# Rock Magnetic Evidence of Tectonic Control on the Sedimentation and Diagenesis in the Andaman Sea over ~ 1 Million Years

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#### **Abstract**

We evaluate the sedimentary (magnetic, mineralogical, geochemical, grain size) records of a long sediment core (NGHP-01-17A) to elucidate the tectonic control on the sedimentation and diagenesis in the Andaman Sea (AS) over the past ~ 1 Ma. Two distinct sedimentary magnetic zones (Z-I and Z-II) are identified based on the rock magnetic signatures. A significant shift in the depositional environment is observed around ~ 90 ka which is marked by a rapid change in magnetite susceptibility ( $\chi_{lf}$ ), Fe, Ti and organic carbon content in Z-I. A trend of upward increase in  $\chi_{lf}$  from ~ 90 ka to 3 ka in core NGHP-01-17A suggest increase in detrital flux to the AS and represents the onset of a new sedimentary regime following most recent event of tectonic uplift in Indo-Burman Ranges (IBR) which occurred at ~ 150 ka. Methane influenced progressive diagenetic dissolution of detrital iron oxides and subsequent precipitation of iron sulfides in Z-II is clearly evident in the rock magnetic and mineralogical data. Multiple bands of diagenetically formed iron sulfide nodules at different periods in Z-II indicate episodic intensification of anaerobic oxidation of methane (AOM) due to enhanced methane-flux. We propose that fluid migration forced by tectonic processes facilitated episodic release of methane from deep reservoir that led to the formation of iron-sulfide nodules in the AS.

**Key words:** Magnetic Minerals; Magnetic properties; Grain Size, Inorganic Geochemistry, Methane Hydrates, Iron Sulfide Nodules

# **Key points:**

- A major shift in the depositional environment near ~ 90 ka is marked by a rapid change in magnetite concentration (χ<sub>lf</sub>), Fe, Ti and organic carbon content. A trend of upward increase in χ<sub>lf</sub> from ~ 90 ka to 3 ka in core NGHP-01-17A indicate increase in detrital flux to the Andaman Sea (AS) and represents the onset of a new sedimentary regime following most recent event of tectonic uplift in Indo-Burman Ranges (IBR) which occurred at ~ 150 ka.
- We provide evidence of tectonically-driven episodic fluid migration in the Andaman Sea (AS).
- Rock magnetic properties effectively recorded the geological processes that controlled sedimentation over ~ 1 Ma in the Andaman Sea.

#### 1. Introduction.

The Andaman Sea (AS) is unique in terms of the complex tectonics, geology, climate and oceanography. Over geological time scales, tectonic activities coupled with changes in the strength of summer monsoon rainfall significantly influenced the physical and chemical weathering intensities, erosional processes in the source areas, and sediment depositional pattern in nearshore and Andaman Continental Shelf (ACS) (Awasthi et al., 2014; Cochran, 2010; Colin et al., 1998, 1999, 2006; Clift et al., 2008; Kuehl et al., 2019; Rashid et al., 2007). Tectonic uplift in the source areas coupled with climatic conditions provides important constraints on the evolution of marine sedimentary systems (Armitage et al., 2011; Zhang et al., 2018). Influence of tectonic activities and climatic changes on the Bengal fan sedimentation was unravelled using Sr-Nd (IODP-353; Peketi et al., 2021) and <sup>10</sup>Be (IODP-354; Lenard et al., 2020) isotopic compositions of the sediment cores. Exhumation and erosion of the Indo-Burman Ranges (IBR) followed by multi-phase upliftments (at ~8 Ma, ~3.6 Ma, ~2.6 Ma, 1.8–1.7 Ma, 1.2–0.6 Ma and 0.15 Ma) and sediment provenance changes in the AS have been reported (Li, 1995; Li et al., 1996, 2014; Najman et al., 2012; 2016; 2019, 2020; Zhang et al., 2015).

Previous studies documented the paleoceanographic and provenance related changes in the AS using mineralogical, elemental, inorganic geochemistry, sedimentological, isotopic, environmental magnetic, and foraminiferal records. Ali et al. (2015) observed no significant change in the sediment provenance in the AS during the past 60 ka. A linkage between variability in the sediment provenance, rapid deviation in the monsoon pattern, and changing pattern of ocean surface currents over 70 ka was also established (Awasthi et al., 2014). Changes in the weathering and erosion pattern in source regions and fluctuations in sediment contribution of terrigenous flux to the AS from the mid-late Holocene were controlled by climatic shifts, source area changes and tectonic processes (Achyuthan et al., 2014; Colin et al., 2006; Damodararao et al., 2016; Kuehl et al., 2019; Miriyala et al., 2017). Multi-proxy records of paleoclimatic, monsoonal and productivity changes are wellreported from the AS (Cao et al., 2015; Gebregiorgis et al., 2016; Kumar et al., 2018). Provenance, weathering pattern, erosion and monsoonal history in the Indo-Burman Ranges (IBR) and Indo-Burman-Arakan (IBA) of Myanmar over the past 21 ka were recorded in a sediment core from the AS (Colin et al., 2006; Najman et al., 2008, 2012). IBR and proximal volcanic sources were the major contributor of sediments in the AS during this period (Najman et al., 2019, 2020; Sebastian et al., 2019). A recent magnetic study on a sediment core from the AS spanning 21 ka tracked the provenance, weathering and history in IBR (Sebastian et al., 2019). They further established a magnetic grain size based proxy to detect the intensities of chemical weathering and Indian summer monsoon (ISM) during the late glacial, early Holocene and mid-Holocene period (Sebastian et al., 2019). A comprehensive understanding of the Himalayan tectonic evolution was developed from the sedimentary records based on the interpretation of the isotopic data of detrital minerals (Najman, 2006). More recently, McNeill et al. (2017) closely examined the stratigraphic and chronostratigraphic data from Bengal fan and Ninetyeast Ridge. They highlighted that an interplay between tectonic, climatic and sedimentological processes in the Himalayan–Tibetan Plateau controlled the evolution of Bengal–Nicobar submarine fan sedimentation. The tectonic-sedimentary history of Gonjo Basin, southeast Tibet in response to India-Asia collision was constrained using magnetostratigraphy (Xiao et al., 2021). Control of climatic and tectonic activities on the Bengal fan sedimentation was recently revealed using Sr-Nd isotopic compositions of the Hole U1445A (International Ocean Discovery Program-353) from the western Bay of Bengal (Peketi et al., 2021). Despite the global importance of AS in context with strong tectonic and climatic forcing, detailed sediment records documenting the linkage between tectonics, changing fluvial supply, diagenesis and its sedimentary signal in marine sediments still remains elusive.

Marine sedimentary deposits from AS represent an important archive of how tectonic, climate and post-depositional (diagenetic) conditions have changed over geologic time. The storage and supply of sediments from source to marine sink depends on how landscape respond to climate and tectonic perturbations (Fildani et al., 2016). An understanding of the time lag between tectonic upliftment, erosion, transport and final sedimentation is important to track the climatic and tectonic signals in the marine sediment records (Burbank et al., 1996; Clift and Giosan, 2014; Kumar and Srivastava, 2017; Kumar et al., 2020). Sediments deposited in the AS potentially record the signatures of provenance changes due to upliftment, re-organization of drainage network (Colin et al., 1999; Najman et al., 2019, 2020), climatic conditions (Sebastian et al., 2019), sediment dispersal and accumulation (Kuehl et al., 2019), diagenetic processes (Kurian et al., 2008), and the evolution of Andaman Basin (Roy and Sharma, 1993). Geological setting assures a high supply of magnetic minerals to the AS through Irrawaddy, Salween and Sittang (ISS) rivers (Colin et al. 1998; Licht et al., 2013; Robinson et al., 2007; 2014) and these minerals archive the signatures of source-to-sink related processes. A sediment core NGHP-01-17A (spanning ~ 1 Ma; Gebregiorgis et al., 2018) retrieved from the AS during the first Indian National Gas Hydrate Drilling Expedition preserves an excellent record and is well-suited for studying tectonic-controlled sediment depositional and diagenetic processes. In this study, we examined the sedimentary (magnetic, mineralogical, geochemical, grain size) records of a long sediment core (NGHP-01-17A) to elucidate the tectonic control on the sedimentation and diagenesis in the AS over the past ~ 1 Ma. The present study provides the longest and most detailed sediment magnetic record available so far from the AS.

