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Original Paper

PRELIMINARY STUDY ON EVOLUTION OF METALS CONTAMINANT RECORDED IN THE COASTAL SEDIMENTS OF SEMARANG WATERS

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ABSTRACT

The evolution of heavy metals contaminant of Cu, Zn and Hg at two main river mouths of Semarang waters have been carried out on June 2009 through sediment cores, ages determination and supporting with population and industry statistical data background. Heavy metals found at West Banjir Kanal river mouth may be associated with natural origin on the contrary the increase of metals concentration found at East Banjir Kanal river mouth is strongly correspond with population and industrial increases since industrial decades. Objective of research is to collect heavy metals contaminant concentration changed data over 30 years which have been recorded and saved in the layers of sediments in Semarang coastal waters. The goal of research is to reconstruct the evolution of contaminant concentration chronologically since industrialization decade. Method used in the research is data analysis through sediment cores supported both population and industrial statistical data.

Keyword : Heavy metals, sediment, river mouth, Semarang waters

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INTRODUCTION

The contaminants especially heavy metals are present in the marine sediments as a result of urban discharge and industrial waste water (anthropogenic) beside of natural sources (Buccolieri *et al.,* 2006, Balkis *et al.,* 2007) . In fact, during the last few decades, industrial and urban activities have contributed to the increase of heavy metals contamination into marine environment and have directly influenced the coastal ecosystems (Buccolieri *et al.,* 2006).

Many heavy metals concentrations in sediment are at least three orders of magnitude higher than the same metals in surrounding water (Karbassi *et al.,* 2005; Balkis *et al.,* 2007; Ahdy & Khaled, 2009. The analysis of heavy metal levels in sediment samples can also helps when the concentrations in water are undetectable with present methods of analysis (Davis *et al.,* 1991; Borretzen and Salbu, 2002 *in* Karbassi *et al.,* 2005; Soares *et al.,* 1999 *in* Balkis *et al*. 2007; Gagnon *et al*. 1997, Bloon *et al.,* 1999 and Mason & Lawrence 1999 *in* Ram *et al.,* 2009).

River mouth is an environment where the water masses from the land meet with the seawater masses. Suspended load that carried by river from various sources in the land, through the oceanographic processes in marine environment to form turbidity zone, partially and selectively will be deposited and the others are carried away to the open sea (Inman & Jenkins, 1999). Land sources are catchments area surface runoff, human intervention in natural resources, building and

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industrial areas, deforestation, spread of agriculture, etc. Suspended sediments are an important component of the river mouth environment harboring a nutrient supply, impacting light penetration through the water column and adsorbing potential contaminants (Cloern, 1987; Domagalski and Kuivila, 1993).

Uchiyama (2007) reported due to height fertility, the river mouths are ecologically significant, because a large fraction of biota depends on these areas for shelter and nourishment even play a key role in providing habitat for resident infauna, which act as a food resource for large communities of shorebirds. Most adsorbed pollutants on the sediments are not readily available for marine organisms. Due to environment changes of some physical and chemical characteristics (pH, salinity, redox potential and the content of organic chelators) of the overlying water may provoke the release of the metals back to the aqueous phase, hence under changing environmental conditions sediments may become themselves important pollution sources (Siaka 2008; Ahdy and Khaled, 2009)

Metals are natural constituents of the sediments in coastal zones and they may be oligo-elements, such as zinc for example, which is a natural element essential for living organisms when present in small amounts, or do not have any known biological role, as in the case for lead. However, both can become toxic for living organisms at high concentrations (Prego *et al.,* 2008). Much of those human activities located in the fluvial watersheds and in the margins of estuaries being important areas for the concentration of contaminants, due to coastal industrial activity and human settlement (Karbassi *et al.* 2005; Meybeck *et al.,* 2007 and Lestel *et al.,* 2007).

