

RESIDU ALDRIN DAN ENDOSULFAN PADA AIR SUNGAI DAN LAHAN PERTANIAN DI DAERAH ALIRAN SUNGAI CITARUM TENGAH, KABUPATEN CIANJUR

ALDRIN AND ENDOSULFAN RESIDUES IN RIVER WATER AND AGRICULTURAL LAND OF THE MIDDLE CITARUM WATERSHED, CIANJUR DISTRICT

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Abstract: *The use of fertilizers and pesticides excessively and continuously will give a negative impact in the water and soil, the contamination of residual nitrate and pesticide. The organochlorine is contaminant that it had persistent, bioaccumulation and toxic to humans, animal and others. Organochlorine is not reactive, stable, very high fat solubility, and low degradation. This study aimed to obtain the data and the distribution of residual Persistent Organic Pollutants (POPs, aldrin and endosulfan) in river water and agricultural land of the middle Citarum Watershed, Cianjur District. This study was conducted in 2011, with a field survey method. Determination of the location of the soil samples is done with a modified grid system. One composite soil sample represents a homogeneous area of 50-100 ha. The results of this study showed detectable pesticide residues of endosulfan and aldrin in river water respectively from 0.001 to 0.005 and 0.003 to 0.043 mg/L and in the soil, respectively from 0.001 to 0.027 and 0.004 to 0.039 ppm. Based on 21 soil samples were analyzed, detected 43% have exceeded the Maximum Residue Limit (MRL) of endosulfan and 14% has exceeded the MRL of aldrin.*

Keywords: *Citarum watershed, POPs, contamination, and MRL.*

Abstrak: *Penggunaan pupuk dan pestisida secara berlebihan dan terus menerus akan memberikan dampak negatif terhadap air dan tanah berupa pencemaran residu nitrat dan pestisida. Organoklorin adalah kontaminan yang mempunyai sifat persisten, bioakumulasi dan beracun bagi manusia, hewan dan lain-lain. Organoklorin tidak reaktif, stabil, kelarutan dalam lemak sangat tinggi dan degradasi rendah. Penelitian ini bertujuan untuk memperoleh data dan distribusi residu Persistent Organic Pollutants (POPs, aldrin dan endosulfan) dalam air sungai dan lahan pertanian di DAS Citarum Tengah, Kabupaten Cianjur. Penelitian ini dilakukan pada tahun 2011 dengan metode survei lapangan. Penentuan lokasi sampel tanah dilakukan dengan sistem grid yang dimodifikasi. Satu sampel tanah merupakan komposit dari daerah homogen 50-100 ha. Hasil penelitian ini menunjukkan terdeteksi residu pestisida endosulfan dan aldrin dalam air sungai masing-masing 0,001-0,005 dan 0,003-0,043 mg/L dan di dalam tanah, masing-masing 0,001-0,027 dan 0,004-0,039 ppm. Berdasarkan 21 sampel tanah yang dianalisis, terdeteksi 43% telah melampaui Batas Maksimum Residu (BMR) endosulfan dan 14% telah melampaui BMR aldrin.*

Kata kunci: *DAS Citarum, POPs, kontaminasi, dan BMR.*

INTRODUCTION

Watershed is generally defined as the area bounded by a stretch of barrier topography (ridge) which receive and collect rain water, sediment and nutrients as well as pass it through the river and out into the main river to the sea or lake Linsley (1980). The Citarum river is divided

into the upstream (from water source of the Citarum river to the Saguling reservoir), the midstream (from the Saguling reservoir to the Jatiluhur reservoir) and downstream (from the Jatiluhur reservoir to the Karawang beach) (Paimin, 2006).

Improvement of critical land degradation showed that the problems associated with environmental degradation can't be solved completely. Negative impacts on the watershed is very detrimental to human life, such as floods, droughts, erosion, sedimentation, decreased soil fertility, decreased agricultural production, agricultural land and water pollution and other effects (Prasetyo, 2006).