#### 2. Materials and Methods

#### 2.1. Geological Setting

The AS is located in a back-arc basin formed by the oblique subduction of the Indian Plate beneath the Sunda Plate (Kamesh Raju et al., 2007; Pal et al., 2003; Rodolfo, 1969a). The Andaman Island Arc complex is located within the central portion of the 5000 km long Burma-Sunda-Java subduction complex (Pal et al., 2003). The ISS are the major rivers, which efficiently transport the eroded continental sediments into the AS (Kurian et al., 2008). Estimated annual suspended sediment load supplied by the ISS river system is more than ~ 350 million tons to the Andaman Sea (Ramaswamy et al., 2004). The terrigenous sediments derived from ISS Rivers are routed through Gulf of Martaban to the deeper region of Andaman basin (Ramaswamy et al., 2004). Source rocks in the catchments areas of Irrawaddy River are dominated by Cretaceous-Cenozoic flysch of the IBR, Eocene-Miocene-Quaternary sediments of the Myanmar Central Basin, Late Precambrian, and Cretaceous-Eocene metamorphic/basic/ultrabasic rocks of the eastern Himalayas (Figure 1c; Bender, 1983; Robinson et al., 2014). The Salween and eastern tributaries of the Irrawaddy River drains through the Precambrian, Oligocene- Tertiary sedimentary, and acidic and metamorphic rocks of the eastern Shan Plateau (Robinson et al., 2007). The catchment areas of IBA and IBR receive heavy rains during the southwest (SW) monsoon (Garzanti et al., 2013). The sediments derived from these ranges are transported through several small rivers systems and get deposited along the western continental shelf of Myanmar (Garzanti et al., 2013; Kuehl et al., 2019). These sediments are further transported into the AS by the east flowing surface currents during the summer monsoon (Awasthi et al., 2014). Dominance of basaltic, granitic and sedimentary (shale) rocks in the source regions suggest that the geological setting around the AS assures a high supply of magnetic minerals to the study area through ISS rivers. An environmental magnetic investigation on a short sediment core (GC-3A) proximal to Site 17 (present core location) (Sebastian et al., 2019) and distal site (MD77-169, Sewell Seamount, Colin et al., 1998) reported that the sediment magnetic mineralogy is dominated by magnetite, titanomagnetite and hematite (Figure 1a,c).

# 2.2. Core Location and Age-Depth Model

As part of the Indian National Gas Hydrate Program (NGHP), a 700 m long sediment core NGHP-01-17A from Site 17 was collected onboard D/V JOIDES Resolution from the AS. The sediment

core NGHP-01-17A (10°45.1912'N, 93°6.7365'E) was raised from a water depth of 1356 m (Figure 1a) (Collett et al., 2008). Age model of the sediment core (NGHP-01-17A) was established by Gebregiorgis et al., 2018 using a high resolution benthic foraminifer (*C. wuellerstorfi* and *C. mundulus*)  $\delta^{18}$ O record with equivalent features in the LR04 global benthic  $\delta^{18}$ O isotope stack (Lisiecki and Raymo, 2005) using Analyseries (Paillard et al., 1996). Furthermore, age model was constrained by Accelerator Mass Spectrometry (AMS) <sup>14</sup>C dates on planktonic foraminifera (Gebregiorgis et al., 2018). An age-depth model is presented as supplementary Figure S1. Numerous volcanic ash layers were found in the sediment core NGHP-01-17A. Sediment lithology at this site is dominated by pelagic/hemipelagic sedimentation (Cawthern et al., 2014; Collett et al., 2008). Upper 50 m of the sediment core which spans ~ 1 Ma was used in the present study.

# 2.3. Sub-Sampling and Measurements

The sediment core was sampled at 5 cm interval which produced 594 sub-samples. Iron sulfide nodules were hand-picked from different sediment core depth intervals during sub-sampling.

# 2.4. Rock Magnetic and Sediment Electrical Resistivity Measurements

Sediment samples for bulk magnetic measurements were dried at 40 °C, weighed and packed in 25mm cylindrical sample bottles at the paleomagnetic laboratory of CSIR-National Institute of Oceanography (CSIR-NIO), Goa, India. Frequency-dependent magnetic susceptibility measurements were performed using the Bartington MS2B dual frequency ( $\chi_{lf} = 0.47$  kHz and  $\chi_{hf} = 4.7$  kHz) susceptibility meter. Anhysteretic remanent magnetisation (ARM) was imparted using 100 mT AF field superimposed with a fixed DC bias field of 50 mT. Isothermal remanent magnetisation (IRM) of 700 mT, 1000 mT, and 2000 mT in the forward direction and -20, -30, -100, and -300 mT in the backward direction was imparted using MMPM10 pulse magnetizer. All remanences were measured using AGICO automatic JR-6A spinner magnetometer. Mass-normalized IRM acquired with a peak field of 2T is considered to be the saturation isothermal remanent magnetization (SIRM). S-ratio is calculated as the ratio between the IRM at -300mT and SIRM (IRM-300mT/IRM2T; Stober and Thompson, 1979). Specialized magnetic measurements were carried out at Environmental Magnetism Laboratory of Indian Institute of Geomagnetism, Panvel, India. Temperature dependence of magnetic susceptibility (γ-T) was measured in a field of 300 A/m at 875 Hz by a CS-3 furnace unit attached to an AGICO (KLY-4S) Kappabridge. Electrical resistivity measurements were made on the split cores using Wenner array onboard D/V JOIDES Resolution.

## 2.5. Sedimentological analyses

## 2.5.1. Sediment Grain Size Measurements

Sediment grain size was measured using a Laser Diffraction Particle Size Analyzer (Beckman Coulter LS 13320) at National Centre for Polar and Ocean Research (NCPOR), Goa, India. Prior to analysis, sediment samples were first desalinated and were later decarbonated using dilute HCl (1N). Sediment suspensions were treated with 10% H<sub>2</sub>O<sub>2</sub> to remove organic carbon, and 300 mg of dispersing agent (sodium hexametaphosphate) was added to the suspension, which was then ultrasonicated before analysis. Grain size values are presented as volume %.

# 2.5.2. Mineralogical Analyses

Mineralogical and microscopy analyses were conducted on selected bulk sediment and iron sulfide nodule samples respectively. Hand-picked nodules were large enough to allow multiple analyses. The magnetic mineral components were identified using a Rigaku X-Ray Diffractometer (Ultima IV). The samples were run from 15° to 65° of  $2\theta$  at 1°/min scan speed using Cu K $\alpha$  radiation ( $\lambda$  = 1.5414 Å). The composition of bulk sediments and iron sulfide nodules was determined using an energy dispersive X-ray spectroscopy (EDS) probe attached to the microscope. Electron microscopic observations were made on the iron sulfide nodules using scanning electron microscope (JEOL JSM-5800 LV). Images were captured in secondary electron imaging mode at energy levels between 15 and 20 keV). Chemical composition of the nodules was determined using an Oxford Instruments Ltd. INCA-300 X-ray energy dispersive spectrometer (EDS) attached to the SEM.

# 2.6. Sediment Geochemistry

Most of the elements reported in the study area were determined using Inductively Coupled Plasma Optical Emission Spectrometer (Perkin Elmer, OPTIMA 7300 DV Simultaneous ICP-OES) at CSIR-NIO, Goa, India. A total of 85 samples were analyzed by ICP-OES for the determination of Fe, Al, Ti, and K. Around 50 mg of bulk sediment sample was digested in 10 ml of acid mixture (HF, HNO $_3$  and HClO $_4$ ) in a proportion of 7:3:1 in a PTFE beaker by heating on a hotplate. Additional 5 ml of acid mixture was added to ensure complete dissolution (Chester and Hughes, 1967). The semi-dry solution was re-dissolved in 4 ml of 1:1 suprapure HNO $_3$  and subsequently diluted to 100 ml using deionized water. These solutions were analyzed using an ICP-OES for the analysis of Fe, Ti, Al and K. The accuracy of the data found better than  $\pm$  5 % was assessed by the International reference material (MAG-1 Marine Sediment and SGR-1 Green River Shale) of United States Geological Survey.

Major element such as Si concentration was determined for 39 samples using PANalytical X-ray Fluorescence Spectroscopy (XRF) Model Axios at NIO Goa, India. For XRF analyses, a two cm thick subsection at every 5 cm sediment interval was sampled through the entire core length. Salt free fraction of the 2-cm-thick subsections of the sediment core were used. About 2 grams of powdered sample was treated with 1N Hydrochloric (HCl) acid to remove the carbonates associated with the sediments. The decarbonated sediments were dried and powdered. An aliquot (~1.5g) of the decarbonated sample was heated in a hot air oven at 110°C for at least 12 hours. This moisture free sample (~ 0.55g) was mixed with 5.5 g of fusion mixture (A-100 Di-Lithium Tetra Borate with 0.07% Lithium Bromide) at a ratio of 1:10, homogenized, converted to melt at 1100°C in a platinum crucible (MiniFuse2) and allowed to cool to become a glass bead. Prepared glass beads were analyzed by XRF to determine the concentration of SiO<sub>2</sub>, which is then converted to Si by using conversion ratio. Accuracy of the data was assessed using the international standard reference material (JMS-1) and found to be better than ± 5%.

