

Geospatial distributions of organochlorine insecticide in shallot agriculture land at Wanasari sub-district, Brebes regency, Central Java, Indonesia

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Abstract. The organochlorine insecticide is a POPs compound that was monitored according to the Stockholm convention in 2003 because it can cause environmental damage and threaten human health. The study of the distribution of organochlorine insecticides on onion farms has been carried out in the village area in Wanasari sub-District and Brebes Regency. This study was conducted in March-June 2019, with the scope of soil sampling, organochlorine insecticide residue analysis, and spline interpolation. To determine the number of organochlorine insecticide residues using the SNI 06-6991.1-2004 method, while to determine the distribution pattern using the Spline interpolation method with the ArcGIS 10.4. In preparing geospatial residues, we use the ratio of the maximum residual limit (MRLs) of each organochlorine active ingredient. These research sites can be categorized as severe pollution of OCPs insecticides. In this study OCPs residues (Aldrin, Lindane, and Heptachlor, DDT, Endrin, and Dieldrin) can be found with concentrations above the MRLs requirements. OCPs residues with very high range characteristics are shown by Aldrin, DDT, Endrin, and Dieldrin each of more than 50% of the total observed land area. Point numbers six detected all OCPs residues were observed with concentrations exceeding MRLs.

Keywords. Organochlorine, concentration and distribution, shallot agriculture, Brebes regency

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1. Introduction

Organochlorine (OCPs) insecticides were widely used by agricultural systems in the past (during the green revolution). The use of OCPs insecticides is used massively through the Mass Guidance (BIMAS) and Mass Instruction (INMAS) programs. At that time the procurement of pesticides was subsidized by the government through the State Budget. The results are also speculatively proven by Indonesia's success in achieving food self-sufficiency in 1984. Now the insecticide has been banned in Indonesia since 2009 [1], except DDT which is still used limitedly to control mosquitoes that cause dengue fever in various countries [2,3].

OCPs insecticide has a long half-life and therefore can last a long time in the [4] environment. OCPs insecticides are difficult to dissolve in water and fat-soluble. Because it is soluble in fat (lipophilicity), it is very potential for bioaccumulation in living things [5,6]. OCPs insecticides can cause cancer, genetic and teratogenic mutations, damage the immune system and endocrine system [6,7]. Therefore identification of the presence of OCPs insecticides is important to prevent negative impacts on the environment and also human health [8-11] and aquatic living things [12,13].

Brebes Regency is known as a producer of shallots in Indonesia and is recorded as a supplier of Indonesian onions around 60% [14]. Wanasari Subdistrict is the largest shallot planting area which is 7,094 (24.44%) of the planting area in Brebes with a production rate of 587,900 tons (21.56) of the total production of Brebes Regency [15].

Brebes Regency is known as the highest insecticide user in Indonesia even in Southeast Asia. [16] in its press release stated that as much as 30% of the circulation of pesticides in Indonesia circulated in Brebes. Edy Kusmartono (Head of Environment Office) of Brebes Regency stated that 50% of the shallot fields in Brebes had been damaged due to the use of pesticides for shallots that were very high [17].

Considering the effects caused by the OCPs insecticide residues, it is necessary to identify their presence on agricultural land. Monitoring the distribution, and effects of OPCs residues inland ecosystems, is considered important for assessing environmental ecological impacts [18-21]. Every pollution can be spatialized using geospatial model [22-25], so that policies can be taken to mitigate pollution in the environment.

This study aims to determine the distribution of OCPs residue in shallot agricultural in Wanasari, Brebes Regency.

2. Research Methods

Description of the research location

The research area is an intensive area for planting shallots [15]. This region receives irrigation water flow from the Pemali river and therefore can be cultivated throughout the year. In general in this region, shallots are planted 3 times and corn or rice once and shallots twice. For most of the year, the land has never stopped planting except the time of tillage..

Sampling Method

Soil samples were taken at a depth of 0-20 cm, with a stainless steel soil drill. Each coordinate sampling point was taken as many as 5-7 sub-sample points, then from the sub-points were mixed in a large bucket of 10 kg volume, then taken as much as 0.5 kg and contained plastic to be brought under to the laboratory for analysis of OCPs insecticide residues. Each plastic must be given a sample identity label such as Sample number,

coordinates, date of collection. Every coordinate point is recorded with its coordinates with the help of GPS MAP 78S type. The coordinates and village administrative areas are presented in Table 1.

