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Organophosphate Pesticide Residue in Fruits and Vegetables

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Abstract

The use of the pesticide which does not follow the procedure may potentially cause residue of pesticide to be left on vegetables and fruits sold in supermarkets and traditional markets. We aimed to find the pattern of pesticide usage and levels of residue found in plants. This study was an analytical observation with a cross-sectional design using detailed sampling procedure. We obtained the data of the pesticide residue concentration by conducting laboratory examinations on eight samples of cabbage and eight samples of tomatoes from traditional markets and supermarkets in the city of Solo. The data were tested using simple linear regression testing. The organophosphate measurement results showed no residues were detected, because it was below the Limit of Detection (LOD). We concluded that pesticides containing active substances were not used in the vegetables we tested or the active substances were no longer contained in the vegetables after harvesting.

Introductions

One of the problems complained by farmers is the existence of Plant-Disrupting Organisms (PDO). There was never any solution that could solve the PDO problem completely, therefore causing the resulting crops to be below expectation. The low number of crops produced means a low income for farmers. The leading cause of the problem is the incomplete implementation of the Integrated Pest Control (IPC) system. One of the IPC components is a prudent, environmentally sound, and sustainable use of pesticide. A pesticide is all of chemical compounds, along with microorganisms and viruses, which are used for controlling various pests.

In recent years, there has been an increasing use of pesticide because it is considered the most effective way to control PDO, raising the demands for pesticides among farmers. Multinational pesticide companies dominated 64% of the total market, about 5.7 trillion rupiahs. The largest pesticide user segments were paddy (41%) and palm plantation (27%) segments. The majority in pesticides market were herbicides (42.5%), pesticides (37.5%), fungicides (18%), and other groups (2%) (BPS, 2012).

Today farmers still depend on pesticide use because it is a fast, efficient, simple, safe, and useful method even though the PDO problems were never solved completely. Some assume that pesticide is an efficacious drug and it is hard to increase their plant productivity without it. It provokes overuse, imprecise, and unwise use of a pesticide. As Untung stated in 2004, Indonesian farmers tended not to take a risk, specifically plant commodities that are prone to

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attack from PDO such as the vegetables. Many of them still relied on periodic pesticide spray.

Uncontrolled and overuse of pesticide cause negative effects on the environment and humans. Pesticide intoxication is one of the negative impacts on humans. Some of the organophosphate components will enter the body through oral, inhalation, and skin absorbtion. Organophosphate pesticides (DDPV, diazinon, malathion, and parathion) cause intoxication by ChE (Cholinesterase) enzyme blocking. It ties ChE in the blood plasma, red blood cell, and nerve synapse making it inactive and unable to hydrolyze Acetylcholine (Ach). Consequently, it provokes paralysis of the body because of Ach deposit in the muscle cell receptor and gland. It also causes stimulation of nicotinic and muscarinic cholinergic because of Ach accumulation in cholinergic nerve-ends, both neuro-effector junction and ganglion.

Muscarinic receptor stimulation causes hypersalivation, hypermotility of the digestive system, hypersecretion of the airway, and bronchoconstriction. The sympathetic nerves innervate the sweat glands and has cholinergic fiber. Therefore, there is a presence of excessive sweating as the result of organophosphate intoxication. On the other hand, nicotinic receptor stimulation in the autonomic ganglion will trigger excitation symptoms from both the nervous systems, although muscarinic symptoms will dominate it in the end. Muscarinic stimulation in the end-plate motor will provoke muscle spasm, fasciculation, and airway paralyzing, with the presence of dyspnea and cyanotic. Also, muscarinic stimulation in the central nervous system will cause mental tension, anxiety, agitation, insomnia, unstable emotion, neurosis, and convulsion. Mortality of this intoxication is caused by respiratory failure, as the result of airway muscle paralyzing.

