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ORGANOCHLORINE PESTICIDE RESIDUES ON THE HORTICULTURAL LAND OF THE BANDUNG REGENCY, INDONESIA: ASSESSMENT OF SPATIAL DISTRIBUTION AND HUMAN HEALTH RISKS

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Abstract

Introduction: *Agricultural land in the Bandung Regency, particularly horticultural land, is contaminated with organochlorine pollutants. This research aimed to determine the extent of organochlorine contamination, acute and carcinogenic health issues, and their spatial distribution.* ***Methods:*** *The study was undertaken by collecting 163 tillage-layer soil samples. The sampling site was determined using the 1.3 x 1.3 km grid method. Using the QuEChERS method, soil samples were extracted, and the residual amounts of endosulfan, dieldrin, and chlordane were determined using GC-MS.* ***Results and Discussion:*** *Endosulfan, dieldrin, and chlordane residual levels in the soil were determined to be 0.00463, 0.00935, 0.01509, 0.02295, and 0.14432, 0.18602 mg/kg, respectively. Acute health risks for adults and children indicate that unfavorable non-cancerous health outcomes are unlikely. For adults, the overall lifetime cancer risk is 12.27% very low risk, 87.12% low risk, and 0.61% moderate risk. The total lifetime cancer risk for children is 8.59% very low risk, 90.18% low risk, and 1.23% moderate risk.* ***Conclusion:*** *The presence of the three organochlorine residues discovered in agricultural soil samples does not threaten human health, but research is still needed on organochlorine residues as a whole.*

INTRODUCTION

The rise in population and the expansion of the agricultural industry have increased the application of pesticides for crop protection. The agriculture industry saw both positive and harmful effects due of the green revolution from 1970 to 1980 (1). The positive result is an increase in productivity and food production until there is no longer a need for external food sources. The usage of agrochemicals is growing at an alarming rate, which has a detrimental impact since it leaves residues in the agricultural environment. These residues can include substances that belong to the organochlorine group. Organochlorine pesticides, also known as OCPs, have a long history of application thanks to their high effectiveness and low cost. However, on the other hand, they pose hazards such as toxicity, can spread over long distances, persist in the environment, and bioaccumulate in food webs (2), which is why it is considered to be a threat to humans as well as the environment (3). As a result, the OCPs are included in the Stockholm Convention's list of 12 Persistent Organic Pollutants (POPs), which prohibits the use of these substances (4).

Organic pesticides, especially organochlorine insecticides, are widely used in all ecosystems, not only agricultural environments. Organochlorine chemicals were observed to be distributed in residential areas and the ginseng processing sector (5). There are organochlorine and organophosphate compounds in various environments, including agricultural land, farmers' communities, water bodies, and sediments (6). The strong resistance of organochlorine compounds to a breakdown in the soil enables the residue to be persistent, which in turn causes it to be absorbed by plants, subsequently causing it to penetrate the food chain, which presents a greater risk (7). Therefore, soil can be a storage place for the long-term accumulation of organochlorine residues and a source of secondary pollutants by releasing them

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into the air, water bodies, sediments, and other media even though these compounds have been banned and are no longer in use. In addition, soil can also be a storage place for the short-term accumulation of organochlorine residues.

According to studies conducted in developing nations, the desire for more food as a consequence of rapid population increase results in the uncontrolled use of organochlorine compounds and causes pollution of the land. Iran is responsible for 0.2% of the world's total pesticide consumption, and there is evidence to imply that OCPs are used in agricultural activities (8-9). Even though all organochlorines have been rendered illegal in Indonesia, farmers continue to use “akodan,” which contains endosulfan compound (10). Studies conducted in various parts of Indonesia have shown that a variety of organochlorines are present in the soft tissues of blood clams (11), in the blood of farmers (12), and even in breast milk (13) which is associated with their use. Many factors, including a lack of adequate training for farmers and a lack of knowledge among farmers about the environmental risks posed by pesticides, contribute to the uncontrolled use of pesticides on agricultural land. This problem is worsened because agricultural land is not adequately monitored (14).

As a consequence of this, the objective of this research is to evaluate the level of contamination caused by the presence of organochlorine compounds on horticultural land in the Bandung Regency. In addition, research on the acute/non-carcinogenic and carcinogenic health concerns, in addition to the regional distribution, was carried out. The results of this research can provide scientific backing for regulating pollution brought on by organochlorine residues and protecting the environment in which agriculture is carried out.

METHODS

Research Locations and Soil Sampling

Land in Bandung Regency with GPS coordinates 107°32'30"– 107°47'30" east longitude and 7°2"– 7°7'30" south latitude was used for the study. This area includes 16 districts (Figure 1). Bandung Regency is one of the central producing districts of vegetables and fruits in West Java Province. The most cultivated commodities include potatoes, cabbage, carrots, celery, shallots, chilies peppers (both large and cayenne), cauliflower, tomatoes, lettuce, oysters, strawberries, and now ornamental plants. The horticultural area has a flat to sloping slope with an elevation of 500-1,812 meters above sea level.