The total organic carbon was analysed on bulk powdered samples by a flash dynamic combustion method using the Thermo Scientific FLASH 2000 Series Nitrogen and Carbon (CN) analyser at CSIR-NIO, Goa, India. The total inorganic content was determined using UIC CM 5014 Coulometer. The CaCO<sub>3</sub> weight per cent was calculated by multiplying the values of inorganic carbon with the ratio of molecular weight of CaCO<sub>3</sub> to C (i.e. 8.33). CaCO<sub>3</sub> % = Total Inorganic Carbon× (Molecular weight of CaCO<sub>3</sub>/Atomic weight of Carbon; Johnson et al., 2014). The total organic carbon content was calculated by substracting the total inorganic carbon from the total carbon.

#### 3. Results

#### 3.1. Rock Magnetic Properties of Sediment Core NGHP-01-17A

Two distinct magnetic zones were defined based on the down-core variations in  $\chi_{lf}$  and SIRM (Figures 1b and 2a-2b). The uppermost magnetic zone (Z-1: ~ 90 ka to 3.0 ka) showed a trend of gradual decrease in magnetic susceptibility ( $\chi_{lf}$ ), and SIRM values indicating the downcore decrease in magnetic mineral concentration in this zone (Figures 2a and 2b). In Z-I (~ 90 ka to 3.0 ka), we observed mixed trend in magnetite concentration ( $\chi_{lf}$ ) and magnetic grain size diagnostic proxy (ARM/SIRM) downcore indicating the presence of both fine and coarse-grained magnetic particles in varying concentration in these samples (Figures 2a and 2c). The Z-II (~1021.79 ka to 91.92 ka) is marked by a notable decrease in magnetic mineral concentration as seen through lower values of  $\chi_{lf}$  and SIRM compared to Z-I (Figures 2a and 2b). A systematic trend of downcore rise in ARM/SIRM in Z-II suggests the dominance of fine-grained magnetite or authigenic greigite (Figure 2c; Lin et al.,

2020; Rodelli et al., 2019). A bivariate plot of  $\chi_{ARM}$  and  $\chi_{If}$  showed the magnetic grain-size dependency (Figure S2: King et al., 1982). In general, magnetic mineralogy diagnostic parameter (S-ratio) is close to unity, but showed drops at four different sediment ages (137.02 – 55.7 ka, 745.3 – 710.1 ka, and 783.07 – 776.1 ka, and 1004.1 – 999.8 ka) suggesting four episodes (marked by S1, S2, S3, and S4) of reduced supply of magnetic particles during these periods (Figure 2d).

# 3.2. Temperature-Dependent Magnetic Susceptibility Analyses

Temperature dependence of magnetic susceptibility ( $\chi$ -T) has been widely used to determine changes in magnetic mineralogy of marine sediments (Liu et al., 2005). Temperature-dependent magnetic susceptibility ( $\chi$ -T) curves for selected samples representing different depth intervals are shown in Figures 3a-3h. A sharp decrease in  $\chi$  between 569°C and 605°C in majority of the samples imply that the bulk magnetic mineralogy is governed by titanomagnetite (Figures 3a-3h; Dunlop et al., 1997). A minor increase in  $\chi$  between 309 °C and 522 °C could be either due to the presence of titanomagnetite with a wide range of Ti-contents (Lattard et al., 2006), or conversion of pyrite into magnetite during heating process (Hirt and Gehring, 1991; Passier et al., 2001; Figures 3a-3h). In the samples from low S-ratio zones (S1, S2), a major drop in  $\chi$  at 630°C could be due to the presence of maghemite (Figures 3a and 3f).

# 3.3. Geochemical, Particle Size and Sediment Electrical Resistivity Data of Core NGHP-01-17A.

Two distinct patterns in the variation of geochemical parameters showing a major shift at  $\sim 90$  ka are observed. The TOC and Fe contents are relatively high upto  $\sim 90$  ka and decreases further downcore and showed less fluctuations (Figures 2f and 2g). CaCO<sub>3</sub> content is low in Z-I and showed large fluctuations in Z-II (Figure 2h). An interesting feature of high mean grain size and distinct low TOC and Fe content is noticed at  $\sim 783.07$  ka (Figures 2e-2g). In the low S-ratio intervals, we noticed that zone S1 is marked by low TOC and Fe content, while S2 showed low TOC and high Fe content (Figures 2d, 2f, and 2g). The S3 is marked by increase in mean grain size and low TOC and Fe content (Figures 2d, 2e, 2f, and 2g), while S4 showed minor increase in mean grain size and low TOC content (Figures 2d, 2e, and 2f). The sediment core electrical resistivity varies between 0.31 to 1.43  $\Omega$ .m (Figure 2h). Six peaks of increased sediment resistivity were identified (Figure 2i). The reconstructed  $\delta^{18}$ Osw pattern showed weak monsoon during glacial maxima and a strong during interglacials (Figure 2j; Gebregiorgis et al., 2018).

Selected elemental profiles characterizing the sediments of core NGHP-01-17A are shown in Figure 4. Profiles of elemental abundances showed significant variations in sediment inputs. The CaCO<sub>3</sub> values range from 9.24 to 40.92 wt% over the last ~ 1Myr (Figure 4b). Two distinct trend in calcium carbonate content are identified. Within Z-I, majority of samples exhibit low and uniform values of CaCO3, while Z-II samples which showed relatively higher values and large fluctuations throughout (Figure 4b). Elemental profile showed a distinct variation between the Z-I and Z-II (Figures 4c-4f). Two elemental records representing terrigenous input (iron, titanium) showed major variations in Z-I and Z-II (Figures 4c-4d). Fe and Ti content are higher in Z-I compared to Z-II (Figures 4c-4d). Overall, the K profile showed downcore increase, but does not exhibit any clear trend (Figure 7e). Downcore decrease in Si content is seen with a major drop at ~ 291.54 ka (Figure 4f). A marked shift of increase in Si content is seen at ~349.13 ka followed by a trend of relatively high values which fairly remains uniform further downcore (Figure 4f). Interelemental ratios showed variations in Z-I and Z-II (Figures 4g-4h). We record a high Fe/K ratio which mimics Ti/Al ratio in Z-I (Figures 4g-4h). A systematic trend of increase in Ti/Al and Fe/K values from ~ 61.96 to 3.0 ka and ~ 85.6 to 3.0 ka respectively are seen (Figures 4g-4h). A gradual downcore decrease in Fe/K profile within Z-II is noticed (Figure 4h).

# 3.4. Scanning Electron Microscopy (SEM-EDS) and X-ray Diffraction (XRD) Analyses

Magnetic mineralogy diagnostic rock magnetic parameter (S-ratio) indicated that the bulk magnetic mineralogy is dominated by titanomagnetite (Figure 2d). SEM-EDS analyses support our rock magnetic observations and further confirm that titanomagnetite is the dominant magnetic mineral in the studied sediments. Detrital titanomagnetite (Figures 5a-5b) and titanohematite (Figure 5c) grains of various sizes and shapes were identified in Z-I based on the grain morphology and composition. Titanomagnetite grains in Z-I appears fresh, well preserved, and less altered (Figure 5a,b). EDS data for these grains indicate the presence of titanium, iron, silicon, and oxygen with minor traces of potassium, calcium, aluminum, and vanadium (Figure 5a,b). Diagenetically formed pyrite occur as fine (Figures 5d,e) as well as framboidal crystals (Figure 5f) in the magnetically reduced zone (Z-II). EDS data indicate the presence of iron and sulfur along with minor amounts of aluminium, calcium, and silicon (Figures 5d, e, and f). A skeletal type, most probably titanohematite grain, with quadrangle plate-like structure is noticed in Z-II (Figure 5c). Titanomagnetite grain (Figure 5g) showing features of partial diagenetic dissolution was found in the bottom-most sediment interval of Z-II (~1021.79 ka to 91.92 ka).

The X-ray diffraction curves of representative bulk sediment samples from Z-I and Z-II are shown in Figure 6. Quartz (Qz) and calcite (Ct) are the dominant minerals identified in all samples (Figures 6a-6l). XRD peaks also confirmed the presence of magnetite (M) and hematite (H) in minor amounts in all the samples (S1, S2, S3, S4) exhibiting low S-ratio (Figures 6g-6l). In all the analyzed iron sulfide nodules, pyrite (Py) and quartz (Qz) are common, while aluminium (Al) is generally present in all nodules (Figures 7a-7f). Elemental sulfur (S) was also abundant in all the samples. SEM-EDS data of nodules confirms the presence of diagenetically formed iron sulfide mineral (Py) of various sizes and shapes in the analyzed samples below present-day sulfate-methane transition zone (SMTZ) (31.98 mbsf, 40.65 mbsf, and 43.61 mbsf) (Figures 8g-8l). EDS data indicate the presence of sulfur and iron along with minor amounts of aluminium, and silicon (Figures 8j-8l).

