

Lead distribution in coastal and estuarine sediments around IndiaSuchararita Chakraborty¹, Parthasarathi Chakraborty^{1*} and B. Nagender Nath¹

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Abstract:

This study describes the geochemical distribution of lead (Pb) and identifies the critical factors that significantly control Pb distribution and speciation in coastal and estuarine sediments around India by using published data from the literature. Crustal sources influence the abundance of Pb in coastal sediment from the south-east and central-west coast of India. Parts of north-east, north-west, and south-west coast of India were polluted by Pb. Distribution of Pb in sediments, from the north-east and north-west coasts of India, were controlled by Fe-Mn oxyhydroxide mineral phases of the sediments. However, organic carbon (OC) seemed to be a dominant factor in controlling the distribution of Pb in sediments from the central-east and south-west coasts of India. The outcome of this study may help in decision-making to predict the levels of Pb from natural and anthropogenic sources and to control Pb pollution in coastal and estuarine sediments around India.

Key Words: Lead, India, coastal and estuarine sediments; Pb geochemistry; Lead pollution

Introduction

Dissolved lead (Pb) is quickly scavenged by negatively charged solid surfaces like clays, carbonates, oxides and hydroxides of Fe, Mn as well as organic carbon (OC) in water column. As a consequence, non-chelated or dissolved Pb has a short water-column residence time in marine environments. Settling of Pb associated particulate matters usually control the distribution of Pb in a particular ocean basin. However, total concentration of Pb in coastal/estuarine sediments does not provide useful information about its speciation, bioavailability and geochemistry in the system. Quality and quantity of different metal binding ligands present in coastal sediments has been reported to control physicochemical forms of metals in the system (Chakraborty et al., 2015, 2014a, 2014b, 2014c; Chakraborty 2012; Chakraborty et al., 2012a, 2012b, 2012c). In coastal sediments, Pb can be present in different physicochemical forms (water soluble and exchangeable Pb complexes; Pb associate with carbonates phase; Pb bound to oxyhydroxide of Fe and Mn; Pb bound to organic matter and sulphides etc). Distribution of Pb in different binding phases of sediment has been used to understand Pb-geochemistry in a sediment system. There are very few speciation studies have been performed in coastal sediments to understand Pb-geochemistry in the system. However, several studies have been carried out to understand the distribution of total Pb content in coastal sediments around India over the last few decades; nevertheless, the information is quite limited and discrete to understand Pb distributions and their geochemistry in the coastal sediments.

Pb has been reported to have different affinities for different binding-phases of coastal sediments. It has been reported that carbonate phase in coastal sediments plays a key role in controlling Pb distribution (Fulghum et al., 1988) in sediments. Scavenging property of Pb by Fe/Mn oxyhydroxide phase in sediment has also been identified as an important process in controlling speciation and geochemistry of Pb in sediments system (Jones and Turki, 1997; Tessier et al., 1979). Organic binding phase has been reported to control Pb distribution and its speciation in coastal sediment (Chakraborty et al., 2012 and Krupadam et al., 2007). However, poor correlation between Pb and sedimentary organic matter (SOM) has also been reported by Abaychi and Douabul, (1986).

In addition, the composition of source rocks of coastal sediments also influences Pb geochemistry in sediment of a particular coastal area (Loring, 1991; Selvaraj et al., 2004). The diverse geology, the long coastline (~7000 km), climatic variability, oceanographic settings, and anthropogenic activities along the coastline of India make it extremely difficult to predict distribution patterns of Pb in coastal and

estuarine sediments. The aim of this study was to understand the distribution pattern of Pb in coastal sediments and identify the factors that control Pb distribution and speciation in coastal sediments around India by using published data from the literature (see **tables 1 and 2** in the supplementary material). The diverse geology of the adjacent mass and source rocks around the coastal India are described below.

1. Geological setting of India:

The diverse geological setting of India has been described (Briggs, 2003; Krishnan, 1968 ; Krishnan, 1960) in three physiographic units as shown in Figure 1. The main physiographic units are: (i) the peninsula, (ii) the Indo-Gangetic alluvial plain and (iii) the extra peninsula. (i) The Indian peninsula primarily composed of Deccan Traps of cretaceous to Eocene period and metamorphic rocks of Archean and Pre-Cambrian age. The north western part (one third) of Indian peninsula is covered by Deccan Trap basalt, it is believed that the Deccan Trap was formed as result of sub-aerial volcanic activity associated with the continental deviation in this part of the Earth during the Mesozoic Era (Todal & Edholm, 1998). The rocks found in this region are generally igneous type (Lightfoot et al., 1987). The type of rock is generally fine grained, non-porphyrific doleritic or basaltic (tholeitic type) in composition. A major part of Maharashtra, a part of coastal Gujarat, Karnataka, Madhya Pradesh and Andhra Pradesh (partially) are made up by this type of rocks. The rest of the peninsular shield (Bihar, a marginal part of west Bengal, Odisha, Tamilnadu, Karnataka, Kerala and Goa also fall under the same physiographic unit) is majorly composed of Precambrian rocks, principally the Dharwar, Peninsular granitic gneiss, charnockites and the closepet granite (Krishnan, 1968). The Dharwar metamorphic are comprised of phyllites, slates, schists with chlorite, biotite, garnet, staurolite, kyanite, sillimanite and hornblende (Srinivasan and Sreenivas, 1977; Krishnan, 1968; Naqvi et al., 1978; Naqvi et al., 1974). (ii) The Indo-Gangetic alluvial plains is the second physiographic unit of India belongs to the Quaternary era. The alluvial deposits were formed by the erosion of the Himalayan rocks by rivers and monsoon (Gibling et al., 2005). Part of Rajasthan, Punjab, Haryana, Uttar Pradesh, Bihar, Assam and West Bengal come under this second physiographic part.

(iii) The third physiographic unit of India is the extra peninsula -the Himalayas and their eastern extensions including Andaman and Nicobar Islands. Most of the northern and north eastern states including Jammu and Kashmir, Himachal Pradesh, Uttarakhand, Sikkim, Arunachal Pradesh, Manipur and Mizoram belong to this extra peninsula region.-These parts of India are primarily comprised of igneous plutonic rocks and sedimentary rocks (Debon et al., 1986). Specific igneous plutonic rock types

in these zones include granite, diorite, gabbro, tonalite, monazite, alunite and pegmatite. The sedimentary rocks found in this region include marl, dolomite, greywacke, siltstone, shale and limestone (Yin and Harrison, 2000).

