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DISTRIBUTION AND SOURCES OF PERSISTENT ORGANOCHLORINE PESTICIDES IN SEAWATER AND SEDIMENTS IN TRANSITIONAL SEASON FROM BANTEN BAY

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ABSTRACT

Studies on distribution of organochlorine pesticides in water column and sediments from Banten Bay were conducted during transitional season on April and October, 2001. The objectives of the study were to determine the concentration and distribution of total organochlorine pesticides in water column and sediments in transitional seasons, and to identify its sources. Our study showed the concentration of pesticides organochlorine in water column ranged between 0.366 and 4.391 ng/l with an average of 1.952 ng/l in April (location 1), and 0.357 and 2.998 ng/l with an average of 1.203 ng/l in October (location 2). And then pesticide concentration in sediment were ranged between 0.263 and 2.090 µg/l dry weight (dw) with an average of 1.281 µg/l in June (location 1), and 0.068 to 10.095 µg/l dw with an average of 1.775 µg/l in October (location 2). The result indicated different influence of the season on organochlorine pesticides concentration and there was fresh input of DDT in the study area.

Keywords *:* Organochlorine pesticide; Banten Bay; persistent organic pollutant

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INTRODUCTION

Banten is located in the western area of Java Island which has potential for industry, tourism, and conservation. Industry areas are focusing from Banten bay to Cilegon coast, tourism areas are from Cilegon coast to Anyer coast, and conservation areas are focusing in south areas of Banten around Ujung Kulon area (Manuputty, 2001). This condition will be possible discharge of such as contaminants to environment, e.g. organochlorine pesticides and then will influence to marine culture and fisherman activity in Banten bay waters.

Java Island as major population over 60% of Indonesian society usually uses organochlorine pesticides (OCPs) particularly for agricultural and human health direction (Sudaryanto *et al*., 2007). The residues of organochlorine still exist and detect in Indonesia coastal waters by several researcher although OCPs was banned by the late 1990s. This condition due to property of OCPs as POPs compounds are have characteristic such as slow rate of degradation in sediment, extremely stable and persist in the environment, high solubility in fatty substances and tissues, accumulate in organisms and food chains, toxic and chronic to humans and animals. And then, it is transported in the environment over long distances to places far from the points of release (Morner, *et al.*, 2002; Zitko, 2000). Besides that, OCPs fate and distribution in tropical environment water and air has influenced by weather (Tanabe, *et al.*, 1991). Temperature is limiting factor in behavior of OCPs long range-transport and its distributed in air, water, and sediment (Rui, *et al*., 2005).

Generally, this present study was collected water and sediment samples from Banten bay especially from the north coast of Banten bay to Anyer beach waters. The objective of the study were determining the concentration and distribution of total organochlorine pesticides in water column and sediments during transitional seasons, measuring

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influence of transitional season on OCPs concentration and identification sources of pesticides contamination for monitoring pesticides used.

MATERIALS AND METHODS

Sampling

Water column and sediments samples were collected around Banten coastal waters during two periods covering the transitional season II on April 2001 from Merak harbour to Anyer beach (Location 1) and transitional season I on October 2001 from Pujut cape to Banten beach (Location 2).

Water samples were taken from station near with coastline are st. 1, 5, 6, 9, but st. 21 far from coastline (April 2001/location 1) and station near with coastal line are st. 66, 67, 68, 69, 70, 71, 78 and 79 (October 2001/location 2). Sediment samples were sampled from st. 1, 3, 5, 7, 9, 21(April 2001/location 1), and station near with coastline (st. 66, 67, 68, 70, 78, 79, 85), station far from coastline (st. 73, 74, 80, 81, and 86) (October 2001/location 2) (**Fig. 1**).

