Risk level mapping of organophosphate pesticides application in agricultural area of Cangkringan Subdistrict, Yogyakarta, Indonesia

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Abstract. Farmers in Indonesia widely use organophosphate pesticides (OPPs) to increase crop production, despite their risk to human health and the environment. Objective of this study is to estimate the loading rate and risk level of OPPs based on OPPs characteristics, farmers' knowledge, and the behavior of OPPs application in agriculture area in Yogyakarta Special Region in Indonesia. Data collected through interview and questionnaire distribution revealed three types of OPPs used in study area; profenofos, chlorpyrifos and diazinon. Data on application dose and crop area were used to estimate loading rate. Risk scores can be calculated by multiplying loading rate with toxicity data of each type of OPPs used. Box-Cox transformation was then used to normalized rightly skewed risk score data to get 4 (four) levels of risk classification range; low, medium, high and very high. It was found that there were areas classified as very high risk in wet season, while it is not the case in dry season. However, in total loading per year, areas with high and very high risk level were found for more than >50% in study area.

1. Introduction

The population growth leads to increasing demand for food supply. As an agricultural country, Indonesia handles this issue by focusing on the expansion of crop production. The Ministry of Agriculture stated that from 2015 to 2019, the productivity of paddy and other commodities were increased [1]. This increasing growth of crop production was achieved through intensification, extensification, rehabilitation and diversification programs [2]. Among these, the intensification program was more widely implemented through the use of high-yielding varieties of seed, increasing fertilization input and pesticide application. In the previous agriculture development program, the most widely used types of pesticides were organochlorine and organophosphate. However, since organochlorine was banned by the government, organophosphate pesticides (OPPs) became extensively used by the farmers.

In Indonesia, Ministry of Agriculture Regulation No.39 of 2015 permitted the use of OPPs use within certain limits. From the environmental point of view, OPPs are less persistent and decomposed faster than organochlorine [3]. However, OPPs have acute neurotoxicity at high doses. It inhibits the performance of acetylcholinesterase which can accumulate in the nervous system [4]. Once the pesticide is applied, its fate is affected by its characteristic and interaction with soil, weather and agricultural usage. Various studies have been conducted related to OPPs residue in Indonesia such as residue in soil.
Currently, OPPs threaten the aquatic ecosystem due to their intensive application in the watershed, then transported to the river system, shallow ground water, and subsequently into the estuaries. Some studies tried to use spatial approach to estimate pesticide loading in watershed and calculate its ecological risk. Some researchers have attempted to implement this approach in Indonesia. However, it has not been widely used due to lacking data quantity and consistency. Thus, this study's objective is to estimate the loading rate and risk level of OPPs based on OPPs characteristics, farmers' knowledge, and the behavior of OPPs application in agriculture area in Yogyakarta Special Region.

2. Materials and Methods

2.1. Study location
The upper part of Opak Watershed, which is dominated by agricultural areas was selected as the location of this study. Agriculture Technical Implementation Unit (UPT) of Sleman Regency has recommended two villages: Wukirsari village and Argomulyo villages as more specified study locations. Wukirsari and Argomulyo Village have an area of 1,456 and 847 hectares, respectively. Residents of these two villages were mainly farmers. Paddy and vegetables were predominant cultivation commodities; therefore, the application of pesticides was expected to be high.

![Study location](image)

**Figure 1.** Study location

2.2. Data collection
Primary data was collected directly from March to May 2017 through interviews and questionnaires distributed to farmers. Collected data includes the source and type of OPPs used for the last five years,
the dose and frequency of application. Variety of crops and planting seasons were also recorded. The location of the plantation area of each respondent was marked by using the global positioning system (GPS). Information on the type of OPPs that most farmers have used was derived from pesticides stores in Wukirsari and Agromulyo villages. Number of farmers as samples were determined based on Slovin equation:

\[
 n = \frac{N}{1 + Ne^2}
\]

\(n\) = total of samples  
\(N\) = population  
\(e\) = error (10%)  

2.3. Loading rate estimation
To estimate OPPs loading rate, crop type, the active ingredient of OPPs used, and planting season per year became the basis for its calculation by using equation below [18]:

\[
 C_i = D_i \times S_f
\]

\(C_i\) is the loading of the active ingredient of certain organophosphate (gr). \(D_i\) is the total dose of organophosphate application (gr/ha) and \(S_f\) is cropped area (ha).

