate rocks using a correction method by Mn/Sr ratio (after Otsuji et al., 2013). Carbonate rock sample from the same region is used in the linear regression.

900 and 850 Ma is relatively constant between 4 and 6‰ (Halverson et al., 2010). There is a possibility that the $\delta^{13}C$ values of carbonates were lowered by the fractionation of isotopes with organic materials within the carbonate sediments, but this alteration should be very limited, given the low content of graphite in the samples. As discussed in earlier reports such as Frimmel (2010), local variations in biologic productivity might have been responsible for such differences in carbon isotope composition of SRM carbonates.

A comprehensive study on similar lines on metamorphosed carbonate rocks from different crustal terrains that formed part of the East African Antarctic Orogen, now distributed as stable continents in southern India, Sri Lanka, Madagascar, southwestern Africa, is now underway. It is expected that more details of paleo-oceans and tectonic activities prior to the amalgamation of Gondwana will be in light in the near future.

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