Native aluminium (spherules and particles) in the Central Indian Basin sediments: Implications on the occurrence of hydrothermal events.

Sridhar D. Iyer, M.B.L. Mascarenhas-Pereira and B. Nagender Nath*

National Institute of Oceanography, Dona Paula, Goa 403 004, India.

Corresponding author: iyer@nio.org, Fax: (91) 0-832-2450602; Phone: (91) 0-832-2450363

Abstract

We have sampled particles of native aluminium (Al^o) in two sediment cores from the Central Indian Basin (CIB). The cores are geographically separated but are located at the base of two seamounts. The native Al^o particles occurring as grains and spherules, have an average Al content of ~95 % and are associated with volcanogenic-hydrothermal material. Morphologically and compositionally, the specimens are similar to those reported from the East Pacific Rise. After ruling out several processes for the presence of the native Al^o, we hypothesize that during progressive melting of magma, a basaltic magma is produced which has high contents of reductants such as methane and hydrogen, and a low oxygen fugacity. During the upward migration of such magma, reduction to metallic aluminium and the formation of native Al^o particles takes place.

Keywords: Native Aluminium, Central Indian Basin, Hydrothermal.

Introduction

Metalliferous sediments, massive sulphide deposits and high-temperature vents occur at the major tectonic boundaries in the oceans (Houghton et al., 2004). Hydrothermal deposits at intraplate seamounts are less common although the seamount morphology, associated fracturing and volcanism could favour hydrothermalism (Alt, 1988). Much of the hydrothermal or mineralised areas in the Pacific seamount areas (Scholten et al., 2004) are remarkably iron-rich e.g., Fe and Si oxy-hydroxides ironstones, spongy iron oxides and Fe-rich slabs (Hein et al., 1994). Hydrothermal deposits have also been discovered along the Central Indian Ridge (Plüger et al., 1990; Nath et al., 1997; Gamo et al., 2001; Halbach et al., 2002).

The occurrence of native metals in oceanic hydrothermal areas is sporadic relative to terrestrial locations because pelagic sediments generally form under oxidizing conditions that hinder the growth of native metals. However, wide range of native metals occurs in Recent and Meso-Cenozoic oceanic sediments (Shterenberg, 1993). Native metals are reported to occur near transform faults, ridges and back-arc spreading centers, for example near the sulphide mounds at 21^{o} N East Pacific Rise (EPR, Cronan, 1979, Oudin, 1983), in the Clarion fracture zone (FZ) (Shterenberg et al., 1980; 1986), in the metalliferous sediments of the EPR (Shterenberg et al., 1981; Marchig et al., 1986) and near the Marcus-Nekker seamounts and the Hess rise (Shnyukov et al., 1981). Native metals are also found in manganese nodules (Baturin and Dubinchuk, 1984).

Native aluminium (Al^o) has been reported in Siberian trap rocks (Okrugin et al., 1981), skarn-ore deposits (Novgorodova et al., 1982), in potash rhyolites (Filimonova and Trubkin, 1996) and in metasomatised sediments of the EPR (13^o N, Davydov and Aleksandrov, 2001). So far there are no reports of the occurrence of native Al^o in seafloor rocks such as basalts, gabbros and serpentinites.

Previous investigations in the Central Indian Basin (CIB) have revealed the presence of volcanogenic-hydrothermal material (vhm) consisting of magnetite spherules, Ti-rich particles and Fe-Si rich metalliferous sediment, derived from localized episodes of hydrothermal activity albeit on a minor scale (Iyer et al., 1997a, b, 1999). From a study of a sediment core (AAS 61/BC 8) from the flank of a seamount (16^o S, 75.5^o E) Mascarenhas-Pereira and Nath (2005) reported several geochemical features that indicate a mixed volcanogenic-hydrothermal process. These sediments are compositionally similar to EPR metalliferous sediments in which native Al^o has been discovered (Shterenberg et al., 1986; Dekov et al., 1995).