Every year about 1000-1500 new chemicals are produced. Almost all of these produce are organic compounds. Many of them have potential as a pollutant or contaminant. If organic compounds can undergo transformation, which gradually turned into the final compounds are relatively harmless, is not the case with organochlorine compounds. Organochlorine compounds naturally will continue to be in nature, because it does not change (persistent), so it holds the potential poisoning of the latent (Solomon and Stigliani, 1995). Aldrin is a chlorinated derivative of naphthalene closely related to dieldrin. It was the recommended active ingredients for control of root feeding larvae during this period. Endosulfan is a chlorinated hydrocarbon used as a broad spectrum contact insect/acaricide. It is registered for use in commercial agricultural settings on a wide variety of vegetables, fruits, cereal grains, cotton and ornamental plants (USEPA, 2002).

Organochlorines are grouped into 3, namely: dichlorodifenil etane (eg, DDT, DDD, portan, metocychlor, and metiochlor), cyclodiene (example: aldrin, dieldrin, heptachlor, chlordane, and endosulfan), and cyclohexane chlorinated benzenes (eg, HCB, HCH, and lindane). Organochlorines are the main pollutants in Persistent Organic Pollutant group that are questioned in the world due to its chronic toxic, persistent and bioaccumulative characteristics

(Zhou *et al.*, 2006). For more than 40 years, organochlorines are still found in the environment and biota, and globally distributed including remote areas where organochlorines have never been used. Since late 1990, all kinds of organochlorine insecticides have been banned in Indonesia. However, because the price is cheap, easy to use, and effectively eradicate the pest, then some kind of organochlorines such as DDT is still used in Indonesia, as well as lack of firmness applicable laws and regulations (Sudaryanto *et al.*, 2007). Although organochlorine pesticides were already prohibited for agricultural purposes (Untung, 1999), however some studies indicate that these pesticides are still used by the farmers in the Citarum watershed. (Rochmanti, 2009) revealed that aldrin, dieldrin and endrin were detected in spray samples collected from farmers with concentration 1800 µg/L; 9.6 µg/L; and 8.5µg/L respectively. Some of pesticide shops still had the stock of endosulfan and some farmer still used it for dry crops (Pratama, 2009; Rahmawati *et al.*, 2010; Rochmanti, 2009).

In the upper of the Citarum watershed in the Bandung district, especially sub district of Pacet and Kertasari, organochlorine insecticide residues have been found in the compounds of heptaklor, aldrin, dieldrin, and DDT in the water channel, respectively 0.133-1.67; 0.161-0.56; 0.171-1.791 and 0.2-1.794 ppb and in the soil, respectively 0.3-25.1; 0.2-1.1; 0.6-1.8 and 0.3-3.0 ppb (Ramadhani and Oginawati, 2009).

Data of organochlorine insecticide residues in river water and agricultural land is limited and partial nature, therefore it is necessary to study. This study aims to determine the contents of organochlorine aldrin and endosulfan in river water and agricultural land of the middle Citarum watershed.

METHOD

The experiment was conducted in the Middle Citarum Watershed of the Cianjur district in 2011. The study used a survey method, which begins with the exploration of data from relevant agencies. In the implementation of the field survey, the type of samples taken were samples of water and soil. Water samples were taken from the Citarum river and theb Cirata reservoir, while the soil samples taken from paddy soil.

In soil sampling, map unit boundaries delineated in the field with ArcGIS programs are based on land slope. Land flat (slope less than 3%) of the sampling point may represent an area of 50-100 acres, and land with a slope more than 3% of the sampling points representing an area of 50 hectares (Hazelton and Murphy, 2007).

One soil sample consisted of 10 individual samples (sub-samples). Each sub-sample was taken at a distance of 25-50 m. soil

sampel was taken on the top soil (20 cm) by using a drill ground. 10 sub-samples put into a bucket and mixed until homogeneous, and then 0.5-1 kg taken and put into a plastic bag.