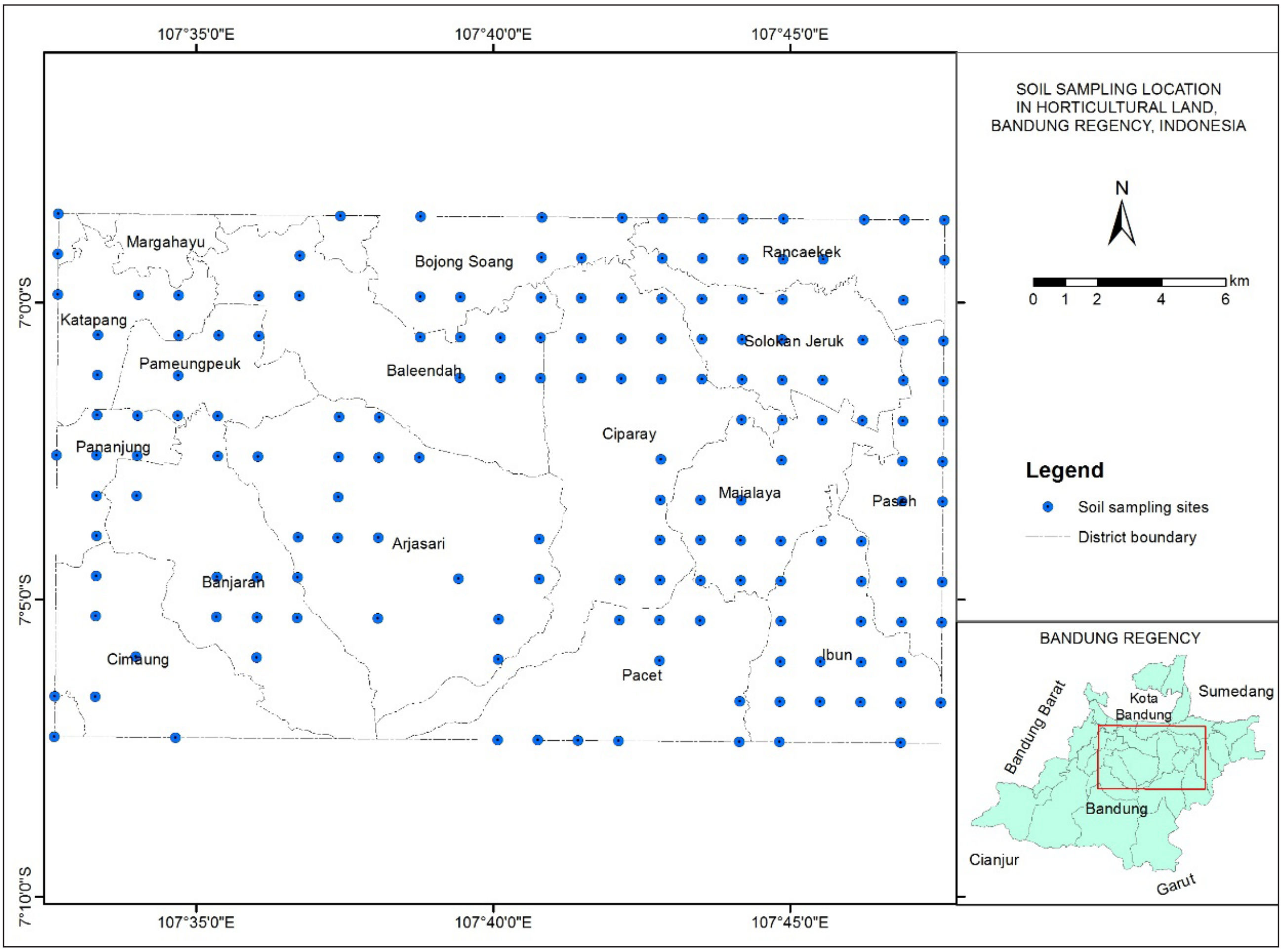


Figure 1. Soil Sampling Location

A grid-based sampling of soil samples (topsoil layer 0-20 cm) in 1.3x1.3 km squares. Each site in the selected field was sampled with approximately 500 gr of dirt (Figure 1). All individual samples in the area are 25 to 50 meters away from one another, and the sampling point has 5 to 10 sub-sampling points (sampling points). A soil drill is the equipment of choice for collecting soil samples; 0.5 kilogram of the homogenized samples were placed in a 15 x 25 cm plastic bag, labeled, and knotted with wool yarn after they had been placed in a bucket and mixed thoroughly.

Pesticide Residue Analysis

The QuEChERS method was used to analyze soil samples (15-16). The technique calls for the sample (either soil or plants) to be weighed using a scale that may go up to 10 grams, then placed in a glass bottle (volume 50 ml). The next step is to incorporate 10 ml of acetone p.a.; however, acetonitrile p.a. may be substituted if desired. Following a minute of vigorous shaking to ensure that the solution was thoroughly mixed, four grams of anhydrous sodium sulfate ($NaSO_4$) powder and one gram of sodium chloride were each added to the mixture (NaCl). After that, the solution was centrifuged at a speed of 3,000 revolutions per minute for two minutes (17). Before the product was used, it was filtered through filter paper that had been treated with anhydrous $NaSO_4$ powder so that it may be placed into a test tube with a volume scale of 10 milliliters. After cleaning the filter paper with acetone p.a. until the volume of the extractant reaches 5 milliliters, the extractant was injected into the GC instrument so it could measure its concentration (18). When the peak chromatograms had been read, the results were compared with the peak chromatograms of the standard solutions for each organochlorine pesticide ingredient. Before using the analysis, the QuEChERS method's ability to determine the levels of pesticide residue was tested and verified (the validation value was more excellent than 80%).