Table 1. Point of Coordinate for soil sampling

Point number	Latitude	Longitude (Village)	Government Administration
1	06 52 631	109 03 522	Wanasari
2	06 53 348	109 01 261	Siasem
3	06 53 202	109 01 283	Pebatan
4	06 52 671	109 01 307	Siasem
5	06 52 944	109 01 276	Wanasari
6	06 53 408	109 01 423	Pebatan
7	06 52 817	109 01 459	Pebatan
8	06 52 718	109 01 391	Pebatan
9	06 52 533	109 01 390	Pebatan
10	06 52 486	109 01 484	Pebatan
11	06 52 319	109 01 483	Pebatan
12	06 52 255	109 01 566	Siasem
13	06 53 015	109 00 263	Siasem
14	06 53 308	109 01 276	Wanasari

Analysis of residues of OCPs insecticide in laboratory

Analysis of insecticide residues was carried out at the Integrated Laboratory of the Indonesian Institute for Agricultural Research (Accreditation of Testing Laboratory Number: LP-556-IDN). Analysis residues of OCPs insecticide are based on SNI 06-6991.1-2004 [26] method. With the following order:

- (1) Preparation of standard solutions and manufacture of working solutions for calibration curves.
- (2) Extraction of the test material, ie Weigh \pm 20 g of the test sample, insert it into the centrifuge tube. Then add 40 ml of high purity acetone, shake using a whisk for 30 minutes. Rotate using a centrifuge at 2500 rpm for 10 minutes. Transfer the liquid into a 300 ml heart flask. Concentrate the liquid by using a vacuum system concentrator at a temperature $<35^{\circ}\text{C}$ and a speed of 25 rpm, to a volume of \pm 30 ml then transfer it to a 500 ml separating funnel. Add 100 ml of 10% NaCl solution to the separating funnel. Add 50 ml of high-purity n-hexane, shake for 20 minutes using a whisk. Let stand until it forms 2 layers, namely the n-hexane layer and water. Fill a layer of water into a 250

ml separating funnel, let the n-hexane layer. Add 50 ml of high-purity n-hexane to the separating funnel containing a layer of water, shake for 20 minutes using a shaker. Let stand until it forms 2 layers, namely the n-hexane layer and water. Discard the water layer, combine the n-hexane layer with n-hexane. Add 100 ml of distilled water and shake. Leave it to form 2 layers and discard the water layer Add + 5 g of anhydrous sodium sulfate powder until all the water is bound. Transfer n-hexane to 300 ml heart flask. Concentrate n-hexane by using a concentrated vacuum system at a temperature of less than 35°C to a volume of + 5 ml.

(3) Washing process (clean up).

(4) Determination of the residual content of the insecticide using Gas Chromatography (GC). To calculate the amount of OCPs residues determined by the formula:

$$A = \frac{c \times v \times f_p}{B} \quad (1)$$

Where:

A = insecticide residue level, µg / g;

B = weight of test sample, g (10 g);

C = insecticide residue level obtained from the calibration curve, µg/ml; V = final dilution volume, ml (2ml);

fp = dilution factor (if without dilution then fp = 1).

Mapping the distribution residues of OCPs insecticides

To maps the distribution of OCPs residues using ArcGIS software series 10.4 with a map scale of 1: 30,000. Distribution pattern distribution uses the spline interpolation method, with five categories of scale range, namely: (1) very low, (2) low, (3) moderate, (4) high, and (5) very high. To determine the magnitude of the range of values is to subtract the value of BMR-LoD divided by two for each active ingredient. The amount of BMR according to [27] is 0.0290 (aldrin); 0.0100 mg/kg (Lindane); 0.0390 mg/kg (heptachlor); 0.0110 mg/kg (dieldrin); 0.0075 mg/kg (endrin); and 0.0150 mg/kg (DDT).