There is correlation between population growth and increasing of contaminants concentration. Dynamics of the contaminants concentration deposited in the sediment layers will correlate with population dynamics: population increase and industrial growth (Hirao *et al.,* 1986). Coastal sediments that include: leftovers of marine plants, marine animals, human activities with their domestic waste and contaminant products since the beginning of industrialization decade were deposited and recorded in layer by layer in the successive coastal sediments. Karbassi *et al.,* (2005) used to trace the source of heavy metals in Persian Gulf based on sediment cores. Reconstruction of metal contaminants concentration increase over 65 years chronologically based on sediment cores was also carried out by Meybeck *et al*., 2007.

Semarang is a capital city of Central Java and belonged to the 5th biggest coastal cities in Indonesia after Jakarta, Surabaya, Medan and Makassar. The city identified began to grow and develop since 1980's, new residence and industry areas have been opened. Portraits of coastal areas at big cities especially in developing countries are poorly of waters quality and waters management. After three decades of industrialization no more environment research data about Semarang coastal waters.

Objective of research is to collect heavy metals contaminant concentration changed data over 30 years which have been recorded and saved in the layers of sediments in Semarang coastal waters. The goal of research is to reconstruct the evolution of contaminant concentration chronologically since industrialization decade.

MATERIALS AND METHODS

There is correlation between population growth, human activities and increasing of contaminants concentration. The contaminant products together with sediment materials are carried by rivers and deposited layer by layer in the river mouth and spread out to the open sea. Relics of the dynamics and evolution processes on coastal environment in the past are recorded and saved in the succession layers of sediment, there are leftovers of marine plants, marine animals and human activities with their contaminant products. The oldest sediments are at the bottom

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successively to the youngest at the top.

Methods used in the research divided into two activities, are primary data collection and secondary data collection.

Primary data collection

Samples of sediment stratification layers were collected at West Banjir Kanal and East Banjir Kanal rivers by using of coring method on 23 – 30 June 2009. The aim of coring method is to collect sediment materials in the stratification column of the sea bottom, in normal condition the oldest in the lower then successively the youngest in the upper. Method used for collection and preparation in sediment cores adopt from Meybeck *at al.,* 2007.

Sediment corer is a poly vinyl chloride (pvc) pipe of 4 inch diameter and 2 m length was slowly pushed down into sediments surface in 1 – 1.5 m waters depth. The sediments recovered were segmented at 20 cm intervals at the lower part, 10 cm at the middle part and 5 cm at the upper part. Sediment segmentations are put in polystyrene containers and stocked frozen and divided for determinations of contaminants concentration, sediment properties analysis and age determination of sedimentation.

Stations which ideally used for coring position in the river mouth are continuously of sedimentary processes and undisturbed from dredging activities. Areas with such characterized above can be obtained on field observation and also be informed from the local fisherman.

To determine the absolute ages of sediments deposition were processed at National Atomic Energy Agency / Badan Tenaga Atom Nasional (BATAN) Jakarta based on the ^{210}Pb isotope with using γ-ray spectroscopy. To determine concentration of heavy metals contaminant (Cu, Zn and Hg), samples were stored and analyzed based on Atomic Absorption Spectroscopy at Geological Survey Institute of Indonesia, Bandung.

Secondary data collection

Population data at Semarang city and Semarang regency 1983-2008 and industrial data at Semarang city 1983-2007 and Semarang regency 1990-2007 were collected from Badan Pusat Statistik Kota Semarang (1983 – 2005) and Badan Pusat Statistik Kab. Semarang (1983 – 2005). Map of recent land cover mainly distribution of building areas, fishpond and other coverages of Semarang and its surroundings was constructed through landsat imagery taken on August 2004. Scheme of research methods and activities are illustrated in **Fig.1**