Spatial Analysis

Using the ArcGIS 10.2 program, an analysis was conducted to determine the geographical distribution of organochlorine residues on horticultural land in the Bandung Regency. To establish the distribution of organochlorine residues and to generate a raster layer containing the average concentration of those residues, the kriging interpolation method was utilized. To map the distribution of environmental pollutants, including organochlorines in topsoil (19) and organochlorines in drinking water (20), as well as heavy metals in soil, the kriging approach has been utilized (21). Kriging interpolation is a method that is used in environmental research that does not include the use of statistics. Its purpose is to forecast pollutant concentrations at sites that have not been measured using optimal spatial prediction methods.

Potential Health Risks

Characterizing health risks associated with three organochlorine compounds, endosulfan, dieldrin, and chlordane, in horticultural land used the methodology recommended by USEPA (22). The approach enables the computation of the cumulative risk or Chronic Daily Intake (CDI) through all three possible modes of exposure (ingestion, skin contact, and inhalation), as will be discussed in the following paragraphs:

$$CDI_{ing} = \frac{(Cs\ x\ CF\ x\ IRs\ x\ EF\ x\ ED)}{BW\ x\ AT} \quad (1)$$

$$CDI_{derm} = \frac{(Cs\ x\ CF\ x\ SA\ x\ ABS\ x\ EF\ x\ ED)}{BW\ x\ AT} \quad (2)$$

$$CDI_{inh} = \frac{(Cs\ x\ IRa\ x\ ET\ x\ EF\ x\ ED)}{PEF\ x\ BW\ x\ AT} \quad (3)$$

CDI_{ing} is the chronic daily intake of metals that occurs as a result of ingestion by a child or an adult and is measured in milligrams per kilogram of body weight per day. CDI_{derm} is the chronic daily intake of metals that occurs due to skin absorption and is also measured in milligrams per kilogram of body weight per day. CDI_{inh} is the lifelong daily intake of metals that arises from inhalation and is also measured in milligrams (mg per kg day). The exposure variables utilized in these calculations, which vary depending on the age of the person being calculated, are the same as those suggested by the USEPA (22) and can be found in Table 1.

Table 1. The Constants of Organochlorine Residue Exposure Factor

Symbol	Description	Unit	Adults	Children
Cs	Concentration of organochlorine	mg/kg	-	-
IRs	Average exposure through digestion	mg/day	100	200
SA	Surface area exposed	cm²/day	6032	2373
AF	Absorption constant of soil to skin	mg/cm²	2	0.07
Ira	Respiratory rate	m³/hour	0.83	0.53
ED	Length of Exposure	year	30	10
BW	Weight	kg	70	30
AT	Exposure time	day	10950	3650
PEF	Emission constant from soil to air	m³/kg	1.36×10^9	1.36×10^9
CF	Correction factor	kg/mg	1.00×10^6	1.00×10^6
ET	Exposure time	hour/day	24	24
EF	Exposure frequency	day/year	350	350

Symbol	Description	Unit	Adults	Children
ABS	Absorption factor	-	0.13	0.13
RfD	Reference dose for endosulfan	mg/kg/day	5×10^{-4}	5×10^{-4}
	Reference dose for diendrin		5×10^{-5}	5×10^{-5}
	Reference dose for Chlordane		6×10^{-3}	6×10^{-3}
CS	Cancer slope factor for endosulfan	mg/kg/day	-	-
	Cancer slope factor for dieldrin		16	16
	Cancer slope factor for chlordane		0.35	0.35

It is possible to reduce the non-carcinogenic risk for children or adults associated with each route of exposure to each organochlorine compound by calculating a value. This number is calculated by dividing the CDI value for each exposure route by the Reference Dose (RfD) for that chemical, which is measured in milligrams per kilogram per day (mg/kg/d), according to equation (4):

$$HQ = \frac{CDI}{RfD} \quad (4)$$

A cumulative HQ value is produced by adding up the individual HQ values for each possible route of exposure (separately for children and adults). An overall Hazard Index (HI), which is represented as a number without units and is calculated by adding the cumulative HQ values for each organochlorine compound, is then determined. The formula for this calculation is illustrated in equation (5): The cumulative HQ values for each organochlorine are added together to characterize the overall non-carcinogenic risk linked with multiple organochlorine exposures in either children or adults.