Most of the pesticides used by the farmers will reside in the fruits and vegetables as a pesticide residue. One of the leftover residues is the organophosphate. Improper use of pesticide may leave high amounts of the residue. According to Dasika, Tangirala and Naishadham in 2012, there were many fruits (green apple, yellow apple, crisp pink apple, green grape, black grape, pear, and guava) and vegetables (red paprika) that contain beyond-threshold pesticide residue (chlorpyrifos, thiabendazole, and malathion) in India. Another study in Indonesia showed that the dosage of malathion consumed from contaminated vegetables for 60 consecutive days caused a clear damage in the liver but a less clear damage to in the mice brain (Hamzah, 2009). Ray *et al.,* (2008) stated that carbaryl organophosphorus residue in the meat (0.0541 mg/mL), egg (0.0506 mg/mL), and milk (0.0453 mg/mL), which is exactly at the threshold.

Farming results such as fruits and vegetables are also sold in the market, instead of being directly consumed. Markets in Solo City area stock their agricultural products from its nearby area, like Karanganyar and Boyolali Regency. It was assumed that the fruits and vegetables in Solo were contaminated by organophosphate. Many traditional markets in Solo, such as Gede, Legi, Hardjodaksino, Danukusuman Markets, and some supermarkets collected their vegetables and fruits supplies from mentioned regencies. We do not know whether the vegetables contain pesticide residue or not. There has been no calculation of pesticide contained in the plants, which could potentially affect the health of the consumers.

From the description above, it raises the question of how much organophosphate pesticide residue is found in the fruits and vegetables that were sold in supermarkets and traditional markets in Solo City. We also question whether there were any difference in residue level between produces sold in a supermarket and traditional market. Our study aimed to observe the amount of organophosphate residue in the fruits and vegetables and the difference in residue level between produces sold in supermarkets and traditional markets.

Methods

We used an observational, analytical study using cross-sectional with classified sampling technique. Our research took place in Solo City area, where the traditional and supermarkets supplied their fruits and vegetable from Boyolali and Karanganyar Regency. Their supplies were suspected to be contaminated by organophosphate pesticide.

We collected cabbages and tomatoes form eight locations, four of them were traditional markets in Solo which were the Gede, Gemlegan, Karangasem, and Legi Markets. The four supermarkets were LotteMart Tipes, Hypermart Grand Mall, Carrefour Paragon and Super Indo, so we collected eight cabbages and eight tomatoes.

The chemical ingredients used were the solvent (acetone, CH2Cl2, and petroleum ether), sodium sulfate (anhydrous), floristic (particle size 0.150-0.250 mm for column chromatography), and pesticide.

The analytical tools for this study were blender, Erlenmeyer (size of 125 and 250 ml), beaker glass (volume of 25 and 50 ml), funnel, absorbent paper, measuring cups (size of 10 and 100 ml), micropipette, syringe (10 µl), scale (Mettler Toledo), evaporator (Airflow monitor, Mach aire Ltd), testing tube, chromatography column, and gas chromatography (model 8000 TOP).

We collected the samples from the traditional and supermarkets. The procedures were similar to pesticide residue testing in agricultural products. The samples were sent to the laboratory and measured using thin-layer chromatography method (Kanasawa, Sutrisno, and Otorita, 1983).

Later, the organophosphate pesticide analysis was conducted using the gas chromatography (model 8000 TOP) which equipped with an Electron Capture Detector, according to AOAC (1990). The steps were: extracting the cabbage, cleaning up, making the standard solution, and doing quantitative analysis (residue level count). Recovery test was a standard to determine whether the used methods were well enough.

Results and Discussions

The traditional and supermarkets in Solo City supplied their vegetables and fruits from the farmers in Central Java (Boyolali and Salatiga) and East Java (Magetan and Jombang) regions. Karanganyar Regency is one of the agricultural products-maker which is sold in Solo City. Two Sub-districts, Mojogedang and Karangpandan, are the tomatoes and cabbages producing area.

There are many benefits from the use of pesticide, even though they are unknown to the public. Its clear and a more accessible to count advantage is the economic benefits from the farmers in the form of product protection and quality. Without it, the number of food products will decrease, and the price will continue to rise. Consequently, the farmers will not be competitive in the market. The use of pesticide is a method to reduce loss caused by pests. It may increase the number of crops with an accessible price for the customers, which is necessary for them (Damalas, 2009). Nevertheless, many farmers in Karanganyar and Boyolali used pesticide to control PDO.