Extraction of water samples:

Examples of 100 ml filtered water put into a round flask of 500 ml, plus 30 ml of a solvent mixture of n-hexane and dichloromethane 90:10, shaken and allowed to stand for 3 minutes to form 2 layers (n-hexane at the top and water at the below). Water samples collected in erlenmeyer, n-hexane is collected in a round flask. Repeated extraction with water to re-enter example erlenmeyer separator funnel and added to 30 ml of n-hexane, shaken 3 minutes and allowed to stand strong.

Bottom layer discarded, upper layer is collected in a round flask. Extract was evaporated and purified in a chromatography column filled with Florisil and anhydrous sodium sulfate while eluted with 50 ml of n-hexane solvent. Examples evaporated again to ± 1 ml, then rinsed flask gradually acetone, collected in a test tube to a volume of 10 ml. Examples of ready injected to gas chromatography. After the

extraction process is complete, each sample is injected into the gas chromatography to identify aldrin and endosulfan.

Extraction of soil samples

Soil samples were dried, finely ground and sieved with a 2 mm sieve. A total of 25 g of sample in a tube inserted into a vertical shaker and shaker, add 100 ml acetone, and allowed to stand for 20 minutes. The results are filtered through Buckner column and plus cellit, then evaporated with a rotary evaporator and purified in a chromatography column containing florisil and anhydrous sodium sulfate while eluted with 50 ml of n-hexane solvent, housed in a round flask.

The result is evaporated again to ± 1 ml, then rinsed flask gradually acetone, collected in a test tube to a volume of 10 ml. Examples ready to be injected into the gas chromatograph. After the extraction process is complete, each sample was injected into a gas chromatograph instrument for identifying additional POPs: Endosulfan and Aldrin. Soil samples were also analyzed pH, CEC, organic C and soil texture.

DISCUSSION

General description of the Middle Citarum watershed

Topography of the the middle Citarum watershed, Cianjur district mostly mountainous, hilly-hill and partly lowland, with a height of 0-2,962 meters above sea level. Rainfall per year on average 1,000 to 4,000 mm and the number of rainy days on average 150 per year. With a

tropical climate, it makes natural conditions Cianjur fertile and contains a diversity of natural resources potential as the basis of development and potential capital investments promising. Agricultural lands and horticultural crops, livestock, fisheries and plantations is the source of life for the community (Distan Cianjur, 2010).