$$HI = \sum_{i=1}^{3} HQ_i \quad (5)$$

Where HQi is shorthand for the hazard quotient of the it-h organochlorine compound that can be discovered in the ground. When the HI value is 1, it is thought that there would be no non-carcinogenic negative impacts on one's health; however, when the HI value is more than 1, it suggests there is a probability that there may be unfavorable effects on one's health. After carrying out the series of steps that were just described (equations (2) to (6)), which were carried out independently for the risk characterization of children and adults (using their respective distinctive parameters from Table 1, the results of which were summed up to give the HI_{total} value. Following that, this value was utilized to determine the overall number of cases. This total HI value is then projected spatially to produce a spatial depiction of the risk unrelated to cancer. The cancer risk for organochlorines was calculated by multiplying the child or adult CDI value for each route of exposure by the Cancer Slope Factor (SF) for that compound and exposure route. Risk was unitless and could be interpreted according to equation (6):

$$Risk_i = CDI_i \ x \ SF_i \quad (6)$$

The term "chronic daily intake" (CDI_i) refers to the "i-th dose" of organochlorine compounds (in milligrams per kilogram per day). In contrast, "cancer slope factor" (SF_i) refers to the "cancer slope factor" of the "i-th compound" when exposed via a particular pathway. The formula for this is (milligrams per kilogram per day) multiplied by 1. This equation modifies the calculation of the mean daily intake throughout a lifetime of exposure to the individual's unitless risk of acquiring cancer. When calculating the total lifetime cancer risk, childhood and adult cancer risks are added together, just as they are when calculating the risk of other types of cancer, to quantify the overall cancer risk during a person's lifespan that is related to numerous exposures to organochlorine substances, as illustrated in equation (7):

$$Risk_{total} = \sum_{i=1}^{3} Risk_i \quad (7)$$

$Risk_{Total}$ is the total lifetime cancer risk from exposure to the three organochlorine compounds in the soil. $Risk_i$ is the lifetime cancer risk from exposure to these compounds individually. $Risk_{Total}$ is the entire lifetime cancer risk. Very low, with a value of < 10^{-6}; low, with a value of 10^{-6} - 10^{-4}; medium, with a value of 10^{-4} - s 10^{-3}; high, with a value of 10^{-3} - 10^{-1}; and very high, with a value of > 10^{-1}. These are the subcategories that make up the cancer risk category (23). Table 2 outlines the RfD and SF considered for use in this investigation (22).

Table 2. The Concentration of Organochlorine Residue and Correlation Analysis of Organochlorine Compounds in Horticultural Land, Bandung Regency

Description	Endosulfan	Dieldrin	Chlordan
Mean (mg/kg)	0.00463	0.01509	0.14432
Standard Deviation (mg/kg)	0.00935	0.02295	0.18602
Detected	70	108	135
Range (mg/kg)	nd-0.081	nd-0.170	nd-1.573
MRLs	0.04*)	0.05*)	0.05*)
Endosulfan	1		
Dieldrin	0.579**)	1	
Chlordane	0.519**)	0.307**)	1

nd – not detected
*) CME (24)
**) significant at 0.01 level

Table 3. The Comparison of Organochlorine Residue Concentrations Globally

Research Location	Location Characteristic	Endosulfan	Dieldrin	Chlordan	Reference
Bandung, Indonesia	Agriculture	nd-0.081	nd-0.170	nd-1.573	This research
Chakwal, Pakistan	Industry and Residential	0.002–0.089	-	nd-0.056	(25)
Azad Jammu & Kashmir, India	Residential and agricultural	0.03–0.78	-	0.004–0.08	(26)
Central–southern Italy	Agricultural area	0.120-3.020	0.240-9.800	0.110-3.400	(27)
Kuttanad agroecosystem, India	Agriculture	0.740-8.900	1.290–3.720	0.330–9.070	(28)

