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HEALTH RISK ASSESSMENT OF PYRETHROID PESTICIDE RESIDUES IN PRODUCES AND SOILS IN A FARMLAND FROM ABEOKUTA, OGUN STATE, NIGERIA

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ABSTRACT

Despite being useful in pest control, pyrethroids have been found to bioaccumulate in aquatic organisms and in human breast milk and research has shown that pyrethroids are neurotoxic, affect reproductive health and have ability to interfere with the immunological and endocrine systems, they also function as cancer precursors. This study sought to determine the amounts of pyrethroid residue in the soils and some of the most popular food crops, yam and cassava, in order to evaluate food safety using Gas chromatography Mass spectrometry (GC-MS). Cyfluthrin, alpha cypermethrin and beta cypermethrin are pyrethroids found in all the yams and cassava and soils analysed in this study and their values ranged from 9.98mg/kg to 14.45mg/kg in yam farm soil, 8.1mg/kg to 18.46mg/kg in yam samples, 10.25mg/kg to 14.94mg/kg in cassava samples and 6.13mg/kg to 30.6mg/kg in cassava farm soils. The pyrethroid with the highest residue levels in all the samples taken was beta cypermethrin. The health risk was assessed by calculating bioaccumulation factor and comparing Acceptable Daily Intake (ADI) with Food and Agricultural Organization (FAO) recommendations. Bioaccumulation factor of the foods analysed ranged from 0.49 (beta cypermethrin) to 1.68 (alpha cypermethrin) in cassava and 0.56 (cyfluthrin) and 1.07 (alpha cypermethrin) in yams. The pyrethroid residues discovered in the foods in this study exceeded the ADI of pesticides established by FAO, 0.02mg/kg per day; a significant health risk indication. Care should therefore be taken when using these pesticides and consumption of food exposed to pesticides should be discouraged.

Keywords: Bioaccumulation, Cyfluthrin, Cypermethrin, Insecticide, Pyrethroids

INTRODUCTION

Insecticides, herbicides, fungicides, rodenticides, wood preservatives, garden chemicals, and household disinfectants that either kill or protect against pests are together referred to as pesticides (Bhatt *et al*., 2019, Udoh and Gibbs 2022). These pesticides vary from one class to another in terms of their physical, chemical, and similar qualities (Rajveer *et al*., 2019). One of the most effective insecticides now in use is pyrethroid, they are chemically produced from pyrethrin I, one of the six active ingredients in pyrethrum, a substance obtained by extracting the dried flower heads of Chrysanthemum genus (Kaur and Sinha, 2019). Natural pyrethrins are good insecticides with little mammalian toxicity, but their application is constrained by their high biodegradability and low photostability (Oshatunberu *et al*., 2023). Pyrethroids, on the other hand, offer a great deal of agricultural potential, highly toxic to a variety of insects, and are largely non-toxic to mammals. Moreover, pyrethroids appear to not accumulate in the environment and are considerably less persistent than organochlorine insecticides like DDT and dieldrin (Jayaraj *et al*., 2016).