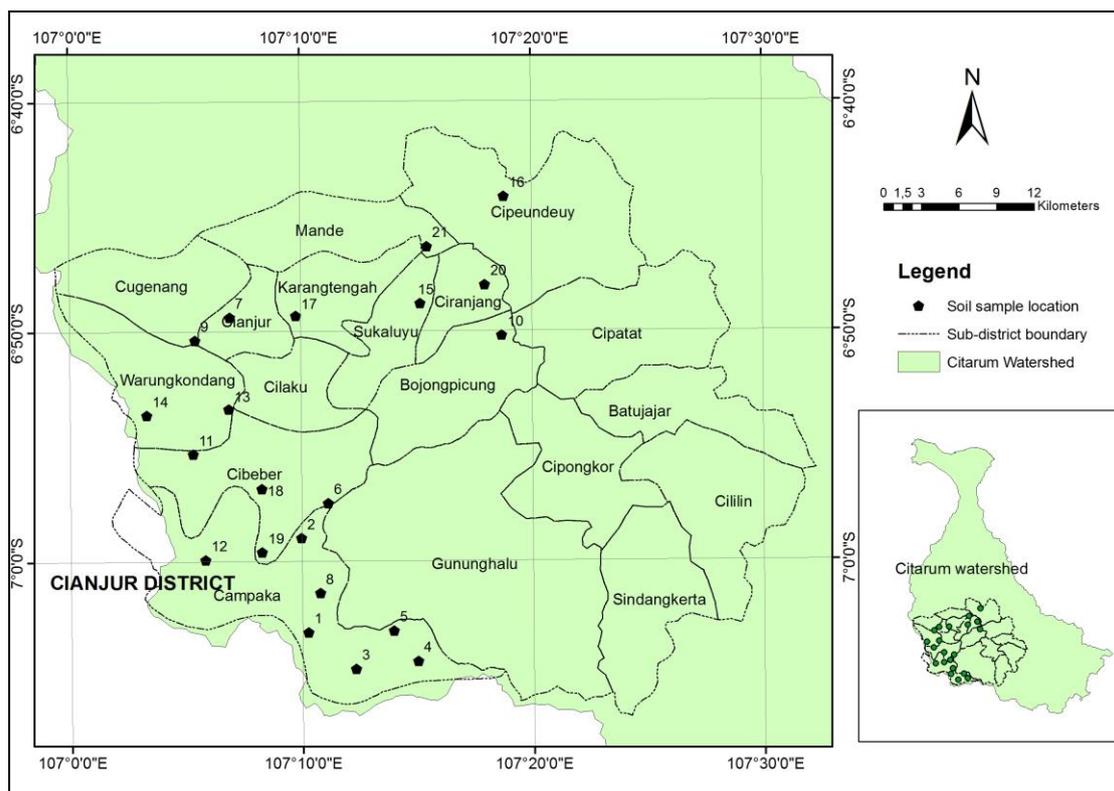


Figure 1. Soil sample location.

River Water Quality of The Middle Citarum Watershed

The physical properties of the middle Citarum river water still meets the appropriate water quality requirements under Government Regulation No. 8, 2001. Physical properties

measured were temperature, Total Dissolved Solids (TDS) and Total Suspended Solid (TSS). Water temperature in the range of 28.6 to 31.9 °C, TDS and TSS values respectively in the range of 106-176 and 2-20 mg/L (table 1).

Table 1. Physical properties of water from the Citarum watershed Central district of Cianjur 2011.

No.	Location	Temp (°C)	TDS (mg/L)	TSS
1	Toc Cisokan river downstream	28,6	116	2
2	Toc Cibalagung river downstream	28,9	124	4
3	Toc Cikundul river downstream	29,8	119	6
4	Cirata Reservoir	31,9	106	5
5	Upper middle Citarum River	30,8	176	3
6	Bottom middle Citarum River	31,7	152	20
Coefficient of variance (%)		4,63	20,02	100,24
Range		28,6-31,9	106-176	2-20
Average ± SD		30,28±1,40	132,1±26,4	6,67±6,68
Maximum limits*			2000	400

Note: * = (government regulations No.82 Th.2001 for irrigate crop); n = 6
Toc = Tributary of citarum

The Chemical properties of Middle Citarum river water is observed pH, DO, COD and CEC. water pH in the range of 6.6-7.2°C, the value of the DO, COD and CEC respectively in the range of 5.07-5.10 mg/L; 8-31 mg/L and 211-

352 µmhos/cm (table 2). Thus, the middle Citarum river water has chemical properties in accordance with the requirements of river water quality based on Regulation No. 08 in 2001 to irrigate crops.

Endosulfan was detected at all locations, while aldrin was detected 2 of 6 locations of water sampling locations (table 2). The same thing was reported by Somah and Ibrahim (2009), In Pattani River the longest river in the south of Thailand and Saiburi river, almost all locations (10 points) detected residues of endosulfan in water, while aldrin detected only 2 points. This is due to there are still people using akodan the active ingredient endosulfan is used

to catch fish in the river. The presence of POPs in the river water caused by runoff water flowing on the surface of the ground have the energy to scrape the surface soil layer, so that organochlorines can be carried over to the receiving water bodies such as rivers, and contaminating aquatic ecosystems including fish. Volatilization of agricultural land also contributed to the presence of pesticides in water bodies. (Bakre *et al.*, 2005).

Table 2. Chemical properties of water from the Citarum watershed Central district of Cianjur 2011.