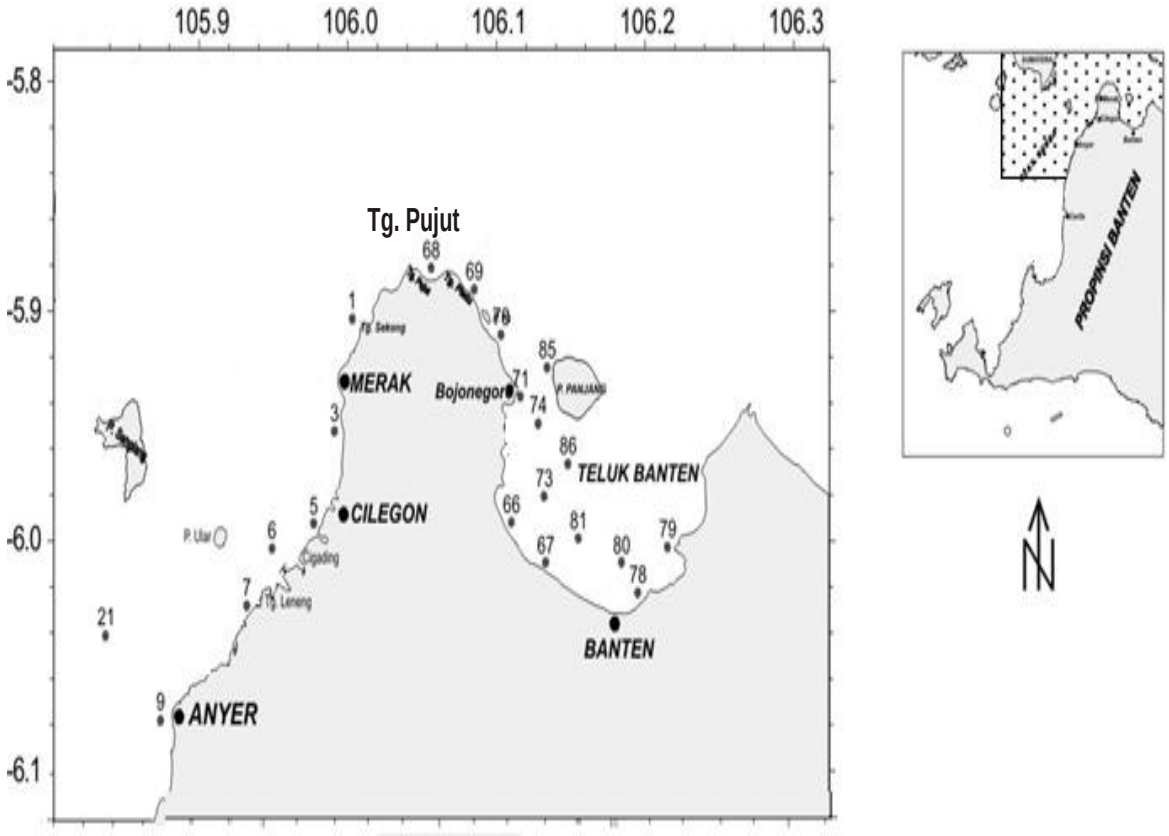

Fig. 1. Sampling location from Merak to Anyer (April 2001/location 1) and Banten beach to Pujut Cape (October 2001/location 2).

Each water samples were taken in 20 liters and directly put in stainless steel drums. Sediment samples were taken by grab sampler and directly put in glass bottle which were covered by aluminum foil. The next processes followed Holden and Marsden (1969), Grave and Gravenstuk (1975), Duinker and Hillebrand (1978). The water samples were filtered using GF/C filter paper in close system and using pressure N_2 gas UHP. The filtrate was extracted using hexane pro analyses in continuously liquid-liquid extractor using ISSABELLE extractor and concentrated to 1 ml using Kuderna Danish evaporator. After that, concentrated extracts was cleaned over 4 gram basic alumina (Alumina WB 5 basic from SIGMA) packed in glass micro-column, to remove interfering substances prior to gas chromatography (GC) analyses. Organochlorine pesticides was pre-separated by elution with n-

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hexane and 10 % diethyl ether in n-hexane over micro-column containing 4 gram silica (Silica powder Merck 7754) and then measured by Gas Chromatography with electron capture detection (ECD) Hp 5890 series II.

Sediment samples were dried in oven at temperature 50-60^{o}C and sub sample to determine concentration of water were dried in oven at temperature 105 ^{o}C. Organochlorines compound was measured as follows, the samples were mixed with Na_2 SO_4 anhydrous until a dry powder remains. After that, dry samples sediment was extracted by dichloromethane in continuous flow soxhlet extraction for 8-10 hours. Raw extract was evaporated in Kuderna Danish equipment to 1 ml. The next step processes was followed by water sample methods. The concentration of organochlorine pesticides was measured by Chromatography Gas HP 5890 series II equipped by Electron Capture Detector (ECD).

Specifications of Chromatography Gas HP 5890 series II / Electron Capture Detector (ECD) were as follows, capillary column used was Cp-Sil 8 CB type with 50 meters length, inner diameter 0.25 mm, outer diameter 0.39 mm and thick of film 0.12 um. The concentration of organochlorines in water was calculated in ng/l (ppt) and in sediment was calculated in µg/l (ppb).