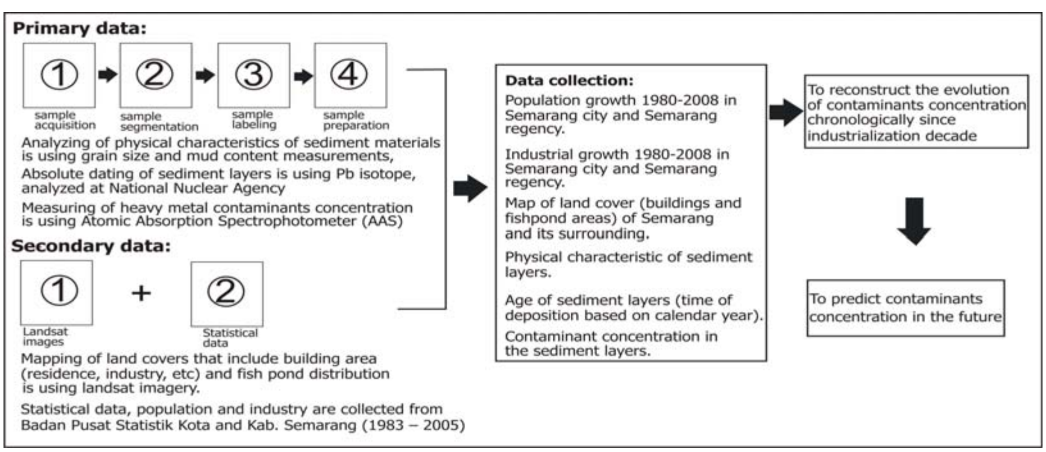

Fig 1. Scheme of research methods and activities.

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RESULTS AND DISCUSSION

Geographic setting

Semarang area discussed in the paper is classified in to two parts administratively, there are Semarang city located at the north side on the coastal plain area and small new spreading area located on the high land at the south area and Semarang regency which whole area is located on the high land at the south side of Semarang city. Geographically the widest part of Semarang city is a coastal plain area ranging from 1 m to 3 m height above sea level. When spring tide some areas of Semarang city are covered by sea water, which flood tide has reached up to 1 km distance from the coastline.

Due to the increasing of population, Semarang city administratively spread to the south direction and new residences are continuously opened. There are two main drainages on the Semarang city, West Banjir Kanal river is sourced from Ungaran volcano flows at the west part of Semarang city to the west Semarang waters and the other is East Banjir Kanal river flows on the east of Semarang city flows to the east Semarang waters (**Fig. 2**). Both of the two rivers above flows through the crowded population and industries area carry sediment materials, domestic sewages and industries pollutant to the Semarang waters. Those of sediment materials are a part will be deposited layer by layer at the river mouth and the other will away to the open sea. In normal condition, sedimentary materials in the river mouth, the oldest located at the lower part successively the youngest at the upper part.

Population and industrial growth

Population growth of Semarang city and Semarang regency collected from Badan Pusat Statistik Kota Semarang (1983 – 2005) and Badan Pusat Statistik Kab. Semarang (1983 – 2005) shows that total population of Semarang city in 1983 was 1072324 and Semarang regency was 727431. In 2000 population of Semarang city and Semarang regency increase continuously, it was 1309667 in Semarang city and 831282 in Semarang regency. Population growth of Semarang city and Semarang regency from 1983 – 2007 is shown in **Fig 4**.

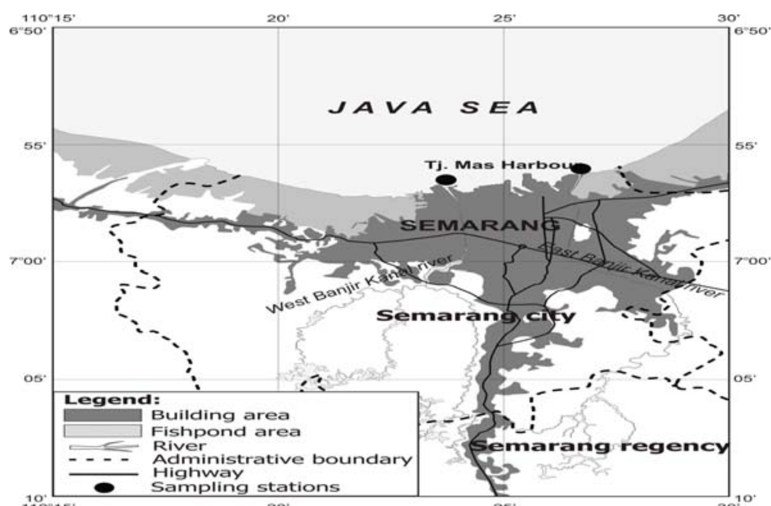

Fig 2. Map of land cover of Semarang city and Semarang regency.

