Asian Journal of Water, Environment and Pollution, Vol. 10, No. 1 (2013), pp. 95–115.

Sources and Degradation of Sedimentary Organic Matter in Coastal Waters off the Brantas River, Java, Indonesia

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Received November 5, 2012; revised and accepted November 16, 2012

Abstract: Organic matter (OM) processing in estuaries is crucial in the marine environment as significant quantities of OM are buried or modified in these land-ocean-interaction zones. Southeast Asia is globally important in this regard because of high sediment inputs to the ocean and intense human modifications in the coastal zone, as exemplarily can be observed in the catchment of Java's second largest river, the Brantas. In order to investigate sedimentary OM processing, surface sediments and short sediment cores were sampled in its estuary in the rainy and dry seasons of 2008. $\delta^{13}C_{org}$, $\delta^{15}N$ and C/N ratios as well as amino acids and hexosamines were used to determine the sources, transformation and fate of estuarine sedimentary organic matter. Ranges in $\delta^{13}C_{org}$ of –24.9 to –20.1‰, in $\delta^{15}N$ of 3.5 to 5.4‰ and a C/N ratio of 7.9 to 16.5 in the sediments indicate a mixture of freshly produced marine algae and degraded terrestrial soil organic matter.

The relative contributions of the autochthonous and allochthonous OM in the estuarine sediments differed according to the amount and dispersal of the land-derived material. As the discharge of the two main river arms, the Porong and the Wonokromo River, showed strong differences with upto five-fold higher values in the Porong River in the rainy season, the highest proportion of terrestrial OM was found off the Porong river mouth that received the highest riverine runoff. Also the lowest sedimentary reactivity was detected in this region as displayed by amino acids (AA) and hexosamines (HA). AA+HA ranged between 0.76 and 5.25 mg g^{-1}, amino acid bound carbon between 5.9 and 22.6% and the AA/HA ratio between 4.2 and 13.0. Furthermore, a reduced intensity of OM degradation was observed in front of the Porong River outlet, which has been attributed to the high quantity of deposited material and the low reactivity of the surface sediments. In a global context, the reactivity of sedimentary OM from the Brantas estuary was in the range of degraded sediments from offshore regions or stations at greater water depth. It indicates that severe OM degradation based on a strong tidally induced resuspension of sediments in the turbid and well mixed waters of the shallow Brantas estuary is responsible for burial of refractory carbon.

Key words: Sediments, organic matter, degradation, stable carbon isotopes, amino acids, Indonesia, Java, Brantas estuary.

Introduction

Estuaries and their adjacent areas are the main connection between terrestrial and marine environments. Significant quantities of terrestrial and marine organic matter (OM) are deposited in these regions of high primary production that is largely sustained by the riverine nutrient inputs (Prahl et al., 1994; McKee et al., 2004; Bianchi, 2007). Thus, estuaries and coastal zones are a significant sink of OM in the marine environment and play an important role in the global organic carbon cycle (Berner, 1989; Hedges and Keil, 1995; Hedges, 1997). Determining the origin and composition of OM and processes that affect its distribution, degradation and preservation in coastal zones is therefore fundamental for a comprehensive understanding of the fate of OM in the marine environment. In this respect, recent studies revealed a strong degradation of OM along tropical coasts and

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shelves that is based on intensive resuspension of sediments caused by high tidal energy and coastal currents (e.g. Aller and Blair, 2006). The identification of OM provenances of estuarine sedimentary OM can be based on elemental, isotopic and molecular biomarkers, whereby a simultaneous use of two or more tracers can considerably improve the determination (Thornton and McManus, 1994).

Organic carbon/nitrogen atomic ratios (C/N ratio) and carbon and nitrogen isotopic composition have been widely used to define the origin of sedimentary OM in estuarine sediments, which predominantly derives both from terrestrial and marine sources (Shultz and Calder, 1976; Peters et al., 1978; Fry and Sheer, 1984; Meyers, 1994). The application of these tracers relies on fundamental differences in the use of carbon and nitrogen sources during biosynthesis of OM in terrestrial and aquatic ecosystems which result in clearly distinguishable $\delta^{13}C$ and $\delta^{15}N$ (Degens, 1969; Peterson and Fry, 1987; Meyers, 1994; Kendall, 1998). The C/N ratios differ due to the abundance of cellulose in vascular plants and its absence in marine produced OM that is instead rich in proteins (Meyers, 1994). Nevertheless, especially the C/N ratio and $\delta^{15}N$ signal of OM can change during various processes like the ammonium adsorption by sediments or degradation processes (Müller, 1977; Rosenfeld, 1979; Thornton and McManus, 1994).

Amino acids (AA) and hexosamines (HA) account for significant parts of the organic carbon and organic nitrogen in most organisms (Lee, 1988) and are labile relative to bulk nitrogen and carbon (Ittekkot, 1988; Cowie and Hedges, 1992). The analysis of these labile compounds provides a useful tool to evaluate the degradation status or reactivity of particulate OM in estuaries that can be either influenced by decomposition in general, or in particular by a different reactivity of OM from land-derived and marine sources (Cowie and Hedges, 1992; Dauwe and Middelburg, 1998). Due to the generally refractory character of riverine material, which predominantly results from high contributions of strongly degraded soil, a distinction can be made towards the freshly produced marine phytoplankton, e.g. by its high contributions of amino acids to bulk OC (Ittekkot et al., 1984; Cowie and Hedges, 1992). Based on changes of the amount and composition of AA and HA occurring during decay, degradation processes can be detected, for example by the calculation of the degradation index (DI) (Dauwe and Middelburg, 1998) or the reactivity index (RI) (Jennerjahn and Ittekkot, 1997) but also AA/HA ratios can be used to determine the reactivity of OM as decreasing values indicate increasing OM degradation based on the fact that AAs are preferentially lost during degradation compared to HAs (Müller et al., 1986; Haake et al., 1992; Unger et al., 2005).

The intensity of OM degradation during early diagenesis in sediments determines the OM preservation and does strongly depend on specific local environmental conditions. It is influenced by organic carbon flux, bulk sedimentation rate as well as oxygen concentration in the bottom water and is related to the extent of bioturbation of surface sediments (Aller et al., 1985; Hartnett et al., 1998). The rate of degradation slows down with increasing sediment depth. This is primarily due to the decreasing OM reactivity as the remineralization of the more labile OM, which breaks down easily, proceeds first and the more refractory material concentrates with depth (Canfield, 1993; Hulthe et al., 1998; Thamdrup and Canfield, 2000; Burdige, 2007). An appropriate way to detect these downcore variations is the measurement of the more labile compounds of OM, like amino acids and hexosamines.

Due to the fact that tropical and subtropical estuaries receive ~70% of the freshwater and ~74% of the sediment discharge to the world's oceans (Milliman and Meade, 1983), investigation of sources and fate of the OM in these coastal areas is especially important for the global carbon cycle. This knowledge is fundamental to understand coastal ecosystem processes as sediments play a very important role in shallow, coastal environments due to their large storage capacity and their buffering function considering the retention and release of nutrients (Jørgensen, 1996). The islands of Indonesia are particularly relevant in this respect as they presumably contribute 20 to 25% of the global sediment export (Milliman et al., 1999). Furthermore, the coastal zones of Southeast Asia are among the regions with the strongest human modifications around the world (Nicholls and Small, 2002), which most probably strongly affect the ecology and elemental cycles of the adjacent coastal ecosystems. However, far less is known about tropical coastal ecosystem functions than from temperate regions (Alongi, 1998).

The most urbanized region in Indonesia corresponds to the catchment area of the second largest river of Java, the Brantas River. Its estuary receives high riverine inputs that are on the one hand seasonally varying (monsoonal cycle) and on the other hand disproportionally distributed to the two main river outlets and its near-shore, extensive muddy tidal flats. This study investigates the seasonal and regional variations of sedimentary OM in this tropical estuary and aims to determine the sources of the OM, their mixing and spatial distribution as well as the reactivity and the degradation of the sedimentary OM.

The Study Area

The Brantas River is located at the eastern coast of Java, Indonesia. With a length of 320 km it is the second largest river of the island and drains a catchment area of 11,050 km^2 (Whitten et al., 1996) accounting for approximately 35% of the East Java Province. The Brantas River originates in high volcanic mountains and diverts into the three branches Porong, Wonokromo and Mas in the coastal lowlands (Figure 1a). The overall discharge of the Brantas River fluctuates enormously during the year, depending on seasonal climatic changes which are dominated by a strong monsoonal system. This is characterized by the alteration of a wet season (West-Northwest monsoon) from November to April and a dry season from May until October dominated by the East-Southeast monsoon. The average annual rainfall amounts to 2300 mm yr^{-1} (Aldrian and Djamil, 2008), of which 80% precipitates during the wet season. This coincides with a peak in water and material discharge of the two main river channels Wonokromo and Porong that discharge into the Madura Strait.