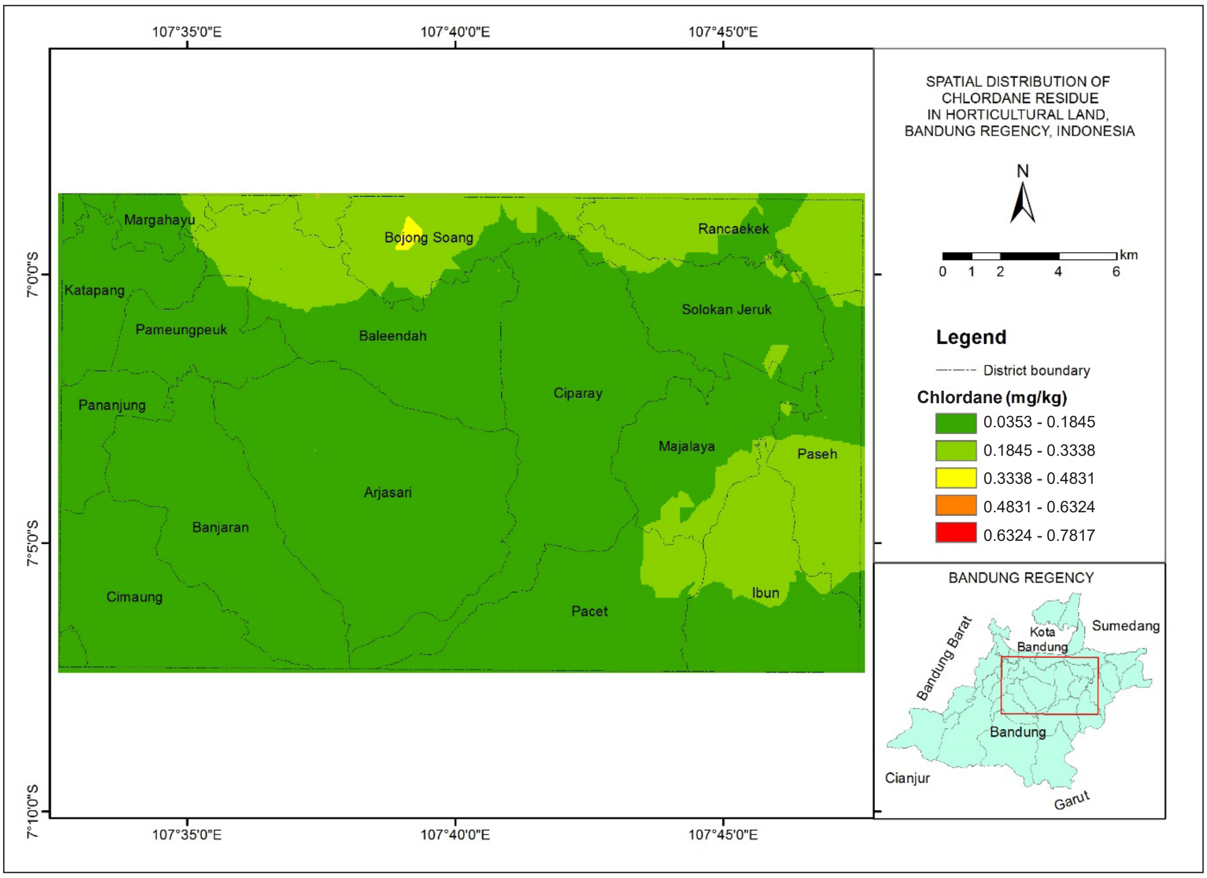
RESULTS

Organochlorine Concentration in Horticultural Land

Organochlorine residues were detected in the agricultural land of the Bandung Regency, as evidenced by the OCPs concentration (Table 2). This finding reflects the past utilization of these compounds in agricultural activities. Chlordane was found to have the lowest average residual organochlorine concentration, followed by dieldrin and then endosulfan in that order. None of the three substances' analytes could be found in some samples. The concentrations of the three organochlorine compounds varied between not detected-0.081 for endosulfan, not detected-0.170 for dieldrin, and not detected-1.573 for chlordane with an average of 0.00463, 0.01509, and 0.14432, respectively. In some of the soil samples, the presence of these three compounds could not be determined. It was determined that 42.9% of the 163 samples contained endosulfan compounds, 66.3% contained dieldrin, and 82.8% contained chlordane.

Spatial Distribution Assessment

The spread of organochlorine residues such as chlordane, dieldrin, and endosulfan is depicted in Figure 2. According to the distribution of organochlorine residue, there are three clusters of places with higher organochlorine residues: Bojongsoang, Rancaekek, and Ibun. Each cluster contains a more significant concentration of organochlorine residues. The Ciparay and Banjaran districts, which have a higher terrain compared to the other districts, have a relatively lower distribution of organochlorine chemicals. A similar level of intensity and dosage was employed in the past for all three of these substances. The notion is supported by discovering a close connection between these three chemicals (Table 2).



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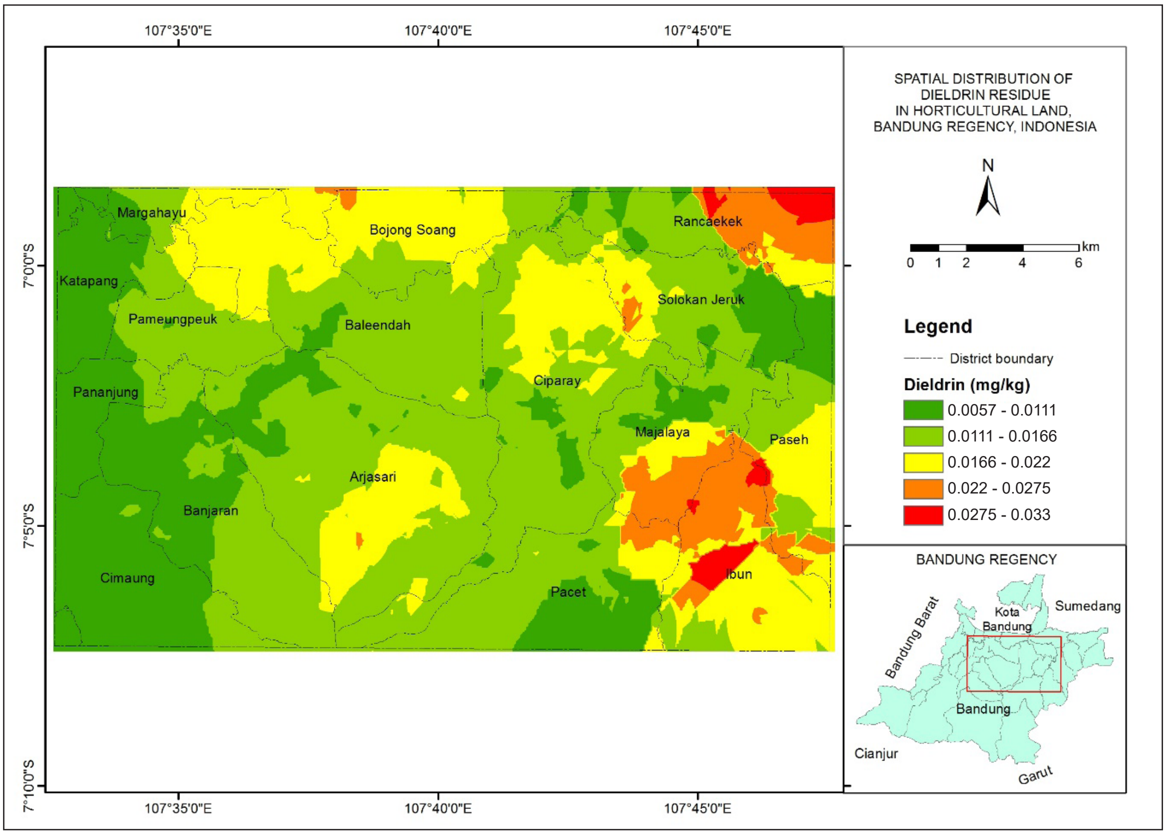