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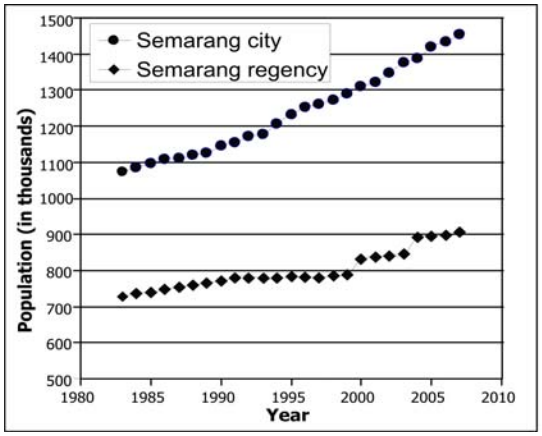

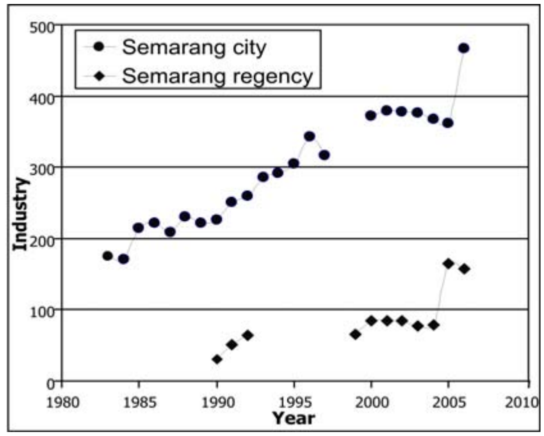

Fig 3. Diagram of population at Semarang city and Semarang regency (left) and industry at Semarang city and Semarang regency (right) 1980-2008.

Based on linear interpolation on the graph above shown that increase percentage of population at Semarang city and Semarang regency from 1983 up to 2007 are constant 1.8 % for Semarang city and 0.34% for Semarang regency. Data for industrial sector documented by Badan Pusat Statistik for Semarang city in 1985 was 200s and in 2005 was 450s while for Semarang regency on 1990 was 30s and on 2006 was 158s. Weakness of industrial statistical data documented by both Badan Pusat Statistik Kota and Kab. Semarang 1983 – 2005 were not classified among various industry which will affect to environment degradation. Graph above also shown average increase of industrial sector at Semarang city is 5% / year and at Semarang regency is 9% / year.

Sedimentary layers and age determination

Sediment samples collected through sediment corer at both West and East Banjir Kanal rivers were 210 cm . Unfortunately 140 cm in the lower part of sediment core at West Banjir Kanal and 175 cm in the lower part of sediment core at East Banjir Kanal could not be identified due absolute ages of their deposition were exceed 160 years Before Present (**Fig 4**). The oldest age determination for sediment deposition using ^{210}Pb isotope is 160 years.

Based on absolute age determination of two core samples and neglected of any processes such as coastal erosion and dredging activities since 160 Before Present, sedimentary processes at West Banjir Kanal river mouth is faster (0.44 cm / yr) than East Banjir Kanal river mouth (0.22 cm/yr).