RESULTS AND DISSCUSIONS

Distribution of OCPs in water column

The concentrations of organochlorine pesticides (OCPs) in water column from Merak harbour to Anyer beach (April 2001/location 1) and from Pujut cape to Panjang Island (October 2001/location 2) are given in **Table 1.** The total concentrations of OCPs in April 2001 and October 2001 were detected between 0.366 to 4.391 ng/l with an average of 1.952 ng/l and between 0.357 to 2.998 ng/l with an average of 1.164 ng/l, respectively.

Table 1. The concentrations of organochlorine pesticides in water column (ng/l) from location 1 (April 2001) and location 2 (October 2001)

Compounds	Location 1 (St. 1, 5, 6, 9, 21)		Location 2 (66, 67, 68, 69, 70, 71, 78, 79)	
	Ranges	Mean	Ranges	Mean
pp-DDT	0.042-0.121	0,019	nd-0.832	0.271
pp-DDD	nd-0.073	0,006	nd-0.045	0.006
pp-DDE	0.008-0.078	0,008	0.007-0.205	0.059
Dieldrine	nd-0.112	0,014	nd-0.036	0.005
Endrine	nd-0.06	0,003	nd-0.034	0.009
Aldrine	0.028-0.1	0,011	nd-0.017	0.004
Heptachlorepoxyde	0.005-3.681	0,157	nd-0.026	0.008
Heptachlor	0.007-0.142	0,010	nd-2.294	0.297
Endosulphan I	nd-0.092	0,006	nd-0.307	0.044
Endsulphan II	0.012-0.098	0,007	nd-0.213	0.029
α-BHC	0.008-0.072	0,007	nd-0.041	0.014
β- BHC	nd-0.171	0,018	0.052-0.163	0.078
γ- BHC	0.027-0.102	0,012	0.021-0.097	0.042
δ-BHC	0.017-0.119	0,015	nd-0.123	0.043
Metoxychlor	nd-0.693	0,092	nd-0.353	0.044
Endrine Aldehyde	nd-0.054	0,002	nd-0.058	0.016
Endrin Ketone	Na	Na	nd-0.036	0.014
Endosulphan sulphate	nd-0.022	0,002	nd-0.625	0.182
Total	0.366-4.391	1.952	0.357-2.998	1.164

Note: Nd = Not detected, Na = Not available data

Several major contaminants in the Banten bay from present study were heptachlorepoxide (0.157 ng/l) > metoxychlor (0.092 ng/l) > pp-DDT (0.019 ng/l) > β- BHC (0.018 ng/l) > δ-BHC (0.015 ng/l). However, widely discussions in other study about OCPs contaminant are

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DDTs and BHCs (Minh, *et al.*, 2007; Mwevura, *et al*., 2002; Mora, *et al*., 2004). As reported by Sudaryanto, *et al*., (2007) that major contaminant from several study in Indonesia environment are DDT and BHC. Moreover, behavior of DDT in human body has reduction time (half-life) is approximately 4 years (Bhuiyans, *et al*., 2008). Furthermore, present study shown that p,p'-DDT was detected approximately 50% in all station in both location 1 and location 2 except in station 78 (**Fig. 2**). As well as, p, p'-DDE always detected although less than 25 % in all station. However, p, p'-DDD only detected in station 1, 5, 6, 9, and 78, with higher concentration detected in station 78 approximately 75% (2.998 ng/l). Higher pp-DDE in station 78 indicated that condition of waters in this area were oxidative which mostly oxygen dissolved so pp-DDT likely degraded to pp-DDE (Muhayimana, *et al*., 2009). This condition could be high risk of human health, because pp-DDE as major ubiquitous environmental pollutants represents a significant risk to wildlife and human health due to persistence in the environment, more accumulative in marine biota and xenoestrogenic effects (Won, *et al*., 2009; Svobodova, *et al*., 2003).

