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ORGANOCHLORINE AND ORGANOPHOSPHORUS PESTICIDE RESIDUES IN GRAINS, VEGETABLES AND FRUITS: A REVIEW

[*1]Okereafor, S., [2]Garba, S., [3]Okunola, O. J., and [4]Adamu, H. I

[1]Department of Chemistry, Air Force Institute of Technology, Kaduna, Kaduna State
[2]Department of Chemistry, Nigeria Defence Academy, Kaduna
[3]Department of Applied Chemistry, Federal University Dutsin-Ma, Katsina State
[4]Department of Environmental Management Kaduna State University, Kaduna State

*Corresponding authors' email: okereaforstella577@gmail.com Phone: +2348034181861

ABSTRACT
Organochlorin and organophosphorus pesticides are broadly applied in agriculture, primarily to boost crop yields and protect crops from pests to reach the needs of a growing global population. This review aims to assess levels of pesticide residues in grains, fruits, and vegetables from already established secondary data from different authors or journals. The data obtained adequately confirmed the presence of concentrations of Organochlorin and organophosphorus residues in vegetables, grains and fruits. Both high and low concentration levels of pesticide residue were documented based on data obtained which showed application of the synthetic pesticides by farmers. The high or minute concentration of synthetic pesticides poses adverse health effect on human. This review suggest that consumers of these type of pesticides contaminated food are liable to contact diseases associated with pesticides. Pesticides have been related to damage to the kidneys, liver, and nervous system, birth defects, cancer, immunodeficiency, reproductive process disruption, and altering or interfering with normal endocrine system function. Thereby, the use of natural pesticides as an alternative to synthetic pesticides for pest control is highly recommended for food security and sustainable environmental practices.

Keywords: Organochlorine, Organophosphorus, Pesticides, levels of residues

INTRODUCTION

Pesticides are chemicals or mixtures of chemical compounds used to defend crops against pests like fungi, weeds, and insects US Environmental, (2007). Chemical substances, biological agents (such as viruses or bacteria), or any pest-control device can be used. Pesticides can be defined as any chemical, or combination of chemical compounds, used to eradicate, repel, or manage pests.

These include unwanted insects, animals and plants that interfere with storage, transportation as well as processing commodities FAO, (2002). The term "pesticide" refers to a broad range of toxicants used to destroy, prevent and mitigate pests, including insecticides, herbicides, virucides, fungicides, crops_growth regulators, and other toxicants are used in agriculture Randall et al., (2014). Pesticides' persistence, or how long they stay active to control pests, varies. As a result, some residual pesticides control pests for weeks, months, or even years. Pesticides are therefore widely used because of the benefits they provide. Pesticides are efficacious and highly trusted at keeping yield healthy and preventing pest destruction. They respond quickly in an emergency, especially when crops are in danger of infestation. Farmers applied various pesticides to mitigate pests to reach the demand for quality as well as quantity in agricultural products Tilman et al.,(2002). Residues of pesticides within fruits, vegetables, and other agricultural products are common after harvest due to the use of pesticides during production WHO, (2005). Pesticides are toxic, ecologically unstable, and mobile chemicals in the ecosystem, despite the benefits they offer. Their presence in food is especially hazardous, and their effects on environmental quality and human health have been extensively studied around the world, Huber et al., **(2**000).

As a result, it is a significant issue that has led to calls for the local, regional, and global elimination of pesticides from foods and indeed the environment (Cerejeira *et al.,* 2003; Huber *et al.,* 2000; Kidd *et al.,* 2001;Ntow, 2001; and Planas *et al.,* 1997). Pesticides can volatilize or drift off from the treatment site and contaminate the environmental compartment (soil, water, and air) as well as the vegetation thereby advancing the risk of human exposure.

Pesticides can penetrate the skin of a human being in several ways**,** including inhalation (breathing) of polluted air, eating contaminated foods, drinking contaminated water, and products applied to the body such as cosmetics, in a situation that acute and chronic pesticide poisoning are documented Barnett et al., (1997). Humans are mostly exposed to pesticides via pesticides contaminated food as well as the quantity of exposure is influenced by both the amount of food available for consumption and the presence of contaminants within the food (Andrew, 2002 and Hassal, 1999). Breast milk or the placenta are two additional routes by which maternal transmission to the foetus may occur. Pesticides can be held to account for biodiversity loss and habitat degradation Sattler et al., (2006).

PESTICIDE CLASSIFICATION BASED ON CHEMICAL NATURE

Inorganic

Mercury, copper, arsenic, and lead are inorganic pesticides naturally mined minerals obtained from the earth and ground into powder. Mercury, copper, arsenic, and lead were broadly used to control pest until the early twenty (20th) before the banned of inorganic pesticides due to their high level of poisoning (toxicity) to humans and wildlife which resulted to death in most cases.