As a less dangerous alternative to the more hazardous organophosphate and carbamate insecticides, pyrethroids were initially created in the late 1940s (Oshatunberu *et al*., 2023). Currently, pyrethroids are frequently utilized in household and agricultural pesticides, veterinary medications, and pet items (Matsuo, 2019; Oshatunberu *et al*., 2023). Based on their chemical makeup, pyrethroids can be divided into Type I and Type II pyrethroid structures, which cause T and CS syndromes, respectively. Type II pyrethroid pesticides have an alpha-cyano moiety at the phenyl benzyl alcohol position, but type I pyrethroid pesticides do not have one at the alpha-position. The choreoathetosis syndrome, often known as "CS syndrome," is characterized by symptoms in insects such as hyperactivity, a bent back, salivation, tremors, and lack of coordination that proceed to sinuous writhing motions. T syndrome is noted for symptoms such as tremors, incoordination, prostration, seizures, and death (Litwin *et al*., 2023). Although they are typically thought to be less harmful to man and animals than the older insecticides they replaced, they can nevertheless found to be harmful to health if applied incorrectly (Ariyani *et al*., 2020). The metabolites of pyrethroids were proven to be present in the urine following consumption of semolina (pasta), rice, whole-wheat bread, breakfast cereals, and fruits from sprayed areas and, due to their lipophilicity, pass through the blood-brain barrier at levels deemed neurotoxic (Costa 2015, Chrustek *et al*., 2018). Recent research has also shown that pyrethroids can bioaccumulate in aquatic organisms and in human breast milk (Litwin *et al*., 2023). They have also been proven to be transmissible through the mother in marine animals and poultry (Litwin *et al*., 2023). Additionally, numerous studies have emphasized the neurotoxicity of pyrethroids, their risks for both male and female reproductive health, their potential to disrupt the endocrine and immune systems, and their role as cancer precursors (Chrustek *et al*., 2018; Wang *et al*., 2020; You and Song, 2021). Morgan (2020) suggested that a major source of exposure to pyrethroids in adults is food, pyrethroids are quickly absorbed, broken down, and mostly eliminated through the kidneys after food consumption and urine elimination half-lives are typically under 15 hours. However, recent cross-sectional studies have raised questions about the possibility that pyrethroid exposure in the environment may harm adults' neurological, endocrine and heart health (Hołyńska-Iwan *et al.*, 2020). In addition, Tang *et al*. (2018), reports that long-term, low-dose exposure to pyrethroids can cause chronic diseases and have toxic effects on the nervous, immune, cardiovascular, and genetic systems of organisms, inducing teratogenicity, carcinogenicity, and mutagenicity.

Available knowledge on pyrethroids is limited as compared to other types of pesticides and understanding of health risks of pyrethroids is quite recent. As exposure to knowledge about pyrethroids increase, regular environmental toxicology studies in areas where pesticides are used, such as farmlands, barns, poultry facilities, ranches pig pens, to mention a few is crucial. This research seeks to assess the level of pyrethroid residue in the soils in Wanderland farms, a settlement in Odeda local government area of Abeokuta, Ogun State and the crops grown on the soils; Yams and Cassava which major staples in Nigeria and evaluate the potential risk to consumers.

MATERIALS AND METHODS

Study Area

The study area is a farmland located between latitudes 7.179853/N7º10ˊ47.472" and longitudes 3.438319/E3°26ˊ17.948" (Figure 1). It is located in Kotopo area of Obantoko, Odeda Local Government Area, Abeokuta, Ogun State. Major food crops cultivated on this Wonderland community farm settlement include *Manihot esculenta* (Cassava), *Discorea spp* (Yam), *Zea mays* (Maize), and *Musa paradisiaca* (Plantains).

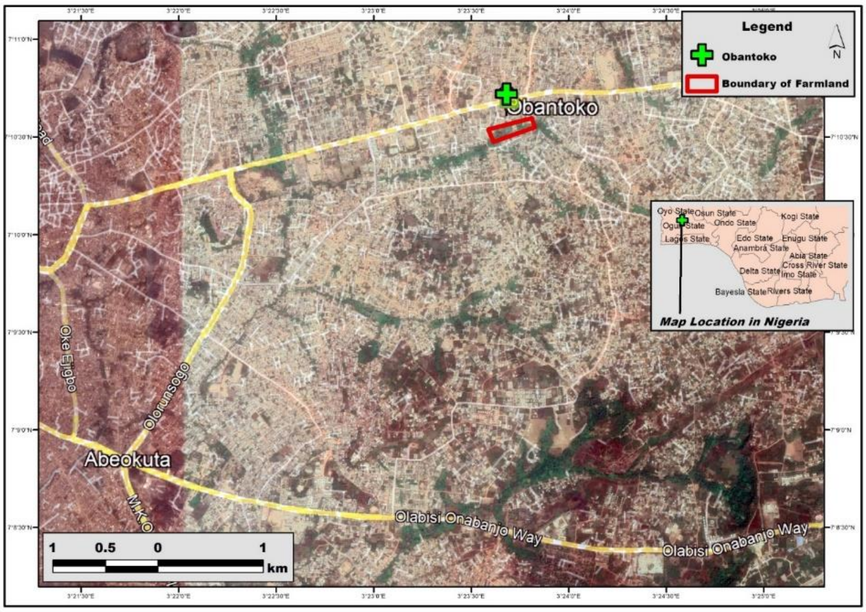

Figure 1: Wonderland community settlement Farmland, Obantoko, Abeokuta, Nigeria

Sample Collections

Collection of Soil Samples

Soil samples were collected similar depths of 0-20cm from 5 random locations on the cassava farm plot. The soil samples were then thoroughly mixed to ensure that the sample was a good representation of the various sections where the soil was collected. The same procedure was applied for the collection of soils from the yam farm plot. The soil samples were collected in plastic bags and tagged appropriately.