2.4. Risk Level
Risk level is determined based on risk score. Risk score is calculated by multiplying loading rate estimation \((C_i)\) and risk value \((K_{di})\). This formula was adapted from Houdart [19].

\[
 Risk\ score = C_i \times K_{di}
\]

\(K_{di}\) is the conversion of risk factors \((F_{di})\) which is calculated by using equation below:

\[
 F_{di} = K_{ti} \times K_{ei}
\]

\(K_{ti}\) is toxicity value of OPPs' active ingredient. It is mainly based on LD50 value in rats taken from Material Safety Data Sheet (MSDS). This toxicity value is divided into four groups representing the level of toxicity according to the International Manual of pesticide [16] as described in the table below.

| Toxicity value \((K_{ti})\) | LD50 (mg/kg) | Toxicity level |
|--------------------------|-------------|----------------|
| 1                        | 0           | Not toxic      |
| 2                        | >1000       | Slightly toxic |
| 3                        | 100-1000    | Toxic          |
| 4                        | <100        | Very toxic     |

On the other hand, \(K_{ei}\) is the exposure level related to pesticide solubility. Data on the solubility of pesticides were collected from MSDS and then converted to \(K_{ei}\) as presented in Table 2 below [20].

| Exposure value \((K_{ei})\) | Solubility (mg/L) | Solubility level |
|-----------------------------|-------------------|-----------------|
| 1                           | 0                 | Not soluble     |
| 2                           | <1                | Slightly soluble|
| 3                           | 1-100             | soluble         |
| 4                           | 100-1000          | Highly soluble  |

Table 1. Toxicity value based on LD50 value

Table 2. Conversion of active ingredient solubility with \(K_{ei}\)
Correlation between risk factor ($F_{di}$) and risk value ($K_{di}$) ranges between 1-2.5. The lower $K_{di}$ means lower risk factor and vice versa (Table 3)[20].

Table 3. Relationship of $F_{di}$ value with $K_{di}$ value [20]

| Risk Factor $F_{di}$ | Risk value ($K_{di}$) |
|----------------------|-----------------------|
| 1-4                  | 1                     |
| 5-8                  | 1.5                   |
| 9-12                 | 2                     |
| 13-16                | 2.5                   |

3. Results and discussion

Data on pesticides application were collected through interviews with farmers of Wukirsari and Agromulyo villages. According to Agriculture Technical Implementation Unit (UPT) of Sleman Regency, in 2017, there were 131 and 171 farmers in Wukirsari and Argomulyo Village, respectively. The number of samples were calculated by using Slovin equation resulted in total respondents of 94.

According to the questionnaire, the area for vegetable plantation and paddy fields range from 500 to 17,900 m$^2$ and from 700 to 7000 m$^2$, respectively. Eighty percent (80%) of these cropped areas were self-owned, while the rest (20%) were rented. Types of vegetables cultivated in the study area were red chili, tomato, and cucumber. In addition, farmers also grew several types of fruits, such as melon and watermelon. In general, there were two or three planting seasons per year with different types of crops for each season, as presented in Table 4.

Table 4. Planting season and harvest time

| Crops       | Planting season / year | Harvest time  |
|-------------|------------------------|---------------|
| Paddy       | 2-3 times              | ±3 months     |
| Red chili   | 1-3 times              | ±3 months     |
| Tomato      | 1-3 times              | ±3 months     |
| Cucumber    | 1-5 times              | ±3 weeks      |
| Eggplant    | 1-3 times              | ±3 months     |
| Watermelon  | 1-3 times              | ±80 days      |

Farmers in the study area applied pesticides to their crops to help to improve their productivity. They obtained these pesticides from agrovet shops near their villages. Pesticide application had been practiced for more than ten years by only 15% of respondents, while the rest practiced it for less than ten years.