The Porong represents the major water and sediment transporting branch. During the rainy season, about 85% of the runoff is discharged through the Porong channel. In contrast, during the dry season the significantly lower river discharge occurs in a large part via the Surabaya River and the Wonokromo (Hoekstra, 1989b). These natural discharge trends were enhanced by river regulations that result in an average discharge of 47 and 20 m^3 s^{-1} in the Wonokromo and 264 to 50 m^3 s^{-1} in the Porong River during the rainy and dry seasons, respectively (2003-2007) (Jasatirta Public Corporation, pers. comm.). The Brantas River has a very high sediment load, especially during the rainy season (Hoekstra, 1989b). This is promoted by very high erosion and denudation rates (Lavigne and Gunnell, 2006) resulting from generally favourable natural conditions for high mechanical and chemical weathering on the one hand (Gaillardet et al., 1999) and human interventions on the other hand, e.g. severe deforestation for the benefit of cash-crop growing areas and the absence of protecting riverbank stripes (Römer-Seel, 2003). The different water runoff via the two main river channels indicate a lower and more stable material supply to the Wonokromo estuary throughout the year compared to the Porong estuary, which receives the predominant part of the high river material discharge in the rainy season. The riverine material is distributed to the extensive intertidal flats at the Brantas estuary. Their depth averages around 1-2 m below sea level and water level fluctuations occur on a micro- to mesotidal scale during mixed diurnal-semidiurnal tidal cycles (Hoekstra et al., 1989).

The Brantas catchment is nowadays the most urbanized region in Indonesia (approximately 16 million inhabitants) and it is one of the nation's major regions of cultivation where nearly half of the area is used for agriculture, mainly rice cultivation (Booth et al., 2001). Numerous efforts have been made to regulate the water resources. This involves the building of dams, reservoirs or irrigation installations for the purpose of power generation as well as flood control and in order to meet the increasing demand of water for domestic, industrial and agricultural water purposes. In this context, a declining water quality has been noticed that is mainly caused by domestic wastewater disposal and occurs especially in urban areas (Ramu, 2004).

Materials and Methods

Stations and Sample Treatment

In the rainy (February) and the dry (July) season of 2008 surface sediments and short sediment cores were sampled in the coastal waters of the Brantas estuary. In water depths between 0.5 and 25 m, 46 (rainy season) and 48 (dry season) surface sediment samples were collected during each field campaign using a Van Veen grab. Samples were taken along transects perpendicular to the coast in front of the Wonokromo river mouth (= Wonokromo estuary, three transects) and the Porong river mouth (= Porong estuary, six transects) (Figure 1b). The transects were sub-divided into proximal and distal regions relative to the coastline (Figure 1b). The boundaries between proximal and distal stations were set according to the respective river plume extension that was defined by satellite images, total suspended matter (TSM) contents and salinity values (Figure 1a, b). Additionally, a further transect was sampled parallel to the coast between these two estuaries (intermediate region). The grab was opened with utmost caution and the surface sediment layer (1 cm) was sampled and split, whereby the surface was easily recognized by a thin brownish layer. One half, taken for geochemical analyses, was frozen, freeze-dried (Alpha 1-2 LDplus, CHRIST) and ground (RETSCH Planetary Ball Mill PM 100, 500 rpm). The other half was stored cool and dark in small plastic bags for grain size measurements.

Short sediment cores were taken at 11 stations in the rainy season and at 12 stations in the dry season of 2008.

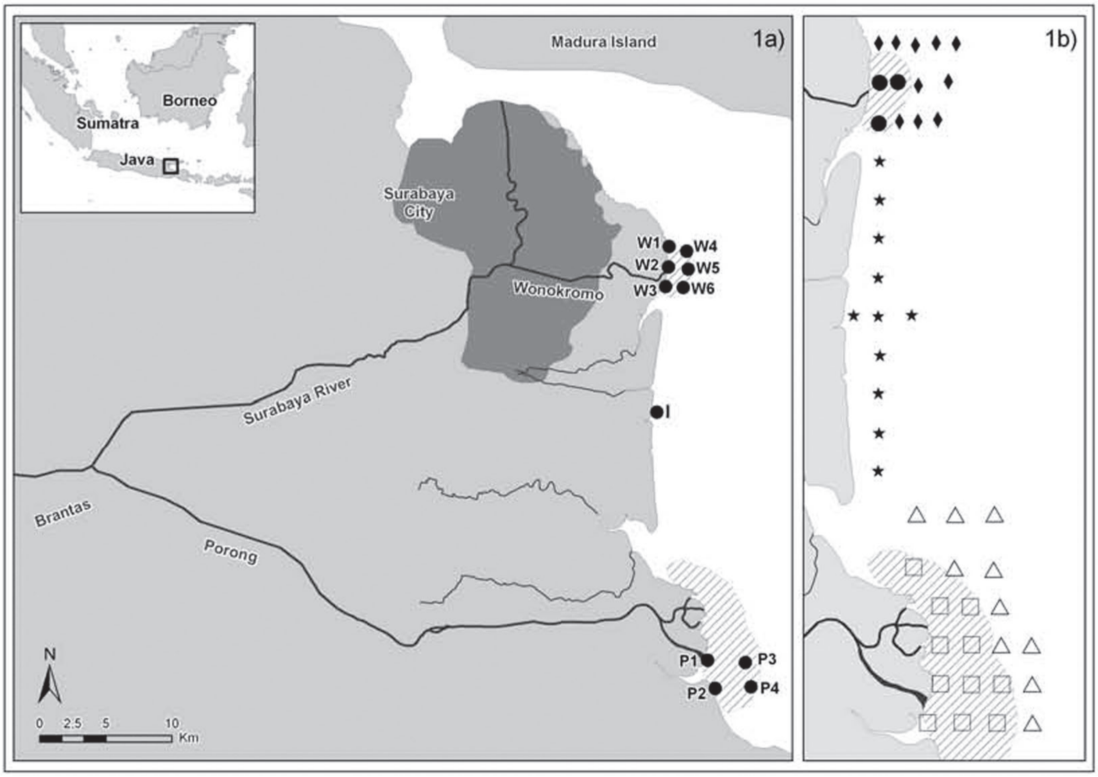

Figure 1: Sampling sites of (a) sediments cores and (b) surface sediment samples including river plume extension in the rainy season. Sediment cores are distinguished in cores taken at the Wonokromo estuary (W1-W6), the Porong estuary (P1-P4) and the intermediate region (I). Surface sediment sampling sites are subdivided into five regions (□ Porong proximal, Δ Porong distal, ● Wonokromo proximal, ◆ Wonokromo distal, ✶ intermediate region). River plume extensions are indicated by hatched areas.

Four cores were situated at the Porong estuary (P1-P4) and five (rainy season, except W5)/six cores (dry season) at the Wonokromo estuary (W1-W6) (Figure 1a). Within ~500 m and ~2 km distance to the coast they were taken from water depths between 0.3 and 1.4 m. One station was located in the intermediate region close to the coastline (I). For sampling, a hand-corer (HYDRO-BIOS) and plastic liners with a length of 60 cm and a diameter of 7 cm were used. The cores, whose length varied between 25 and 45 cm, were sliced in 5 cm intervals. Samples were split for grain size analyses and geochemical analyses and further processed like the surface sediment samples.

Analytical Methods

Total carbon (TC), total nitrogen (TN) and total organic carbon (TOC) concentrations were analyzed by high-temperature combustion in a Carlo Erba NA 2100 element analyzer (Verardo et al., 1990). TOC measurements took place after the removal of carbonate by acidification with 1N HCl and subsequent drying at 40ºC (analytical errors OC <0.05%, N <0.01%). The total organic carbon is reported in weight percent (%) and the ratio of OC and TN is discussed as C/N ratio in the following.

The organic carbon ($\delta^{13}C_{org}$) and nitrogen ($\delta^{15}N$) isotopic composition were determined in a Thermo Finnigan Delta Plus mass spectrometer after high temperature combustion in a Flash EA 1112 elemental analyzer. Sediment material was decarbonated with 150 µl of 1N HCl and dried at 40ºC for the $\delta^{13}C_{org}$ determination. $\delta^{13}C_{org}$ and $\delta^{15}N$ results are given as per mil relative to Vienna PDB standard and nitrogen isotopic composition of atmospheric air, respectively, based on the following equation:

$$\delta R = (R_{sample}/R_{standard} - 1) \times 1000$$

with $R = {}^{13}C/{}^{12}C$ or $R = {}^{15}N/{}^{14}N$

The amount and monomeric composition of total hydrolysable amino acids (AA) and total hydrolysable hexosamines (HA) were analyzed with a Biochrom 30 Amino Acid Analyzer after hydrolysis with 6 N HCl for 22 h at 110°C. The individual monomers were separated with a cation exchange resin and detected fluorometrically. A detailed method description is given by Jennerjahn and Ittekkot (1999).

The grain size distribution was determined using a laser particle analyzer HORIBA LA-300. For homogenization, 0.1 to 0.4 g of sediment were suspended in ~30 ml distilled water and sodium phosphate ($NaPO_4$) was added (1-2 g). Further disaggregation of the particles was achieved with ultrasonication (15 seconds). The measurements of the suspension were performed in water dispersion according to the Mie Scattering Theory (Mie, 1908).

The grain size is commonly strongly correlated to the content of OM that adsorbs onto mineral surfaces (Keil et al., 1994). Since fine particles have the largest surface area and due to the fact that the fine grain size fraction <20 µm (clay, fine and medium silt) showed the best correlation with the OC content in our data set (data not shown), we used this fraction for the characterization of the grain size distribution.

Statistics

For a synoptic presentation of the data, sediment core characteristics were consolidated via *locally weighted scatter plot smoothing* (LOESS) (Hastie and Tibshirani, 1990). The analyses were performed with the program *R*.