Collection of Food Samples

Manihot esculenta (Cassava) and *Discorea spp* (Yam) samples were harvested and collected from the same farms where the soil samples were collected, tagged and taken to the laboratory.

Gas Chromatography–Mass Spectrometry (GC–MS) Determination of Pyrethroids in Samples

10g ± 0.05g of each tuber sample was weighed and cut into pieces into 250mL Teflon bottle, at room temperature. 15ml of 6N potassium hydroxide was also added to the sample in the Teflon bottle. About 3 spatula full of activated sodium sulphate was added to the samples in the Teflon bottles in order to eliminate water/aqueous portions. 20mL of 1:1 acetone: hexane was used for extraction procedure thrice, giving ~60mL of final extracting solvent. The covered Teflon bottles were then sonicated in an ultrasonic bath at 70 °C for 30 min (Akinsanya *et al.,* 2020). Organic layer was decanted into a clean beaker/round-bottom flask, further dried with sodium sulphate and clean-up procedure using silica gel column carried out. The sample extract was then concentrated to 2 mL using a rotary evaporator prior to Pyrethroids analysis using gas chromatography mass spectrometer (GC-MS).

Organophosphorus Pesticides (OPPs) standard, 100ppm (Catalog Number: M-8080) containing 3 Pyrethroids components was purchased from AccuStandard. Five (5) point serial dilution calibration standards (0.25, 0.50, 1.00, 2.00, 4.00ppm) was prepared from the stock and used to calibrate the GC-MS (Okeagu *et al.,* 2021).

Prior to calibration, the mass spectrometer (MS) was auto-tuned to perfluorotributylamine (PFTBA) using already established criteria to check the abundance of mass to charge ratio (*m/z)* 69, 219, 502 and other instrument optimal and sensitivity conditions. Determination of the levels of OPPs in the sample was carried out using GC-MS by operating MSD in selective ion monitoring (SIM) and Scan mode to ensure low level detection of the target constituents.

Purge flow to spilt vent was 30.0 mL/min at 0.35 min with a total flow of 16.36 mL/min; gas saver mode was switched off. Oven was initially programmed at 80 °C (1 min) then ramped at 10 °C/min to 300 °C (10 min). Run time was 33 min with a 3 min solvent delay. The mass spectrometer was operated in electron-impact ionization mode at 70eV with ion source temperature of 230 °C, quadrupole temperature of 150 °C and transfer line temperature of 300 °C. Acquisition of ion was via Scan mode (scanning from *m/z* 50 to 500 amu at 2.0s/scan rate) and selective ion mode (SIM). After calibration, the samples were analysed and corresponding OPPs concentration obtained.

Acceptable Daily Intake (ADI)

The ADI is expressed in milligrams of the chemical, as it appears in the food, per kilogram of body weight per day (mg kg^{-1} day^{-1}).

$$= \frac{concentration\ of\ pesticide\ \times average\ consumption\ of\ tuber}{1000}$$

Bioaccumulation Factor The bioaccumulation factor (BAF) was used to evaluate the effectiveness of a plant in metal accumulation and translocation, It is well known that plants acquire metals from contaminated soil and from deposits on plant parts exposed to air from polluted surroundings (Adedokun *et al.,* 2016). Biological accumulation factor (BAF) was defined as the concentration of heavy metals in plant shoots divided by the heavy metal concentration in soil

$BAF = \frac{Metal\ in\ shoot}{Metal\ in\ Soil}$ ……..by (Adedokun *et al.*, 2017).