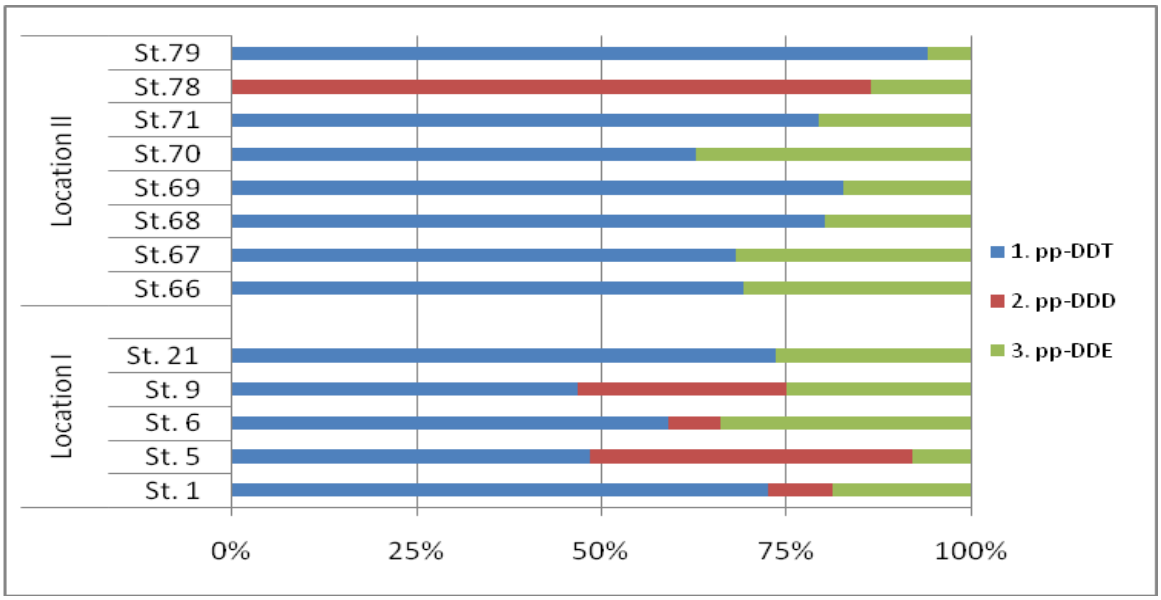

Fig. 2. Composition of DDTs in water column from location 1 and location 2.

As DDTs compound, BHC compound was still discussed and detected in environment. BHC is a isomer names of HCH which consist of 5 other isomer (α, β, γ, δ, and ε) (Ozer, 2005). Present study was detected α, β, γ, and δ-BHC in all station in location 1 and 2, except in station 71 (α-BHC), station 9 (β-BHC), and station 66, 67, 68, 78 (δ-BHC). Isomer compounds of BHC as mostly detect is β-BHC approximately 50 % (**Fig. 3**). Possible high risk to biota and human health because characteristic of β-BHC are higher bioconcentration, slower rate biodegradation and elimination (Yang, *et al.*, 2005). Moreover, as note by Sudaryanto, *et al*., (2011) that β-BHC could from isomerization processes of α-BHC and γ-BHC in environmental system.

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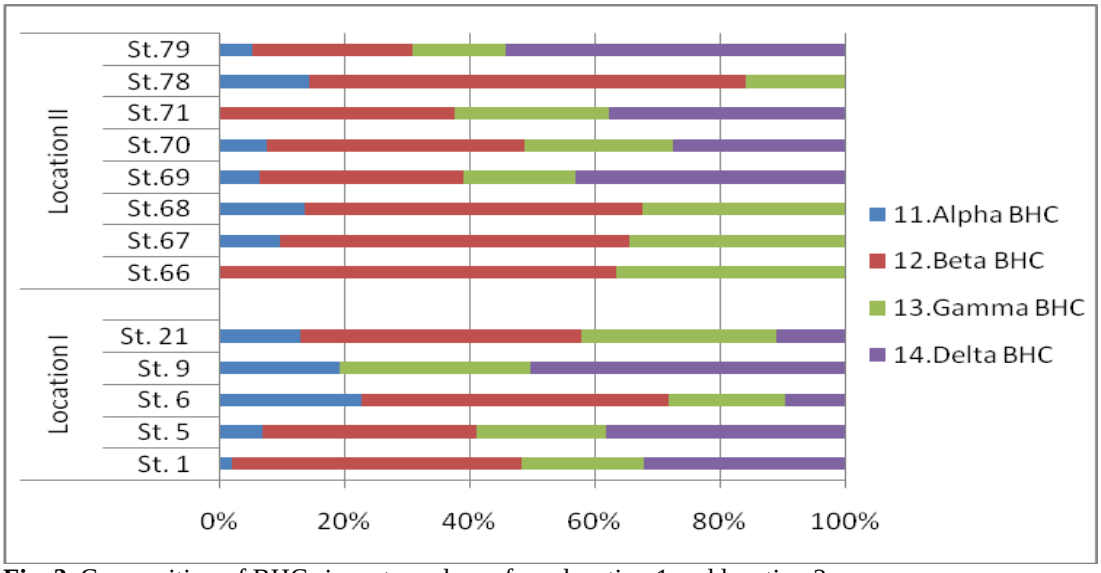

Fig. 3. Composition of BHCs in water column from location 1 and location 2.