Spline interpolation is a method for analyzing geostatistical data in interpreting values based on data from sampling points. This method is used to estimate the number of organochlorine residuals at the unsampled point based on information from organochlorine residues from the surrounding sampled points. Mentioned by [28] the influence of the value of the closest point is greater than the value of the far point.

Spline interpolation can be used to estimate values using mathematical functions and can minimize the curvature that connects the values of the observed sample point [29]. By doing mapping using the help of ArcGIS spline interpolation can be easily done [30]. Geospatial can provide an overview of each observation and can be associated with the extent of the impact caused [31-34].

3. Results and Discussion

Of the nine active organochlorine insecticide ingredients tested, six of them were detected in the test sample. The six active ingredients are aldrin, lindane, heptachlor, dieldrin, endrin, and DDT. All samples that have detected active residues of their active ingredients show residual rates above BMR. Aldrin residues were detected between <LoD-0.4442 mg / kg detected at 11 observation points. Dieldrin was detected at ten points with residues between <LoD-0.0818 mg/kg. Meanwhile DDT and Endrin residues were detected at nine and eight observation points ranging between <LoD-0.1728 mg/kg and <LoD-0.0771 mg/kg. Whereas Lindan and Heptachlore residues were only detected at one observation

point that was equal to 0.3222 mg/kg and 0.2144 mg/kg. Point number 6 was detected to contain six active ingredients above the MRLs determination based on [27]. The performance of organochlorine residues is presented in Table 2.

The discovery of organochlorine insecticide residues in shallot planting soils provided certainty that OCPs residues could be bioaccumulated (6, 7], as well as providing information that organochlorine insecticides were used at least at this location.

Organochlorine insecticides in Indonesia have been banned from limited use since 1989 and has been permanently banned since 2009 [1].

Table 2. Residu of OCPs insecticide in the shaloot fields of Wanasari, Siasem, and Pebatan village, at Wanasari subdistrict, Brebes regency 2019

Point Number	Residues					
	Aldrin	Lindane	Heptachlore	Dieldrin	Endrin	DDT
	----- mg/kg -----					
1	0.1389	<LoD	<LoD	0.0365	0.0373	<LoD
2	0.2611	<LoD	<LoD	0.0235	0.0242	0.0720
3	0.2681	<LoD	<LoD	0.0487	<LoD	0.1387
4	0.2134	<LoD	<LoD	<LoD	<LoD	0.1031
5	0.0961	<LoD	<LoD	0.0195	<LoD	<LoD
6	0.1057	0.3222	0.2144	0.0465	0.0471	0.1048
7	0.1311	<LoD	<LoD	0.0483	0.0538	0.0992
8	0.1836	<LoD	<LoD	0.0537	0.0603	0.1728
9	0.4442	<LoD	<LoD	0.0491	0.0519	0.0953
10	0.2924	<LoD	<LoD	0.0818	0.0771	0.1174
11	0.1883	<LoD	<LoD	0.0588	0.0547	0.1310
12	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
13	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
14	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
LoD ^{*)}	0.0029	0.0024	0.0024	0.0042	0.0045	0.0039
MRLs ^{**)}	0.0290	0.0100	0.0390	0.0110	0.0075	0.0150

Note: *) LoD = Limit of Detection
 **) MRLs = Maximum Residual Limit

The total area of this study ± 1,853 ha, consisting of 14 soil sampling points from three villages (Wanasari, Siasem, and Pebatan), Wanasari Sub-District, Brebes Regency.

After interpolation, most of the land area is categorized as heavily polluted. Distribution of Aldrin contamination, the widest area experiencing pollution is a very high category of 1183.51 ha. The same thing also happened to the distribution of pollution in Dieldrin, DDT, and Endrin each covering 1178.06 ha; 1120.76 ha; and 1084.42 ha. Meanwhile, Lindane and Heptachlore residues mostly showed very small residues of 1239.26 ha and 1241.50 ha. The low contamination of Lindane and Heptachlor is estimated that indeed these two OCPs are not widely used in this land.

A large number of residues OCPs of DDT were found by [35] in Wonosobo vegetable agriculture, and even residues of OCPs insecticides were also found in carrot, cabbage and potato products [36]. Soil and vegetable products in Magelang Regency were found by

OCPs residue [37]. Not only in Indonesia, OCPs residues around the city of Zhangzhou China by [38], Sokoto Nigeria [39].