Results

Surface Sediments

The organic carbon content of surface sediment samples varied from 0.3 to 1.5% (Figure 2a). Sediments at the Porong estuary showed values between 0.3 and 1.5%. An increase in sedimentary OC was observed towards the offshore sampling sites. This gradient was more distinctly pronounced in the dry season. Except for significantly higher values directly in front of the Wonokromo river mouth in the rainy season (average of the proximal area = 1.1%), lower values were found at the Wonokromo estuary compared to the Porong estuary (Table 1). The sediments in the intermediate region displayed higher OC contents towards the Porong estuary and lower values in northward direction to the Wonokromo (Figure 2a). Highest C/N values were found at the proximal area of the Porong estuary in the rainy season (mean 11.4, Table 1, Figure 2d). Besides an overall decline of values during the dry season, lower C/N ratios were observed with increasing distance to the coastline in both seasons. At the Wonokromo estuary, this gradient was only apparent during the dry season (Table 1). During the entire year, the intermediate region had the lowest values (annual average: 9.2).

The $\delta^{13}C_{org}$ of the sediments ranged between –24.9‰ and –20.1‰. The Porong and the Wonokromo estuary had higher values in offshore direction, with the proximal samples showing slightly higher values during the dry season (Table 1, Figure 2c). Highest $\delta^{13}C_{org}$ values in the coastal sediments were found in the intermediate region, with a slight decline during the dry (–20.8‰) compared to the rainy period (–21.5‰). Amino acid and hexosamine contents in sediments were found in quantities of 0.62 to 5.67 mg g^{-1} and 0.09 to 0.75 mg g^{-1}, respectively. The concentrations of both compounds were correlated (rainy season $R^2 = 0.78$, $p < 0.05$; dry season $R^2 = 0.79$, $p < 0.05$) and are discussed as the sum of AA+HA. Lowest values were found at the Porong estuary, where higher AA+HA contents were observed in the distal (annual average = 3.93 mg g^{-1}) compared to the proximal sediments (annual average = 2.18 mg g^{-1}) (Table 1). This spatial trend was more pronounced in the dry season. Such a gradient was not found at the Wonokromo estuary, where the highest values were detected in the proximal stations during the dry season (Table 1, Figure 2b). The sediments in the intermediate region exhibited an annual average of 3.93 mg g^{-1} AA+HA with slightly higher values in the dry season.

Based on the fact that the factor coefficients of a PCA carried out with our amino acid data were not in agreement with the factor coefficients used by Dauwe and Middelburg (1998) to calculate the DI, which implies that our data was not determined by degradation alone, we decided not to apply this indicator. Furthermore, strongly varying non-protein amino acids in our data prohibited the calculation of reliable (robust) RI's that could be compared with each other. Instead, we use the percentages of amino acid bound carbon and the ratio of amino acids to hexosamines (AA/HA ratio) that both decrease with OM degradation (Cowie and Hedges, 1992; Unger et al., 2005). This is based on the fact that these sum parameters are more robust as compared to variations of single monomers like the non-protein amino acids and therefore provide a more consistent data set.

Despite the general correlation of AA and HA, their relative contents varied within the sediments leading to differing AA/HA ratios. They ranged between 4.2 and 13 and were highest in the sediments of the intermediate region and lowest at the Porong estuary (Figure 2e). These ratios were higher in the entire coastal region during the dry season. During both seasons, sediments of the Porong estuary had higher ratios in offshore direction, which was more pronounced in the rainy season (Table 1). This gradient could not be found at the Wonokromo estuary, where to the contrary strongly elevated values were

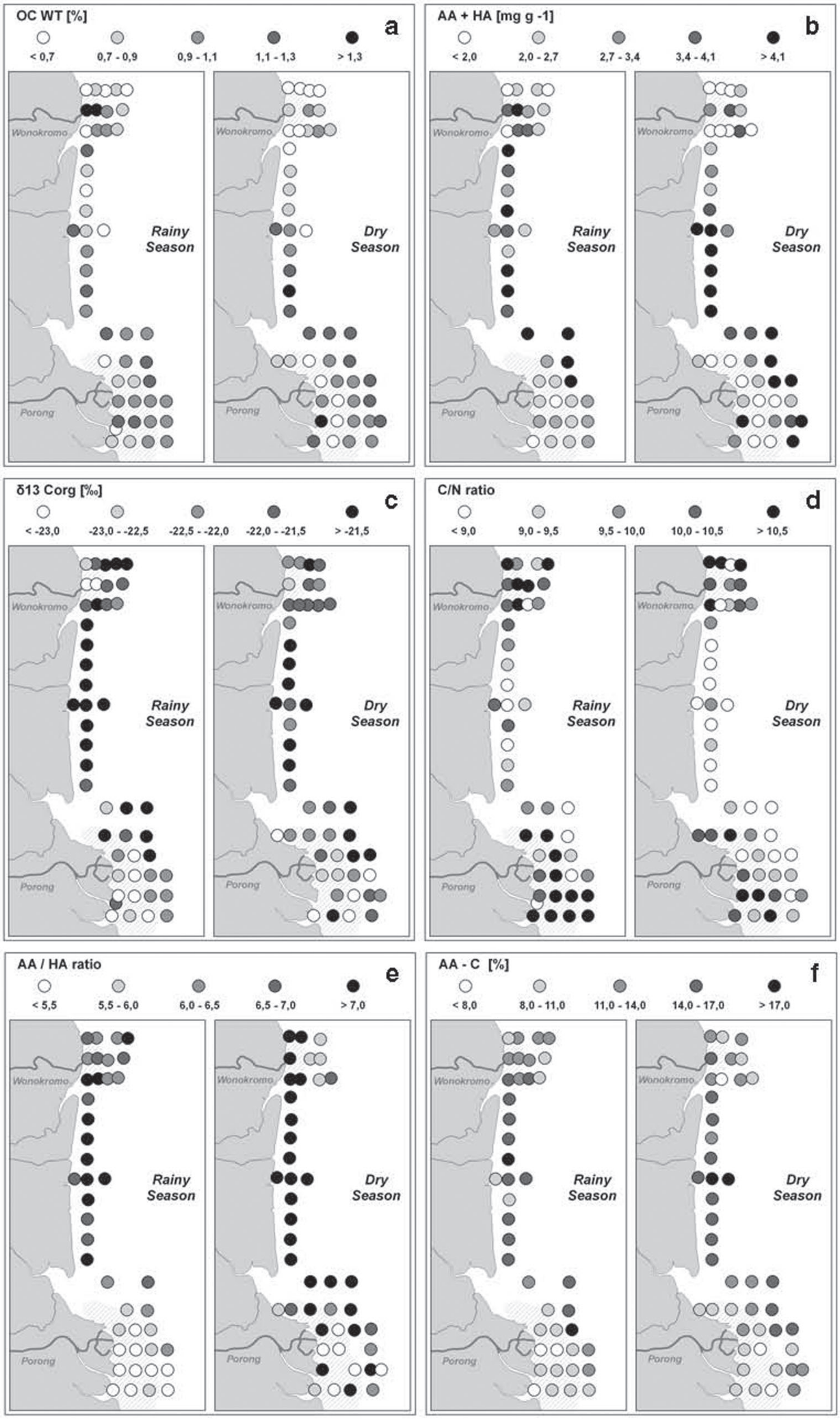

Figure 2: Spatial distribution of (a) organic carbon, (b) total hydrolysable amino acids + hexosamines, (c) carbon isotopic composition, (d) C/N ratio, (e) amino acid/ hexosamines ratio and (f) amino acid bound carbon in surface sediments at the Brantas estuary in the rainy and dry season of 2008.

Table 1: Mean value, standard deviation and range of total organic carbon, C/N ratio, carbon and nitrogen isotopic composition, amino acid characteristics and grain size of the surface sediment samples at the Brantas estuary.