No	Location	pH	DO	COD	EC	Endosulfan	Aldrin
			(mg/L)		$\mu\text{mhos/cm}$	(mg/L)	
1	Toc Cisokan river downstream	7,2	7,59	8	232	0.001	ND
2	Toc Cibalagung river downstream	6,95	7,61	10	245	0.005	ND
3	Toc Cikundul river downstream	6,78	6,20	15	238	0.003	0.003
4	Cirata Reservoir	6,84	5,07	15	211	0.003	0.043
5	Upper middle Citarum River	6,67	7,90	31	352	0.001	ND
6	Bottom middle Citarum River	6,78	5,10	8	331	0.003	ND
Coeff of variance (%)		2,70	19,74	59,92	21,74	62,57	
Range		6,6-7,2	5,07-7,9	8-31	211-352	0.001-0.005	
Average \pm SD		6,8 \pm 0,18	5,5 \pm 1,2	14,5 \pm 6,6	268,1 \pm 58,3	0.003 \pm 0.001	
Maximum limits*		5-9	0	100	2250	(-)	(-)
Maximum limits**						0,00002	0,0007

Note: * = (Government regulations. No.82 Th.2001 for irrigate crop);

** = Soil And Groundwater Remediation GUIDELINES Alberta Environment, Canada, 2009

$n = 6$; Toc = Tributary of citarum; ND = not detected

Soil Properties of Agricultural

The results of analysis of soil samples in the middle Citarum watershed based assessment criteria of soil chemical properties Soil and

Agro-climate Research Center, 1983, dusty clay texture with sand ratio of 13.41%, 35.56% and 51.03% clay dust. Tends to slightly acid soil with organic C content and high CEC (table 3).

Table 3. Description of soil properties the Citarum watershed middle Cianjur.

Description statistics	of pH	CEC	C-org	Texture		
		Cmol(+)/kg	%	Sand	Dust	Clay
Maximum	7,06	40,96	3,70	22,91	42,46	70,32
Minimum	4,61	16,97	1,84	5,46	24,22	38,42
Average	6,26	25,77	2,77	13,41	35,56	51,03
Stad Dev	0,49	7,00	0,53	6,24	5,03	8,63
Numb of samples	21	21	21	12	12	12

Endosulfan was detected in all sampling sites, with a minimum concentration of 0.001 ppm and 0.027 ppm maximum concentration. While aldrin was detected 52.17% with a minimum concentration of 0,004 ppm and a maximum concentration of 0,039 ppm respectively (table 4). It is suspected endosulfan

is still used by the majority of farmers in the field with the trademark Akodan (the active ingredient endosulfan) even though the book is not already registered pesticide in Indonesia (Directorate General of Agricultural Infrastructure, 2012) and has been banned since 1996.

Table 4. The results of statistical analysis of description Endosulfan and Aldrin the soil in the middle Citarum watershed Cianjur 2011.

Value of statistical description	Organochlorines POPs	
	Endsulfan	Aldrin
Count	21	10
Maximum (ppm)	0.027	0,039
Minimum (ppm)	0.001	0,004
Average (ppm)	0.008	0,021
Frequency detected (%)	100	52,17
Satd Dev	0,007	0,012
CV (%)	92.952	58,192
Varian	5.996E-05	0,0001
VMR	0,00719	0,00715

*VMR : Varian Mean Ratio, if the value of VMR > 1 the data clumped but if <1 the data spread.

The existence of residues of endosulfan and aldrin in the soil may be derived from the use of previous years. POPs insecticides generally decomposes very slowly in the soil, air, water and living organisms, and settled in the environment for a long time (Gorman & Tynan, 2003). Kusnaedi (2001) in Sinulangga (2006) reported that the persistence of aldrin resistance in the ground long enough, namely 14 years and after application of pesticide residues remaining in the soil was 40%.

Distribution of Endosulfan in the soil

Endosulfan detected in all locations of 21 sampling location, 43% already exceeds the Maximum Residue Limit (MRL) (figure 2). MRL in soil according to Alberta Environment, Canada (2009), endosulfan contamination

(MRL=0.0085 ppm). It is suspected endosulfan is used by most farmers in the Cianjur district. Based on field survey found that farmers still use Akodan (the active ingredient endosulfan) even though the book is not already registered pesticide Indonesia (Directorate of Infrastructure, 2012) and has been banned since 1996.