3. Source rocks to coastal sediment:

As the river dominated environment prevails, the sediment contributed by rivers becomes most predominant in the coastal areas around India (Bay of Bengal and Arabian Sea). These diverse geological setting alters the concentrations of geogenic Pb in coastal sediments around India. The concentrations of Pb in different source rocks are presented in **Table 1**. It shows that the concentrations of Pb is not constant in different lithological units but varies from one to another.

The composition and relative abundance of clay minerals in coastal sediments are controlled by the source rocks and weathering conditions. Mineralogical studies on the coastal sediments from the east coast of India have a geographical zoning related to the adjacent crustal rocks, soils and to the weathering processes that have occurred there (Goldberg and Griffin, 1970; Kessarkar et al., 2003; Nair et al., 1982; Rao and Rao, 1995).

The sediments deposited by the Himalayan rivers (Ganges, Brahmaputra, Gandak and Yamuna) have large amounts of illite consisting of equal quantities of kaolinite and chlorite with little or no mixed-layer clays and montmorillonite. These minerals are derived from the weathering of Himalayan rocks and are found in the sediments from the northeastern coastal region of West Bengal and parts of Odisha. The average concentration of Pb in the Himalayan rock is $\sim 15.0 \text{ mg.kg}^{-1}$ with a range from 3.5-32 mg.kg^{-1} (Table 1). Himalayan rock is made up of granite, shale, greywacke, limestone, basalt and gabbros etc.

Sediments from the central and southern part of the eastern continental margin are mainly derived by the Mahanadi, Godavari, Krishna and Cauvery Rivers. These coastal sediments are characterized by the presence of high concentrations of montmorillonite or smectite, trace quantities of mixed-layer clays, small quantities of illite and absence of chlorite. The Deccan Trap basalts have been reported to be one of the major sources of detrital solids for the shelf sediments in this region. The concentration of Pb in basaltic rock from the Deccan Trap has been found to be $\sim 3.5 \text{ mg.kg}^{-1}$ (Table 1).

Mineralogical surveys of sediments from the west coast show geographical zoning related to the adjacent rocks and soils (Gourbunova, 1966; Paropkari et al., 1992). Three distinct clay mineral

assemblages have been reported: (i) Illite-chlorite-rich sediment - from Himalayan rock derived by Indus River, (ii) smectite-rich sediment- from Deccan Traps derived by Narmada Tapti and (iii) kaolinite-smectite-rich sediment- from the Gneissic province (Rao and Rao, 1995). The illite and chlorite-rich assemblage derived by the Indus River (Indus Province) is the predominant feature found in continental margin sediments to the north of the Gulf of Kachchh. The average concentration of Pb in source rocks of this region is $\sim 15 \text{ mg.kg}^{-1}$ typical of Himalayan rock (Table 1).

An assemblage of smectite with minor kaolinite, illite and chlorite, mostly derived from the Deccan Trap basalts (Deccan Trap Province) occurs all along the inner shelf of northern to central part of the west coast of India (from Saurashtra to Goa). Illite dominates smectite in the outer shelf of the Saurashtra (coastal areas of Gujarat) and on the continental slope from the Saurashtra to Goa. The concentration of Pb in the source rocks of this region is ranging from 3.5-15 mg.kg^{-1} .

Smectite and kaolinite-rich assemblage, illite, chlorite and gibbsite derived from the Gneissic Province occur both on the central to southern part of west coast of India. A geochemical study by Kurian et al. (2013) along the western continental shelf has shown distinct north-south differences consistent with the source signatures. The Deccan Trap basalt characterizes the sediments in the north (from coastal Gujarat to Goa) and gneisses and granulites in the south (from Goa to southern most part of Kerala). The average concentration of Pb in the source rock of the sediments in the southern part of west coast ranges from 9.8 to 16 mg/kg .

It is difficult to relate the source rock composition with the abundance of total Pb found in the coastal sediments because several other factors (such as, local pollution sources, the intensity of coastal currents; waves ; climatic factors that are specific for pre-monsoon, monsoon, post-monsoon conditions etc.) come into play. In this investigation we related the influence of diverse geology on Pb distribution in the coastal sediments around India. An effort was made to identify the factors that control the distribution of Pb in coastal sediments around India. Enrichment Factor (EF), was used from the available data in the literature to predict the extent of Pb accumulation and pollution in the coastal sediments. With this additional information the local pollution sources can be placed in context with the overall levels predicted from geochemistry.

4. Enrichment Factor (EF)

Measuring enrichment factor (EF) is an essential part of geochemical studies and is generally used to differentiate between the metals originating from anthropogenic (non-crustal) and geogenic (crustal)

sources, and to assess the degree of metal contamination (Adamo et al., 2005; Chakraborty et al., 2014d; Darvish et al., 2012) in sediment.

Enrichment factor of heavy metals was calculated as the ratio of elemental concentration of sediment normalized to immobile Fe or Al: (Landsberger et al., 1982; Loring, 1991).

The EF was calculated by using the following equation:

$$EF = \frac{C_a \times N_b}{C_b \times N_a}$$

Where, C_a and C_b are the concentrations of examined metal in sediment and the background reference sample respectively. N_a and N_b are the concentrations of the normalizing elements in the sediment and the background reference respectively.

Concentrations of Fe or Al were used as normalizing element to calculate EF of Pb in the coastal and estuarine sediments. However, it is well known that the use of Fe as a normalizing element is restricted in anoxic sediment due to the remobilization and dissolution of Fe in anoxic sediment which in turn changes Pb/Fe ratio in the sediment (Schiff and Weissberg, 1999). Since in this study anoxic surface sediments are not expected to be found in coastal and estuarine areas of India, we have used Fe along with Al (as per the availability of the data) to normalize Pb concentration in sediments. The world average concentrations of Pb (20 mg.kg⁻¹); Al(81 gm.kg⁻¹) and Fe(47 gm.kg⁻¹) reported for shale (Turekian and Wedepohl, 1961) were used as the background values in this analysis.

Based on the Enrichment Factor (EF), Pb contamination was classified into the following groups: : (a) EF ≤1: no enrichment; (b) 1 < EF ≤3: minor enrichment; (c) 3 < EF ≤5: moderate enrichment; (d) 5 < EF ≤10: moderately severe enrichment; (e) 10 < EF ≤25: severe enrichment; (f) 25 < EF ≤50: very severe enrichment; (g) EF > 50: extremely severe enrichment (Ho et al., 2010). Correlation matrix and regression analysis were also performed to identify the factors that control Pb distribution in coastal and estuarine sediments.