Sediment cores collected both from West and East Banjir Kanal river mouth are consist of alternating repetition of medium – fine sand and mud. Based on sediment characteristic, sediment material found at West Banjir Kanal river mouth shows grayish in color with small shells. Similar sediments characteristic was also collected from East Banjir Kanal river mouth, especially in layers formed on Calendar Year 1905 (25 cm depth from surface sediment) shows black color, abundant shells and hydrogen sulfide (H_2S) smell. Previous study explained the presence of black color of hydrogen sulfide in the sediment layer due to rapidly of deposited sediments. Recent study (Alves *et al.,* 2007) explained that hydrogen volatile sulfide were identified a non steady process with instabilities during deposition or a depositional process in steady state characterized by a low average deposition rate. Preservation of acid volatile sulfide is

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favoured by low oxygen concentrations in the overlying water.

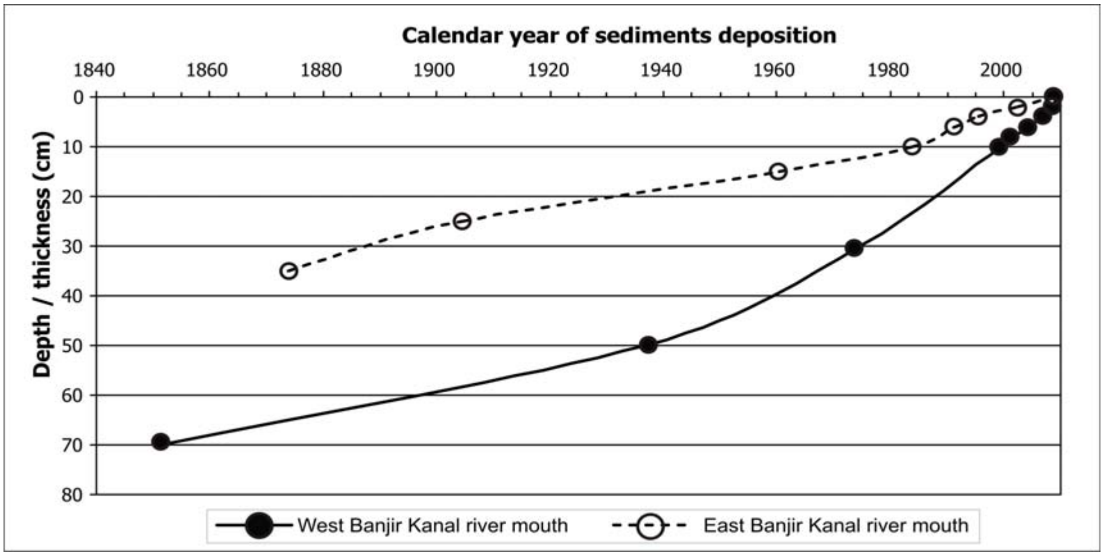

Fig 4. Diagram of sediments age determination (deposition year of sediments) collected from sediment corer at West Banjir Kanal river (solid line) and East Banjir Kanal river (doted line) versus depth / thickness.

Heavy metals contaminant (Cu, Zn and Hg) recorded in the sediment layers

Heavy metals contaminant together with sediment and other materials carried by surface runoff into the sea waters are deposited, dilution, dispersed and absorbed by marine organism. Heavy metals concentration in the coastal sediment depend on various factors, there are natural source from rocks weathering processes, human activities / industrial activities with domestic and industrial wastes, absorption of heavy metals dissolved in the nearest water. Concentration of heavy metals can also be enriched in the area where there are high sediment re-suspension occurs as well as high organic matter production and accumulation, fine sand surface sediment which heavy metals are easily tend to bound and in reducing waters environment which metals are easily dissolved (Veeck *et al.,* 2007, Rochyatun and Rozak 2007).