Here we present new data on the occurrence and possible source of native Al^o particles recovered from the CIB in the vicinity of seamounts. These particles occur as individual entities and are associated with the vhm in the sediment cores.

Geological setting

The CIB is bounded by the Ninety East ridge, the Central Indian Ridge

and the Southeast Indian Ridge and is open and connected to the Bay of Bengal in the north. The basin is between 50 and 60 Ma in age and there exists three major FZ that trend along 73^{o} E (Vishnu or La Boussole), 76^{o} 30' E and 79^{o} E (Indrani or L'Astrolabe) and the Triple Junction Trace on the Indian Plate (TJT-In) between 76^{o} and 78^{o} E longitude (Dyment, 1993). A number of seamounts host N-MORB (Das et al., 2005) and are similar to the young, slow-generated Mid-Atlantic Ridge (MAR) and fast-generated EPR seamounts (Mukhopadhyay et al., 1995).

In considering the prevailing morphological features and the associated volcanics, the CIB seems to be a vulnerable area for tectonic and magmatic activity. The basin is prone to intraplate seismic activity triggered by magmatism due to their association with pre-existing zones of weakness during a phase of re-activation. Several of the CIB seamounts have an E-W elongated base due to addition of magmatic mass from subsequent eruptions that occurred after the seamounts were emplaced thus, hinting at multiple volcanic episodes. For instance, the seamount near S657, where abundant vhm has been recovered, has an E-W bulge and the site is ~50 km from the TJT-In, near about which ferromanganese crusts with incipient hydrothermal signature were recovered (Iyer, 2005).

To the north (1^{o} S /81^{o} E, ODP Site 717-719) of our study area, Boulegue and Mariotti (1990) have detected two hydrothermal events corresponding to ~7.5-9 Ma and 0.5 Ma ago. According to these authors the high temperature fluids that up welled along a fault plane and spread laterally were probably

derived from the basaltic basement and the immediately overlying sediments. The process of concurrent precipitation and deposition of inorganic calcite and sulphide minerals (FeS_2 and $FeCuS_2$) suggest that this ongoing hydrothermal activity is closely linked with the intraplate deformation. The present study reports on the occurrence of elemental Al particles that are associated to event of volcanogenic-hydrothermal activity in the CIB that are much younger than the above reported ages.

Methodology and Analytical Techniques

Native Al^o grains have been observed in two sediment cores (S657 which is 37 cm long; AAS 61/BC 8 which is 20 cm long) from the CIB (Fig. 1). Core S657 was taken at the base of a seamount at 14^o S and 75.935^o E, water depth of 4440 m and ~ 50 km from the TJT-In. The seamount is 800 m high, has an area of 37.56 km^2, a summit width of 0.18 km and overlies magnetic anomaly A23b. In this core the vhm is concentrated within the upper 6 cm of the core and the Al^o particles were observed at depths of 17-18 cm, 22-24 cm and 25-26 cm. Core AAS 61/BC 8 was taken from a seamount flank (16^o S and 75.5^o E) located near the TJT-In (Fig. 1).The particles of Al^o were observed at depths >8 cm in the core.

Twenty-one specimens from the non-magnetic fraction of S657 were picked out and mounted and polished for electron microprobe analysis using a JEOL 6880 Super probe. Additionally, 4 specimens from the sand-sized fraction of AAS 61/BC 8 were studied using a JEOL JSM-5800LV SEM coupled with an

OXFORD Link-ISIS EDS. The operating conditions used were a voltage of 20 kV, sample current of 55 μa and a beam diametre of 3-5 μm.