5.Coastal areas of India:

In this paper, the vast east and west coastlines of India were further subdivided into the following six coastal regions to provide a better classification to relate to the understanding of Pb-geochemistry. **(Figure 1).**

East coast: East coast was divided into three parts:

- North-east coast - This part includes the coastal parts of West Bengal and Odisha. There are two major estuaries in this region (the Ganges and Mahanadi estuaries)
- Central-east coast -This part includes the coastal region of Andhra Pradesh. The Godavari and Krishna estuaries are in this region ()
- South-east coast - This part includes the coastal region of Tamil Nadu where the Cauvery estuary is located.

West coast: The west coast was divided into three parts:

- North-west coast - This includes the coastal region of Gujarat. The two major estuaries in this region are the Narmada and Tapti estuaries.
- Central-west coast - This includes the coastal region of Maharashtra, Goa and northern part of Karnataka including three major estuaries the Ulhas, the Mandovi and the Zuari estuaries.
- South-west coast - This coastal region of Kerala includes one major estuary called the Cochin estuary.

North-east coast

Coastal parts of West Bengal and Odisha belongs to this region. Considerable data for Pb concentrations in sediments are available in the literature from the estuarine regions of Ganges/Hooghly and Mahanadi estuaries (Subramanian, 1993; Subramanian et al., 1988; Sarkar et al., 2004; Chatterjee et al., 2007; Banerjee et al., 2012; Mukherjee and Kumar, 2012; Saha et al., 2001; Ramesh et al., 1999; Sundaray et al., 2012; Raj et al., 2013). However, data from the coastal areas of this region are scarce. The concentrations of Pb in the surface sediment from this region were found to vary from 6.8-214.4 mg.kg⁻¹ with a median value of 24.3 mg.kg⁻¹ (**Table. 2**). The primary sources of sediment deposit in the Hoogly estuary and nearby coastal areas are principally due to the erosion of the Himalayan rocks by the Ganges–Brahmaputra river system. The concentration of Pb in the Himalayan rocks ranges from 5-22 mg. kg⁻¹. Relatively lower concentration of Pb has been reported in the sediments from the Ganges/Hoogly estuarine region compared to the sediments from the Mahanadi estuarine region. The concentrations of Pb in sediments from the Mahanadi estuarine region have been reported (Sundaray et al., 2012) to vary from ~105-214 mg.kg⁻¹. Erosion of Precambrian rock of Indian peninsula is the

primary source of sediments deposited in the Mahanadi estuary and its nearby coastal areas. Sediments of this region are composed of the minerals eroded from the major rock types such as khondalites, charnockites, leptynites, granites, gneisses, etc. The average concentration of Pb in these source rocks ranges from 5-32 mg. kg⁻¹. However, much higher Pb content in the sediments from the Mahanadi estuary compared to the Ganges/Hooghly estuary is due to the presence of the richest mineral belt of the subcontinent (consisting of Fe ore, coal, lime-stone, dolomite, bauxite, Pb and Cu deposits) which fall within the Mahanadi basin (Chakrapani and Subramanian, 1993). It has also been documented that there are 15 large industries in the Mahanadi basin (Jena, 2008) and industrial effluent from these industries is released into the Mahanadi River (Jena, 2008). Several medium and small industries on the Mahanadi basin discharge 1,00,000 m³ of waste water every day; 12 coal mines discharge 14,000 m³ mine water daily during non-monsoon months(Jena, 2008).

The Pearson correlation and statistical regression analysis (based on the available literature data from this region) show a significant positive correlation between Pb and Fe/Mn in the sediments from this region (**Table 3**) and a weak correlation between Pb and organic carbon (OC) (**Table 3**). This is in agreement with the findings of Subramanian et al, (1988) and Banerjee et al., (2012) that OC does not control metal distribution in sediment of this region. It has been reported that high microbial activity is responsible for low OC content in sediment (Canuel and Martens, 1993; Chatterjee et al., 2007) from this region. Poor correlation between Pb and OC content in sediment may also reflect the low availability of OC content in the sediment. It is well known that Fe/Mn-oxyhydroxide phase is a good scavengers of Pb. Significant positive correlation and strong regression between total Pb content and Fe/Mn in sediment suggest that Fe–Mn oxyhydroxide might be one of the major controlling factors for Pb distribution in the sediment of this region (**Figure 2(a,b)**). A poor correlation between Pb and Al (**Table 3**) indicates that anthropogenic sources were the major sources for Pb in the sediments. Moderately high EF values of Pb in sediment also indicate that anthropogenic activities were responsible for Pb enrichment in these sediments (**Figure 3**).

Central-east coast:

Coastal part of Andhra Pradesh belongs to this region. Geochemical information of Pb in coastal sediment from this region is limited. The median concentration of Pb in estuarine and coastal surface sediments was found to be 12.7 mg.kg⁻¹(**Table 2**) (Satyanarayana et al., 1994; Krupadam et al., 2007, 2003; Ray et al., 2006 ;Chakraborty et al., 2012; Ramesh et al., 1999). The concentrations of Pb in

estuarine and coastal sediment were in the range of ~10 - 14 mg.kg⁻¹ except few elevated concentrations of Pb reported in literature (Satyanarayana et al. 1994; Ray et al, 2005; Chakraborty et al, 2012b) in the estuarine sediment. High concentration of Pb was found in the coastal sediment collected adjacent to harbor region (Satyanarayana et al 1994). Pb from natural sources in the coastal and estuarine sediment of this region has been reported to originate from the adjacent land mass including some major geological formations such as the Tertiary Deccan Traps, Archean granites, Precambrian and Gondwana sedimentary rocks and recent alluvials (Krishnan, 1968). However, the mineralogical studies indicate that majority of the sediments are eroded from basaltic rock (depleted in Pb) of Deccan Trap. The geochemical fractionation (Krupadam et al, 2007; Chakraborty et al., 2012) studies of Pb also indicate the same. The geogenic contribution of Pb in the total Pb of sediment in this region was found to be little.

High concentrations of Pb in the sediments from this region has been reported (Satyanarayana et al. 1994 and Chakraborty et al.,2012b) in the literature and indicate that anthropogenic input of Pb in the sediments were higher than geogenic sources. The EF of Pb also indicates that there was anthropogenic input of Pb in the coastal sediment. **(Figure 3)**. It was not possible to determine the predominant geochemical carrier of Pb in these sediments due to the lack of data. However, OC has been reported to play a key role in controlling Pb distribution and speciation in estuarine sediments from this region (Krupadam et al., 2007; Chakraborty et al., 2012).