Concentration of organochlorine pesticide in water from location 2 was smaller than location 1 (1.203 < 1.952 ng/l). Those conditions because influence of different transitional season. Location 1 influenced by transitional season one from rainy season in March to dry season in June, while location 2 influence by transitional season two from dry season in August to rainy season in November. So, condition of transitional one also influences by rainy season which mostly agricultural activity in Indonesia (Paramitha and Oginawati, 2010). Furthermore, high pesticides were entered into waters by runoff processes during rainy season from agricultural land and channels, and from urban city sewage sites (Ratnaningsih, *et al*., 2002). Besides that, OCPs concentration was increased because likely to be contributed by suspended particles, as the water samples were analyzed without being filtered (Won, *et al.*, 2009).

Compared with average concentration of organochlorine pesticide in other study, such as with Mouth rivers in east Sumatera (Siak-Riau, Kuala Tungkal-Jambi and Musi-Palembang) still lower (Munawir, 1997; Munawir, 1998; Hutagalung, *et al*., 1997) also with Way Kambas Mouth River (Munawir, 2001). Besides that, concentration of organochlorine pesticides was low compared with quality standard of the seawater from Minister of Environment decision no. 51/2004 which organochlorine pesticides concentration in marine waters not higher than 10 ng/l (KLH, 2004).

Distribution of Organochlorine Pesticides in Sediments

Investigation of organochlorine pesticides in sediments was conducted to record of contamination levels in the aquatic environment in Banten bay, especially in location 1 (Merak harbor to Anyer beach) and location 2 (Pujut cape to Panjang Island). The concentration of organochlorine pesticides from location 1 was detected between 0.263 to 2.090 µg/l dry weights (dw), with an average of 1.281 µg/l dw, while in location 2, total concentration of OCPs between 0.068 to 10.005 µg/l dw, with an average of 1.775 µg/l, respectively (**Table 2**).

The average of total concentration of OCPs in location 2 was higher than location 1 (1.775 > 1.281 µg/l). This condition because location 2 was influenced by industry activity in land and influenced of season wherein transitional seasons two after dry season has high temperature ranged from 29.7 – 30.7 ^{o}C with an average of 30.2 ^{o}C (Manuputty, 2001). Temperature has a limited factor for differences of the physicochemical and biochemical properties of several OCPs compound such as water solubility, vapor pressure, and bioavailable to degradation in natural (Rui, *et al*., 2005; Shiu, *et al*., 1997). Location 2 with high level of temperature was caused evaporation in surface water hence alter behavior of OCPs compound likely to transport

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to other location or link with particles and sink to surface sediment via leaching (Blenkisop *et al.*, 2008; Sudaryanto, *et al.*, 2007). Besides that, increases in salt concentration during high evaporation in dry season might be caused of increases the gas solubility such as OCPs compound in the seawater (Ozer 2005; Xie, *et al.*, 1997, Rodríguez-Liébana, *et al*., 2011). Other researcher reported that transport of pesticides to depth influence by climate characteristics (Blenkisop, *et al*., 2008).

Distribution of pp-DDT compound in sediment samples from location 1 and 2 was still detected over 60%, with higher composition between 90-100% are in station 1, 5, 9, 14 and station 79. Four stations have higher pp-DDT composition located in location 1 wherein influence by rainy season and there was indicated recent input of DDT contamination. DDT contamination might be carried from other location agricultural lands in Banten with rainfall and storm water.

Besides that, Java island as major user of DDT for controlling the disease vector in agricultural area or human disease by malaria and therein was formulated of DDT in Bogor (West Java) until late 1990s (Sudaryanto, *et al.*, 2007).

Table 2. Organochlorine pesticide concentration in the sediments samples (µg/l) from location 1 (April 2001) and location 2 (October 2001).

Compounds	Location 1 (St. 1, 3, 5, 7,9,14)		Location 2 (66, 67, 68, 70, 73, 74, 78, 79, 80, 81, 85, 86)	
	Range	Mean	Range	Mean
pp-DDT	nd-0.102	0.047	nd-0.234	0.046
pp-DDD	nd-0.047	0.008	nd-0.085	0.026
pp-DDE	nd-0.017	0.003	nd-0.76	0.018
Dieldrine	nd-0.111	0.027	Nd	0
Endrine	nd-0.058	0.021	nd-0.041	0.003
Aldrine	nd-0.116	0.025	nd-0.016	0.001
Heptachlorepoxyde	0.009-0.418	0.087	nd-0.023	0.004
Heptachlor	0.018-1.043	0.442	nd-9.816	1.043
Endosulphan I	0.006-0.111	0.025	0.028-0.537	0.237
Endsulphan II	0.017-0.054	0.045	nd-0.411	0.157
α-BHC	nd-0.072	0.029	nd-0.03	0.013
β- BHC	nd-0.235	0.091	nd-0.092	0.041
γ- BHC	0.011-0.091	0.041	nd-0.053	0.02
δ-BHC	0.008-0.133	0.04	nd-0.041	0.003
Metoxychlor	nd-0.784	0.315	nd-0.597	0.129
Endrine Aldehyde	Nd	0	nd-0.065	0.024
Endrin Ketone	Na	Na	nd-0.054	0.01
Endosulphan sulphate	0.01-0.036	0.025	nd-0.051	0.005
Total	0.263-1.970	1.27	0.064-10.055	1.779