Season		*OC [%]*	*C/N ratio*	*$\delta^{13}C_{org}$ [‰]*	*$\delta^{15}N$ [‰]*	*AA+HA [mg/g]*	*AA-C [%]*	*AA/HA ratio*	*Grain size [<20 µm]*
Rainy Season 2008	Porong-proximal n=10-12								
	Average	**0.84**	**11.4**	**–22.8**	**4.6**	**2.28**	**8.6**	**5.0**	**77.4**
	Stdv	0.26	2.1	0.9	0.3	0.43	1.6	0.5	23.6
	Range	0.27–1.15	8.3 - 16.5	–24.3–21.0	4.1–5.0	1.53–2.74	5.9–10.7	4.2–5.8	15.1–95.5
	Porong-distal n=9-10								
	Average	**1.06**	**9.7**	**–21.7**	**4.4**	**3.80**	**12.9**	**5.9**	**87.9**
	Stdv	0.10	0.8	0.7	0.3	0.97	2.8	0.5	5.1
	Range	0.93–1.28	8.6–10.6	–22.7–20.7	4.0 –4.9	2.68–5.21	9.4–17.2	5.3–6.7	77.7–95.5
	Wonokromo-proximal n=3								
	Average	**1.05**	**10.4**	**–22.8**	**3.9**	**3.61**	**13.1**	**6.7**	**58.8**
	Stdv	0.48	0.5	0.9	0.3	1.48	1.7	0.4	29.8
	Range	0.49–1.33	10.1–11.0	–23.5–21	83.6–4.1	1.95–4.82	11.3–14.7	6.3–7.0	24.8–80.0
	Wonokromo-distal n=9-10								
	Average	**0.74**	**10.4**	**–21.7**	**4.5**	**2.63**	**12.1**	**6.6**	**53.0**
	Stdv	0.24	0.5	0.5	0.2	0.87	1.8	0.6	22.0
	Range	0.30 - 0.97	8.6 - 13.0	–22.7–20.7	4.2–5.1	1.2–4.1	9.4–15.7	6.0–7.9	16.6–80.3
	Intermediate region n=11								
	Average	**0.95**	**9.4**	**–20.8**	**4.3**	**3.74**	**14.9**	**7.6**	**69.4**
	Stdv	0.19	0.7	0.4	0.3	0.93	3.3	1.4	13.6
	Range	0.65–1.20	8.5–10.5	–21.5–20.3	3.8–4.7	2.37–5.23	8.5–21.0	6.7–11.5	49.8–88.7
dry Season 2008	Porong-proximal n=12-13								
	Average	**0.80**	**10.5**	**-22.7**	**4.1**	**2.09**	**9.3**	**6.5**	**57.5**
	Stdv	0.35	1.3	1.0	0.4	0.96	0.2	2.3	19.4
	Range	0.27–1.47	8.1–13.1	–24.9–21.0	3.5–5.1	0.76–4.26	6.0–13.6	4.6–13.0	24.3–84.0
	Porong-distal n=11–12								
	Average	**1.11**	**8.8**	**–21.8**	**4.4**	**4.05**	**13.4**	**6.9**	**87.3**
	Stdv	0.07	0.6	0.7	0.4	0.74	2.1	1.0	7.6
	Range	096–1.19	7.9–9.7	–23.2–20.8	4.0–5.1	2.69–5.25	8.8–16.9	5.2–8.2	69.2–96.1
	Wonokrono-proximal n=2								
	Average	**0.57**	**10.7**	**–22.2**	**4.1**	**2.11**	**13.7**	**8.7**	**25.6**
	Stdv	0.40	0.9	0.6	0.1	1.65	1.4	0.5	21.5
	Range	0.29–0.86	10.1–11.4	–22.7–21.8	4.0–4.2	0.94-3.27	12.7-14.7	8.3-9.0	10.4-40.9
	Wonokromo-distal n=8-10								
	Average	**0.73**	**10.1**	**–21.8**	**4.5**	**2.23**	**11.1**	**6.6**	**58.3**
	Stdv	0.14	0.7	0.6	0.5	0.89	2.3	1.0	26.0
	Range	0.56–0.95	8.8–10.9	–22.4–20.1	3.7–5.1	1.16–3.56	7.2–14.0	5.5–8.3	2.7–82.7
	Intermediate region n=11								
	Average	**0.97**	**9.0**	**–21.5**	**4.8**	**4.12**	**16.2**	**8.3**	**70.1**
	Stdv	0.26	0.5	0.5	0.4	1.31	2.8	0.7	19.0
	Range	0.57–134	8.4–9.9	–22.3–20.9	4.3–5.4	2.3–6.27	12.0–22.6	7.6–9.5	33.7–89.7

observed in the proximal compared to the distal stations in the dry season. The percentages of amino acid bound carbon (AA-C%) showed similar seasonal and spatial trends as detected for the AA/HA ratio (Table 1, Figure 2f). The contribution of AA-C% ranged between 5.9% at the Porong estuary and 22.6% in the intermediate region.

The grain size of sediments from the Brantas estuary was dominated by the clay and silt size fractions (<20 μm) (Table 1). The most fine-grained material was found at the distal stations in the Porong estuary, where it amounted to 87% on an average. At the Wonokromo estuary, this grain size fraction accounted for an average of 57% in both seasons except for the proximal samples taken in the dry season, which exhibited the overall most coarse-grained sediments. In the sediments from the intermediate region the proportion of the <20 μm fraction was ~70%.

Sediment Cores

Organic carbon in the sediment cores ranged between 0.3 and 2.0% (Figure 3). Values were in the same range in all three areas. The cores in the Wonokromo and the Porong estuary showed strong fluctuations with depth (ranges: *P* = 0.3-1.8%, *W* = 0.3–1.4%). Several cores displayed a downcore increase at both estuaries but no significant overall trend could be observed as the confidence intervals of the upper and lower sediment layers overlapped. The core taken at the intermediate region exhibited a small range of values (0.8-1.3%) and a marginal decrease with depth.

Total AA+HA were found in quantities of 0.85 to 6.24 mg g^{-1}. The core in the intermediate region exhibited generally the highest values (3.42-6.24 mg g^{-1}) and showed a distinct decline of AA+HA contents with depth. This was more pronounced in the dry season due to higher values in the upper sediment layers (Figure 3). Like the OC content, the AA+HA content varied strongly and ranged between 1.02 and 4.97 mg g^{-1} off the Wonokromo and 0.85-4.21 mg g^{-1} off the Porong. Slight changes with increasing depth were observed at both estuaries in the dry season, when several cores showed a slight increase of AA+HA at the Porong estuary and a decline of values was detected in most of the cores of the Wonokromo estuary. AA-C% ranged between 5.3 and 19.8% and the AA/HA ratio between 2.9 and 10.4. Both parameters revealed a significant decrease with depth in all cores taken at the Wonokromo estuary (Figure 3). In contrast, at the Porong estuary, most of the sediment cores showed constant values with depth. The core of the intermediate region showed a significant decline of AA-C% and AA+HA in the dry season.

The grain-size fraction <20 μm varied between 14 and 98% and fluctuated strongly in most sediment cores. No significant trends were observed at the Wonkromo and the Porong estuary. However, many cores at the Porong estuary showed a decline of the fraction <20 μm in the upper half of the cores in the rainy season and an opposite trend in the dry season. At the Wonokromo estuary, the sediment became more fine-grained with increasing depth. Within core I, overall high values were detected.

Discussion

Sources of Organic Matter

Porong Estuary

The $\delta^{13}C_{org}$ of the surface sediments at the Porong estuary ranged between –24.9 and –21‰ and the C/N ratio between 7.9 and 16.5 in both seasons of 2008 and showed higher ($\delta^{13}C_{org}$) as well as lower (C/N ratio) values in the distal compared to the proximal stations (Table 1, Figure 4). $\delta^{15}N$ values varied between 3.5 and 5.0‰ and revealed no spatial pattern. It has been widely observed that the OM in estuarine sediments is a mixture of terrestrial and marine sources (Thornton and McManus, 1994; Middelburg and Nieuwenhuize, 1998; Yu et al., 2010). The land-derived OM that is transported to estuaries via rivers can be a very heterogeneous composition of autochthonous and allochthonous material (Fry and Sherr, 1984; Mook and Tan, 1991). Freshwater phytoplankton shows $\delta^{13}C_{org}$ values lower than –26‰, C/N ratio from 4 to 10 and a $\delta^{15}N$ signal between 5 and 8‰ (Sigleo and Macko, 1985; Cai et al., 1988; Meyers, 1994).

Other potential major sources of riverine particulate OM in the agricultural dominated Brantas River catchment area are vascular plant debris and underlying soils from the main crop rice (C_3 plant) and other important crops like maize and sugarcane (C_4 plants) as well as soybean and cassava (C_3 plants) (Badan Pusat Statistik Republik Indonesia). Vascular plants exhibit C/N ratios above 20 and the C_3 and C_4 type differ in their carbon isotopic composition (C_3 plants $\delta^{13}C_{org}$ ~– 27‰, C_4 plants ~– 14‰) (Rullkötter, 2006; Meyers, 1994). Rice and sugar cane plants sampled in the Brantas river basin exhibit $\delta^{13}C_{org}$ values of –28.1 and –12‰, C/N ratios of 29 and 54 as well as $\delta^{15}N$ values of 1.2 and –2.7‰, respectively (Jennerjahn et al., 2004). Soils from freshly sewn and harvested rice fields in the Brantas region are characterized by $\delta^{13}C_{org}$ of –26 and –23.4‰,