South-east coast:

Coastal part of Tamil Nadu belongs to this region. The median concentration of Pb in the coastal sediment from this region was calculated to be ~16 mg. kg⁻¹ (**Table 2**) and ranged from 0.1 - 130 mg.kg⁻¹ (Raju et al., 2011; Ravichandran and Manickam, 2012; Raj and Jayaprakash, 2007; Achyuthan et al., 2002; Selvaraj, 1999; Satpathy et al., 2010; Dhanakumar et al., 2013; Ramesh et al., 1999; Kumar and Edward, 2009; Magesh et al., 2013, 2011; Venkatramanan et al., 2014; Subramanian and Mohanachandran, 1990; Anithamary et al., 2012; Jonathan et al., 2004; Sundararajan and Srinivasalu, 2010; Palanichamy and Rajendran, 2000). Charnockites acidic rock to intermediate composition associated with granites (Chattopadhyay and Sarkar, 1999) are the major natural sources of Pb in the coastal and estuarine sediments of this region. Achyuthan et al. (2002) have reported that coastal sediment of Tamil Nadu is depleted in Pb. The bulk concentrations of Pb in some of the places of south east coast of India were found to be much lower than the average values found in earth crust (13 mg. kg⁻¹

¹) or shale (20 mg. kg⁻¹) reported by Turekin and Wedepohl (1961). On the other hand, in few places, some of the estuarine as well as the coastal sediments from this region have been reported to be moderately enriched with Pb. The EF values were found to vary from 0.18 to 8.43. However the calculated EF for the compiled data as shown in **Figure 3** reflects that most of the places of this region showed minor enrichment of Pb with EF values from 1 to 3. The Pearson correlation and regression analysis on the available literature data indicate a strong positive correlation between Pb and Al (**Figure 4, Table 4**). This finding suggests that most of the Pb in coastal sediment from this region was probably from the crustal sources.

North-west coast:

Coastal parts of Gujarat and northern Maharashtra belong to this region. The available literature data indicates that the concentrations of Pb in estuarine and coastal sediments from this region varied from 8.9 - 220 mg. kg⁻¹ (Rokade, 2009; Reddy et al., 2004 ;Tewari et al., 2001;Sahu and Bhosale, 1991; Fernandes et al., 2012, 2011; Fernandes and Nayak, 2014; Patel et al., 1985; Dilli, 1986; Zingde et al.,1988). The median concentration of Pb in coastal sediment from this region was calculated to be ~58.3 mg. kg⁻¹ (Table 2) and was found to be higher than the rest of the coastal areas of India. The concentration of Pb in estuarine sediments was found to be higher than that of coastal sediments.

The natural sources of Pb in coastal sediment from this region are from the Himalayan rocks eroded by Indus River and the Deccan basalt drained by Narmada and Tapti River. However, the source rocks for most of the study areas (reported in literature) were mainly influenced by the basaltic rocks from the Deccan Trap terrene. The Pb content in basaltic rock is 3.5mg. kg⁻¹(Table 1)). Thus, geogenic contribution of Pb was very low to the coastal sediment of this region. The high value of EF in coastal sediment from this region indicates that heavy industrialization of Gujarat and Maharashtra was probably responsible for this enriched concentration of Pb in the coastal sediment of this region (**Figure 5**). Pearson correlation and regression analysis showed significant positive correlations between Pb, and Fe/Mn (**Figure 6, Table 5**). Concentrations of OC in coastal sediments from this region has been reported to be relatively low compared to the rest of the west coast of India (Paropkari et al., 1993, 1992) and thus, Pb was expected to be associated with Fe /Mn-oxyhydroxide in these sediments.

Central-west coast:

Coastal regions of southern Maharashtra, Goa and Mangalore belong to this area. The median concentration of Pb was calculated to be ~10.6 mg.kg⁻¹ (Table 2) and it ranged from 4.5 - 46.5 mg. kg⁻¹

(Alagarsamy, 2006; Karbassi and Shankar, 2005; Rokade, 2009; Pandarinath and Narayana, 1992; Krishnakumar et al., 1998). The coastal and estuarine sediments of this region are mainly the weathering product of Precambrian granites, gneisses, schists and charnockites (Krishnan, 1969; Rao and Rao, 1995). Concentrations of Pb in estuarine and coastal sediments indicate that there was more accumulation of Pb in estuarine sediments than marine sediments. The strong positive relation with Al indicates the abundance of Pb in sediment is principally from crustal sources (**Figure 7, Table 6**). The low average values of EF support the fact that there was no (or little) anthropogenic input of Pb in this region (**Figure 5**)

South-west coast:

Coastal part of Kerala belongs to this region. The median concentration of Pb in estuarine and coastal sediment from this region was 33.3 mg.kg^{-1} (Table 8). The total concentration of Pb in the sediments were found to vary from $0.15 - 176.87 \text{ mg.kg}^{-1}$ (Balachandran et al., 2006; Sengupta et al., 2012; Kumar and Edward, 2009; Sudhanandh et al., 2011; Martin et al., 2012; Nair and Ramachandran, 2002; Dipu and Kumar, 2013; Selvam et al., 2012). The highest concentration of Pb in sediment has been reported in Manakudy estuary (Kumar and Edward, 2009) and the lowest concentration of Pb has been found in the coastal sediments from this region (Nair and Sujatha, 2012). The major source rocks of this region are mainly composed of charnockites, khondalities, granite gneisses and hornblende gneisses (enriched in Pb). However, very high enrichment of Pb (EF~9) in some estuarine sediment has been attributed to the point sources (anthropogenic influence). Sediments belong to this area was mostly rich in organic carbon (Balachandran et al., 2006; Kumar and Edward, 2009; Sudhanandh et al., 2011; ; Martin et al., 2012; Nair and Ramachandran, 2002). It has been reported that suboxic environment of the overlying water column specially in the continental shelf of this region preserves carbon content in the bottom sediment (Martin et al., 2012; Balachandran et al., 2006). Therefore, Pb showed strong affinity towards OC within the sediments (Figure 8(a), Table.7). However, Pb showed similar affinity for Fe-Mn oxyhydroxide phases in coastal sediments from this region (Figure 8(b), and Table.7). It has been reported that Fe-Mn oxy-hydroxide phase is relatively low in marine sediment (sub-oxic condition) in this region compared to the estuarine sediment (oxic condition). Therefore, association of Pb was found to be higher with organic carbon in the coastal sediment from this region.