Note: Nd = not detected, st. = station, Na = not available data

Distribution by product of DDT such as DDE and DDD not significant detected neither in location 1 nor location 2. However, pp-DDD was higher composition in station 78 approximately to 75% (**Fig. 4**). Dominance of pp-DDD in station 78 could probably less dissolve oxygen so degradation processes of DDT to DDD would be done (Qui, *et al*., 2010).

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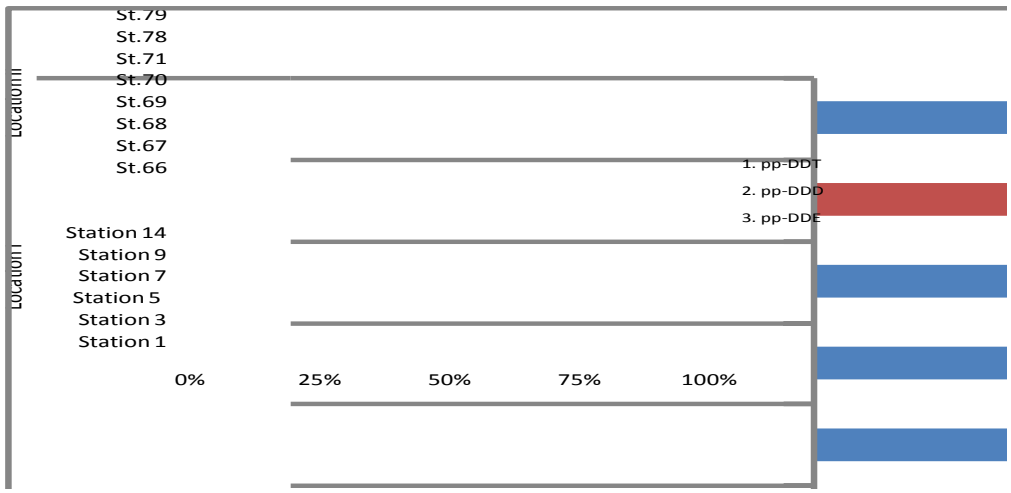

Fig.4. Composition of DDTs in sediments from location 1 and location 2

As for BHC, several isomers was detected in all sediments sample with higher composition is β-BHC. This condition similar with BHC's water composition that β-BHC was high composition until 50% (**Fig. 5**). This result suggest that β-BHC still stable in environment and have high risk for biota and human life because bioaccumulative of the isomers and a possible endocrine disruptor (Ozer, 2005). Besides that, β-BHC was most detected accumulating as metabolically stable isomer in organism tissue (biota and human) and sediments (Willet, *et al.*, 1998; Sun, *et al.*, 2010). Moreover, γ-BHC as presentation of lindane still detected in all station sediments sample although no high concentration detect and no more information usage of BHC in Indonesia, but several study was reported that BHC still detected in water and sediments (Sudaryanto *et al.*, 2007). That is could probably sink from atmosphere as long range transport property of BHC from other county, such as from Northeast Asian region as large amounts user of these chemicals (Won, *et al.*, 2009).

Compared with other location in Indonesia waters, range levels of total OCPs sediments in location 1 and II from Banten Bay were lower than other location in Indonesia waters such as Way Kambas river mouth in Lampung estuarine (1.06 to 2.69 ppb), Way Sekampung in Lampung estuarine (2.11 to 2.19 ppb), Siak Estuarine, Riau (1.48 to 10.76 ppb), and Jakarta Bay (0.44 to 4.34 ppb) (Munawir, 1997; Munawir, 2001; Razak and Munawir, 1994).