This compound is still used by farmers in the field, although in 2011 it was decided the Stockholm Convention banning the use of endosulfan in 2012. Due to its potential to evaporate and migrate long distances, causing endosulfan as the most widespread pollutants in the world. Endosulfan can be found extensively in water, soil and air. Endosulfan affects the environment through aquatic and food chains whose effects are very harmful to the environment.

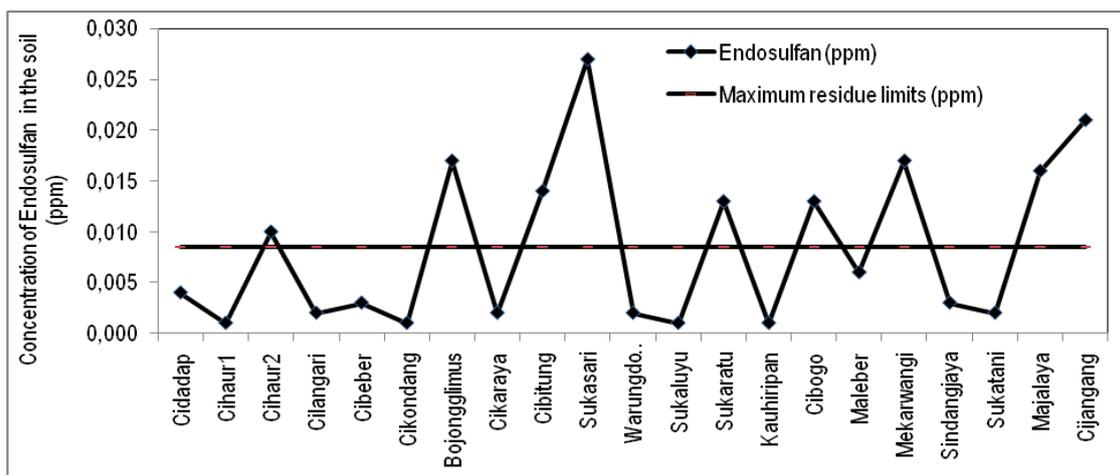


Figure 2. Distribution of Endosulfan residues in the middle Citarum watershed.

The government already banned organochlorine pesticides (OCPs) for agriculture purposed and OCPs residue in the enviroment decreased significantly, but OCPs residue was still detected sediment and waters of Citarum River, fish samples from Citarum River, spray samples, vegetables from the upper area of Citarum Watershed, West Java, Indonesia (Paramita, 2010; Pratama, 2009; Rahmawati *et al.*, 2010; Rochmanti, 2009).

Surveys conducted in West Java in 1999 includes Subang, Indramayu, Cirebon, Falkirk, Cianjur, Sukabumi and Pandeglang, POPs have been found in rice. Similarly, in Central Java, including Rembang, Grobogan, Sukoharjo, Bantul, Wonosobo and Cilacap, and Bojonegoro; in East Java include, Ngawi, Magetan, Madiun, Malang, Lumajang, Jember and Banyuwangi in 1999 showed the presence of POPs content in rice (table 5), (Ardiwinata, 2008).

Table 5. POPs compounds in rice in West Java, Central Java and East Java in 1999.

POPs	Concentration (ppm)		
	West Java	Central Java	East Java
Aldrin	-	0,0037-0,0199	-
Endosulfan	0,0002-0,0005	0,0157-0,0357	0,0003-0,0006

Distribution of Aldrin in the soil

Aldrin was detected in the soil, from 21 sampling locations already exceeds the 14% MRL (figure 3). MRL in the soil by USA EPA (2012), pollution of aldrin (MRL=0.029 ppm). Aldrin was not detected even at the ground surface because these compounds are no longer used. Another possibility for the influence of rain that washed into the deeper layers of the soil through the desorption process. Morgan in Notodarmojo (2005) stated that the absorption and desorption processes occur simultaneously, but when high rainfall concentration in the soil solution increases the desorption rate that organochlorine compounds carried in the deeper soil layers increased. Sinulingga (2006) states that within 1-2 hours after application,

approximately 90% of pesticide residues have been lost due to leaching of water-borne rain and water runoff into rivers and empties into the sea.