# 3.6. Correlation between Magnetic, Geochemical and Sedimentological Parameters

We examined the bivariate data plots to evaluate the plausible constraints on the sediment properties (Figure 9). A good covariation between  $\chi_{lf}$  and SIRM is observed only in Z-I samples. The difference between slopes of Z-I and Z-II reflects the presence of ferri-and paramagnetic type mineral assemblages in these samples respectively (Figure 9a). Z-II samples showed much lower values of  $\chi_{lf}$  and SIRM indicating the reduction in concentration of ferrimagnetic minerals and also hint on the presence of either para or anti-ferromagnetic minerals (Figure 9a).

Two different trends exhibited by two distinct population of magnetic particles were observed in bivariate data plot between  $\chi_{lf}$  and ARM/SIRM within Z-I (Figure 9b). Uppermost sediment interval (~34.32 ka to 3.0 ka) within Z-I is dominated by high  $\chi_{lf}$  and shows trend of down-core fining in magnetic grain size (Figure 9b). Comparatively, lower sediment interval (~ 89.55 ka to 35.21 ka) exhibited relatively low  $\chi_{lf}$  and showed a trend of down-core coarsening in magnetic grains (Figure 9b). In general, S-ratio ranges between 0.73 and 0.99 and show wide variations in  $\chi_{lf}$  (Figure 2d and Figure 9c). Few samples from both zones exhibited low S-ratio indicating the presence of high coercive minerals (Figure 9c). Coupling between  $\chi_{lf}$  and mean grain size is strong only for Z-I samples, with highest values of  $\chi_{lf}$  dominated by coarse-grained magnetic particles (Figure 9d). Z-II samples showed much narrow range in  $\chi_{lf}$  and wider variations in mean grain size (Figure 9d). Coupling between magnetic grain size and physical grain size is generally weak for all samples. (Figure 9e). As seen through magnetic parameters (Figures 2a, 2b), Z-II samples are dominantly composed of paramagnetic minerals. High Fe content in few Z-II samples (Figures 2g, 9f) might be due to the presence of paramagnetic minerals (chlorite, pyrite or clays). Based on these co-variations, two distinct groups exhibiting similar characteristic are identified (Figure 9f). Z-I samples are

relatively rich in Fe content and exhibited higher  $\chi_{lf}$  (Figure 9f), while few samples in Z-II showed relatively low  $\chi_{lf}$  and Fe content. A cross plots (Figures 9g-9h) between Fe/K (proxy for intensity of chemical weathering; Govin et al., 2012) and magnetic susceptibility (concentration dependent; Thompson and Oldfield, 1986) provided important constraints on the weathering and erosional process controlling the magnetic grain size variations within Z-I.

#### 4. Discussion

# 4.1. Rock Magnetic and Geochemical Evidences of Tectonic Control on the Sedimentary Evolution of the Andaman Sea

Tectonic uplift in source regions coupled with regional monsoon climate may affect the sediment provenance by modifying the topography, erosion regime, reorganization of the river drainage pattern and sediment routing system and consequently provide major constraints on the evolution of sedimentary system (Armitage et al., 2011; Clift et al., 2002, 2010; 2017; Matenco and Haq, 2020; Najman et al., 2019, 2020; Tang et al., 2016; Zhang et al., 2018a). Previous studies have demonstrated that tectonic and climatic changes in the source regions can be very well inferred from sediment rock magnetic, geochemical, grain size, mineralogical, and chronological records. For instance, a dedicated mineral magnetic, geochemical, sedimentological and mineralogical studies recorded the signals of tectonic uplift in the Himalayan region (Phartiyal et al., 2003), southern Alborz Mountains, northern Iran (Ballato et al., 2008), Tibetan Plateau (Govin et al., 2018; Hu et al., 2019; Tan et al., 2020; Tang et al., 2016; Yang et al., 2019), Back-Arc Basins, Western Pacific Ocean (George DeVries Klein, 1985), Kumano basin, Japan (Shi et al., 2017), Izu-Bonin forearc, northwest Pacific (Robertson et al., 2018), Ganges-Brahmaputra-Meghna delta (Grimaud, J.-L., et al., 2017), Pannonian Basin, Hungary (E Thamó-Bozsó et al., 2002), and Al Rehaili area, Northern Jeddah, Saudi Arabia (Algahtani and Khalil, 2019). More recently, Peketi et al (2021) revealed the control of climatic and tectonic activities on the Bengal fan sedimentation using Sr-Nd isotopic data of the hole U1445A from the western Bay of Bengal (Peketi et al., 2021).

Magnetic minerals are capable to record a wide variety of geological processes including tectonics, provenance, weathering/erosion and diagenetic changes (Hatfield and Maher, 2008, 2009; Oldfield et al., 1985). IBR situated in the southern branch of the Himalayan–Tibetan Plateau has undergone multiple upliftments including the rapid rise at ~8 Ma, ~3.6 Ma, ~2.6 Ma, 1.8–1.7 Ma, 1.2–0.6 Ma and 0.15 Ma (Li, 1995; Li et al., 1996, 2014; Najman et al., 2019, 2020). Geology of IBR is dominated by igneous, metamorphic, and sedimentary rocks (Robinson et al., 2007). A well-developed ISS river systems flowing through these terrains assure high supply of magnetic minerals

to the AS (Bender, 1983; Colin et al 1998; Kurian et al., 2008; Licht et al., 2013; Robinson et al., 2007; 2014). In the studied core, a significant shift in the depositional environment is observed around  $\sim 90$  ka and is marked by rapid change in magnetite concentration ( $\chi_{\rm lf}$ ), Fe, Ti and TOC content in Z-I (Figures 2a, 2f and Figures 4c-4d). A linkage between magnetic enhancement followed by compositional, chronological (detrital zircon U-Pb, Ar-Ar ages), and isotopic changes in the terrestrial and marine sediments with the shifting of source areas due to discrete erosional unroofing events and drainage reorganization constrained by tectonic uplift is well-established. For example, in the Bengal fan (Peketi et al., 2021), Indus Basin, Arabian Sea (Kumar et al., 2020), Kameng River Section, eastern Himalaya (Chirouze et al., 2013), Celano catchment, central Italy (Forzoni et al., 2014), Miocene foreland basin, northern Iran (Ballato et al., 2008), Nanga Parbat massif, western Himalaya (Clift, 2017), NW Borneo (Menier et al., 2019), and Gulf of Mexico (Smith et al., 2019). In context of regional geology of IBR, increased Fe and Ti content (Figures 4c-4d) observed in Z-I are mainly derived from the Late Precambrian and Cretaceous-Eocene metamorphic/basic/ultrabasic rocks (Bender 1983; Robinson et al., 2014). Age of most recent tectonic upliftment in the IBR is ~ 150 ka (Li and Fang, 1999; Li et al., 2014; Najman et al., 2006, 2008, 2012; Qingsong Z, 2000). A recent study based on high-resolution seismic, water column, sediment core, and chronology data delineated the control of tectonic and oceanographic processes on the sediment dispersal and accumulation off ISS delta (Kuehl et al., 2019), Bengal fan (Peketi et al., 2021), and Indus Basin, Arabian Sea (Kumar et al., 2020). A trend of upward increase in χ<sub>If</sub> from ~ 90 ka to 3 ka in core NGHP-01-17A indicate increase in detrital flux to the AS and represents the onset of a new sedimentary regime following most recent event of tectonic uplift in IBR which occurred at  $\sim 150$  ka. A similar  $\gamma_{\rm lf}$  enhancement accompanied by coarser grain size was recorded in the sediment core (IODP Site U1457) from Indus Fan, Arabian Sea (Kumar et al., 2020). They attributed the magnetic enhancement to the rapid change in erosional processes and sediment supply linked with tectonic-climatic interaction.