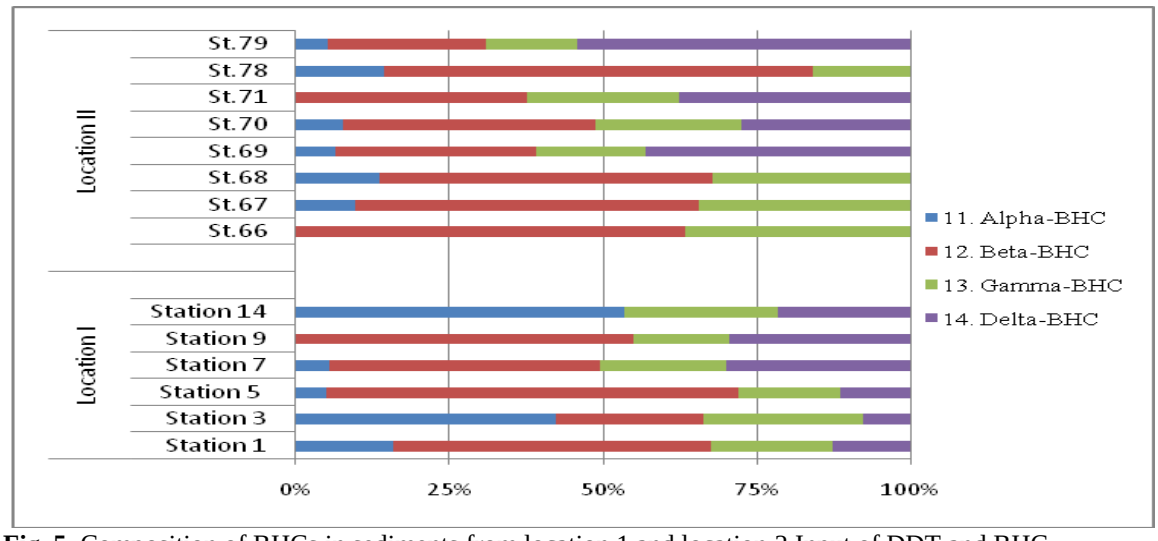

Fig. 5. Composition of BHCs in sediments from location 1 and location 2 Input of DDT and BHC compounds in water and sediment samples

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The ratios of some organochlorine pesticide compounds were applied to identify the old and new of its contaminated in the environment (Muhayimana, *et al.*, 2009; Qui, *et al.*, 2010; Guo, *et al.*, 2008). Especially if there are usually detected of those compounds. The OCP compounds were still detected in all station samples are DDT and BHC. That condition similar as note by Arai (2009) Indonesia uses a large quantity of OCPs, particularly DDT for increasing agricultural production as well as for vector-borne disease eradication program. Moreover, various ratios of DDT and BHC were used to identify the sources of contaminant in Banten bay.

Composition of commercial DDT as one of the oldest group of the OCPs has contain of 75% p,p'-DDT, 15% o,p'-DDT, 5% p,p'-DDE, <0.5% p,p'-DDD, <0.5 o,p'-DDE and <0.5% of unidentified compounds (WHO, 1979). However, environmental condition might degraded of DDT to its derivatives, such as DDE (aerobic condition), and DDD (anaerobic condition) (Qui, *et al.*, 2010). Therefore, ratio DDT and its derivatives such as DDD/DDE and DDT/ (DDD+DDE) were used to identify the source of DDT contaminant and condition of the waters. Ratio of DDD/DDE > 1 indicates condition of waters was anaerobic so DDD compound was higher and ratio less than 1 indicated the waters was contaminated by DDE and waters condition was contain more oxygen (Sun, *et al.*, 2010). Ratio of DDT/(DDD+DDE) was applied to know age of DDT contaminated, ratio score more than 0.5 indicated recent input of DDT, while those less than 0.5 indicated long term weathering of DDT or old input of DDT (Wasswa, *et al.*, 2011).

Assess input of DDT contamination in this study was showed in **Fig. 6**. DDT input was higher in locations, location 1 and 2, except st. 78 and st. 79 in location 2. Those mean there is new input of DDT from agricultural activity in land. However, DDT source not only from agricultural activity as note by Li and Macdonald (2005) in Guo, *et al.*, (2008) that antifouling paint has high concentration of pp-DDT. Ratio of DDD/DDE was high in st. 9 (1.1), st. 5 (5.5), and st. 78 (6.4 in water and sediment samples. (**Fig.6.)** Guo, *et al.*, (2008) was reported that DDT would be deoxidized to DDD compounds under anaerobic or reduction environment after that would be oxidized to DDE in aerobic or oxidation environment. Besides that, the higher ratios of DDD/DDE in seawater and sediment samples in that station may partly stem from direct inputs of p,p-DDD (Guo, *et al.*, 2008).