The frequency of pesticides spraying depends on various factors, including the type of crops and crop age. Vegetables got more often spray than paddy, and red chili got even more near harvest time than other crops. Season also influenced as the frequency in rainy seasons was almost three times that in the dry season (Figure 2).

Regarding dose application, only 53% of respondents follow dosing instructions on the package. Around 32% prepared the mixture based on experience, while the rest of respondents applied both practices. Almost 60% of respondents use hand sprayer to apply pesticides on their crops, while the rest used machine type.
Pesticides used by farmers in the study area belong to around 20 groups including avermectin, organophosphate, dithiocarbamate, carbamate, organomanganese and organozinc. Organophosphate was the second most commonly used type of pesticides (16%) after avermectin (19%). This figure would put organophosphate as one of the important contributors to environmental pollution in study area.

OPPs comprises compounds with various active ingredients. OPPs used in the study area contain to three active ingredients; profenofos, chlorpyrifos, and diazinon. Market brand name, concentration of active ingredients (AI), data on solubility and toxicity of each AI are presented in Table 5. Farmers sprayed most of the crops with at least two types of active ingredients. Profenofos was used on almost all types of crops, followed by chlorpyrifos and diazinon (Figure 3). The interview found that mixing different types of pesticides or mixing pesticides with fertilizer was sometimes practiced by farmers. It should be noted that combining pesticides may result in interactions that increase its risk to human health and the environment. Furthermore, improper use or disposal of agricultural pesticides can lead to severe environmental damage [21]. In this study, loading rate estimation was calculated based on single or mixed OPPs application. However, the effect of mixed OPPs application was neglected.

**Table 5. Organophosphate pesticide and its active ingredient**

| Active ingredient (AI) | Trade name     | AI Concentration (gr/L) | LD50 (mg/kg) | Solubility (mg/L) |
|------------------------|----------------|-------------------------|--------------|-------------------|
| Profenofos             | Anwafin 500EC  | 500g/L                  | 358          | 28                |
|                        | Curacron 500EC | 500g/L                  | 358          | 28                |
|                        | Sidacron 510 EC| 510g/L                  | 358          | 28                |
| Chlorpyrifos           | Durshan 200EC  | 200g/L                  | 230-310      | 1.4               |
|                        | Smesban 590EC  | 530g/L and Sipemetrin 60g/L | 800          | 1.4               |
| Diazinon (21%)         | Diazinon      | 600g/L                  | 1600         | <1                |

**Figure 2. Spraying frequency in rainy season and dry season**

**Figure 3. Dose application and sprayer type**
Based on LD50 value as presented in Table 1, both profenofos and chlorpyrifos have toxicity value ($K_{ti}$) of 3 hence categorized as "toxic". Meanwhile, diazinon is "slightly toxic" with lower $K_{ti}$ value of 2. On the other hand, the exposure level ($K_{ei}$), ranges from slightly soluble ($K_{ei}$=2) to soluble ($K_{ei}$=3). Risk factor ($F_{di}$) can then be calculated which ranges between 4 to 9 or with risk value ($K_{di}$) between 1 to 2. To determine risk score then, the loading rate ($C_i$) should first be estimated. Table 6 presents the range of OPPs application dose in different season in gram/hectare in one year. By multiplying this application dose with area per crop's type, loading rate of OPPs in each village per season per year can be estimated as presented in Table 7, and Figures 5 and 6.