Drainage reorganization and sediment routing system provides important constraints on the evolution of source-to-sink depositional systems (Chirouze et al., 2013; Govin et al., 2018; Lenard et al., 2020; Najman et al., 2016). Hence the sedimentary record from those systems provide vital insights on the tectonic and climatic forcing on the sediment delivery to the marine basin (Kuehl et al., 2019; Lenard et al., 2020; Smith et al., 2019). Several studies reported that the rates of tectonic uplift coupled with monsoonal changes in the source region determines the time required to develop an efficient drainage system to produce large sediment load into the marine sinks. For example, delayed response of sedimentation and provenance changes in terrestrial and marine basin due to hinterland tectonics,

drainage evolution, and change in sediment routing system are well-recorded in the Bengal fan (Lenard et al., 2020; Peketi et al., 2021), Ganga-Brahmaputra drainage basin (Chirouze et al., 2013), Neogene Siwalik foreland basin of the Himalaya (Huyghe et al., 2014), off ISS delta (Kuehl et al., 2019), Central Myanmar Basin (Najman et al., 2019, 2020), Canada (Shugar et al., 2019), Gulf of Mexico (Smith et al., 2019), Back-Arc Basins of the Western Pacific (George DeVries Klein, 1985), and offshore western Norway (Sømme et al. 2019). Thermochronological and provenance based studies provided evidence on the multi-phase upliftment of IBR (Najman et al., 2006, 2019, 2020). Their findings revealed that tectonic upliftment in the IBR significantly modified the ISS drainage network and constrained the delivery of continental sediments to the AS (Cochran, 2010; Derry and Lanord, 1996; Kuel et al., 2019; Najman et al., 2019, 2020; Rodolfo, 1969b; Roy and Sharma, 1993; Zhang et al., 2015). Response of landscape to tectonic and climatic forcing control the source to sink sediment transfer (Fildani et al., 2016). Based on the detrital zircon U-Pb and (U-Th)/He dating analyses on the sediment cores from Mississippi deep-sea fan, Fildani et al. (2016) reported that the response of landscape to tectonic and climatic forcing controlled the sediment accumulation and movement. However the timing and magnitude of sediment storage can cause lag in the transfer of sediments from terrestrial to marine sink (Kumar et al., 2020; Romans et al. 2016). For instance, Clift and Giosan, 2014 reported largest time-lag in the Indus submarine fan sedimentation after the tectonic uplift in the Himalayan-Tibetan plateau (Clift et al., 2010; Jonell et al., 2017; Lavo and Avouac, 2001). We hypothesized that a time-lag between recent tectonic uplift in the source region (IBR ~ 150 ka; Li and Fang, 1999; Li et al., 2014; Qingsong Z, 2000; ) and onset (~ 90 ka, present study) of increased sedimentation in Z-I (recorded through increase in  $\chi_{lf}$ , Fe, Ti and TOC; Figures 2a, 2g, 2f, and 4d) could be attributed to the delay in sediment delivery caused by the development and re-organization of drainage basin, and sediment routing system of ISS rivers in IBR region after the recent uplift event (Najman et al., 2016; 2019, 2020). However additional chronological and numerical modelling (geomorphological, erosion and deposition of sediment) data over longer timescales is needed to clearly attribute the influence of tectonic forcing on the sedimentation in the AS. Increase in Fe and Ti content in Z-I ( $\sim 90 \text{ ka} - 3 \text{ ka}$ ) is also indicative of strong weathering and erosion induced by tectonic uplift in IBR and is well recorded in the elemental data (Figures 4c, 4d, 4g, and 4h). A bivariate plots (Figures 9b, 9g, 9h) between magnetic (ARM/SIRM; Sebastian et al., 2019) and inorganic geochemical (Fe, Fe/K, Ti/Al) based proxy parameters provided additional constraints on the weathering regime (Colin et al., 1998; Govin et al., 2012; Kissel et al., 2003) controlling the magnetic grain size variations within Z-I. Two distinct magnetic grain size populations (group-A and group-B) exhibiting different characteristic are identified (Figure 9a).

Samples from group-A are distinctly marked by higher  $\chi_{lf}$ , Fe, Fe/K, Ti/Al and relatively lower ARM/SIRM values compared to group-B samples which showed relatively lower  $\chi_{lf}$ , Fe, Fe/K, Ti/Al and higher ARM/SIRM values (Figure 9). Magnetic grain size proxy (ARM/SIRM; Evan and Heller, 2003) has been successfully utilized to identify the weathering regime (Kissel et al., 2003; Sebastian et al., 2019) and track the monsoonal changes (Colin et al., 1998). Relatively low  $\chi_{lf}$  manifested by the dominance of coarser magnetic particles in group-B (~ 90 – 33.21 ka) samples within Z-I most likely reflect the period of increased physical weathering and erosion in the catchment (Figure 9b). Enhanced increase in the supply of fine-grained magnetic particles (as seen through elevated ARM/SIRM values; Figure 2c) manifested by high Ti/Al values (Figures 4g and 9h) in group-A (~ 34.32 – 3 ka) samples within Z-I is indicative of period of increased chemical weathering in IBR. This is also well reflected in bivariate plots between Fe/K versus  $\chi_{lf}$  and Ti/Al versus  $\chi_{lf}$  (Figures 9g-9h). Higher values of Fe/K (Figure 9g) in group A is also indicative of the presence of more humid climatic conditions within the catchment area (Mulitza et al., 2008).

Previous studies reported that decline in vegetation cover, aridification, intensification of erosion in the catchment areas during drier period could result in higher supply of detrital hematite (Clift et al., 2017; Hu et al., 2019; Phartiyal et al., 2003; Shen et al., 2017; Wan et al., 2010; Yang et al., 2016). Aridification has been attributed to the interaction between the uplift of the Tibetan Plateau and global cooling. For example, intense arid condition in central Asia induced by the uplift of the Tibetan Plateau has been established from multi-proxy records (Kutzbach et al., 1989; Li and Fang, 1999; Liu and Yin, 2002; Manabe and Terpstra, 1974; Tada et al., 2016). Four major episodes (S1, S2, S3, S4) of low S-ratio are identified in the studied core (Figure 2d). SEM-EDS observations on the magnetic particles S1 (~ 137.02 ka to 55.7 ka, Figure 5c) showed the presence of titanohematite grain which might have been formed either due to increased arid conditions in IBR (Clift, 2017; Hu et al., 2019; Phartiyal et al., 2003; Shen et al., 2017; Wan et al., 2010; Yang et al., 2016;) or progressive reductive dissolution of ferrimagnetic minerals at sulfidic fronts (Nowaczyk, 2011; Rodelli et al., 2018; 2019).

Presence of authigenically formed magnetic iron sulfides in S2 and S3 (magnetically reduced zone) is indicative of intense magnetic mineral diagenesis in Z-II (Figures 2a, 2b, Figures 5e-5f, Figures 8d-l). Multiple bands of diagenetically formed iron sulfide nodules at different periods in Z-II indicate episodic intensification of AOM due to enhanced methane-flux (Figure 8). Previous studies have reported that sedimentary greigite and pyrrhotite can also exhibit higher coercivities which could result in low S-ratio (Kars and Kodama, 2015; Horng, C.S., 2018). Authigenically formed

pyrite were noticed in S2 and S3 (Figures 5e-5f). We hypothesize that these low S-ratio intervals (S2, S3) contained previously formed authigenic ferrimagnetic iron sulfide (greigite), which were no longer stable and subsequently got converted into pyrite (Kars and Kodama, 2015a,b). Recent magnetic mineral diagenetic study by Gaikwad et al., 2021 reported similar mechanism explaining the transformation of greigite to pyrite in a newly discovered active methane seep site from the Bay of Bengal. The low S-ratio (S2, S3) intervals are also manifested by low  $\chi_{lf}$  and SIRM values (Figure 2a and 2b) in Z-II and supports our interpretation.

# 4.2. Diagenetic Modulation of Magnetic Mineral Properties

Drilling and coring activities conducted under the aegis of first National Gas Hydrate Expedition -01 (NGHP-EXP-01), onboard D/V JOIDES Resolution confirmed the presence of subsurface gas hydrate deposits manifested in the form of a bottom simulating reflector (BSR) at Site 17 in the AS (Collett et al., 2008, Kumar et al., 2014). Gas hydrates occurred as pore-filling material ranging from disseminated to highly saturated in volcanic ash beds (Kumar et al., 2014; Winters et al., 2014). Sediment core NGHP-01-17A retrieved from potential gas hydrate bearing region (Site 17) provides a unique opportunity to reconstruct a long-term history of methane-influenced magnetic mineral diagenesis in the AS.

Diagenesis of magnetic mineral is one of the vital post-depositional factors which could significantly affect the sediment magnetic record. Detrital iron-bearing minerals reacts with hydrogen sulfide produced via bacterial mediated sulfate reduction processes or AOM and gets further transformed into iron sulfides (Berner, 1970; Canfield, 1989; Jørgensen, 1982). Changes in concentration, grain size and mineralogy of magnetic particles in core NGHP-01-17A from ~1021.79 ka to 504.4 ka appears to be largely influenced by methane-related diagenetic processes induced by AOM coupled sulfate reduction (Figures 2a-2d). Absence of diagenetically magnetic iron sulfides (pyrite, greigite, pyrrhotite) and nodules in Z-I suggests that diagenesis did not have any significant effect on the sediment magnetic record as confirmed through SEM-EDS and XRD data (Figures 5a-5b and Figures 6a, 6b, 6g, and 6h). Interestingly, Z-II is marked by distinct low  $\chi_{If}$  values and showed the presence of numerous bands of iron sulfide nodules below present-day SMTZ (> 504.46 ka) (Figure 8a and Figures 8d-8l). We propose that these nodules were probably formed as a result of multiple methane seepage events, which led to enhanced AOM-coupled sulfate reduction at different times (Figures 8d-8l; Dongen et al., 2007; Horng, 2018; Mei et al., 2018).