Results and Interpretations

We examined 25 specimens of elemental Al occurring in two sediment cores at 17-18 cm bsf, 22-24 cm bsf and 25-26 cm bsf in #S657 core associated with vhm and in 8-20 cm bsf in AAS 61/BC 8 core associated with glass shards, palagonites and volcanoclastic material. The specimens have variable size and morphology (Table 1), and consist of 13 spherules, 6 grains (Fig. 2) and 2 oval shaped particles. In addition, 4 irregular Al-rich growths over a glass shard (?) were observed in S657. While the Al^{o} grains in the core #S657 sediments are dominantly spheroidal, the southern station (AAS 61/BC 8) has three out of four grains of irregular shape (Table 1). Typical shapes of the Al^{o} particles representing both the stations are presented in Fig. 2. The spherules (Fig. 2a) range in size from 40 to 200 μm and have a brownish coating of siliceous sediments. In core AAS61/BC 8, the particles can be classified into two types: (1) massive with irregular shape and sharp edges (Fig. 2b), elongated (60 to 230 μm) along one axis and exhibit cracks resembling hydration features probably formed when lava chilled and spattered on contact with sea water, and (2) particles with a botryoidal structure (Fig. 2c) which also display cracks at the joints of the globules to the main particle. The oval to elongated-shaped particles (125 to 250 μm) have botryoids (Fig. 2c) protruding from the host. The globular shaped botryoids have smooth edges (Fig. 2c) and cracks at the boundaries

suggesting that the spheroids (Fig. 2a) in S657 may have been broken off from similar globules.

The studied particles are mainly composed of Al (85-99 %), with negligible Si, Mg, Ca, Na, K and traces of Ti, Fe, Cu, Zn and Ag and S (Table 1). In one grain of AAS 61/BC 8, S is somewhat higher (~2.33 %) and in another Cu and Zn are higher (3.65 and 5.5 %, respectively; Table 1). Chlorine content measured in the particles of AAS 61/BC 8 range between 0.08 and 2.85 %. Sulphur content in the metalliferous sediment ranges between 0.21 and 4.65 % (one specimen had ~10 % S in S657) while in the magnetite spherules sulphur ranges between 0.21 and 1.55 % (Iyer et al., 1997a). Morphologically and compositionally the Al specimens are comparable to those from the EPR (Dekov et al., 1995).

Biostratigraphic dating of core S657 based on radiolarian assemblages has shown that the core depths of 28, 16 and 6 cm (vhm dominant <6 cm depth) correspond to ages of 130, 70 and 10 kyr, respectively (Iyer et al., 1997a). The vhm is associated with younger sediments of ~10 kyr ago than the Al^o particles that occur in sediments of ~70 kyr. Although Al^o occurs with sediments older than 70 kyr, we cannot ascertain if the formation of Al^o is synchronous with sediment deposition. The sediments of core AAS 61/ BC 8 comprise of 2 lithic units (a) a short sediment surface (~ 6 cm, < 180 kyrs) of radiolarians ooze, underlain by (b) grayish-brown coloured indurated pelagic clay (6-20 cm, > 180 kyrs; age dating described in Mascarenhas-Pereira et al., 2006).

Discussion

The aluminium-rich spherules and particles found by us are nearly pure and considered as native aluminium (Al^o). Furthermore, these native aluminium particles are not intergrowing with any alumino-silicates. There are several reports on the occurrence of native minerals at the EPR. Native copper occurs in the sediments from the EPR black smoker at 18.5^o S (Marchig et al., 1986) and wurtzite, chalcopyrite, pyrite and native sulphur are common at 21^o N (Oudin, 1983). Sediment traps deployed at 13^o N EPR recovered graphite crystals (5-100 μm) with antimony rich overgrowths and crystals are hydrothermal in origin as shown by the occurrence of metallic iron associated with (and deposited on) silica (Jedwab and Boulegue, 1984). Shterenberg and Voronin (1994) reported native copper and copper-zinc alloy clasts co-existing with Fe-Mn micronodules near the EPR. The near-axial metalliferous sediments of the EPR have native tin and tin alloys (Dekov et al., 1996) and also metallic grains of an alloy ($Ag_{71.5}$ $Cu_{28.5}$) which probably formed endogenetically (Dekov and Damyanov, 1997).