Table 6. Organophosphate application dose per active ingredient in Argomulyo and Wukirsari Villages

| Active ingredient | Wet season (gr/hec/year) | Dry season (gr/hec/year) | Total (gr/hec/year) |
|-------------------|--------------------------|--------------------------|---------------------|
| Profenofos        | 0.048-44.064             | 0.036-15.429             | 0.036-44.064        |
| Chlorpyrifos      | 0.010-51.429             | 0.005-17.143             | 0.005-51.429        |
| Diazinon          | 0.367-18.514             | 0.184-10.286             | 0.184-10.286        |

Table 7. Organophosphate loading rate in Argomulyo and Wukirsari Village

| Organophosphate loading rate (gr/year) |
|---------------------------------------|
| Argomulyo                             |
| Wukirsari                             |
| Minimum                               |
| Wet season 0.003                      |
| Dry season 0.001                      |
| Total 0.003                           |
| Wet season 0.025                      |
| Dry season 0.011                      |
| Total 0.038                           |
| Maximum                               |
| Wet season 4.744                      |
| Dry season 0.926                      |
| Total 5.554                           |
| Wet season 2.057                      |
| Dry season 0.686                      |
| Total 2.743                           |
| Average                               |
| Wet season 1.100                      |
| Dry season 0.212                      |
| Total 1.312                           |
| Wet season 0.535                      |
| Dry season 0.171                      |
| Total 0.706                           |
According to Figure 5 and 6, chili had the highest OPPs loading rate among crop's types in both villages. Meanwhile, wet season gives higher loading rate as farmers tend to spray their crops more frequently in that season. The subsequent risk score derived by multiplying loading rate ($C_i$) and $K_{d_i}$ is presented as histogram and normal distribution curve in Figure 7.
Figure 7 Histogram (above) and normal distribution (below) of risk score.

As shown in Figure 7, more than 80% score between 0.006 and 2.106. Only several locations have score above 4. According to Haudart (2009), risk level classification is carried out based on the range of risk score [19]. However, since the distribution of risk score was rightly-skewed with several extreme value, risk level classification should be modified through prior transformation of risk score data. There are several available methods for data transformation; transformation to normality (Box and Cox), transformation to symmetry, and rank transformation [22]. In this study, Box-Cox method was selected to improve symmetry and stabilizes the spread of risk score data [19]. Box-Cox method has been applied in analysis of data over a wide range of topics such as heavy metal pollution in soil core samples [23], spatial data [24], gen analysis based on structural variance heterogeneity [25] and hydro-meteorological data [26]. Histogram of transformed risk score data is presented in figure 8. Risk level classification was then determined after inversion of the transformed data (Table 7). Risk classification for Argomulyo and Wukiran village in dry season, rainy season, and the whole year is presented in figure 9.

Figure 8 Histogram of transformed risk score data

| Risk Classification | Risk score |
|---------------------|------------|
| low                 | 0.00-0.15  |
| medium              | 0.15-0.99  |
| high                | 0.99-3.85  |
| very high           | >3.85      |
Figure 9 shows that for rainy season, both villages have areas classified as very high risk. Almost 50% total percentage areas in Argomulyo Village are classified as high and very high risk, while it is lower in Wukiran Village with only 33%. In dry season, no areas with very high risk level in both villages, while those with high level risk are also found less than 10%. However, in total loading per year, the high and very high level were found for more than >50% in both villages.

Spatial distribution of risk level of each observation point is presented in a map (Figure 10). Different colour represents different levels of risk, while the size of the dots represents cropland area. This map can be used to communicate risk to concerned parties especially to policy maker as one consideration to develop human health and environmental protection program.

Figure 10 Risk level distribution map in study area
4. Conclusions
Organophosphate pesticides (OPPs) was the second most commonly used type of pesticides in study area. The type of OPPs used contain active ingredients of profenofos, chlorpyrifos, and diazinon. Profenofos was used on almost all types of crops, followed by chlorpyrifos and diazinon. Loading rate estimation ranges from 0.003 to 4.74 gr/year in rainy season and 0.001 to 0.93 gr/year in dry season. According to risk score analysis and risk level classification, it was found that in rainy season, there were areas classified as very high risk, while it is not the case in dry season. However, in total loading per year, areas with high and very high risk level were found for more than >50% in study area

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