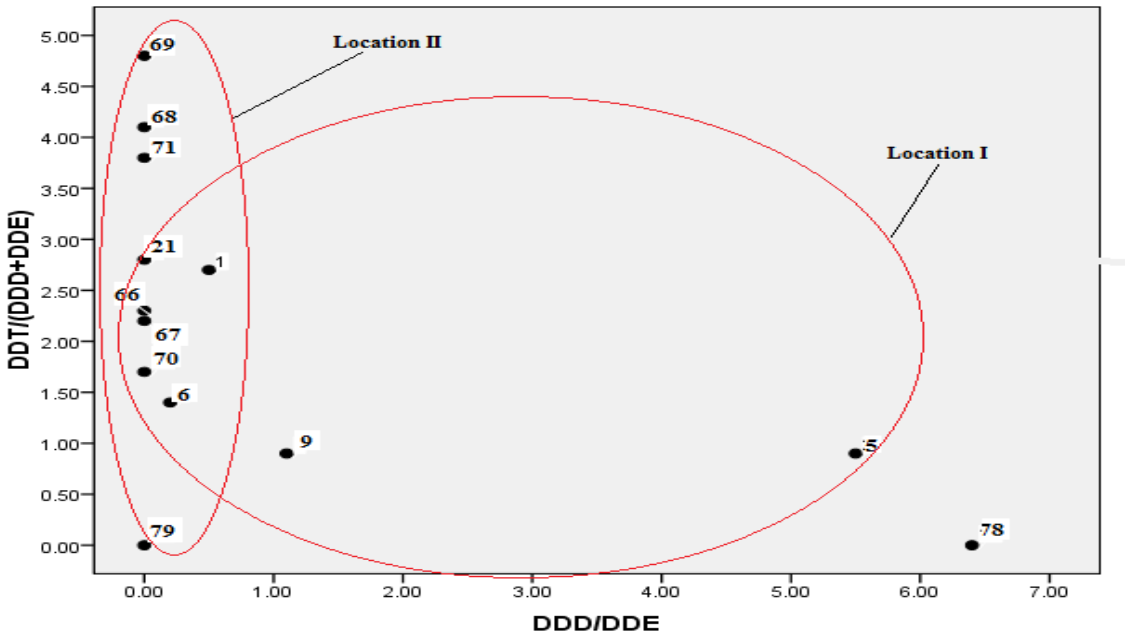

Fig. 6. Ratios of DDD/DDE and DDT/(DDD+DDE) in water of different location

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Isomer compound of HCH is BHC that it was used as two type : technical-BHC that has ratios of alpha-/gamma-BCH between 3 to 7 or has contain composition of α-(60-70%), β-(5-12%), γ- (10-15%), δ-(6-10%) and ε-(3-4%) and lindane has γ-BHC composition > 99% (Guo, *et al.*, 2008; Ozer, 2005). Each of isomer has different property of chemical and physical (Willett, *et al*. 1998). However, only α-BHC and γ-BHC has been used as tool to identify the possible source of BHC, because α-BHC as high composition in technical BHC and γ-BHC in lindane. That is possible to differentiation of source contaminant of BHC. High ratio of α-/γ-BHC indicates the input of technical-BHCs, while low ratio points to the dominant use of lindane (Guo, *et al*., 2008). The ratio of α - to γ –BCH showed between 3 and 7 for fresh input of technical BHC (Yang, *et al.*, 2008) and close or <1 ratios score for lindane source (Willet, *et al*., 1998) However, BHC source may from other source because of their properties (α-BCH) has long-range transport in atmospheric or re-cycling of technical BHC. This source showed by high ratio of α/*γ-B*CH > 7 (Willet, *et al.*, 1998).

Based on that criteria, ratios of α-/γ-BHC in location 1 have ranged between 0.1 to 1.2 and in location 2 ranged between 0.0 to 0.9, while ratio α-/γ-BHC in sediments samples showed that in location 1 (0.0 to 2.1) and location 2 (not available to 1.5). Lower ratios of α-/γ-BHC have indicated possible contaminant of lindane. However, γ-BHC as high contain compound in lindane has potency to be transformed into other isomers in the environment such as into α-BHC. Moreover, α-BHC has most carcinogenic activity and has been classified along with technical-grade HCH as a Group B2 probable human carcinogen by the U.S. EPA (Walker, *et al*., 1999).

CONCLUSION

Concentration of organochlorine pesticides in water column and sediments of Banten bay were below of Quality standard of seawater from Environmental Ministers of Indonesia in 2004 year. Distribution of OCPs were higher in location 1 (Merak harbour to Anyer beach) with average of 1.952 ng/l, and then followed by location 2 (Pujut cape to Banten beach) with average of 0.114 ng/l, while in the sediments samples, the highest average of organochlorine pesticides concentration was in location 2 with an average of 1.281 µg/l dw, followed by location 1 with averaged of 0.330 µg/l dw. Both locations were direct input of new DDT contamination in water column and sediments.

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