5. Quality of coastal sediments of India in terms of Pb :

To assess the quality of coastal and estuarine sediments of India in terms of Pb pollution, the concentrations of Pb in sediments from the six different coastal regions of India (discussed above) were compared with Pb concentrations reported in unpolluted estuarine/coastal sediments from different parts of the world (Table 8). The concentrations of sedimentary Pb around India were comparable with the unpolluted coastal and estuarine sediments from different parts of the world (Table 8). However, there were few sporadic locations (viz., some areas from northern and central parts of east coast and northern parts of west coast of India) where high concentrations of Pb has been reported. The effect range low (ERL) and effect range median (ERM) values proposed by Long et al. (1995) were also used to assess the quality of the coastal sediments around India (Table 8). It was found that the concentrations of Pb in most of the coastal and estuarine sediments were lower than the ERL values (Table 8). Furthermore, the enrichment factor (EF) values also showed that there was “no to minor enrichment” of Pb in coastal areas around India. This study suggests that: (1) coastal sediments were less enriched with Pb than estuarine sediments; (2) sediment from the southeast coast and central west coast of India were not enriched by Pb and there were not much anthropogenic Pb input. The geogenic Pb was probably the major source of Pb in sediments from this region. (3) Anthropogenic input of Pb was high in sediment from the north-east (mainly Mahanadi River basin), north-west, and south-west coast of India. (4) Fe/Mn oxyhydroxide mainly controlled Pb distribution and speciation in coastal sediments around India. (5) OC played dominant role in binding and controlling Pb distribution and speciation in coastal sediment. However, Fe/Mnoxyhydroxide phase in sediment played key role in controlling Pb distribution in presence of little or no OC in coastal sediment.

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Captions and Legends for Figures:

Figure 1. Geological map of India with the studied areas

Figure 2. Variation of total Pb concentrations with varying concentrations of (a) Fe and (b) Mn in the sediment from of the north-east Coast.

Figure 3. Distribution of Enrichment Factor (EF) along the east coast.

Figure 4. Variation of total Pb concentration with Al concentration in sediments from the south-east coast.

Figure 5: Distribution of Enrichment Factor (EF) along the west coast

Figure 6: Variation of total Pb concentration with Mn concentration in the sediments from the north-west coast.

Figure 7. Variation of total Pb concentrations with Al concentrations in sediments of the central-west coast.

Figure 8. Variation of total Pb concentrations with the varying (a) OC content and (b) Fe concentrations in the sediments of south-west coast .

Table.1 Concentration of Pb in different lithologies (Wedepohl, 1978)

Lithology	Concentration of Pb (mg.kg ⁻¹)
Continental crust	15
Oceanic crust	0.9
Basalts,gabbros	3.5
Granites	32
Limestones	5.0
Granulites	9.8
Greywackes	14
Gneisses, mica schists	16
Shale	22

Table 2. The average concentration of Pb and contributing source rock of the different coastal parts of India

Coastal areas of India	Contributing source rock	Median value of Pb (mg.kg^{-1}) in the sediment
Northeast coast	Igneous plutonic (granite, diorite, gabbro, tonalite, monazite and pegmatite), sedimentary rocks (marl, dolomite, greywacke, siltstone, shale and limestone), (Yin and Harrison; 2000) khondalites, charnockites, leptynites granites, gneisses. (Chakrapani and Subramanian; 1993)	24.3 (n=105)
Centraleast coast	Basalt, Granite and sedimentary rock. (Krishnan, 1966)	12.7(n=62)
Southeast coast	Charnockites and intermediate granites. (Chattopadhyay and Sarkar, 1999)	16.0 (n=335)
Northwest coast	Basalt (majorly) and Himalayan rocks(Igneuos plutonic and sedimentary rock)	58.3 (n=186)
Central west coast	Granites, gneisses, schists and charnockite. (Krishnan, 1968; Rao and Rao, 1995)	10.6 (n=41)
Southwest coast	Charnockites, khondalities, granite gneisses and hornblende gneisses. (Nath et al., 2000)	33.3 (n=161)

Table 3. Correlation coefficient matrix of Pb, Fe, Mn, Al, OC, Sand, Silt and Clay in the coastal and estuarine sediments from the Northeast Coast of India.

	Pb	Fe	Mn	Al	OC	Sand	Silt	Clay
Pb	1							
Fe	0.82	1.00						
Mn	0.78	0.86	1.00					
Al	0.23	-0.46	-0.29	1.00				
OC	0.24	0.83	0.45	-0.05	1.00			
Sand	-0.01	-0.61	-0.52	0.20	0.41	1.00		
Silt	0.01	0.65	0.65	-0.29	-0.28	-0.86	1.00	
Clay	0.05	0.02	-0.16	0.25	-0.42	-0.54	0.06	1

Correlation is significant at the 0.01 level (2-tailed).

Table 4. Correlation coefficient matrix of Pb, Fe, Mn, Al, CaCO₃ and OC in coastal and estuarine sediments from the Southeast Coast of India

	Pb	Fe	Mn	Al	OC	CaCO ₃	Sand	Silt	Clay
Pb	1								
Fe	-0.83	1.00							
Mn	0.82	-0.54	1.00						
Al	0.73	-0.41	0.81	1.00					
OC	0.24	-0.49	0.07	0.70	1.00				
CaCO ₃	-0.66	0.51	-0.75	-0.78	-0.45	1.00			
Sand	-0.79	0.48	-0.66	-0.81	-0.51	0.65	1.00		
Silt	0.66	-0.36	0.58	0.72	0.40	-0.56	-0.94	1.00	
Clay	0.82	-0.61	0.68	0.81	0.63	-0.67	-0.85	0.64	1.00

Correlation is significant at the 0.01 level (2-tailed).