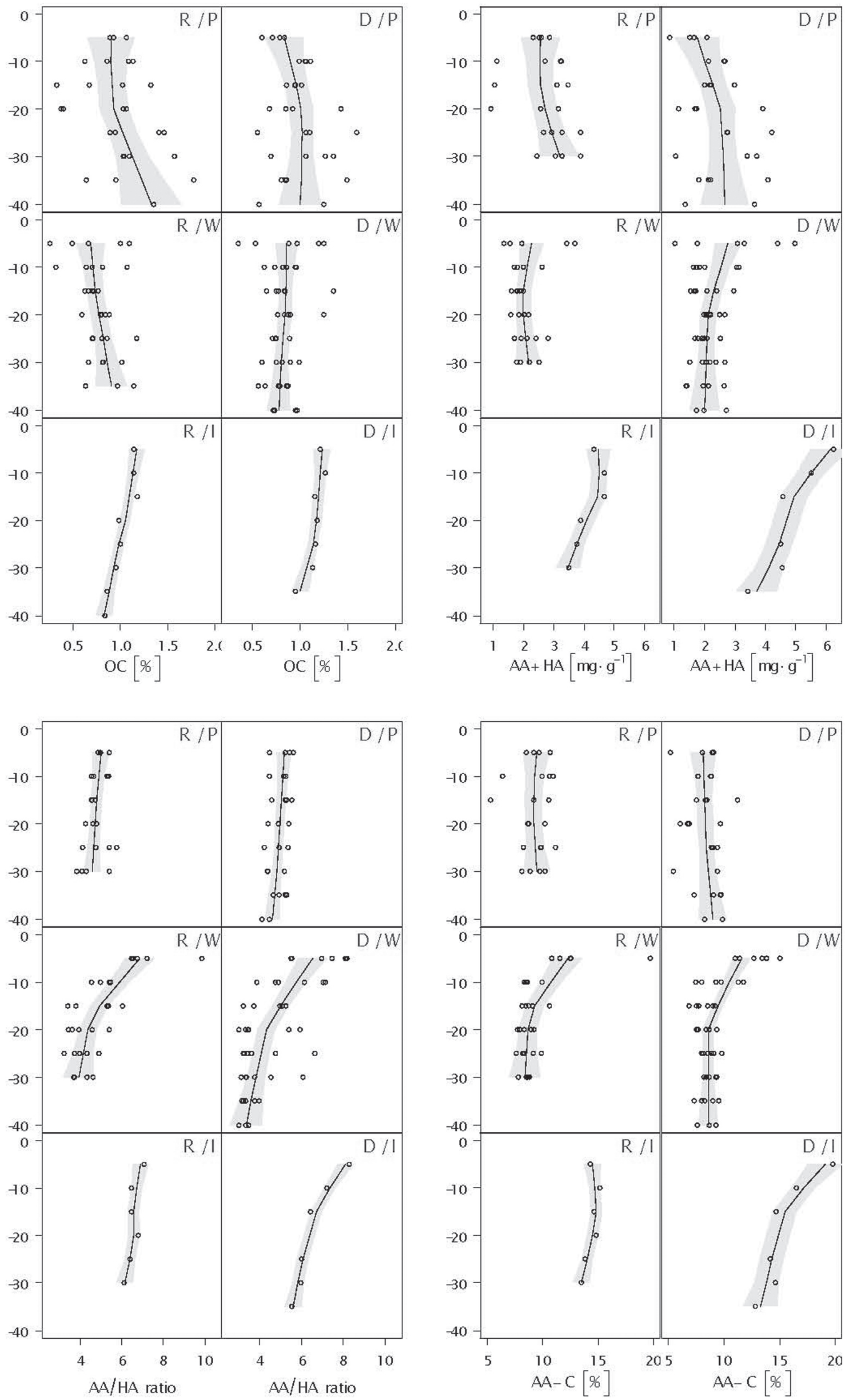

Figure 3: Downcore distribution of total organic carbon (OC), total hydrolysable amino acids and hexosamines (AA+HA), amino acid/hexosamines ratio and amino acid bound carbon (AA-C) in short sediment cores sampled at the Porong estuary (P, four cores), Wonokromo estuary (W, 5-6 cores) and the intermediate region (I, one core) in the rainy (R) and dry (D) season of 2008.

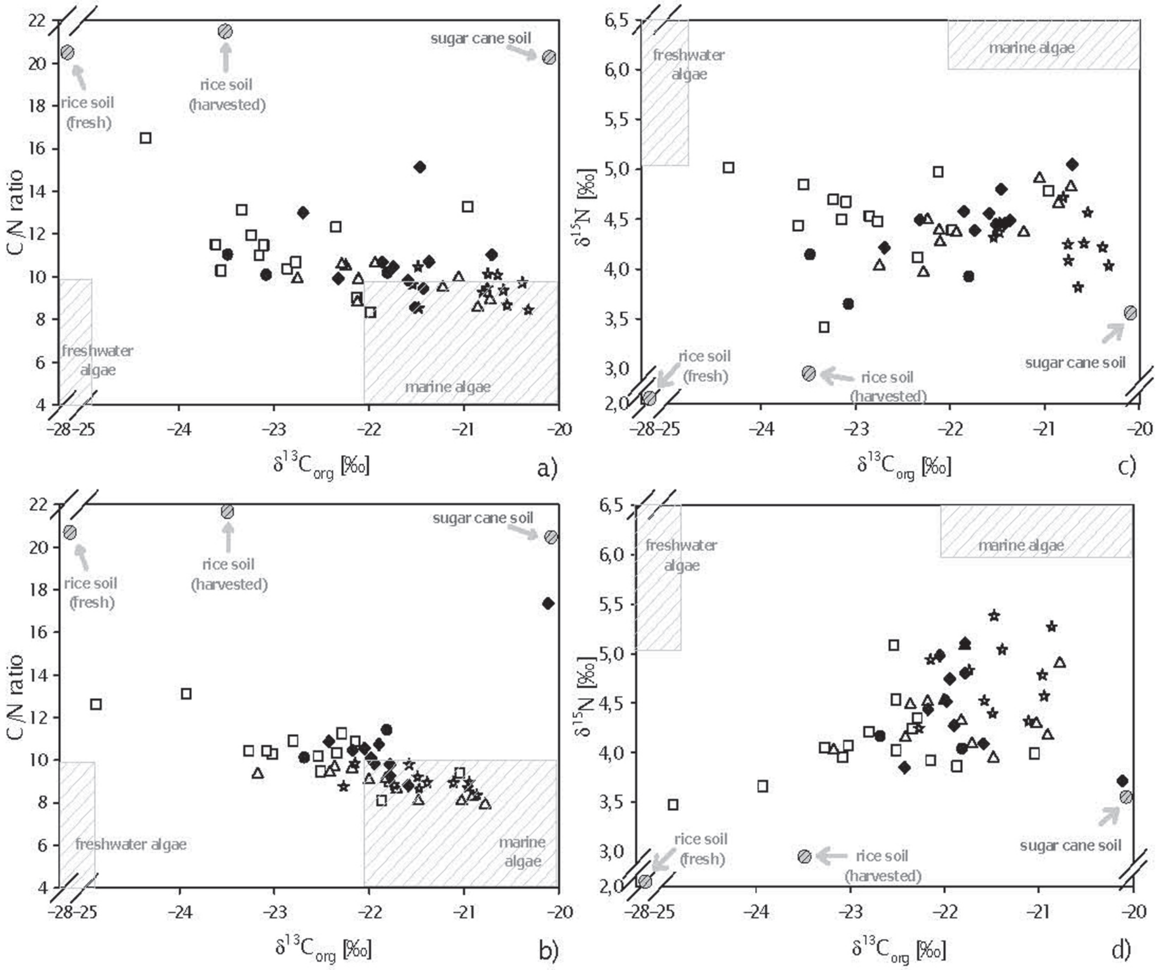

Figure 4: Stable carbon and nitrogen isotopic composition (δ^{13}Corg and δ^{15}N) and C/N ratios of surface sediments at the Brantas estuary. (a) δ^{13}Corg and C/N in the rainy season; (b) δ^{13}Corg and C/N in the dry season; (c) δ^{13}Corg and δ^{15}N in the rainy season; and (d) δ^{13}Corg and δ^{15}N in the dry season. Surface sediment sampling sites are subdivided into five regions (□ Porong proximal, Δ Porong distal, ● Wonokromo proximal, ◆ Wonokromo distal, ★ intermediate region).

C/N ratios of 20.7 and 22.1 and δ^{15}N signal of 2.0 and 2.9‰, the soil of sugar cane fields exhibits values of –20‰ ($\delta^{13}C_{org}$), 20.6 (C/N) and 3.7‰ (δ^{15}N) (Jennerjahn et al., 2004). Due to the large erosion rates in the region (Lavigne and Gunnell, 2006) the soil OM can contribute large amounts. This is indicated by data of suspended matter obtained in the Brantas River that shows a mixture of possible OM sources, but is mainly dominated by terrestrial soil underlying C_3 plants (Jänen, 2012) also supported by a previous study investigating the Brantas catchment that suggests a high input of agricultural soil via the river (Jennerjahn et al., 2004). This is in general agreement with numerous observations of riverine OM composition (Meybeck, 1982; Ittekkot and Arian, 1986; Ludwig and Probst, 1998). Suspended matter taken at high salinity stations in the Brantas estuary exhibited $\delta^{13}C_{org}$ values of –20 and –22‰ and C/N ratios of 8 to 10 as well as a δ^{15}N signal around 6‰ (Jänen, 2012). They are similar to those reported for marine algae from low latitudes, which range between –18 and –22‰ ($\delta^{13}C_{org}$), 4-10 (C/N ratio) and 6-10‰ (δ^{15}N) (Fry and Sherr, 1984; Fischer, 1991; Meyers, 1994; Currin et al., 1995; Middelburg and Nieuwenhuize, 1998).

Due to the extensive cultivation of rice and other C_3 plants in the catchment, we suggest a dominance of terrestrial soil of C_3 plants in the riverine material that is intensively mixed with marine algae OM in the coastal sediments (Figure 4). The gradient of $\delta^{13}C_{org}$ and the C/N ratio in offshore direction point to an increasing portion of marine OM in offshore direction (Figure 5a, b). We ascribe this gradually changing mix of the OM sources to a rapidly near-shore settling of (terrestrial)