Table 1. Organophosphate pesticide residue levels in the tomato samples from the traditional markets of Solo City

No	Test Parameter	TR(B)	TR(S)	SP(B)	SP(S)	Level of Detection (LoD) (mg/kg)
1	Diazinon	<$3.84x10^{-3}$	<$3.84x10^{-3}$	<$3.84x10^{-3}$	<$3.84x10^{-3}$	$3.84x10^{-3}$
2	Parathion	<$0.82 x10^{-3}$	<$0.82 x10^{-3}$	<$0.82 x10^{-3}$	<$0.82 x10^{-3}$	$0.82 x10^{-3}$
3	Ethion	<$2.76 x10^{-3}$	<$2.76 x10^{-3}$	<$2.76 x10^{-3}$	<$2.76 x10^{-3}$	$2.76 x10^{-3}$
4	Profenofos	<$0.80 x10^{-3}$	<$0.80 x10^{-3}$	<$0.80 x10^{-3}$	<$0.80 x10^{-3}$	$0.80 x10^{-3}$
5	Malathion	<$0.50 x10^{-3}$	<$0.50 x10^{-3}$	<$0.50 x10^{-3}$	<$0.50 x10^{-3}$	$0.50 x10^{-3}$
6	Chlorpyrifos	<$0.33 x10^{-3}$	<$0.33 x10^{-3}$	<$0.33 x10^{-3}$	<$0.33 x10^{-3}$	$0.33 x10^{-3}$

Legends:
TR(B) : Tomato samples from Gede, Gemblegan, Karangasem, and Legi Market
TR(S) : Cabbage samples from Gede, Gemblegan, Karangasem, and Legi Market
SP(B) : Tomato samples from LotteMart, Hypermart Grand Mall, Carrefour Paragon, and Super Indo Banjarsari
SP(S) : Cabbage samples from LotteMart, Hypermart Grand Mall, Carrefour Paragon, and Super Indo Banjarsari

They mostly use organophosphate pesticides, such as Azinphosmethyl, Chloryfos, Demeton Methyl, Dichlorovos, Dimethoate, Disulfoton, Ethion, Parathion, Malathion, Diazinon, Chlorpyrifos, Fenitrothion, Temefos, and metil-pirimifos. This pesticide was first introduced in Germany during the World War II. During its early synthesis, the substances tetraethyl pyrophosphate (TEPP), parathion, and schordan were produced which were useful as pesticide, but they were also toxic to mammals (Prijanto, Nurjazuli, and Sulistiyani, 2009). The study regarding this issue is still being developed. They found an active component for the insects, such as malathion, that had a less toxic effect to the humans (Alegantina et al., 2005).

We examined the pesticide residue level by checking the cabbages and tomatoes in eight places; four traditional markets in Solo City: Gede, Gemblegan, Karangasem, and Legi Market, and four supermarkets: LotteMart Tipes, Hypermart Grand Mall, Carrefour Paragon and Super Indo.

The table above showed that all the samples did not contain organophosphate pesticide. The table also showed that organophosphate was not detected in any of the samples. The diazinon level was <3.84x10-3 mg/kg, which was below the threshold, Parathion <0.82 x10-3 mg/kg, Ethion <2.76 x10-3 mg/kg, Profenofos <0.80 x10-3 mg/kg, Malathion <0.50 x10-3 mg/kg, and Chlorpyrifos <0.33 x10-3 mg/kg. All of them were under the threshold level (LoD).

Alegantina e*t al.,* (2005) described a negative result or absence of the organophosphate residue, such as diazinon, chlorpyrifos, metidation, phention, phenitroin, phenthoate, profenofos, protiofos, triazophos, methamidophos and dimethoate, within tomatoes and lettuces from various traditional and supermarkets in Jakarta. Srivastava *et al.,* (2014) showed the presence of low level organophosphate residue, Malathion and chlorpyrifos, in the mango examined in Lucknow, India. Putu *et al.,* (2016) analyzed the presence of benzophenone, 2,6-diisopropylnaphthalene, and ftalat acid, instead of organophosphate, in the strawberry samples from Candikuning Market, Bedugul, Bali. According to our results, organophosphate levels (diazinon, parathion, ethion, profenofos, malathion, and chlorpyrifos) were below the LoD because they were undetected by the tools (Hartini, 2014).