Cu contaminant

Cu concentration found at East Banjir Kanal river mouth is higher than at West Banjir Kanal river mouth. **Fig. 5** (left) shows variation pattern of the contaminant concentration from the oldest up to recent sediment. Concentration of Cu at West Banjir Kanal river mouth is varied in between 31.7 – 46.2 ppm and at East Banjir Kanal river mouth in between 35.7 – 71.2 ppm. Evolution of Cu concentration especially at East Banjir Kanal river mouth shows increasing trend. Cu concentration in the pre industrial sediments suggested a natural origin (45 – 48 ppm) continues to industrial sediments (54 ppm) and even in recent sediment (71 ppm). Small anomaly is shown in the sediments formed on 1960 (represent pre-industrial decade), concentration of Cu reached 58 ppm. Concentration of Cu in recent sediment at East Banjir Kanal river mouth is higher than Cu concentration in the sediments at the Jakarta Bay on September 2003 (13.81-63.45 ppm) (Rochyatun and Rozak, 2008).

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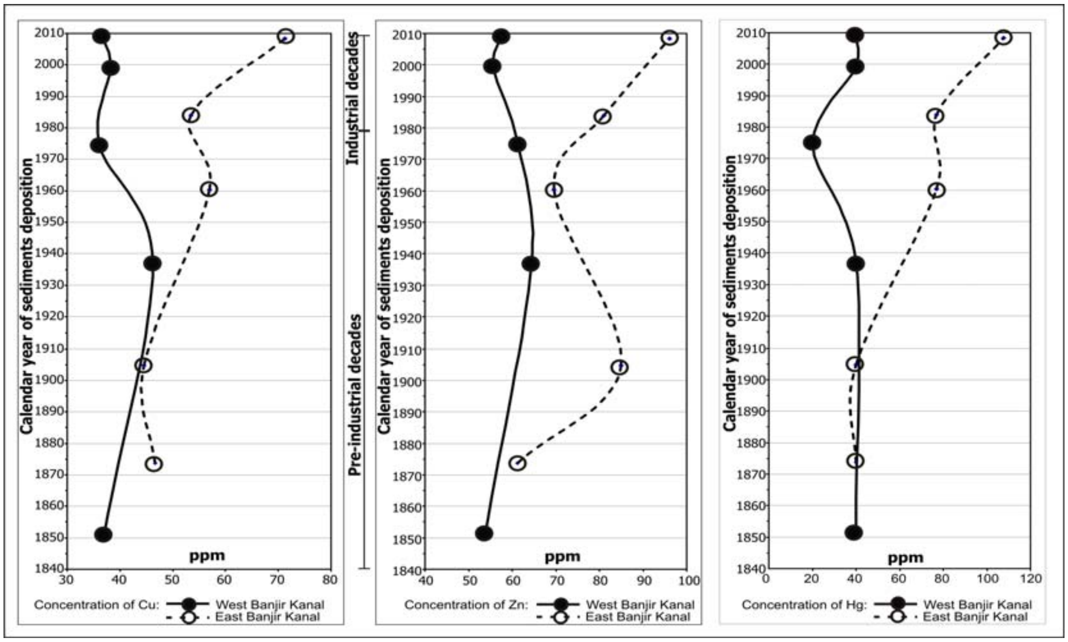

Fig 5. Diagram concentration of Cu (left), Zn (center) and Hg (right) at West Banjir Kanal river (solid line) and East Banjir Kanal river (doted line) versus year of sediments deposition.

Average of natural concentration for Cu in the sediment is 45 ppm (Rubio *et al., in* Lestari, 2008). Based on this reference, concentration value for Cu at East Banjir Kanal river mouth can be classified high. The anthropogenic contributing sources for Cu contaminant in the waters that finally deposited in the sediments are fertilizer plant and copper smelter, iron and steel complex industres, thermal power plant (Balkis *et al.*, 2007), wood industries (Palar 1994 *in* Siaka 2008), paints and antifouling industries (Austin 1984 *in* Siaka 2008).