For this study, it was revised thus;

$BAF = \frac{Pyrethroid\ in\ Tuber}{Pyrethroid\ in\ Soil} - - - - -$ Li and Ai (2023)

Statistical Analysis

The experimental analysis was done using R programming language, version 4.2.2. Descriptive statistics distribution, cumulative frequencies, graphs and percentages were used to describe the findings according to each specific objective.

RESULTS AND DISCUSSION

Three pyrethroids were detected in this study; alpha-cypermethrin, beta-cypermethrin and cyfluthrin. These three pyrethroids were found on the farm soils and food products analysed at various levels as shown in Figures 1 and 2. In both Cassava farm soil and Yam farm soil the level of beta-cypermethrin was found in higher concentrations than the other two.

It is generally expected that the residues levels found in the soil would be more than the levels found in the food grown on them as a result of adsorption and translocation from regions of higher concentration to areas of lower concentrations (Bondareva and Fedorova, 2021) in both yam and cassava farms the soils had more beta-cypermethrin and cyfluthrin than the yam and cassava harvested from the soils. However, alpha-cypermethrin residues were higher in both yam and cassava than the soils from which they were harvested. This could have to do with the way alpha cypermethrin was degraded in the soil. Xu *et al*. (2015) reported significant microbial degradation of alpha cypermethrin, although microbial activities were not considered in this work it is probably responsible for the reaction that allows the yams and cassava degrade and accumulate alphacypermethrin in the tissues.

During verbal interactions with the farmers it was discovered that pesticides were hardly used on the farm, if at all it is used it is once a year while preparing the soil for cultivation herbicides may be used to help clear the weeds if necessary. This information does not tally with the pyrethroid residues found in the soil because half-lives of pyrethroids are from 5 to 170days (Laskowski, 2002) this is one of the advantages of pyrethroids and other organophosphate pesticides over organochlorines. There are probably other factors that need to be investigated in future studies such as topography of the soil and closeness of the farm to other pyrethroid sources.

It was observed in this study that beta cypermethrin residues in both yam and cassava was higher than the other pyrethroids present. According to EFSA 2014, “beta-cypermethrin is of high acute oral toxicity, of moderate toxicity by the inhalation route and of low dermal toxicity”. It remains to be discovered the effects of residues of these pyrethroids taken over varying periods of time.