Table 5. Correlation coefficient matrix of Pb, Fe, Mn, and OC in coastal and estuarine sediments from the North west Coast of India

	Pb	Fe	Mn	OC
Pb	1			
Fe	0.43	1.00		
Mn	0.50	0.62	1.00	
OC	-0.32	0.29	-0.14	1.00

* Correlation is significant at the 0.05 level (2-tailed). Table 6. Correlation coefficient matrix of Pb, Fe, Mn, Al and OC in coastal and estuarine sediments from the Central west Coast of India

	Pb	Fe	Mn	Al	OC
Pb	1.00				
Fe	0.35	1.00			
Mn	0.63*	0.78	1.00		
Al	0.72**	0.82	0.89	1.00	
OC	0.38	0.70	0.52	0.82	1.00

* Correlation is significant at the 0.02 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

Table 7. Correlation coefficient matrix of Pb, Fe, Mn, Al, OC, CaCO₃, Sand, Silt and Clay and in coastal and estuarine sediments from the Southwest Coast of India

	Pb	Fe	Mn	Al	OC	CaCO ₃	Sand	Silt	Clay
Pb	1								
Fe	0.78	1.00							
Mn	0.01	0.32	1.00						
Al	0.72	0.61	0.04	1.00					
OC	0.88	0.82	0.01	0.68	1.00				
CaCO ₃	0.12	-0.02	-0.22	0.07	0.11	1.00			
Sand	-0.95	-0.76	0.04	-0.68	-0.91	-0.18	1.00		
Silt	0.88	0.67	-0.02	0.60	0.80	0.27	-0.93	1.00	
Clay	0.81	0.72	-0.07	0.59	0.84	-0.04	-0.83	0.60	1

Correlation is significant at the 0.01 level (2-tailed)

Table.8 Comparison of the concentration of Pb in the coastal sediments of India with the other parts of world and ERL,ERM values of Pb

	Concentration of Pb (mg.kg^{-1}) in sediment	References
Pearl river estuary and surrounding coastal area, China	21.7-47.9	Ip et.al (2007)
Tees estuary, England	37-680	Jones & Turki (1997)
South west coast of Spain	20-197	(Morillo, Usero, & Gracia (2004)
Suez Gulf, Egypt	70.44-93.74	el-Nemr et.al (2006)
Mangrove sediments along the coastline of French Guiana.	6.56-14.76	Marchand et al (2006)
Swartkops River Estuary, Port Elizabeth South Africa.	9.6-90.7	Binning and Baird (2001)
Mangrove and Lagoonal Sediments from south east Gulf of California,Mexico	14-100	Soto-Jiménez and Páez-Osuna (2001)
Continental Shelf of the North-western Black Sea, Romania	0.5-50.3	Secrieru and Secrieru (2002)
	6.83-214.40 Av=35.81	
North east coast of India	Med=24.28	This study

	5.65-95	
central east coast of India	Av=26 Med=12.69	This study
	1.01-74.20	
South east coast of India	Av=22.70 Med=16	This study
	8.83-130	
North west coast of India	Av=57.50 Med=58.32	This study
	2-46.5	
central west coast of India	Av=14.91 Med=10.60	This study
	0.10-163.30	
South west coast of India	Av=45.03 Med=33.33	This study
Effect range low value(ERL)	46.7	US NOAA's sediment quality guidelines
Effect range median (ERM)	218	US NOAA's sediment quality guidelines