Past studies reported the presence of iron sulfides, greigite and pyrite in methane and gas hydrate bearing marine sediments. For example, in Nankai Trough (Kars and Kodama, 2015a,b), K-G basin

(Badesab et al., 2017, 2019; Dewangan et al., 2013), offshore Taiwan (Horng and Roberts, 2018), and Cascadia Margin (Housen and Musgrave, 1996; Larrasoaña et al., 2007; Musgrave et al., 2006). Mineralogical (SEM-EDS and XRD) analyses of iron sulfide nodules from (Z-II) showed the presence of pyrite (Figures 7a-7f and Figures 8d-8l). However, we did not observe any signatures of greigite in our mineralogical data (Figures 7a-7f and Figures 8g-8l). Lack of greigite in these samples can be explained by the fact that previously formed greigite might have got converted into more stable paramagnetic pyrite due to increase sulfide concentration which favoured pyritization process to complete (Berner, 1984; Kars and Kodama, 2015b).

In gas hydrate settings, formation of SMTZ is primarily dependent on the sedimentation rate, intensity of upward-migrating methane fluxes from deeper sedimentary layers and downward diffusing sulfate concentration (Canfield and Berner, 1987). Occurences of numerous bands of iron sulfide nodules in Z-II provide hints on the strong build-up of paleo-SMTZ fronts formed as a result of long durational AOM-coupled sulfate reduction controlled by enhanced methane flux at Site 17 (Figures 8d-8f, and Table-01). Similar iron sulfide nodules were reported in the sediment cores from Yungan Ridge, Taiwan (Horng, 2018) and Western Siberia (Rudmin et al., 2018). Previous studies have reported the concurrence of distinct magnetic susceptibility minima close to SMTZ for example, in Argentine continental slope and continental margin off Uruguay (Garming et al., 2005; Riedinger et al., 2005). They suggested that enhanced hydrogen sulfide produced at SMTZ led to the dissolution of magnetite and subsequent precipitation of paramagnetic pyrite and produced distinct minima in χ<sub>lf</sub>. In marine sedimentary sequences from southwestern Taiwan, Kao et al. (2004) reported the occurrence of higher susceptibility peaks at SMTZ associated with presence of ferrimagnetic greigite formed as a result of limited availability of hydrogen sulfide, while enhanced concentration favoured the formation of pyrite. In core NGHP-01-17A, a distinct minima in χ<sub>lf</sub> is near ~ 90 ka which is much shallower than the depth of present day-SMTZ (~504.46 ka). (Figure 2a). It is interesting to note that we did not observe any iron sulfide nodules or distinct change in  $\chi_{\rm lf}$ near present-day SMTZ (Figures 8a-8c). In fact the  $\chi_{\rm lf}$  were rather low and showed more uniformity in their variations (Figure 8a). Mineralogical diagnostic (XRD) data of the samples close to SMTZ revealed the presence of both magnetite and pyrite (Figure 6e).

Marine gas hydrate system is sensitive to changing pressure and temperature conditions, and tectonic forcing can cause the migration of gas hydrate stability zone (GHSZ) (Shi et al., 2017). Detailed mineral magnetic and electron microscopic study by Larrasoana et al. (2007) on a sediment core from Southern Hydrate Ridge, offshore Oregon suggested that migration of methane through the

sediment column could result in the formation of iron sulfide nodules. Their work highlighted that tectonically forced fluid migration can significantly affect the normal steady state diagenesis and favour the formation of nodules. The present-day SMTZ in core NGHP-01-17A is at ~ 25 mbsf (Figures 8b-8c). Eighteen bands containing diagenetically formed iron sulfide (pyrite) nodules were found to be spatially distributed below present-day SMTZ (Figures 8d-8l, and Table-01). A recent high resolution rock magnetic and electron microscopy study conducted by Shi et al., 2017 on IODP core Site C0002 in the Kumano Basin, Nankai Trough, Japan established the linkage between tectonically driven methane migration and formation of iron sulfides. They reported that tectonic activities in Kumano Basin triggered methane venting from deeper sediment layers and caused the formation of pyrite aggregates along sediment fractures/faults which acted as conduits for methane migration. Similar observations were reported from Southern Hydrate Ridge by Larrasoaña et al. (2007). The sediment core NGHP-01-17A lies within a seismically active region and the sedimentary sequence at this site provides a record of volcanic activities in the AS (Kumar et al., 2014; Kamesh Raju et al., 2007). Recently, Rose et al. (2014) reported the occurence of gas hydrates in numerous coarse-grained ash layers produced as a result of volcanic activity (Cawthern et al., 2010) in core NGHP-01-17A. High permeability offered by such coarse-grained ash layers provided conducive environment for the methane migration through sediment column and constitute a dominant host lithology for nucleation of gas hydrates at this site (Collett et al., 2008; Rose et al., 2014). Organic geochemistry and pressure-core studies confirmed the presence of abundant methane starting below SMTZ till the bottom of the cored sediment section in NGHP-01-17A (Collett et al., 2008). High electrical resistivity measured in the bottom part of the studied core (~ 954.74 - 924.46 ka) indicate increased gas hydrate saturations (Figure 2i) (Collet et al., 2008; Winters et al., 2014). Tectonically induced migration of methane and shifting of SMTZ front was recorded in pore-water geochemical profiles of sediment cores from the active Ganos Fault in the Sea of Marmara (Halbach et al., 2004) and U-Th dating of seep carbonates, Vestnesa Ridge, off West Svalbard, Arctic (Himmler et al., 2019). We hypothesize that tectonic activities at Site 17 might have facilitated the episodic release of methane from the deeper gas reservoir which caused multiple episodes of enhanced methane flux followed by AOM leading to precipitation of iron sulfide nodules at different intervals in Z-II (Figures 8d-8f, and Table-01). However, additional organic geochemistry investigations are warranted to further confirm this hypothesis.

A plausible mechanism is proposed to explain the controls on the evolution of sedimentary magnetic zones (Z-I, Z-II) over the past ~ 1 Ma based on the magnetic mineralogies, elemental composition, porewater geochemistry, grain size data, tectonic events and age —depth model (Figure 10). Z-I

represent the onset of a new sedimentary regime ( $\sim$  90 ka) following the most recent event of tectonic uplift in the IBR which occurred at  $\sim$  150 ka. A trend of upward increase in  $\chi_{lf}$  from  $\sim$  90 ka to 3 ka in core NGHP-01-17A indicate increase in detrital flux to the AS. In Z-II, tectonically forced methane migration controlled the diagenesis of magnetic minerals which led to the significant dissolution of detrital iron oxides and subsequent transformation to iron sulfides (pyrite). This is clearly evident in the rock magnetic and SEM-EDS data (Figures 2, 5, and 6). Numerous bands of diagenetically formed iron sulfide nodules (Figure 7 and Figure 8) at different intervals in Z-II indicate episodic intensification of AOM due to enhanced methane-flux.

#### 5. Conclusion

Rock magnetic, sedimentological, mineralogical and inorganic geochemistry data of sediment core (NGHP-01-17A) demonstrated the magnetic property variations are controlled by changes in the magnetite content, mineralogy, and grain size, which are linked to tectonic upliftment of the IBR (Z-I), aridification, and tectonically-induced fluid migration in the AS (Z-II). Major change in the depositional environment near  $\sim 90$  ka is marked by rapid shift in magnetite concentration ( $\chi_{If}$ ), Fe, Ti and organic carbon content in Z-I. Enhanced influx of detrital magnetic minerals from  $\sim 90$  ka to 3 ka represents the onset of a new sedimentary regime following most recent event of tectonic uplift in the IBR which occurred at  $\sim 150$  ka.

Multiple bands of diagenetically formed iron sulfide nodules at different periods in Z-II indicate episodic intensification of AOM due to enhanced methane-flux. We propose that fluid migration mainly driven by tectonic processes facilitated episodic release of methane from deep reservoir that led to the formation of iron-sulfide nodules in the AS. Rock magnetic properties effectively recorded the geological processes that controlled sedimentation over ~ 1 Ma in the Andaman Sea. A plausible conceptual model summarizing all possible constraints on the sedimentary evolution of the AS is developed in Figure 10.