Compared to the EPR, there are only a few reports of native minerals from the MAR. Native iron and aluminium in basalts from the Sao Paulo fault in the South Atlantic probably originated during the pre-crystallisation stage of basaltic melt evolution while Zn aggregates formed by hydrothermal processes (Shterenberg, 1993). Mozgova et al. (1996) have reported the occurrence of cobalt pentlandite from a MAR hydrothermal deposit, (14^o 45' N) that occurs as <20 μm, round to irregular grains and aggregates. Dekov et al. (1999) have also reported strands and elongated grains (up to 300 μm in length) of native Cu and α-Cu-Zn collected from the Trans-Atlantic Geotraverse (TAG) hydrothermal field

(26^{o} N, MAR). Subsequent findings in the TAG area revealed a number of small, irregular shaped and spherical metal particles of Sn, Sn-rich Pb, Sn-Cu and native Ni (Dekov et al., 2001; Dekov, 2006). In general, native metals form either as accessory minerals disseminated in the ridge rocks and/or as massive sulphide mounds and through serpentinisation of the upper mantle/lower crust ultramafics/mafics (Dekov, 2006). Degradation of the rocks and mass wasting of the mounds liberated the metallic grains, which were later deposited into adjacent sediments (Dekov et al., 1999).

It appears that native metals and alloys may form as a result of a number of geological processes occurring in reducing environments. The presence of metallic Al^{o} specimens in the metalliferous sediments of the CIB is therefore of relevance to establish their origin. Before investigating the possible ways in which native Al^{o} could form in the CIB, it is necessary to rule out several possibilities, a few of which have previously been noted by Dekov et al. (1995) in their study area.

1. An anthropogenic origin for the Al^{o} specimens is not feasible because the onboard samplers are of stainless steel while the sieves used are of bronze. Moreover, the CIB particles do not contain substantial amounts of Cu and Mn that are diagnostic of aluminium alloys manufactured by industries.

2. Terrigenous input by aeolian or riverine contributions may be sources for native Al^{o} but presently we have no evidences of these processes in the CIB. Although confirmation exist for terrigenous sediment input by the Ganges-Brahmaputra rivers up to 8^{o} S in the CIB (Nath et al., 1989), it is unlikely that the

Al specimens have been brought down from eroded rocks of the Himalayas and buried at the studied site. There are no known reports of native Al^o in the Himalaya and also in the Bengal fan. Additionally, freshness and morphology of the specimens (Fig. 2) do not support a long distance transport.

Clay minerals and quartz are the main forms of aeolian transported material to the Indian Ocean region from distal continental sources (Kolla et.al., 1976). Furthermore, the sampled sites are far from any neighbouring land thus ruling out the possibility of Al^o by aeolian input.

3. An extraterrestrial origin is untenable because cosmic spherules found throughout the ocean are enriched in Fe and Ni, but not Al^o.

4. The role of bacteria in producing native metals in manganese nodules has been reported (Baturin and Dubinchuk, 1984). Presently we have no data from the CIB to support this contention.

5. Serpentinisation of basic and ultrabasic rocks under low oxygen conditions could produce native metals and alloys (Shteinberg and Chashchukhin, 1977; Dekov, 2006). However despite extensive sampling in the CIB no ultrabasic rocks have so far been dredged (Mukherjee and Iyer, 1999).