Av=average, Med=median

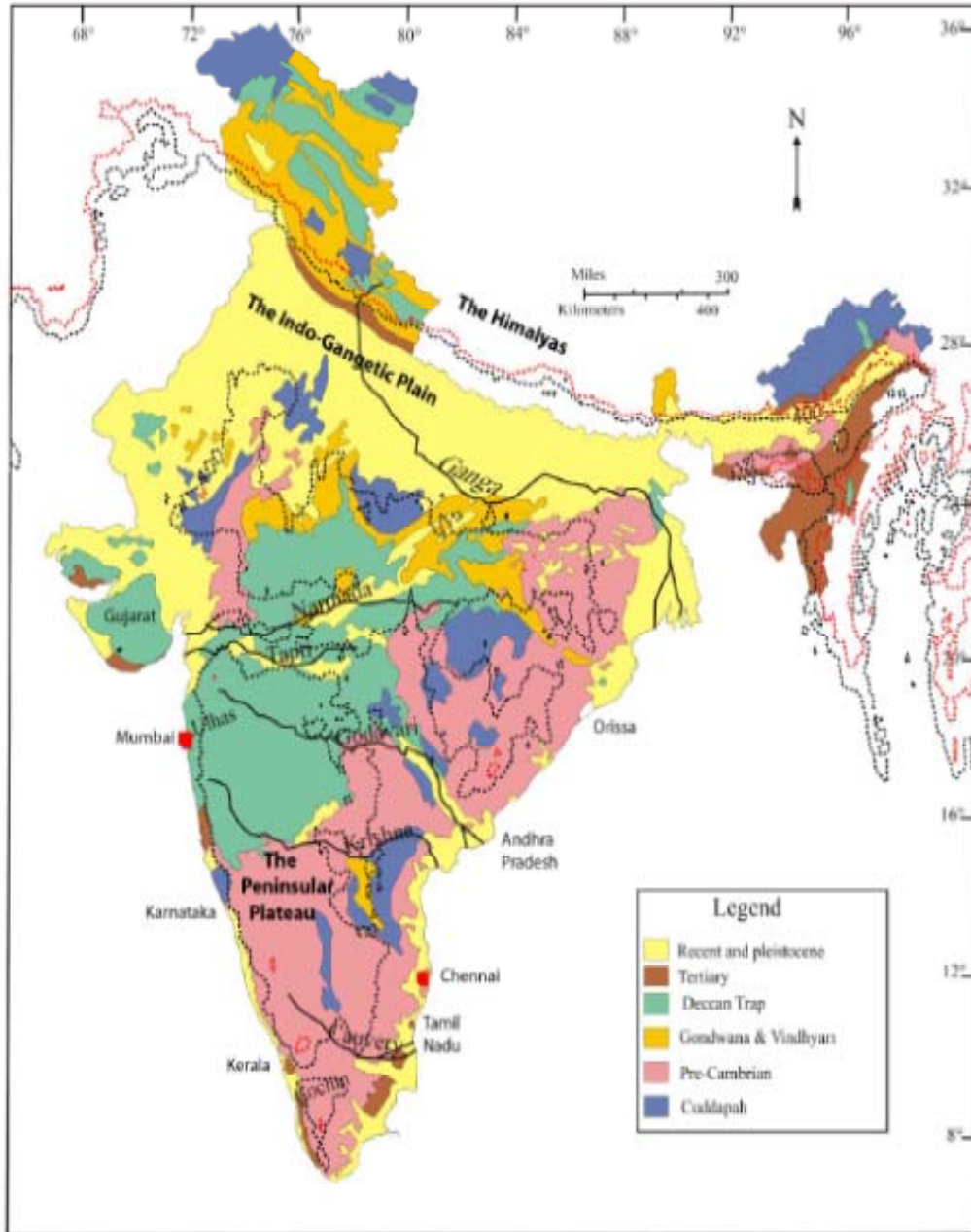


Figure 1

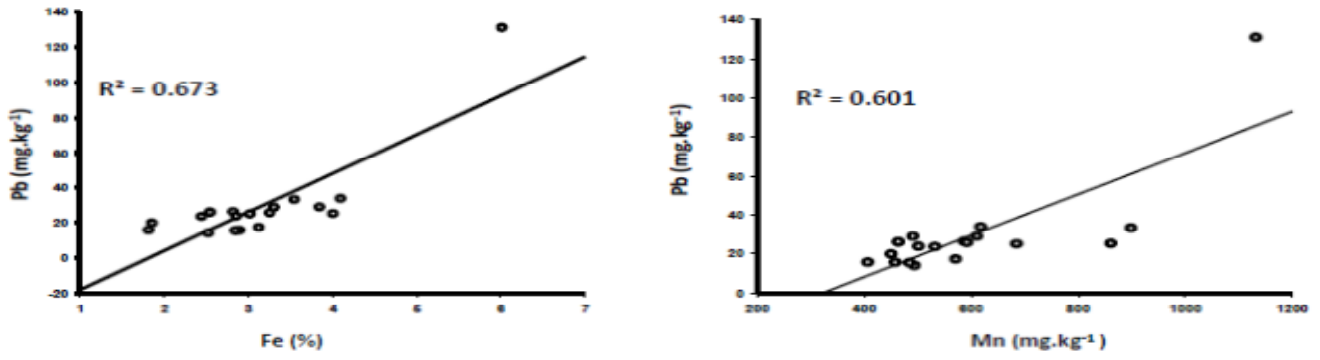


Figure 2

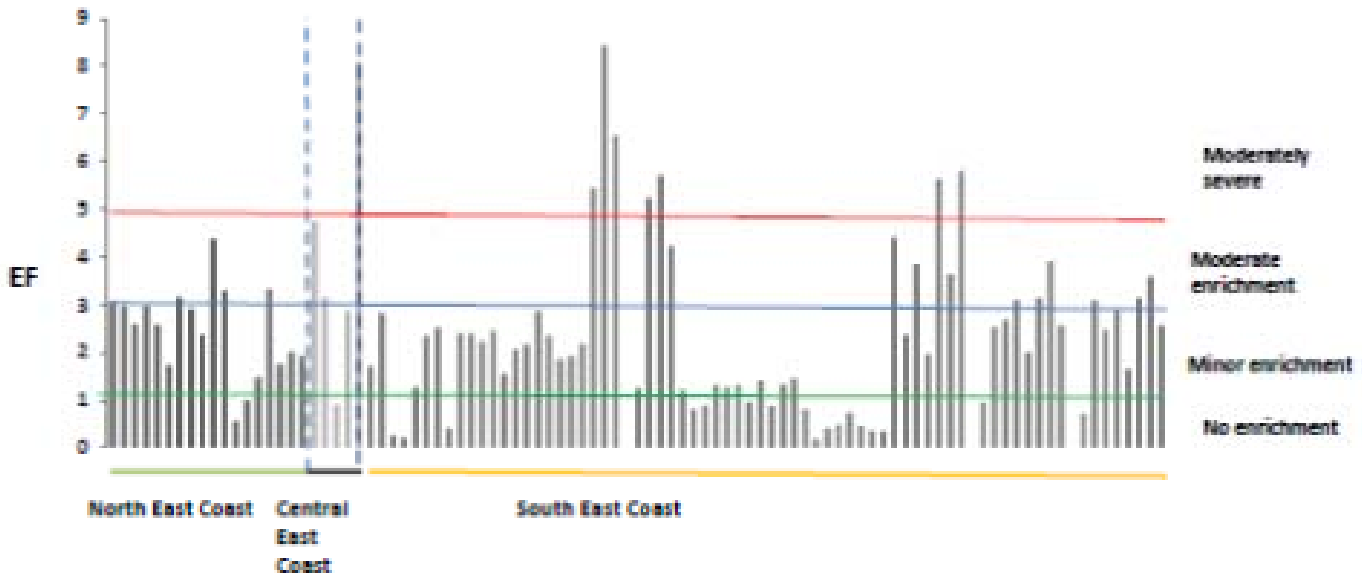


Figure 3

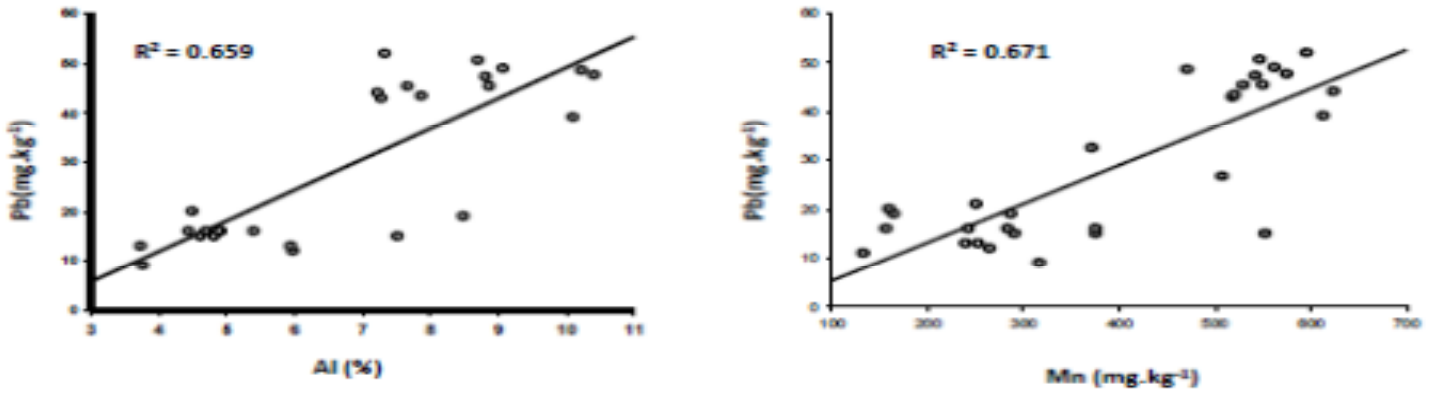