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Table-01: Depth-wise distribution of iron sulfide nodules in sediment core NGHP-01-17A

Sr. No.	Nodule No.	Core depth
		Interval (mbsf)
1.	AN-01	31.56
2.	AN-02	31.98
3.	AN-03	32.22
4.	AN-04	35.07
5.	AN-05	36.83
6.	AN-06	40.50
7.	AN-07	40.49
8.	AN-08	40.61
9.	AN-09	40.63
10.	AN-10	40.65
11.	AN-11	41.08
12.	AN-12	41.13
13.	AN-13	41.94
14.	AN-14	42.03
15.	AN-15	43.59
16.	AN-16	43.61
17.	AN-17	44.57
18.	AN-18	48.46

Figure -1: (a) Bathymetric map of Andaman Sea and the sediment core location (NGHP-01-17A) (modified after Awasthi et al., 2014), (b) Down-core variation in sediment magnetic susceptibility record of NGHP-01-17A, and (c) General geological map of the Andaman Sea sediment source regions modified after Guo et al. (2016). Sediment magnetic zones are marked by red (Z-I) and blue (Z-II) color line respectively. The depth of sulfate-methane transition zone (SMTZ) is marked by the dotted (pink color) line.

Figure-2: Multi-proxy sediment record of core NGHP-01-17A (a-d) rock magnetic ( $\chi_{If}$ , SIRM, ARM/SIRM, S-ratio), (e) Sedimentology (mean grain size), (f-h) Geochemical (TOC, Fe and CaCO<sub>3</sub> content) parameters, (i) Physical property (sediment electrical resistivity; data taken from NGHP Expedition-01 report; Collet et al., 2008), and (j) Ice volume corrected  $\delta^{18}$ Osw record (Gebregiorgis et al., 2018). Sediment magnetic zones are marked by red (Z-I) and blue (Z-II) color line respectively. Please note that four low S-ratio zones are marked as S1, S2, S3, and S4. Distribution of volcanic ash layers (sampled from sediment core) are indicated by arrows (light blue color). Timings of tectonic events and uplift of the NE Tibetan Plateau are presented and marked by dashed lines (for details please see Li and Fang, 1999; Li et al., 2014; Qingsong Z, 2000). Episodes of Kunlun–Huanghe (Yellow River) Movement (KHM, 150 Ka) and Gonghe Movement (GHM, 1200 ka – 600 ka) are mentioned.

Figure-3: Temperature-dependence of magnetic susceptibility ( $\chi$ -T) for selected representative sediment samples (a-d): Zone-I and (e-h) Zone-II of core NGHP-01-17A. Solid red lines indicate heating curves, and blue lines indicate cooling curves.

Figure-4: Vertical distribution of (a) magnetic susceptibility ( $\chi_{lf}$ ), (b) calcium carbonate content, (c-f) selected major element compositions of sediments and (g-h) elemental ratios in the core NGHP-01-17A.

Figure-5: Scanning electron microscope images (secondary electron) on magnetic extracts representative of two different sediment magnetic zones (a-b: Zone-I and c-g: Zone-II) of core NGHP-01-17A. Energy dispersive X-ray spectroscopy spectra are placed adjacent to the respective images (a–g). Iron (Fe), titanium (Ti), sulfur (S), oxygen (O), calcium (Ca), silicon (Si), carbon (C), aluminum (Al) and magnesium (Mg) peaks are indicated.

Figure-6: X-ray diffraction spectra for selected representative bulk sediment samples of core NGHP-01-17A: Zone-I (a,b,g,h) and Zone-II (c,d,e,f,i,j,k,l) of core NGHP-01-17A. M = magnetite, Py = pyrite, Qz = quartz, Calcite = Ct, H=Hematite.

Figure-7: (a-f) X-ray diffraction spectra for selected iron sulfide nodules picked from core NGHP-01-17A. Qz = quartz, S=Sulfur, G = Gypsum, Py = pyrite, Al=aluminium.

Figure-8: (a) magnetic susceptibility ( $\chi_{lf}$ ) (b) pore-water sulfate (c) methane gas profile of sediment core NGHP-01-17A. (d-f) microscopic images of iron sulfide nodules, (g-i) scanning electron microscope images (secondary electron) of iron sulfide nodules. (j-l) energy dispersive x-ray spectra are placed close to the respective images. Iron (Fe), sulfur (S), oxygen (O), silicon (Si), carbon (C), aluminum (Al) peaks are indicated. Brown color dashed lines indicates the depth of nodule occurrence. Pore-water sulfate and methane gas data of core NGHP-01-17A was taken from NGHP Expedition-01 report (Collet et al., 2008).

Figure-9: (a-c): Bivariate scatter plots of magnetic parameters: magnetic susceptibility ( $\chi_{lf}$ ), saturation isothermal remanent magnetization (SIRM), anhysteretic remanent magnetization ARM/SIRM, and S-ratio), (d)  $\chi_{lf}$  versus mean grain size, (e) ARM/SIRM versus mean grain size, (f)  $\chi_{lf}$  versus iron content (Fe %) content, (g) Fe/K versus  $\chi_{lf}$ , and (h) Ti/Al versus  $\chi_{lf}$ . Samples from group-A are distinctly marked by relatively higher  $\chi_{lf}$ , Fe, Fe/K, Ti/Al and relatively lower ARM/SIRM values compared to group-B samples. Clusters of group-A and group-B samples are highlighted in light blue and light green color respectively in Figure 9g and 9h. Note that gray arrows in the scatter plots are used only to highlight trends.

Figure-10: (a-b): A plausible mechanism explaining the controls on the evolution of sedimentary magnetic zones (Z-I, Z-II) in the Andaman Sea over the past ~ 1 Ma.

# **Supplementary Figures:**

Figure S1. Age-depth model for sediment core NGHP-01-17A. Age model was developed based on AMS <sup>14</sup>C dates on planktonic foraminifera by Gebregiorgis et al. (2018). Age depth model indicating a near linear sedimentation rate.

Figure S2. A bivariate plot between  $\chi_{lf}$  and  $\chi_{ARM}$  exhibiting the grain-size control on the iron bearing sediments (King's plot).