Ruling out the above possible sources for the occurrence of native Al^o in the sediments from S657 and AAS 61/BC 8, we now consider alternative mechanisms. There are several ways by which native metals could be produced in the oceanic environment, from magmatic or metamorphic process, from hydrothermal activity or from erosion of altered ore deposits (Dekov et al., 2001). Alternatively, exsolution of metal-rich fluid present in the magma (Yang and

Scott, 1996) and degassing of magmatic vapors during submarine eruptions (Rubin, 1997) may lead to the deposition of certain metals. However this process is probably highly episodic occurring almost entirely during eruptions and involves enhanced and abnormal hydrothermalism as well. The mean hydrothermal effluent concentrations of Al, Zn, Fe, Mn, Au and Ir are predicted to be ~10-10^3 times greater than in a degassed effluent (Rubin, 1997). The acidity (pH 3-6) of the hydrothermal solutions (100-350^o C) not only results in an intense alteration of the basalts (Howard and Fisk, 1988) but also results in the mobilization of Al, until it is enriched 1,000 times in vent fluids relative to normal seawater concentrations (Michard et al., 1984). This could probably explain the Al-Si gels and the hydrothermally derived Al-rich clays. Therefore, the native Al^o may not crystallize in the hydrothermal fluid. On the contrary, the location of the CIB samples at the base and flank of seamounts (that are geographically ~200 km apart), freshness of the particles, co-existence of vhm (Iyer, 2005) and chemical signatures of hydrothermal inputs to core AAS 61/BC 8 (Mascarenhas-Pereira and Nath, 2005), indicate an endogenous origin for the Al^o particles.

Since Al^o is not crystallized directly from the hydrothermal fluid, we need to consider another mechanism by which native Al^o separates from the silicate melt. This could occur under conditions of high pressure-temperature and low fO_2 during the interaction of the magma with intratelluric H_2 coupled with disproportionating of AlCl (the CIB specimens have up to 2.85 % Cl), AlF and Al_2O_3 that are transported in the gas phase and associated with other fairly 'dry' reducing gases (Osadchii and Alekhin, 1984). Additionally, Lukanin and Kadik

(1984) reported that the separation of basaltic magmas during progressive melting could increase CH_4 and H_2 contents and decrease fO_2 of the melt and thus causing reduction of some elements to their metallic state. The native phases (Al, Fe, Zn, Cu, Pb and Cd) commence to crystallize at 1500-1550^o C and continue up till 183^o C (Okrugin et al., 1981). The above process suggests the existence of a mantle fluid that was rich in hydrocarbons, halides and sulphur (Davydov and Aleksandrov, 2001) which creates a reducing environment at different depths. During the upward migration of such an intratelluric fluid, native Al^o could be transported.

Interestingly, in the Indian Ocean the occurrence of an unusual abundance of an highly magnetic mineral (maghemite with some aspects of magnetite) at 23^o 56' S and 73^o 53' E, was ascribed to a hydrothermal origin (Harrison and Peterson, 1965). Similarly, octahedral magnetite, co-occurring with native Al, has also been observed at 13^o N EPR, both in the sediment trap and as a minor phase of a hydrothermal chimney (Jedwab and Boulegue, 1984). Therefore, based on our present observations, i.e., (i) the link between the sampled sites with the associated seamount and FZ, (ii) co-occurrence of vhm in the cores, (iii) the freshness of the Al^o specimens and (iv) their chemical composition, we propose an hydrothermal origin for the native Al^o in the CIB.

Conclusion

- We found the native Al^o particles and spherules to occur in two sediment cores that are geographically apart in the CIB. The native Al^o are

comparable to those discovered in the EPR.

- Hydrothermal process is invoked to account for the formation of the native Al^o.
- Preservation of aluminium in its native form in an oxidizing environment may be due to the formation of a microscopic oxide layer (corundum?) which hampers further oxidation.

Considering the present finding and the co-occurrence of Al^o particles and vhm with a variety of volcanics, it is possible that the CIB might have witnessed localized episodes of hydrothermal events at other seamount environments, albeit minor and focused, as in the Pacific Ocean.

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Captions for Tables

Table 1. Morphologic features and geochemical data of aluminium-rich specimens recovered from two sediment cores in the CIB.