The distribution of the OCPs residual can be mapped as follows (Figure 1). Each pollution has a certain area based on spline interpolation mapping [22, 23, 25, 31, 33]. The very high pollution residue categories interpreted in red are shown by Aldrin 64.90% (Figure 1.a), DDT 61.46% (Figure 1.d), Endrin 59.49% (Figure 1.e), and Dieldrin 64.60% (Figure 1.f). This data is interesting considering the OCPs residue should have been 0 (zero) because it can reduce environmental quality and reduce product quality.

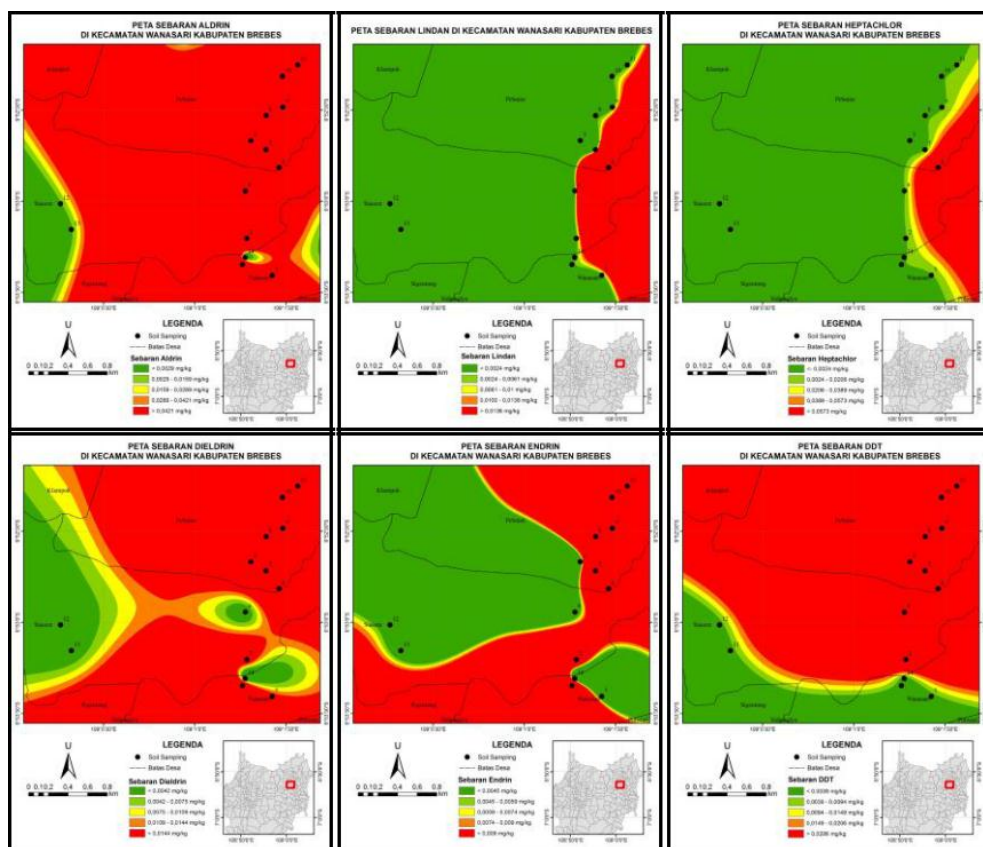


Figure 1. Clockwise in the same direction is the distribution of OCPs insecticide residues (a) Aldrin, (b) Lindane, (c) Heptachlore, (d) DDT, (e) Endrin, and (f) Dieldrin

4. Conclusion

These research sites can be categorized as severe pollution of OCPs insecticides. In this study OCPs residues (Aldrin, Lindane, Heptachlore, DDT, Endrin, and Dieldrin) can be found with concentrations above the MRLs requirements. OCPs residues with very high range characteristics are shown by Aldrin, DDT, Endrin, and Dieldrin each of more than 50% of the total observed land area. Point numbers six detected all OCPs residues were observed with concentrations exceeding MRLs.

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