Zn contaminant

Zn concentration found at East Banjir Kanal river mouth is higher than at West Banjir Kanal river mouth. **Fig. 5** (center) shows variation pattern of the contaminant concentration from the oldest up to recent sediments. Concentration of Zn at West Banjir Kanal river mouth is varied between 53 – 64 ppm and at East Banjir Kanal river mouth between 60 – 96 ppm. Evolution of Zn concentration especially at East Banjir Kanal river mouth shows increasing trend, started in the preindustrial sediments (60 – 70 ppm) continues to the industrial sediments (80 ppm) and even in recent sediments (90 ppm). Anomaly is shown in the sediment dated 1905 which is belonged in to the pre industrial sediments (**Fig. 4** and **5**), concentration of Zn reached 85 ppm. Sediment material in this layer is consist of very fine sand – silt (sediment material with ∅ grain 0.063 – 0.0315 mm), shows black color, abundant shells and hydrogen sulfide (H_2S) smell. Metal especially for Zn, natural and anthropogenic inputs may be of the same order (Ahdy and Khaled 2009). Normal concentration for natural Zn in the sediment is 95 ppm (Rubio *et al.,* 1994 *in* Lestari 2008). Salomons & Förstner 1984 (*in* Prego *et al.,* 2007) explained that since sediment particles smaller than 0.063 present a very strong adsorption potential for trace metals, especially for Zn and Pb are predominantly

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distributed and concentrated in this fraction. Alves *et al.,* (2007) in his recent study showed that sulfide environments seems to be the principal metal-binding phase. From the points viewed above suggested that natural source of Zn was trapped and enriched locally in this layer.

For comparison, concentration of Zn in recent sediment at East Banjir Kanal river mouth is still lower than Zn concentration in the sediments at the Jakarta Bay on September 2003 (84-259 ppm) (Rochyatun and Rozak, 2008). Karbassi *et al.,* 2005 viewed that anthropogenic Zn generally in associated with Pb as an indicator of oil pollution. Siaka (1998) pointed that concentration of Pb in the coastal sediment is close relationship to the harbor activity. From the two points viewed above it is suggested that marine traffic activities and oil pollution in the Jakarta Bay higher than in Semarang waters.

Other significant contributing of anthropogenic sources for Zn contaminant are: domestic sewages, shipyard works (Prego *et al.,* 2008), used tinned food (Palar 1994 *in* Siaka 2008), thermal power plant industries and iron and steel complex industries (Balkis *et al.,* 2007).

Hg contaminant

Hg like other heavy metals discharge into the waters through various sources, there are natural and anthropogenic. Surface runoff, atmospheric deposition and mainly fluvial transport are prominent pathways in the Hg transport from land area to coastal waters (Suseno and Panggabean, 2007). Prevailing distribution of Hg in sediments can be an indicator of extent of contamination in water receiving area. Loring (1975) viewed that mercury derived from natural source and anthropogenic sources migrates in a variety of dissolved and particulate forms. In the surface sediments, Hg is usually retained by these constituents especially in reducing environments where the presence of hydrogen sulphide (H_2S) from organic matter decaying allows some Hg to dissociate and be precipitated as a rather insoluble with little forming of mercury sulphide (HgS) (Veeck *et al.,* 2007; Rochyatun and Rozak 2007). The presence of Hg in pre-industrial sediments is also contributed by clay minerals, organic matters and Mn and Fe oxides in the sediments (Ram *et al.,* 2009).

Similar with Cu and Zn, Hg concentration found at East Banjir Kanal river mouth shows higher (40 – 107 ppm) than at West Banjir Kanal river mouth (20 – 56 ppm). Concentration of Hg contaminant at East Bajir Kanal river mouth tend to increase continuously since industrial decades (**Fig. 5** right). Anthropogenic source of Hg are soap and cosmetic industries (Muwanga & Barifaijo 2006), chlor-alkali plants (Loring 1975; Ram *et al.,* 2009) and pulp mills (Loring 1975).

CONCLUSION

Degree of metals contaminant deposited at West Banjir Kanal river mouth was not affected by human activities, there is shown of constant values since pre-industrial decades. Degree of metals contaminant deposited at East Banjir Kanal river mouth shows increasing values since industrial decade and correspond with the increasing of population and human activities.

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