Captions for Figures

Fig. 1: Location map depicting the seamounts (open star) that that are mostly aligned along eight chains with some isolated edifices. Box in the inset figure represents the study area. The studied sediment cores are marked on the map. The seamounts distribution is from Das et al. (2005).

Fig. 2: Scanning electron micrographs of Al^o in the CIB.

a. Typical spherule of native Al^o (#S657, 17-18 cm). (b).Large, irregular particle with hydration cracks (AAS 61/ BC 8, 8-20 cm bsf). (c) Bulbous or nodular structure with hydration cracks seen at edges of the globule (AAS 61/ BC 8, 8-20 cm bsf). This kind of feature probably represents a later detachment of spherules as seen in (a).

Table 1:

#	SiO_2	TiO_2	Al_2O_3	FeO*	MgO	CaO	Na_2O	K_2O	P_2O_5	SO_3	CuO	ZnO	Ag_2O	Type/Size
Northern area (#S657)														
17-18 cm														
1	1.91	nd	96.77	0.09	nd	0.84	0.07	0.12	0.21	nd	nd	nd	nd	Grain, 175x95 μm
2	0.66	nd	98.4	nd	nd	0.88	0.03	0.02	nd	nd	nd	nd	nd	Spherule, 175x95 μm
3	0.23	nd	98.76	nd	nd	0.76	nd	nd	0.23	nd	nd	nd	nd	Spherule, 115 μm
4	0.3	nd	98.68	nd	nd	0.81	0.02	nd	0.17	nd	nd	nd	nd	Spherule, 60 μm
5	0.46	nd	98.52	nd	nd	0.88	nd	nd	0.13	nd	nd	nd	nd	Spherule, 200 μm
6	0.32	nd	98.55	nd	nd	0.82	nd	0.04	0.28	nd	nd	nd	nd	Spherule, 200 μm
7	0.56	nd	98.08	0.1	nd	0.94	0.19	0.09	0.27	nd	nd	nd	nd	Spherule, 40 μm
8	0.45	nd	98.59	nd	nd	0.85	nd	nd	0.12	nd	nd	nd	nd	Spherule, 50 μm
9	0.46	nd	98.61	nd	nd	0.74	0.03	0.03	0.12	nd	nd	nd	nd	Spherule, 50 μm
10	0.24	nd	98.61	nd	nd	0.76	nd	nd	0.16	nd	nd	nd	nd	Spherule, 0 μm
22-24 cm														
11	0.3	nd	98.84	nd	nd	0.83	0.02	0.02	nd	nd	nd	nd	nd	Spherule, 55 μm
12	0.28	0.02	98.77	nd	nd	0.78	nd	nd	0.13	nd	nd	nd	nd	Spherule, 60 μm
13	0.52	nd	98.36	nd	nd	0.9	0.03	0.03	0.14	nd	nd	nd	nd	Spherule, 80 μm
14	0.13	0.03	99.11	nd	nd	0.62	nd	nd	0.11	nd	nd	nd	nd	Grain, 80x70 μm
15	0.24	nd	98.69	0.12	nd	0.74	0.06	nd	0.17	nd	nd	nd	nd	Oval, 100x70 μm
16	0.53	nd	98.25	nd	nd	0.82	0.05	nd	0.33	nd	nd	nd	nd	Oval, 90x60 μm
17	0.99	nd	97.98	nd	nd	0.9	nd	nd	0.11	nd	nd	nd	nd	Spherule, 120 μm
18	0.35	nd	98.42	nd	nd	0.89	0.06	nd	0.29	nd	nd	nd	nd	Irregular with moss like growth
19	0.34	nd	98.68	0.07	nd	0.77	nd	nd	0.15	nd	nd	nd	nd	Irregular with moss like growth
20	0.3	nd	98.91	nd	nd	0.62	nd	0.03	0.15	nd	nd	nd	nd	Irregular with moss like growth
25-26 cm														
21	0.32	nd	96.73	nd	2.01	0.8	nd	nd	0.15	nd	nd	nd	nd	Irregular with moss like growth
Southern area (AAS 61/BC 8)														
22	1.57	nd	94.48	nd	0.24	nd	nd	nd	nd	1.97	0.68	0.88	0.17	Massive with cracks, irregular, sharp edges. 60 μm
23	1.44	nd	96.65	nd	0.51	nd	nd	nd	nd	nd	0.43	0.53	0.53	Massive, irregular, globule like protrusion. 90 μm
24	nd	nd	84.65	nd	nd	nd	nd	nd	nd	nd	6.14	9.19	nd	Massive, irregular, globule like protrusion. 220 μm
25	3.28	nd	96.72	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	Globular, botryoidal structure, with cracks. 203 μm