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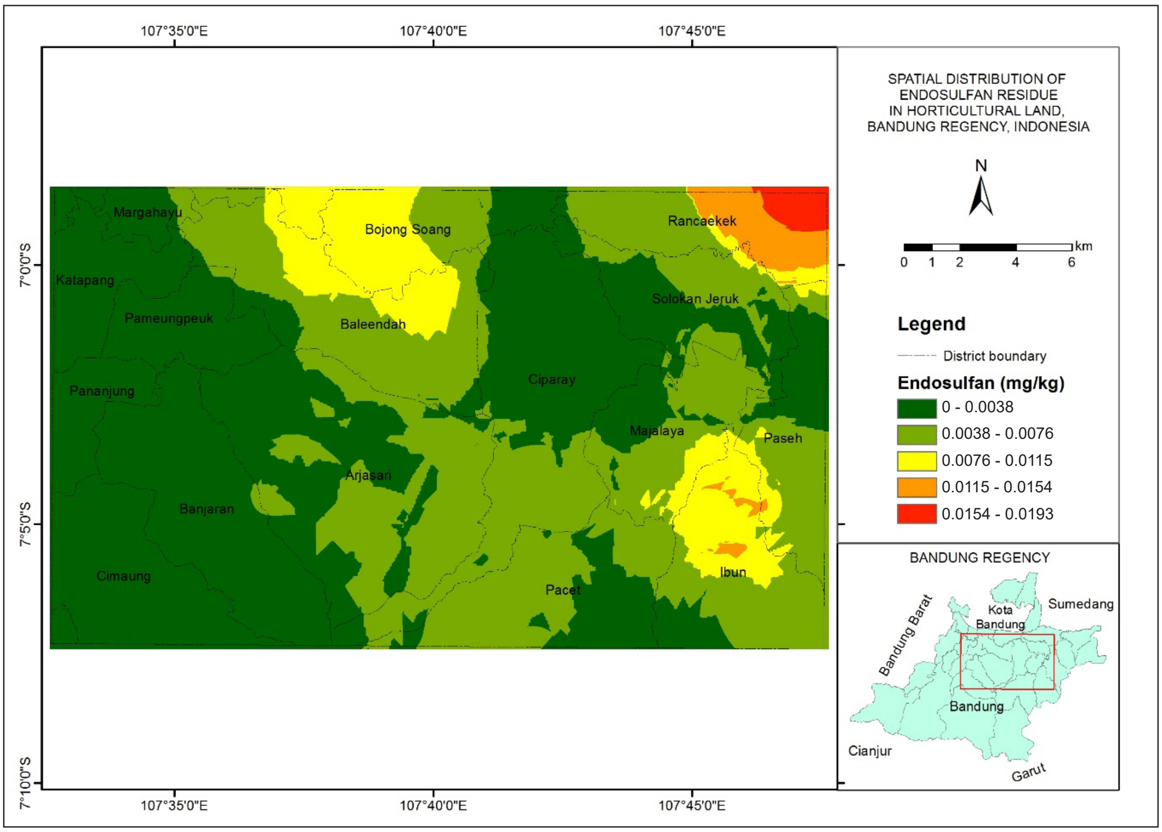


(c)

Figure 2. Spatial Distribution of Residual Chlordane *(a)*, Dieldrin *(b)*, and Endosulfan Compounds *(c)*

Potential Health Risks

For adults, the risk of developing acute health problems as a result of organochlorine exposure was calculated using exposure to concentrations of 0.13% endosulfan, 49.93% dieldrin, and 49.93% chlordane. Children were exposed to endosulfan, dieldrin, and chlordane at concentrations of 0.17%, 36.22%, and 63.61%, respectively, in residue. In the meantime, based on the route of exposure, the gastrointestinal tract was responsible for 38.936% of adults' exposure. In comparison, absorption through the skin accounted for 61.063%, and the respiratory tract accounted for 0.001% of adults' exposure. The route of exposure through the digestive tract is the most common for children, accounting for 98.931% of all exposures. In comparison, the routes of exposure through the skin and the respiratory tract account for only 1.068% and 0.001%, respectively.