Paddy fields with high organic matter content will contain high pesticide residue and soil can absorb chlorinated hydrocarbons such as aldrin. Aldrin degraded to dieldrin through the process of evaporation. Aldrin is found only in low numbers. Based on research in Thailand, aldrin can be converted in a short time become dieldrin (Poolpak, 2007). Aldrin may be physically damaged such as sunlight, moisture, air, and pH. Aldrin may be degraded by soil microorganisms such as *Pseudomonas* sp, *Trichoderma Viridae*, *Micrococcus*, *Bacillus* sp and (Patil *et al.*, 1970).

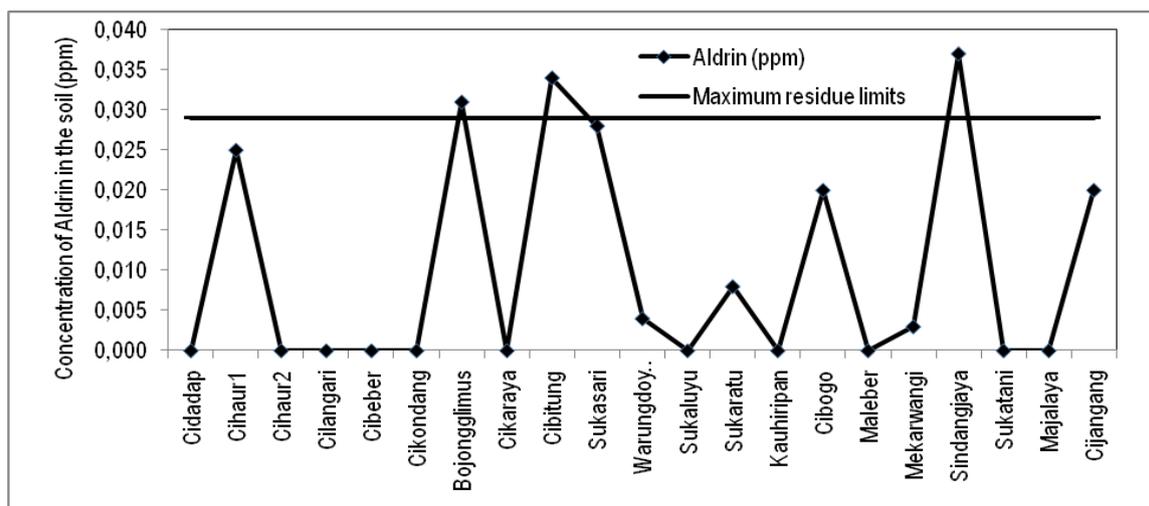


Figure 3. Distribution of Aldrin Residues in the middle Citarum watershed.

Rahmawati *et al.*, (2013) analyzed the content of OCPs in catfish. Heptachlor, endosulfan, and DDT were detected in catfish samples in all sampling points. Heptachlor is the highest average concentration followed by endosulfan and aldrin. In addition, average

concentration of each OCPs in this study is higher compare to the average OCPs concentration of fish samples from Citarum River in 2010. This means that farmers are still using OCPs in agricultural land and leached into the Citarum river.

CONCLUSION

Land and water in the middle Citarum watershed has been contaminated by endosulfan and aldrin compounds. Endosulfan compounds in the soil and in the water was detected in all study sites. The concentration of endosulfan in the soil in the range of 0.001 to 0.027 ppm, with 43% of samples exceeded the maximum residue limit. The concentration of endosulfan in water in the range of 0.001 to 0.005 mg/L, all of the samples exceeded the maximum residue limits.

Aldrin compounds detected in the soil sample locations at 52.17%, while in water were detected in 33.3% of the sample location. The concentration of aldrin in the soil in the range of

0.004 to 0.039 ppm, with 14.29% of the samples exceeded the maximum residue limit. Aldrin concentration in the water is in the range from 0.003 to 0.043 mg/L, with 33.3% of the samples exceeded the maximum residue limit.

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