Figure. 1

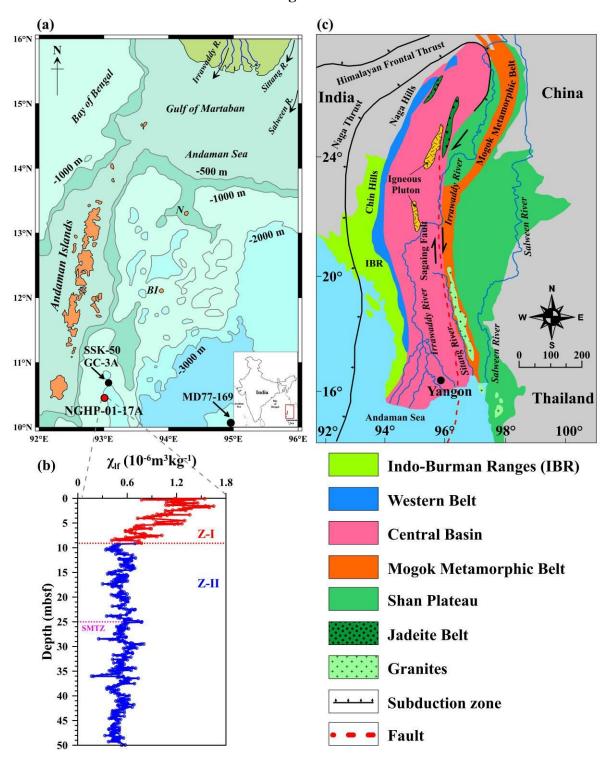


Figure. 2

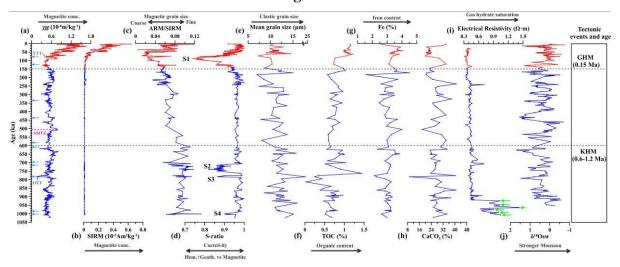


Figure. 3

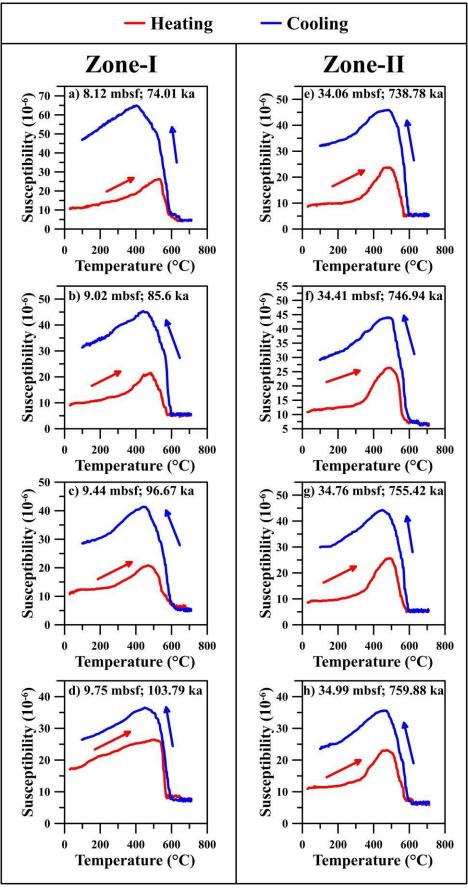


Figure. 4

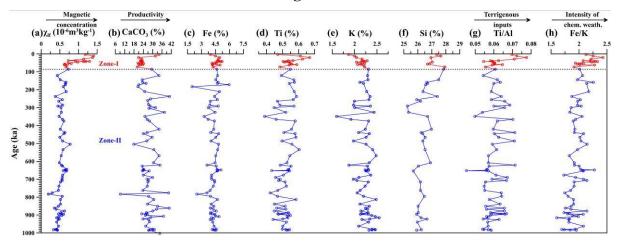


Figure. 5

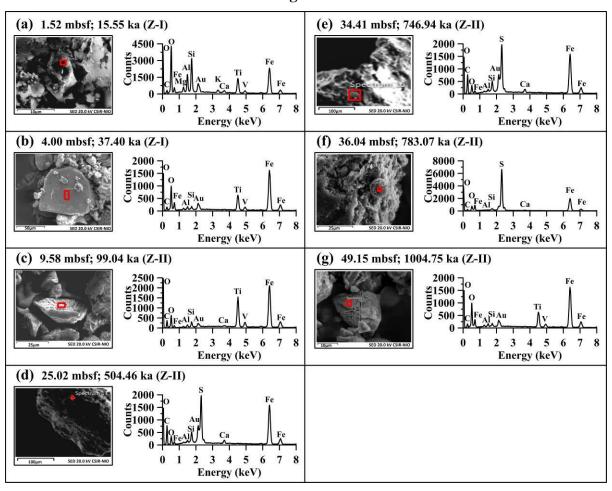


Figure. 6

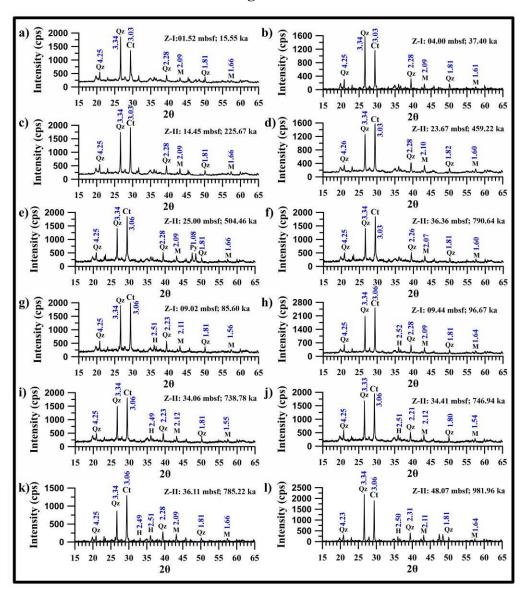


Figure.7

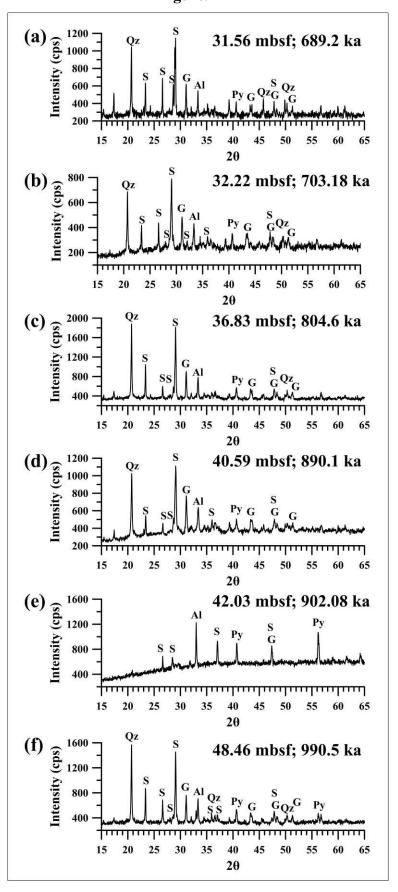


Figure. 8

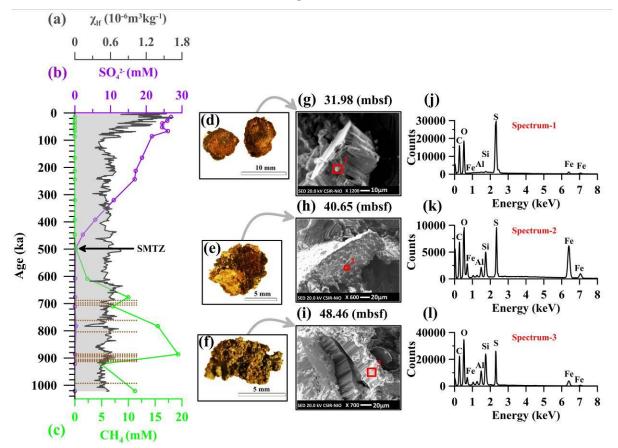


Figure. 9

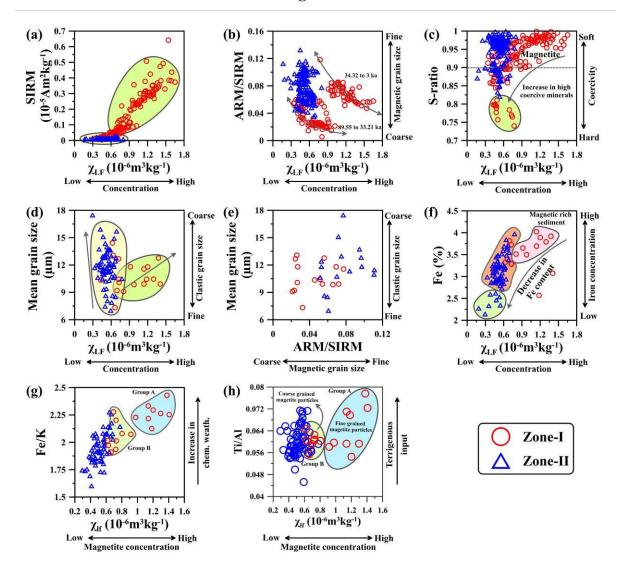


Figure. 10

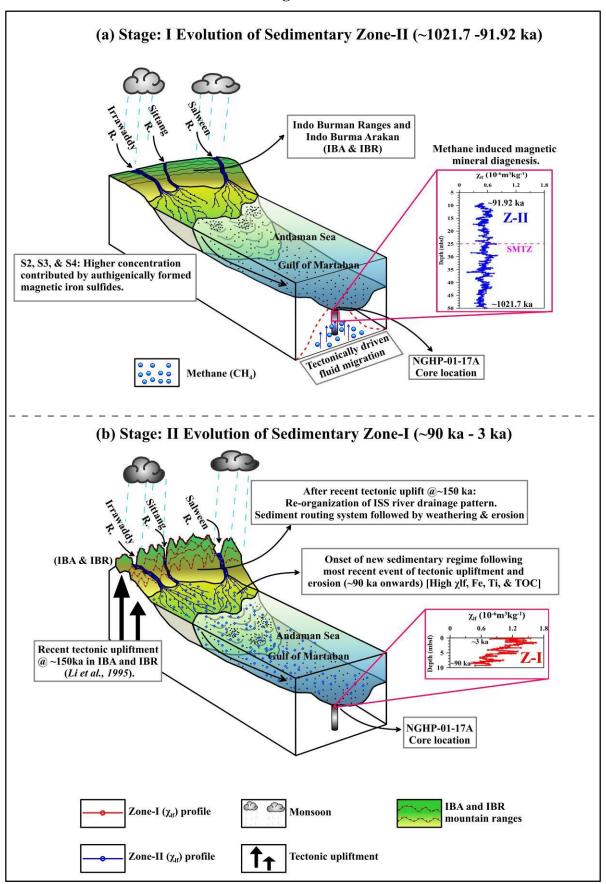


Figure. S1

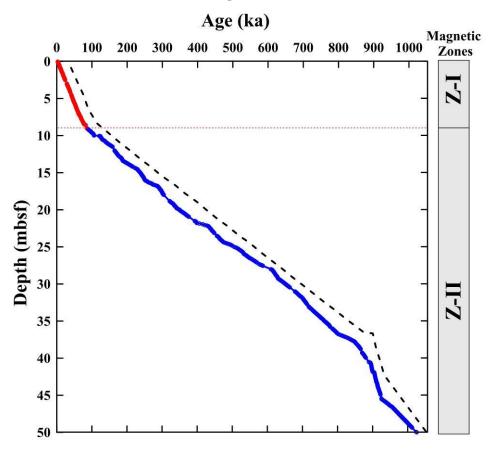


Figure. S2