DISCUSSION

Organochlorine Concentration in Horticultural Land

According to the Canadian Ministry of the Environment (CME) (24), the maximum residue limit is exceeded in some tests, which is relevant when discussing the agricultural soil quality standard. There was 1.8% of samples of endosulfan, 3.7% of samples of dieldrin, and 68.7% of samples of chlordane that contained higher residues than the standard that was required by CME (24). In other nations, a great deal of research has been done on the number of organochlorine residues present in various soil characteristics (Table 2). The levels of organochlorine, endosulfan, dieldrin and chlordan that were found as residues in the soil at the location of the study were significantly higher than those found in investigations conducted in other countries such as in the north of Punjab Province, Pakistan (25), Himalayan Region of Pakistan (26), British Columbia, Central–southern Italy (27), and Kuttanad agroecosystem, India (28). It should serve as a warning to the many stakeholders to develop agricultural land management strategies to reduce the number of pesticide residues.

There are many ways to remove organochlorines from agricultural lands, such as phytoremediation, bioremediation, incineration, adsorption, ozonation, etc. (29). However, using methods that promote a green economy based on local materials is an option that must be considered. Indonesia is rich in biodiversity, which has the potential to become raw material for land remediation. The utilization of agricultural waste is a source of adsorbent material. Likewise, exploration of bacteria capable of degrading organochlorine compounds has also been carried out. Activated charcoal in its many derivative forms, including activated charcoal made from agricultural waste, urea coated with activated charcoal, and urea coated with activated charcoal that has been enhanced with microorganisms, can be used as an alternate method for the management of agricultural land (12). Efficient utilization of bacteria in pesticide degradation can also be employed. Besides being able to decompose pesticide compounds, bacillus aryabhattai bacteria and consortia microbes (*Bacillus aryabhattai*, *Pseudomonas Sp., Azopirillium Sp., Azotobacter Sp., Cromobacterium Sp.*) were also able to increase the growth and production of shallots (30-31).

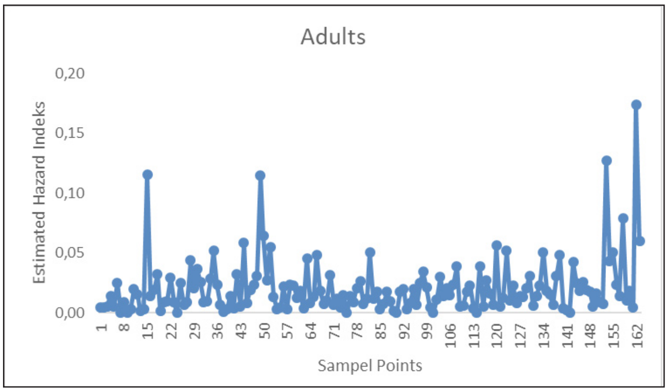
Spatial Distribution Assessment

The increased concentration of organochlorines in the Bojongsoang, Rancaekek, and Ibun areas compared to other regions is believed to be a result of the compound's extensive use in the past. Bojongsoang, Rancaekek, and Ibun are agricultural areas surrounded by industry, so industrial waste contamination increases (32-33), plant growth, is disrupted, and pests and disease attacks increase (14). The use of pesticides will increase along with the increase in pest and disease attacks, which impacts the accumulation of pesticide residues in the soil.

Organochlorine residues can spread due to soil erosion and follow runoff from the water surface (34). Bandung Regency is a mountainous or hilly area with an altitude above sea level varying from 500 m to 1,800 m. The districts of Bojongsoang, Rancaekek and Ibun are located in the lower plains with an average elevation of 681 m, 630 m, and 700 m, respectively. These three areas are suitable as a place for the deposition of sediments that carry organochlorine pollution. It confirms why these three regions have concentrations of organochlorine residues compared to other.

Potential Health Risks

Exposure to organochlorine residue contamination in soil may result in acute/non-carcinogenic health concerns and cancerous health risks in humans. Bandung Regency's horticulture land was examined and evaluated for potential health concerns to children and adults by considering the digestive tract route and absorption through the skin and respiratory tract. Figure 3 shows that health problems are unlikely to arise when the potential non-carcinogenic health risk is less than 1. Because the hazard index is less than 1, there is no need to worry about children or adults developing severe health concerns.