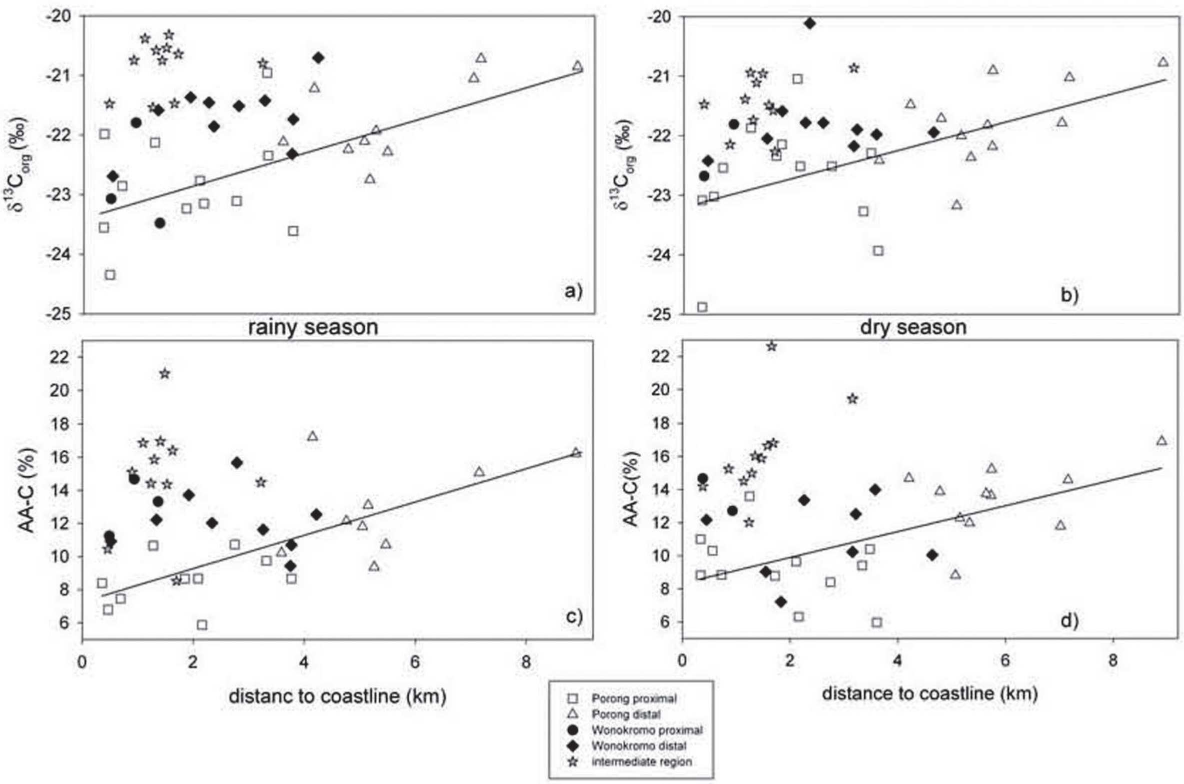

Figure 5: Stable carbon isotopic composition (a = rainy season/ b = dry season) and percentages of amino acid bound carbon (AA-C%) (c = rainy season/ d= dry season) of the surface sediments plotted against the distance to the coastline. Surface sediment sampling sites are subdivided into five regions: ☐ Porong proximal, Δ Porong distal, ● Wonokromo proximal, ◆ Wonokromo distal, ✶ intermediate region. (regression Porong estuary: Figure 5(a) $R^2 = 0.42$, $p < 0.01$; Figure 5(b) $R^2 = 0.33$, $p < 0.01$; Figure 5(c) $R^2 = 0.56$, $p < 0.01$ and Figure 5(d) $R^2 = 0.37$, $p < 0.01$)

particles, which occurs due to a pronounced bottom friction and fast deceleration of the river flow velocity at the shallow Porong river mouth (Hoekstra, 1989b). This is corroborated by a declining content of suspended matter in offshore direction (Jänen, 2012). The decrease in turbidity commonly leads to an increase in light penetration enabling phytoplankton to use the river nutrients more efficiently at the edge of the plume (Aller et al., 1985). Therefore, we infer a much higher input of freshly produced marine OM to the distant Porong sediments. By comparison, the contribution of planktonic material is most likely reduced in the sedimentary OM at the proximal stations based on the high turbidity and the strong nearshore settling of terrestrial organic and lithogenic material. The latter, might also have a quantitative dilution effect on the sedimentary OM of the proximal stations, where lower contents of OC and AA+HA were observed compared to the distal stations (Table 1). This suggestion is based on the fact that a stronger sedimentation of particles is usually accompanied by a higher settling of mineral particles from the river load (Rullkötter, 2006).

A high accumulation of terrestrial organic material can also affect the reactivity of sedimentary OM since degraded soil and vascular plant debris are commonly referred to be more refractory than freshly produced marine OM. They contain less labile compounds like amino acids (Cowie and Hedges, 1992; Jennerjahn et al., 1999; Unger et al., 2005). The percentages of amino-acid bound carbon and the AA/HA ratio of the estuarine sediments at the Porong estuary showed increasing values in offshore direction in both seasons (Table 1, Figure 5c, d) that indicate a higher reactivity with increasing distance from the coast. This could on the one hand result from a higher proportion of labile (more reactive) marine OM in the distal sediments. On the other hand, a less intense degradation of organic material in the slightly deeper offshore region than in the shallow highly turbid proximal waters is conceivable, based on the fact that the latter are characterized by strong resuspension processes (Hoekstra, 1989a) that are known to enhance the degradation of OM (Stahlberg et al., 2006).

Besides these spatial variations, the AA-C% and the AA/HA ratio were higher and the C/N ratio was lower during the dry compared to the rainy season while, in contrast, $\delta^{13}C_{org}$ displayed constant values (Table 1, Figure 2). It points to a more reactive sedimentary OM and a higher proportion of freshly produced phytoplankton during the dry season. This most likely resulted from the varying discharge of the Porong River that decreases from 264 $m^3 s^{-1}$ in the rainy to 50 $m^3 s^{-1}$ in the dry season (2003-2007) (Jasatirta Public Corporation, pers. comm.). Thus, the input of terrestrial material as well as the turbidity was much lower during that time and hence most likely promoted the phytoplankton production in the whole estuary. Furthermore, the reactivity as well as the $\delta^{13}C_{org}$ of riverine suspended matter sampled in the downstream part was higher in the dry season (Jänen, 2012). This indicates a higher production of riverine/estuarine phytoplankton that usually display ranges between –30 and –25‰ $\delta^{13}C_{org}$ (Goni et al., 2006). The transport of this material during less freshwater discharge and its mixing with marine phytoplankton might have led to the constant sedimentary $\delta^{13}C_{org}$ signal in the estuary. Seasonal variations were also observed for $\delta^{15}N$. The values at the proximal stations were higher in the rainy than in the dry season (Table 1, Figure 4c, d). It has been widely observed that, among other factors, $\delta^{15}N$ values increase with degradation (Saino and Hattori, 1980; Peterson and Fry, 1987). Based on the much higher discharge rates in the rainy season, it is conceivable that the seasonal increase at the proximal sites result from a stronger erosion and transport of older, degraded soil from the river bed and the subsequent deposition of this material near the river mouth.

Wonokromo Estuary

The sediments at the Wonokromo estuary showed ranges of $\delta^{13}C_{org}$, C/N ratio and $\delta^{15}N$ similar to those found at the Porong estuary and therefore likewise indicate a mixture of terrestrial and marine OM (Figure 4). During both seasons, sedimentary $\delta^{13}C_{org}$ at the Wonokromo estuary displayed higher values in the distal compared to the proximal sediments, which was more pronounced in the rainy season (Table 1). Although this trend was more distinct at the Porong estuary, we attribute this offshore directed increase to an enhanced proportion of marine OM in the more distal sediments. In contrast, the regional distribution of the OC and AA+HA contents as well as of the AA/HA-ratio and AA-C% differed strongly from the spatial patterns found at the Porong estuary. In the proximal sediments of the Wonokromo estuary we observed notably higher OC and AA+HA contents than in the distal sediments in the rainy season (Table 1, Figure 2a, b). This spatial difference most presumably resulted from a high deposition of riverine particulate organic matter (POM) directly in front of the river mouth, which rapidly decreases in offshore direction due to the small river plume.

A dilution effect, like observed at the Porong estuary, is unlikely in the proximal sediments due to lower TSM and higher POC contents in the Wonokromo compared to the Porong River (Jänen, 2012). In the dry season, the OC and AA+HA content were much lower at the proximal stations and a much lower portion of the <20 µm fraction was observed in these sediments (Table 1). A relationship between OM and grain size has been widely observed in marine sediments and is based on the adsorption of OM to inorganic particles (Keil et al., 1994; Mayer, 1994; Hedges and Keil, 1995). Thus, the loadings of OM increase with increasing surface area of inorganic particles and fine-grained sediments have a higher sorption capacity than coarse grained sediments. Therefore, we conclude that the lower OC and AA+HA contents in the dry season are related to the seasonal grain size difference of the proximal sediments (Table 1).

In contrast to the Porong estuary, the sedimentary AA/HA ratios and AA-C% did not increase with distance to the coast in the rainy season and even showed significantly higher values in the proximal region in the dry season (Table 1, Figure 2e, f). Moreover, the values were generally higher in the proximal sediments off the Wonokromo than off the Porong. These trends most likely result from a higher reactivity of suspended material in the Wonokromo compared to the Porong River, which was detected in both seasons. Furthermore, a strong increase of the reactivity from rainy to dry season was observed for the Wonokromo riverine material (Jänen, 2012). We basically ascribe this seasonal variation and the difference between both rivers to a varying production of fresh, autochthonous OM. The riverine primary production can be strongly controlled by the available amount of light and nutrients. Thus, the much higher TSM concentrations in the Porong River, which reduce the light availability, can result in a lower primary production.

It has been documented that inputs of urban wastewater and sewage can be extensively decomposed in warm tropical waters and the resultant high amounts of remineralized nutrients could trigger an autochthonous riverine primary production (Carreira et al., 2002). Additionally, the sewage material itself might contribute to a higher reactivity of the riverine material, as it is usually rich in proteins (Wu et al., 2007). Within the Brantas catchment, the urban area of Surabaya is the most rapidly industrialized region (Sudaryanti et al., 2001) where the maximum amount of pollution load with the river basin is generated (Ramu, 2004). Therefore, we conclude that a higher input of urban materials to the Wonokromo River lead to a higher primary production, and thus a higher reactivity, of the Wonokromo suspended material. The enhanced reactivity of riverine suspended matter in the dry season is most probably associated with a stronger primary production during the lower river discharge in the dry season (dry season: 20 $m^3\ s^{-1}$, rainy season: 47 $m^3\ s^{-1}$ 2003-2007) (Jasatirta Public Corporation, pers. comm.).