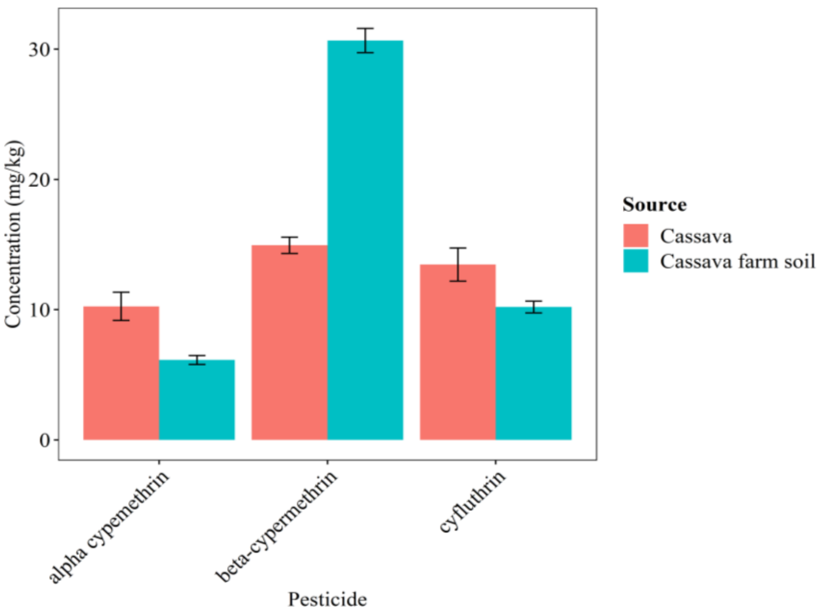

Figure 2: Pyrethroid pesticides in harvested cassava and cassava farm soils

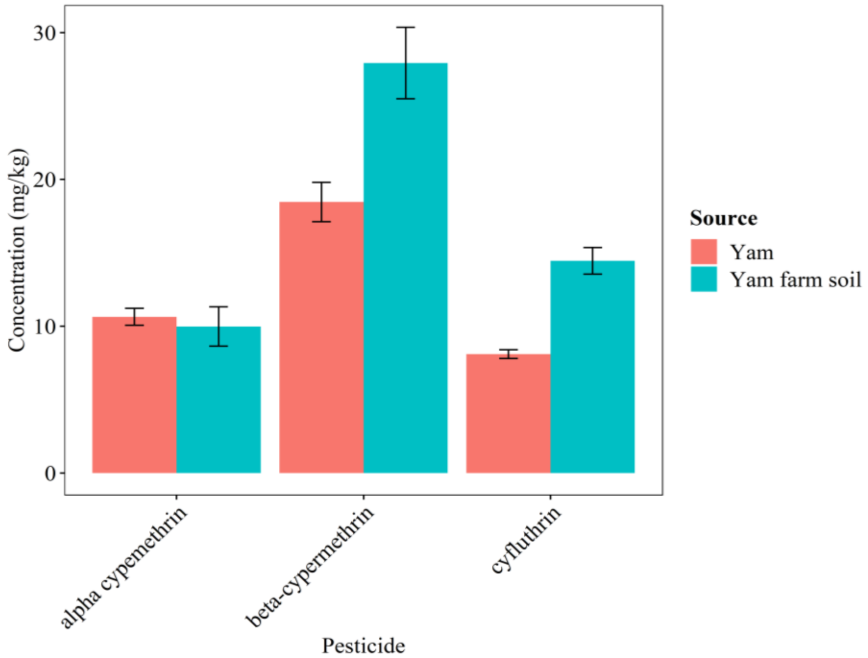

Figure 3: Pyrethroid pesticide in harvested yams and yam farm soils

Bioaccumulation Factor and Acceptable Daily Intake
Documented research has suggested that the general populace is exposed to substantially varied quantities of pesticide residues through food and water, people and livestock who are not in the vicinity of where pesticides are employed are also shown to be exposed to some risks (Morgan, 2020 and Miller *et al.* 2020. Bioaccumulation is a critical concept used in ecological risk assessments to determine the extent of pollutant transport within food webs. According to Miller *et al.* (2020), bioaccumulation is occurring when uptake of a contaminant is greater than the ability of an organism to egest a contaminant, in this study the contaminant is Pyrethroid pesticides. Bioaccumulation and subsequent trophic transfer of a contaminant may result in the biomagnification of these contaminants at higher trophic levels. This understanding led to the assessment of the daily intake of the pyrethroids studied. The acceptable daily intake (ADI) is commonly defined as the maximum amount of a chemical to which a person can be exposed, on a daily basis over an extended period of time, usually without suffering a deleterious effect. Daily intake of the various pyrethroids found in this study; alpha-cypermethrin, beta cypermethrin and cyfluthrin were computed based on the residue levels detected in both the food and the soil and the average tuber consumption. It is estimated that the alpha cypemthrin and beta cypermethrin residue intake from yam and cassava ranges from 2mg/kg per day in cyfluthrin in yam to 4.8mg/kg per day, beta cypemethrin in take from yam tubers as shown in Figures 3 below. Table 1 also shows the Acceptable Daily Intake (ADI) of alpha and beta cypermethrin and cyfluthrin as recommended by Food and Agricultural Organization.

Based on the data we have from Table 1, the pyrethroid residue levels in tubers in this exceed the ADI and from table 2, the bioaccumulation factor of cyfluthrin and alpha cypermethrin in cassava was shown to be higher than in yam. Elobeid *et al.*, 2021 in a similar research conducted on pesticide residues in fruits and vegetables found over 18% of all 49 samples analysed to have exceeded safe limits and maximum residue levels. Okeagu *et al.*, 2022, also did a study on *Amietophrynus regularis(common toad) and reported varying concentrations of pesticides in their liver and intestines. These studies as well as my findings indicate the presence of pesticide residues in the environment and in living organisms and buttress the need for continuous monitoring and further toxicology studies as these pesticide residues have been shown to move through the food chain.*

It is safe to speculate based on this study that from the consumption of yam and cassava alone, unsafe levels of pyrethroids are consumed daily with potentially dangerous accumulation rates.