Figure 4

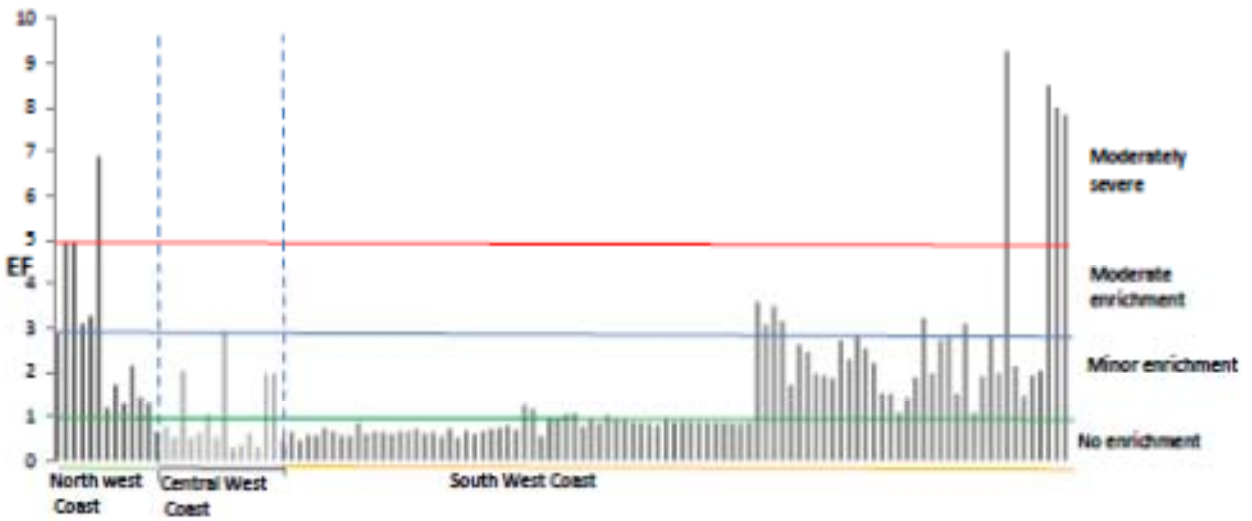


Figure 5

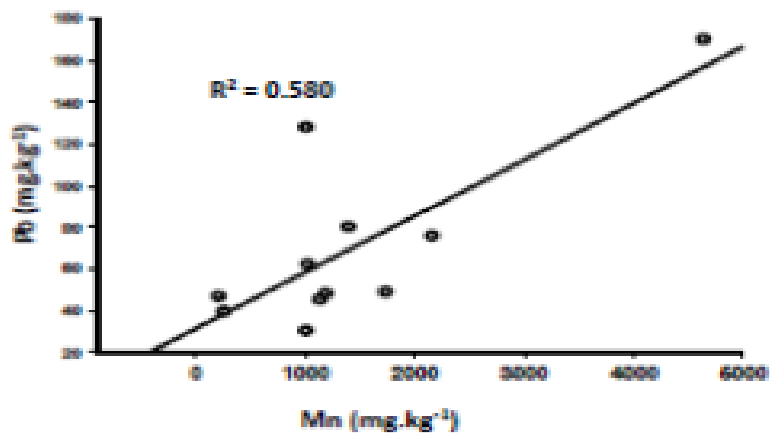


Figure 6

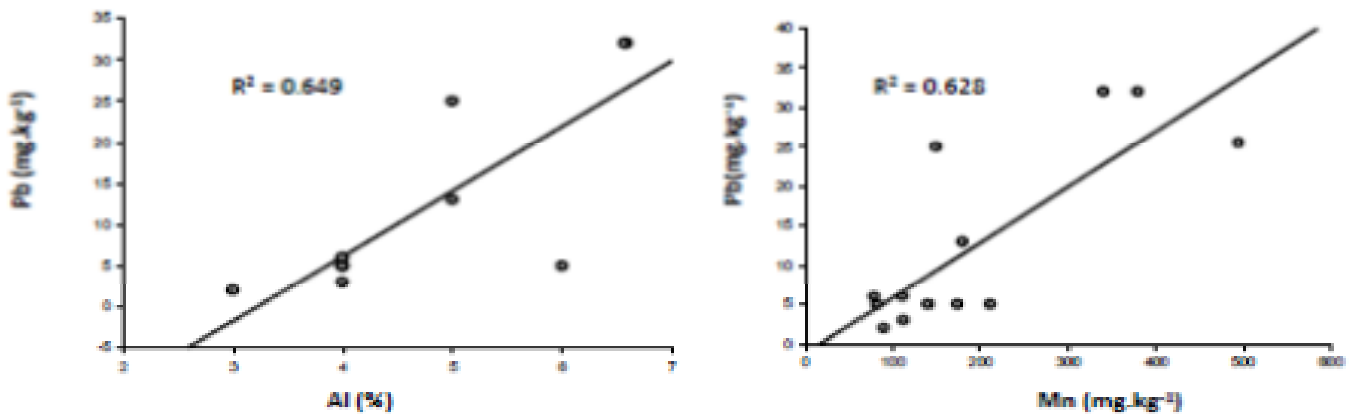


Figure 7

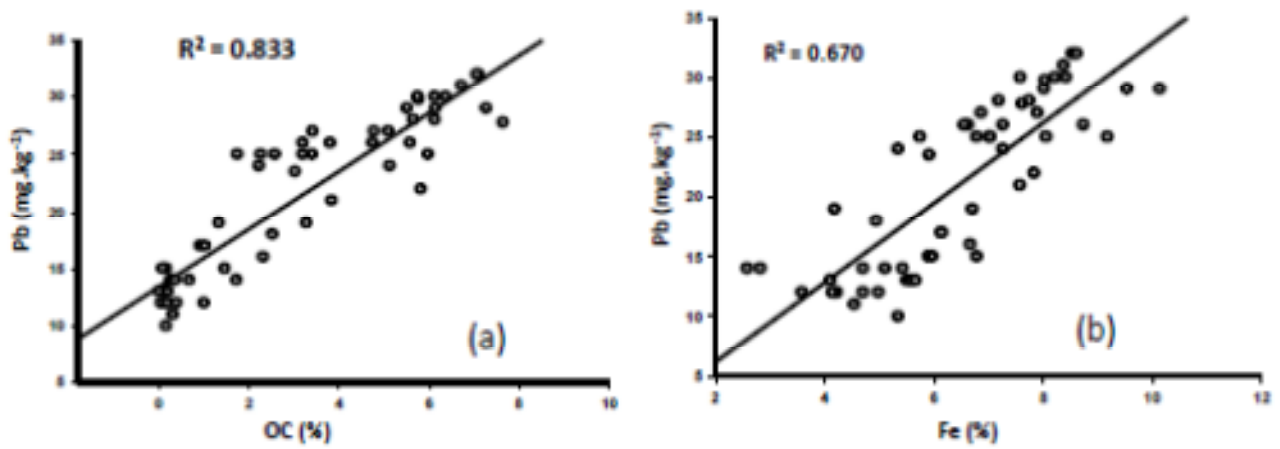


Figure 8