The transport of the freshly produced riverine material is limited to the very proximal regions of the Wonokromo estuary due to the overall lower discharge of the Wonokromo River and the small river plume. This input most probably enhanced the reactivity of the proximal sediments in both seasons. In the rainy season, it resulted in values equal to those at the distal stations that contained a higher proportion of labile marine OM. In the dry season, the deposition of the much more reactive riverine suspended matter result in strongly enhanced reactivities in the proximal sediments.

Intermediate Region

The overall lower C/N ratios as well as higher $\delta^{13}C_{org}$ values, AA+HA concentrations, percentages of AA-C and AA/HA ratios compared to the Wonokromo and Porong estuary (Table 1, Figure 4a, b; Figure 5) indicate a higher portion of marine OM in sediments from the intermediate region. Due to the distance to the main river mouths terrestrial OM can only be transported to the sediments at the intermediate region in a reduced amount via longitudinal coastal currents, by diffusive terrestrial runoff or via smaller drainage channels located along the coastline between the Wonokromo and the Porong river mouths. In the dry season, higher AA/HA ratios, AA-C%, AA+HA contents as well as slightly lower C/N ratios point to less terrestrial influence than observed in the rainy season (Table 1, Figure 2) because of lower precipitation and hence lower diffusive terrestrial input via the small draining channels.

Interestingly, $\delta^{13}C_{org}$ decreased from –20.8‰ in the rainy to –21.5‰ in the dry season although terrestrial input was lower. Suspended matter in the intermediate region also displayed lower $\delta^{13}C_{org}$ values in the dry season and the nutrient concentrations were lower (Jänen, 2012). Despite lower terrestrial runoff nearly constant, chlorophyll-*a* values throughout the year (Jänen, 2012) indicate little variation in primary production in the dry season. This suggests an increased nutrient recycling in the water column at that time. It has been found that the fractionation against the heavier $^{13}C_{org}$ increases with enhanced respiration (Degens et al., 1968), what implies an enrichment of $^{13}C_{org}$-depleted compounds and therefore a lighter isotopic planktonic signal within a more heterotrophic system. It is conceivable that the lower $\delta^{13}C_{org}$ values of suspended matter at the intermediate region in the dry season were caused by an enhanced heterotrophic cycling. Accordingly, seasonal variations of carbon isotope fractionation by plankton were probably responsible for the seasonal changes of the sedimentary $\delta^{13}C_{org}$. In the rainy season, $\delta^{15}N$ values were in the same range as in sediments at the Porong and Wonokromo estuary, but were higher in the dry season (Table 1, Figure 4c, d). $\delta^{15}N$ values vary with the strength of isotopic fractionation. They get lower under abundant nutrient supply due to the higher discrimination against the heavier isotope ^{15}N (Fry and Peterson, 1987). Therefore, we conclude that the seasonal changes of $\delta^{15}N$ can be ascribed to a lower isotopic fractionation during the reduced nutrient supply in the dry season (see above).

Organic Matter Degradation

The OC as well as the AA+HA contents strongly varied within the sediment cores and showed no significant trend with depth at the Wonokromo and Porong estuary, but a slight decrease in the intermediate region (Figure 3). Due to the fact that OM commonly adsorbs onto mineral surfaces (Hedges and Keil, 1995) and therefore OM contents are higher in fine grained sediments because of the higher mineral surface area, the fluctuations of OC and AA+HA contents might be ascribed to co-occurring high fluctuations of the grain-size as indicated by correlation of the grain size fraction <20 µm with the OC content of cores taken at the Wonokromo estuary and the intermediate region (W: $R^2 = 0.6$, $p < 0.05$, I: $R^2 = 0.7$, $p < 0.05$). This correlation diminished in the Porong cores ($R^2 = 0.3$, $p > 0.1$), what could be based on large amounts of discrete terrestrial plant debris deposited at the Porong estuary that can hamper grain-size measurements and indicate that adsorbtion is not the only control of OC in Porong sediments. AA-C% and AA/HA generally decrease with increasing degree of degradation (Cowie and Hedges, 1992; Haake et al., 1992). The AA-C% varied between 5.3 and 19.8% and the AA/HA ratio ranged from 2.9 to 10.4 (Figure 3). Both parameters showed basically the same trend of more constant values in the Porong cores and a stronger decline of values in the Wonokromo cores and the core taken in the intermediate region.

In accordance with the surface sediment data of the proximal subregions, the least reactive material was found in the upper 5 cm of the Porong sediment cores (Figure 3). The uppermost sediment layers in the cores taken at the Wonokromo estuary had a higher reactivity, mainly due to a higher reactivity of the settling riverine suspended matter. The core of the intermediate region showed a high reactivity of the upper sediment as well because of a higher portion of labile marine OM. While the reactivity decreased only slightly downcore in the Porong sediments, a strong decline of the OM reactivity was observed in the Wonokromo estuary. Thus, the degradation of OM appeared to be much stronger in the Wonokromo estuary, hence OM preservation, or burial efficiency (Betts and Holland, 1991; Reimers et al., 1992; Harnett et al., 1998), was higher at the Porong estuary.

Hartnett et al. (1998) discussed that the burial efficiency and preservation of OM depends on its contact time with oxygen ("oxygen exposure time"). Among other factors, primary production rate, bottom water oxygen content and the overall sedimentation rate are important in this respect. The latter controls the rate at which organic material moves out of the oxygenated upper sediment layers and therefore escapes the intense oxic respiration (Canfield, 1993; Hartnett et al., 1998; Henrichs and Reeburgh, 1987). It is conceivable that this influencing factor on the oxygen exposure time is much more pronounced in front of the Porong river mouth based on the huge river discharge difference between both rivers (mainly in the rainy season: Wonokromo = 47 $m^3 s^{-1}$, Porong = 264 $m^3 s^{-1}$). It is likely that the higher sediment accumulation at the Porong estuary results in a higher burial rate than in front of the Wonokromo river mouth which receives less material delivery. This implies that the OM had less time for degradation in the oxygenated upper sediment zone in the Porong estuary. Thus, a higher amount of OM could be preserved and buried in Porong sediments.

Another factor that most probably leads to the higher degradation intensity at the Wonokromo estuary is the higher OM reactivity of the surface sediment in this region. As the upper sediment layers contain a higher proportion of more labile material that breaks down easily, the degradation of OM could proceed faster and stronger than in the more refractory Porong sediments that are more resistant to decomposition. It has often been observed that initially less reactive OM is stored more efficiently than more labile material (Aller et al., 1985). In those cases other boundary conditions were similar, whereas in our study area different sedimentation patterns coincided with a different initial OM reactivity. Therefore, we cannot determine unambiguously which factor primarily causes the different OM preservation.

However, the importance of sedimentary OM reactivity for the intensity of degradation is discernable from seasonal changes occurring at the intermediate region core station. The lower AA/HA ratios in the upper sediment layers indicate a lower reactivity in the rainy season. This was possibly caused by a higher diffusive terrestrial runoff or outwelling of mangrove-derived OM in the high rainfall season (Dittmar et al., 2001). Apparently, the lower reactivity of the surficial sediment in the rainy season led to an overall lower degradation intensity and therefore a better preservation of the less reactive OM compared to the stronger decay of OM in the dry season, when OM reactivity of the uppermost sediment layer was much higher (Figure 3). These changes occurred independently of varying sedimentation rates and therefore point to the importance of OM reactivity as a control factor for OM decay rates at the Brantas estuary.

In addition to the differing burial rates and sedimentary OM reactivity, the activity of macrobenthos might also have influenced the extent of degradation in our study area. Macrobenthic activity generally could enhance the oxygen supply mainly due to porewater irrigation (Archer and Devol, 1992) and therefore increase the degradation of sedimentary OM. The establishment of a well-developed benthic community can be promoted by high fractions of labile organic material and can be impaired by strong material accumulations (Rhoads et al., 1985; Alongi, 1991). This indicates better settlement conditions at the Wonokromo estuary. Benthos data obtained for the Wonokromo and Porong estuary support this assumption as it pointed to a much stronger animal abundance off the Wonkromo (Yusli, University of Bogor, pers.comm). Therefore, we suggest a higher benthic activity and thus a stronger impact of this oxygen-biasing factor at the Wonokromo estuary and the intermediate region which likely have enhanced the OM degradation in these areas. We conclude that the high sedimentation rate of mineral and more refractory material at the Porong estuary and the higher reactivity of the OM at the Wonokromo estuary and in the intermediate region determined the regionally strongly different decline of OM reactivity.

Brantas Sedimentary Organic Matter in the Global Context

To put the characteristic of the sedimentary organic matter in our study area into a global context, we compare them with surface sediment data measured in other coastal environments (Table 2). $\delta^{13}C_{org}$ values are higher than values found in estuarine sediments of the Philippines and Vietnam that are characterized by extensive mangrove stands (Kennedy et al., 2004). $\delta^{13}C_{org}$ of mangroves plants range between –25 and –30‰ (Marguillier et al., 1997; Bouillon et al., 2008) and mangroves soil reveals values around –27‰ (Jennerjahn and Ittekot, 2002). The lower values at sites of the Philippines and Vietnam likely occur by a high input of leaves and soil OM from the mangrove forest, e.g. by outwelling (Dittmar et al., 2001). High inputs of mangrove material cannot be assumed at the Brantas estuary due to scarcely existing mangrove stands along the coastline which results from the strong decrease of the natural mangrove forest for the construction of aquaculture ponds (Römer-Seel, 2003). This and the high inputs from terrestrial OM from the hinterland might be the reason for the comparatively higher $\delta^{13}C_{org}$ values in our study area. In contrast, $\delta^{13}C_{org}$ and $\delta^{15}N$ values are lower compared to sediments of the Delaware estuary (Cifuentes et al., 1988) or the open estuary of the Kallada River, Indonesia (Jennerjahn et al., 2008), most likely a result of the predominantly planktonic origin of the organic matter reported for these estuaries in relation to the high proportions of terrestrial material found in the Brantas estuarine sediments.