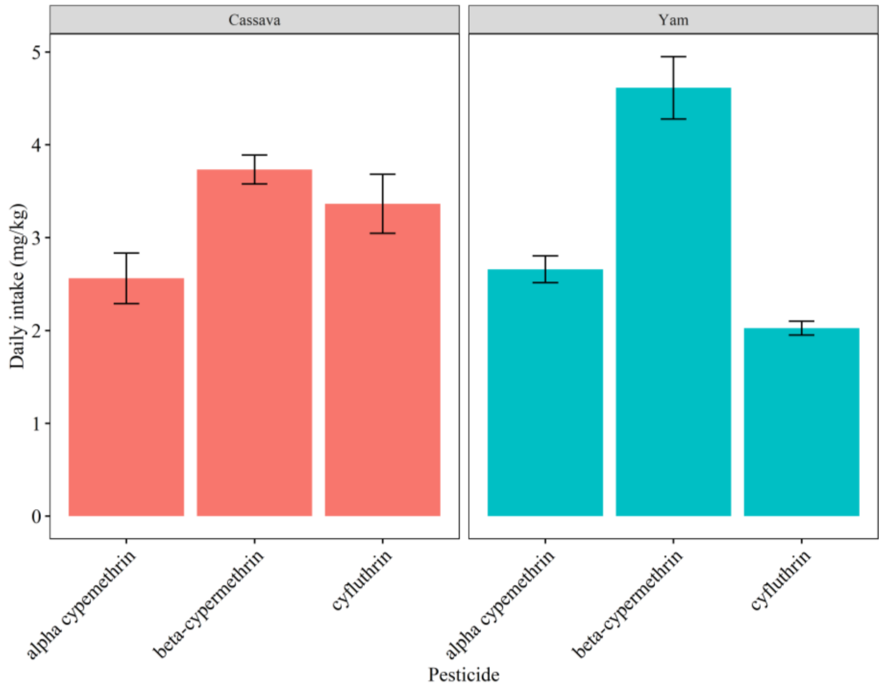

Figure 4: Daily intake of Pyrethroids pesticide in the yam and cassava

Table 1: Acceptable daily intake (mg/kg body weight per day) of pesticides in food

Group	Pesticide	ADI	Reference
Pyrethroids	Cyfluthrin	2e-02	FAO/WHO (2016)
Pyrethroids	Beta-cypermethrin	2e-02	FAO/WHO (2012)
Pyrethroids	Alpha-cypermethrin	2e-02	FAO/WHO (2003)

Table 2: Bioaccumulation factor of Pyrethroids pesticides in yam and cassava tubers

Group	Pesticide	Bioaccumulation factor µg /kg	
		Yam	Cassava
Pyrethroids	cyfluthrin	0.56	1.32
Pyrethroids	beta-cypermethrin	0.66	0.49
Pyrethroids	alpha cypemethrin	1.07	1.68

Figure 5 and Figure 6 are scatter plot representations of the relationship between pyrethroids in cassava and cassava plot and pyrethroids in yam tubers and yam plots. Observing the scatter plots it is seen that the clusters of each pyrethroid observed in this study; cyfluthrin, beta cypermethrin and alpha cypermethrin are in similar clusters both in yam and cassava. Beta cypermethrin was higher than both alpha cypermethrin and cyfluthrin in both the tubers yam and cassava and the soils fem which they were harvested. The cypermethrin in yam plot is significantly higher than the quantity found in the yam itself, this is consistent with what is described by (Bondareva and Fedorova, 2021). The higher quantity in the soil is so most likely because it is the source of the pesticide residue. There is higher quantity of alpha cypermethrin and cyfluthrin in cassava than in the soil. This could be because cassava can accumulate the pesticide more or the cassava absorbed the cypermethrin and cyfluthrin from other sources. Probably water runoff during the rains from other farms. During the period of this study conversations with the farmer revealed that the only pesticide used on the farms are herbicides, (trade name force-up) once a year before planting season. This herbicide is an organophosphate. The question now becomes where did the pyrethroids found in the soil come from? Could it be that the herbicides used contain more than what is written on the label or runoff during the rains as mentioned earlier brought the pyrethroids found from other sources?