There were some studies that showed a different result. Chandra et al., (2014) described chlorpyrifos, cypermethrin, and monocrotophos pesticide residue levels in the eggplants, capsicums, and cabbages using gas chromatography which was detected below the maximum limit. Another study in India revealed the levels were beyond the threshold in some fruits (green apple, yellow apple, crisps pink apple, green grape, black grape, pear, and guava) and vegetables (red paprika) (Dasika et al., 2012). A study using 1,423 samples consisted of 573 fruits and 830 vegetables conducted in Aegean, Turkey, demonstrated as many as 84 (4.8%) of the fruits and 83 (9.8%) of the vegetables contained acetamiprid, carbendazim, and chlorpyrifos pesticide beyond the threshold (Bakirci et al., 2014).

The reason for undetected residue or levels below the LoD in mentioned studies, including ours, was either the absence of pesticide's active ingredients or the harvested vegetables no longer contain residue. The residues were affected by many factors, such as persistence and application technique of pesticide, climate, also the weather.

There is an unstandardized pesticide handling practiced by the farmers in Karanganyar and Boyolali, such as mixing over one type of pesticides with the help of water as a solvent. It is common to find farmers who mix pesticides at their homes, starting from opening its packaging, pouring into the container, until stirring it, which can potentially harm their family members. It can potentially harm the farmers themselves and society (Prijanto et al., 2009). Yuantari et al., (2015) study at Curut Village, Penawangan Sub-district, Grobogan Regency using 54 melon farmers also described the farmers unstandardized behavior, which were combining over one type of pesticide, some even mixed more than ten active ingredients into one tank. Moreover, this action will lead to undetected pesticide residue, as gas chromatography is unable to read the mixed formula (Hartini, 2014).

The farmers in Karanganyar and Boyolali

sprayed the pesticide during the beginning of the cabbage and tomato planting. They did not spray it when the harvesting time had arrived. The last spraying was done way before the harvesting time. This caused a low, or even an absent, pesticide level because it had already dissolved.

The vegetables and fruits were cleaned using water before being distributed to the traditional and supermarkets in Solo City. The cleaning process is not only conducted before harvesting but also during the distribution from the farmers to the merchants or customers. Farmers usually clean the vegetables before handing them to the merchant; and they will clean them again using water to keep its freshness. This action will lead to a decreasing number of pesticide residue. A study by Alen et al., (2015), showed a decreasing number of residue in lettuce after being cleaned by antiseptic solution, as much as 0.061 ppm from 0.204 ppm, reduced by 70.1%.

Rainfall cleansing would also reduce the residue. Furthermore, the organophosphate will evaporate after being sprayed because organophosphate is volatile when exposed to sunlight, photodecomposition, and chemical reaction, and it also easily decompose. The half-life for profenofos pesticide is only one week. It is also volatile when exposed to sunlight. Moreover, the residue may decrease as the distribution process occurs between the farmers and consumers because of sunlight or rain.

The farmers in Karanganyar have also added the limestone into the cabbages which contained calcium carbonate ($CaCO_3$) in the form of calcite mineral. They discarded the outer layer before putting them on sale to remove the limestone preservative. It is also a reason for undetected residue because the inner layer of cabbage is not affected by pesticide spraying.

Conclusions

Our organophosphate residue test revealed all the samples did not contain organophosphate because all of them were below the level of detection (LoD). It is necessary to conduct a further study with a more significant number of samples and different places. We suggest consumers to be more careful in choosing vegetables. We also recommend farmers using pesticide to follow the predetermined rules and give socialization for proper use the pesticide. Lastly, we acknowledged LPPM UNS and LPPT UGM for their generous help in our study.

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