Whereas a high primary production was detected in the Delaware estuary, the low amount of terrestrial material in the open estuary of the Kallada River has been ascribed to a high deposition of river-derived material in the upper parts of the estuary. At the Brantas estuary, a high phytoplankton production is not very likely due to the predominantly very high turbidity. Moreover, a strong settling of huge amounts of material transported before the river outlet like observed for the Kallada River are not very likely due to the much higher discharge of the Brantas River. Furthermore, a lower erodibility in the Kallada catchment compared to the Brantas catchment results in a relatively reduced transport of lithogenic terrestrial material leading to less dilution of the estuarine sediments by mineral material and therefore to comparatively higher organic contents in the Kallada estuary.

The $\delta^{13}C_{org}$ values of the coastal sediments in front of the Ayeyarwaddy River in the northern Andaman Sea (Ramaswamy et al., 2008) range in the same interval, or even slightly higher, as the Brantas estuarine sediments although samples were taken in a broader spatial setting (~50-250 km) where a higher input of marine material

Table 2: Organic carbon content (OC), C/N ratios, carbon and nitrogen isotopic composition ($\delta^{13}C_{org}$ and $\delta^{15}N$) and amino acid characteristics (AA+HA = total hydrolysable amino acids and hexosamine content, AA-C% = amino acid bound carbon, AA/HA ratio) in coastal sediments ("-" = no data; mean value in parentheses)

	Location	*OC [%]*	*C/N ratio*	*$\delta^{13}C_{org}$ [‰]*	*$\delta^{15}N$ [‰]*	*AA+HA [mg/g]*	*AA-C [%]*	*AA/HA ratio*
Indonesia[a]	Brantas River (open) estuary	0.27–1.47	7.9–16.5	–24.9–20.3	3.48–5.38	0.76–6.27	5.9–22.6	4.2–13.0
		(0.88)	(10.1)	(–22.0)	(4.4)	(3.1)	(12.5)	(6.9)
East Coast USA[b]	Delaware estuary	–	–	–21.4–20.0	7.1–8.2	–	–	–
India[c]	Kallada River (open) estuary	2.0	10.8	–20.9	6.6	–	–	–
Philippines Vietnam[d]	mangrove coast	2.1–16.0	23.43.3	–23.6–28.1	–	–	–	–
Thailand, Myanmar[e]	Andaman Sea	0.08–2.66	2.7–34.2	–26.4–20.5	3.2–6.5	–	–	–
Papua New Guinea[f]	Fily river delat	0.8–1.6	12–17	–26.5–25.8	–	–	–	–
East coast USA[g]	Potomac estuary	–	–	–	–	20.5*	–	–
Norway[h]	Bunnefjord	–	–	–	–	19.2	9.9	–
Arabian Sea[i]	Western Arabian Sea,	0.9–3.45	4.8–85	–	–	2.1–8.5	9.8–11.4	–
China[j]	Peari River estuary	–	–	–	–	0.9–2.9	11.7–16.2	–
Brazil[k]	shelf sediments	1.43	9.8	–	–	4.4	12.0	8.0
	slope sediment	1.09	7.7	–	–	3.4	10.4	5.7
Siberia[l]	Kara sea	1.1–2.1		–	–	0.35–8.27*	10.3–14.3	8.0–9.8

[a]This study; [b]Cifuentes et al., 1988; [c]Jennerjahn et al., 2008; [d]Kennedy et al., 2004; [e]Ramaswamy et al., 2008; [f]Goni et al., 2006; [g]Sigleo and Shultz, 1993; [h]Haugen and Lichtentaler, 1991; [i]Suthhof et al., 2000; [j]Chen et al., 2004; [k]Jennerjahn and Ittekkot, 1997 and [l]Unger et al., 2005. *Only AA [mg g^{-1}]

could be suggested. This is based on the higher water and material discharge of the Ayeyarwaddy River that transports the terrestrial material much further offshore than the comparatively lower outflow of the Brantas River. Nevertheless, increasing $\delta^{13}C_{org}$ values and the OC contents indicate an offshore directed increase of marine OM contributions like observed at the Brantas estuary. Another example of intense offshore material transport is the Fly River delta where sediments exhibited $\delta^{13}C_{org}$ values of –26.5 to –25.8‰ even 50 km off the coast (Goni et al., 2006). The strongly different spatial extension of the terrestrial signal between the three coastal areas furthermore indicates how fast the settling of riverine material takes place at the Brantas estuary due to its shallow morphology.

The concentrations of amino acids and hexosamines are lower than at the Potamac estuary (Sigleo and Shultz, 1993) that is characterized by a high primary production and seasonally anoxic conditions. This entails a higher deposition of freshly produced planktonic material and better preservation conditions for OM than in the turbid, well mixed Brantas estuary. The AA+HA contents, AA-C% and the AA/HA ratio of shelf and slope sediments along the Brazilian coast (Jennerjahn and Ittekkot, 1997), in the Western Arabian Sea and the Oman Basin (Suthhof et al., 2000), the Pearl River estuary (Chen et al., 2004) as well as in the Kara Sea (Unger et al., 2005) are similar to those at the Brantas estuary. Compared to the Brantas estuary, all these sampling sites are either located in a larger distance to the coast or in greater water depths. Therefore, it can be suggested that the sedimentary OM deposited in these areas was subject to a notable degradation during redistribution processes or while sinking through the water column or both (Suess, 1980; Thomsen et al., 2002).

The similarity of the AA+HA contents, the AA-C% and the AA/HA ratio of the shallow, nearshore sediments at the Brantas estuary and the sampling sites listed above suggest a relatively strong degradation of the sediments in our study area. This high OM decomposition most likely results from the intense resuspension processes that have been observed at the Brantas estuary. These are based on flat morphology of the extensive tidal and subtidal flats and the strong resuspensive action of tidal currents and waves (Hoekstra, 1989a) that are known to strongly enhance the degradation of OM in shallow coastal areas (Valeur et al., 1995; Pusceddo et al., 2005; Ståhlberg et al., 2006). Furthermore, the year-round high tropical temperature might additionally enhance the microbial-driven decomposition of OM. The data suggests that the "incinerator" function for OM attributed to tropical shelf systems in recent studies (Aller and Blair, 2006) already occurs close to shore in areas exposed to high tidal energy, like intertidal flats.

Summary and Conclusions

Terrestrial soil OM and marine derived organic material are the dominant sources of the sedimentary OM in the Brantas estuary. Their respective contributions in the sediments depend on the seasonal and regional variations of the river discharge that is disproportionally distributed between the two main river outlets, the Porong and the Wonokromo. As the reactivity of these autochthonous and allochthonous sources exhibits strong differences, its final balance determines the magnitude and spatial gradients of the OM reactivity in estuarine sediments. Sedimentary OM at the Porong estuary, which receives the highest terrestrial inputs in our study, was less reactive than sedimentary OM in the intermediate region, where freshly produced marine OM dominates over the terrestrial due to the distance to the river mouths. A reduced river and diffusive runoff in the dry season lead to an overall decreasing input of terrestrial material. Next to a different amount and dispersal of terrestrial material, its composition also has an impact on the estuarine sediments. This is most pronounced at the Wonokromo estuary that receives highly reactive OM in the dry season. The high reactivity of the riverine material is based on a high autochthonous production that is most probably intensified by the inputs and reminaralization of urban wastewaters. The spatial differences in sediment accumulation and OM reactivity of the surficial sediments result in a higher intensity of OM degradation at the Wonokromo estuary and the intermediate region compared to the Porong estuary. This implies a higher preservation and storage of OM in the area of the highest sedimentation, the Porong estuary.

On a global scale, the reactivity of the shallow Brantas estuarine sediments is similar to sediments located in greater depth or distance to the coast that have undergone considerable degradation during redistribution or sinking processes. This points to a substantial degradation of OM during intense resuspension processes in the turbid waters above the extensive tidal flats characterizing the Brantas estuary. The flat morphology also leads to a comparatively rapid settling and thus less extension of terrestrial input into coastal water in comparison with other estuarine systems.

It has been elucidated in our study that the quantity and quality of riverine material can largely influence estuarine sediment characteristics and the intensity of

OM degradation in the Brantas estuary. As the characteristics of land-derived material, in turn, generally depend on the processes in the catchment area, perturbations of the river discharge regime and other anthropogenic influences like the wastewater disposal or intensified agriculture can strongly influence the OM in estuarine sediments. Due to the fact that sediments have an important regulatory effect in coastal ecosystems (Jørgensen, 1996) our results indicate that changes of sedimentary characteristics and processes should be monitored with regard to increasing human impacts in the coastal zone.

Acknowledgements

The authors would especially like to thank Didik Agus Wijanarko and his team from the Environmental Agency of East Java for their invaluable support during the field campaigns in Surabaya. We also thank Seno Adi and the Agency for the Assessment and Application Technology (BPPT) in Jakarta for his support. Special thanks go to Dorothee Dasbach, Christina Staschok and Jana Holler for their support in the laboratory. This work was financially supported by the German Federal Ministry of Education and Research (BMBF) (Grant No. 03F0456C). Additional support from the Bremen International Graduate School for Marine Science (GLOMAR) which is funded by the German Research Foundation (DFG) is gratefully acknowledged.

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Asian Journal of Water, Environment and Pollution

Volume 9 Number 2 **April – June, 2012**

Contents