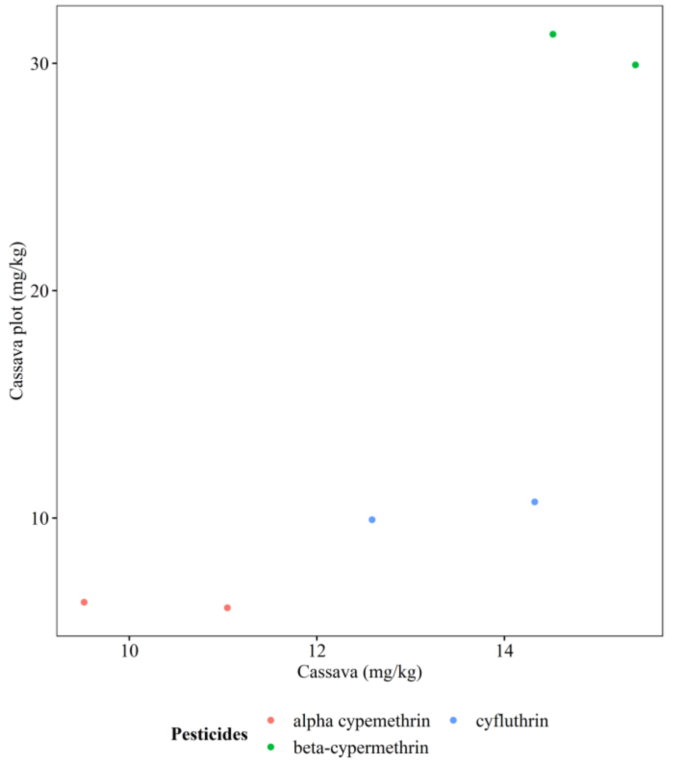

Figure 5: Scatter plot showing a relationship between pyrethroids in cassava and pyrethroids in cassava plot

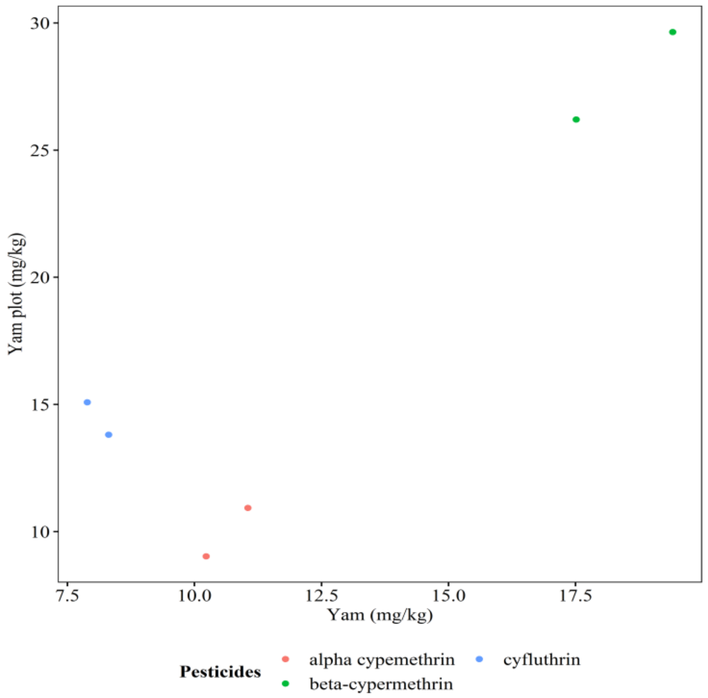

Figure 6: Scatter plot showing a relationship between yam and yam plot in Pyrethroids pesticides group

CONCLUSION

While pyrethroids are often considered safer alternatives to many other pesticides, the presence of pyrethroid residues in the environment and food products underscores the importance of vigilance to prevent potential issues arising from their usage. Our research highlights the necessity for a more comprehensive examination of these pesticides, with the aim of providing farmers and the general public with a better understanding of their usage, associated risks, and potential hazards.

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