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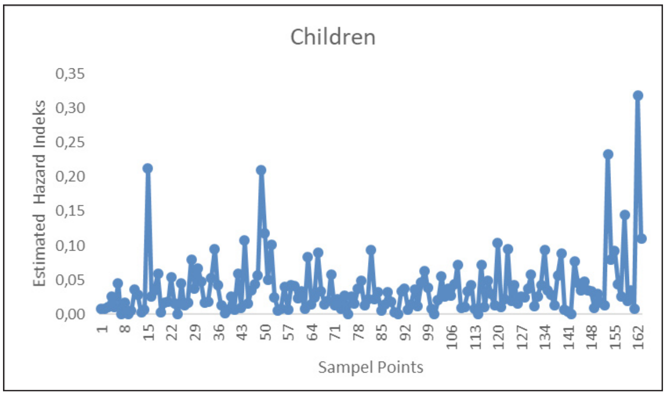
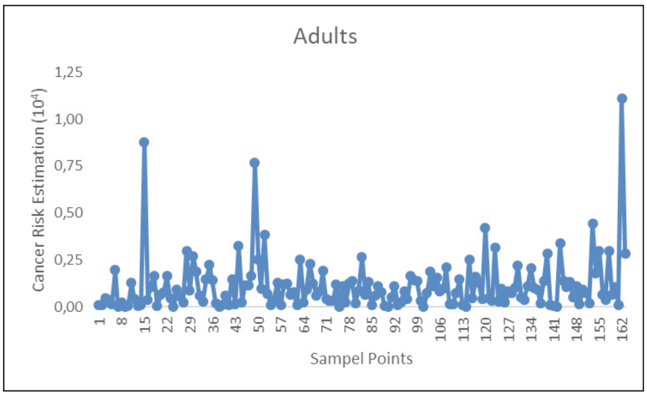


(b)

Figure 3. Acute/Non-Carcinogenic Health Risk Estimation on Adults *(a)* and Children *(b)*

In this study, endosulfan residues contributed the least to the risk of acute health problems, but we need to be careful because farmers are still finding the use of this compound. The Indonesian government has banned the use of the active ingredient endosulfan since 2015 through the Regulation of the Minister of Agriculture of the Republic of Indonesia Number 39 of 2015 concerning Pesticide Registration (17). But, in reality, the use of endosulfan compounds is still found in the trademark Akodan such as in Bandung Regency (35), Yogyakarta and West Java Province (36) and Papua Province (37).



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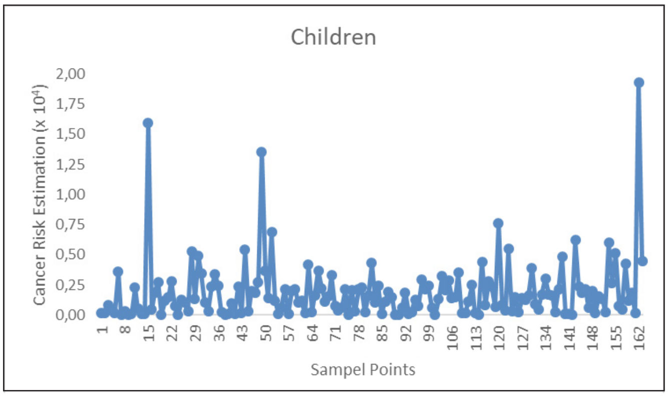

(b)

Figure 4. Estimated Lifetime Risk of Cancer on Adults *(a)* and Children *(b)*

When the risk of acquiring cancer is less than one instance for every million people exposed to it, it is regarded as posing a slight chance of cancer (or less than 1.10^{-6}). As shown in Figure 4, the overall risk of cancer for children and adults working in horticulture land in the Bandung Regency is far lower than the threshold value. This would imply that the majority of the population that is exposed to polluted soil does not have a considerable risk of developing cancer as a result of the exposure. Because there is one sample with values that are higher than the requirement for adults and there are three samples with values that are higher than the threshold for children, it is clear that this matter needs a lot of attention. Identical research with the same findings has been carried out previously (8, 38). These investigations found that assessments of low soil pesticide levels were associated with a negligible cancer risk. Exposure through the skin showed the highest risk for adults at 64.78% of total exposure, while exposure through the gastrointestinal tract showed the highest risk for children at 98.75%. Among all the observed exposure routes, skin exposure exhibited the highest risk. Skin exposure is associated with the highest risk of increased exposure for adults, followed by digestive exposure, respiratory exposure, and skin exposure for children. Children are more susceptible to developing cancer from exposure to pesticides in polluted soil than adults. This disparity is because of the increased risk of the digestive route in children who engage in activities involving the ground (8, 38-39).

ACKNOWLEDGMENTS

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CONCLUSION

Horticultural land in Bandung Regency contains endosulfan, dieldrin, and chlordane residues in the range (mg/kg) nd-0.081, nd-0.170, and nd-1.573, respectively. There is no acute health risk for adults and children that may adversely occur, while for the total lifetime risk of cancer for adults, there are 12.27% very low risk, 87.12% low risk, and 0.61% moderate risk, and for Children, there is 8.59% at very low risk, 90.18% at low risk and 1.23% at medium risk. The content of the three organochlorine residues found in agricultural soil samples is not a concern for human health, but overall organochlorine residues still require further research. However, health risks from organochlorine pesticide residues need to be watched out for because of the movement of organochlorine compounds and the possible impact of the anthropogenic activity of the active ingredient endosulfan, which is still used by farmers, which leads to the release of these compounds into the environment and can accumulate on agricultural land.

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