1) All values in wt %, 2) nd- not detectable, 3) size of the specimens from the southern area (22-25) represent the longest dimension

Figure 1.

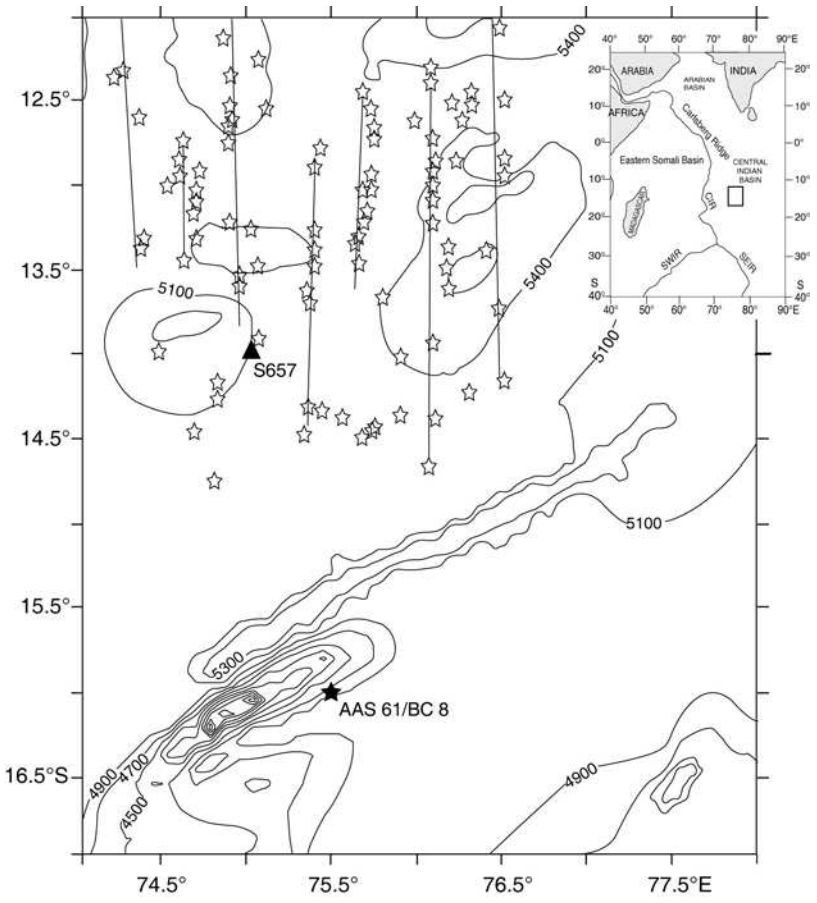

12.5°
13.5°
14.5°
15.5°
16.5°S
74.5°
75.5°
76.5°
77.5°E
5400
5100
5300
4900
4700
4500
S657
AAS 61/BC 8
ARABIA
INDIA
AFRICA
ARABIAN BASIN
Carlsberg Ridge
Eastern Somali Basin
CENTRAL INDIAN BASIN
CIR
SWIR
SEIR
40°
50°
60°
70°
80°
90°E
20°
10°
0°
10°
20°
30°
40°
S

Figure 2.