(Baird and Cann, 2005). Pesticide toxicity can be determined in a variety of ways, but most tests on rats and other animals are used.

Organic Pesticides

These are compounds that primarily consist of carbon and hydrogen, along with other elements like chlorine, nitrogen, sulphur, and phosphorus. They are then divided into two categories: Natural and synthetic organic pesticides (Koehler and Belmont, 1998).

Organic natural pesticides

The term "pesticides" nowadays refers to synthetic chemical compounds. Plant extracts or naturally occurring substances were used by ancient man to control pests before the invention of synthetic pesticides. Sulphurous rock, salt, tobacco extracts (nicotine), neem, rotenone, red pepper, and crushed chrysanthemum plants were among the first pesticides used to control lice and flying insects, Washington, DC, (1993). Still, all of these have the same goal: to kill, repel, or otherwise interfere with insect pests' destructive behavior.

Organic synthetic pesticides.

Synthetic organic pesticides are categorized as "modern" pesticides (i.e., those discovered and used during the World War II to protect troops from typhus and malaria). Synthetic pesticides are chemicals used to neutralize or kill undesirable organisms in gardens, homes, public areas, and crops Bhadekar et al.,(2017). Four major classes of synthetic organic pesticides include organochlorines, organophosphates, carbamates, as well as pyrethroids (Koehler and Belmont, 1998).

Organochlorine (OCPs)

These compound consist of hydrogen, carbon and chlorine, along with other elements like oxygen and sulphur. DDT and other organochlorine pesticides have been used extensively ever since the 1940s due to their high performance in preventing diseases and controlling pests. OCPs were utilized to manage parasites on cotton and other crops Safiatou, (2007). They have been known to have poor water solubility, lipid-soluble, long - lasting persistence, long-range transport nature, toxicities, and bioaccumulation properties (Bulut *et al.,* 2011 and Loganathan, 2012). They can cause chronic toxic effects in living creatures through ingestion of water, food and inhalation of contaminated air. As a result, exposure to organochlorines has been linked to consequences for human health diseases including cancer, endocrine disruption, developmental abnormalities, bone disorder, joint pain, and skin infection. Food is the main source of OCP exposure in man (Hedley, 2010). In her book "The Silent Spring," Rachel Carson discussed how biodiversity is at risk and under threat. As a result, the Stockholm Convention on Persistent Organic Pollutants (POPs) prohibited the use of DDT, dieldrin, aldrin, heptachlor, and several other organochlorine pesticides. However, they are still employed in some developing nations' OCPs to eradicate pests (Lobe, 2006).

Organophosphates pesticides.

Organophosphates are chemical compounds formed when phosphoric acid and alcohol react and hence include all pesticides containing phosphorus. In early 1940s the discovery of organophosphates becomes as a substituent or alternative to persistent organochlorine derivatives due to their less persistence in the environment (Ritter,2009). These chemicals can cause immediate health problems when consumed, inhaled, or absorbed through the skin (McKinney and Schoch, 2003). Organophosphates kill insects by interfering with their brain and nervous system activity. They can affect both humans as well as wild animals' nervous systems due to similarities in brain biochemistry Bjorling-Poulsen et al., (2008).

Carbamates pesticides

Carbamates are pesticides that are carbamic acid derivatives (H_2NCOOH). Broad spectrum insecticides are most commonly used on cotton, fruits, vegetables, and other crops. Carbamates, in general, degrade quickly (i.e., have a short life) in the environment, are less dangerous to humans, and act and persist similarly to organophosphates Baird and Cann, (2005).

Pyrethroide pesticides:

Pyrethroides are synthetic versions of bio-pesticides modified to boost their chemical stability in the ecosystem. Their chemical compositions and action mechanisms are similar.
Some of these synthetic pesticides are poisonous to nervous system (example: permethrin, resmethrine, etc (Baird and Cann, 2005).
In addition, toxicity can be used to classify pesticides: Exceptionally risky (Class 1A), dangerously high (Class 1B), risky in a moderate way (Class II), dangerous just a little (Class III) and in typical use, unlikely to pose a serious hazard (Class IV). Ingestion of 5ml of class Ia pesticides is effective to terminate human lives. Will documented that in developing countries farmers use class Ia, Ib, and II due to their affordability rate (cost effective) WHO (1990), (McConnell and Hruska 1993). As a result, not all farmers apply pesticides according to the law when growing, storing, processing, transporting, importing, and exporting food and other agricultural products. The following range of analytical methods have been developed and applied for the qualitative and quantitative deamination of numerous pesticide residues (Cook *et al.,* 1999; Fillion *et al.,* 1991; Food and Drug Administration, 1999; General Inspectorate for Health Protection, 1996; 2000; Lee *et al.,* Sheridan and Meola, 2000). The Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS) standard method for pesticide analysis was established (introduced) in Anastassiades et al., (2003). It is a rapid, inexpensive, and cost-effective approach that adapts to adequate results. By conducting follow-up studies for the analysis of more than 200 pesticides, the QuEChERS method has been updated and validated further Lehotay et al., (2007). Technique advancements typically take a step-by-step approach, moving from the simple to the complex. The most popular analytical methods used in modern multi-residue target pesticide residue analysis are gas chromatography, liquid chromatography coupled to mass spectrometry (GC-MS, LC-MS), and/or tandem mass spectrometry (GC-MS/MS, LC-MS/MS) with triple quadrupole mass analyzer. The wide range of techniques available for pesticide analysis exemplifies both the importance of this application and the rapid rate of advancement in analytical chemistry. Agura *et al.,(*2000), for instance, described a method (long large-volume GC-MS injection for the analysis of organo-phosphorus and organochlorines in vegetables using a miniaturized ethyl acetate extraction) for the measurement of only ten organophosphorus and organochlorine pesticides by GC-MS, but over the past decade, the number of pesticides typically included in methods has increased. Sample preparation methods have advanced to support analytical techniques, depending on the types of analyte and matrix monitored. The QuEChERS method for pesticide residues in food was cited by Anastassiades (2003) as an illustration of a technique that takes advantage of the strong features of nearly universal selectivity and high sensitivity of contemporary GC- and LC-MS(/MS) instruments. The Association of

Analytical Communities has designated the QuEChERS method as AOAC Official Method 2007.01 after it underwent extensive validation for hundreds of pesticide residues in numerous food stuffs (Lehotay *et al.*, 2007). The QuEChERS method has some advantages over the majority of traditional analytical techniques. High recoveries (greater than 85%) are achieved for a wide range of pesticides with various polarities and levels of volatility, including notoriously difficult analyte due to the removal of organic acids during extract cleanup.

LEVELS OF ORGANOCHLORIN AND ORGANOPHOSPHORUS PESTICIDE RESIDUES IN GRAINS AND FRUITS:

Fruits, grains and vegetables have been sources of nutritional components in human food, and their contamination with synthetic chemicals tends to put people's lives in danger. Many studies have discovered different levels of organochlorine and organophosphorus pesticide residue contamination in foods such as grains, vegetables and fruits. In vegetables, uncooked fruit, and tubers, Adeniyi and Oladele, (1999), found pesticide residues from organochlorine pesticides in selected markets in Nigeria using GC. DDT, aldrin and HCH were found in 30, 38 and 77 % in all fruits respectively. Vegetable samples recorded 53% of DDT, 30% of aldrin and 95%. In 98, 79, and 49 % of the samples, aldrin, dieldrin, HCH, and DDT, respectively, were found. Qualitative and quantitative determination of paraquat resides were carried in 150 samples of vegetables , fruits and other crops using Spectrophotometric method. *Ddioscorea alata, talinum triangulare, amaranthus candatus, zea mays, lycopersium esculentum, corchorus olitorius, cratylia argentea, amaranthus candatus, capsicum frutescens, raphanu sativus* recorded paraquat residues a concentration range of 0.01 to 0.027 within the paraquat residue tolerance or maximum pesticides limits, according to the findings Akinloye et al., (2011). Rice, maize, wheat, and brown beans from Ondo, Nigeria, were tested for carbamate and organophosphate levels. Pesticide residues of Dichorovos, carbonfuran, parathion, chlorpyrifos, malathio, pirimlphos-methyl, diazion, and carbofuran were detected in cereal grains at a low level of FAO/WHO permitted limit. The pirimlphos-methyl highest concentration was 0.75 ppm Akinneye et al.,(2018). In Ibadan, Oyo State, Bamigboye et al., (2017) used an ultraviolet-visible spectrophotometer to qualitatively and quantitatively analyze fruits (banana, cucumber as well as orange) and vegetables (African spinach**,** bitter leaf, jute leaf and pumpkin leaf). The samples were all below the maximum residual limits set by the European Union (EU). Besufekad et al., (2021), used the (QuEChERS) standard extraction technique and gas chromatography mass spectrometry to establish the levels of pesticide residues of organophosphate as well as organochlorine in cereal grain samples from Ethiopia. Endosulfan sulfat was detected at 0.076 whereas DDE, and DDT were at 0.087 0.133 mg/kg concentration respectively. Aldrin and dimethoate (organophoshorus) are pesticides found in the highest concentration level at 0.082 and 0.077 mg/kg in rice, sorghum, and millet respectively. Aldrin levels in rice, sorghum, and common millet exceeded the Codex Alimentarius and European Union maximum residual limits. Determination of organochlorine residues in onion, cucumber green beans, marrow squash, banana pepper, lettuce, cherry tomato and purslane in Kirklareli, Turkey using QuECHERS standard extraction method with GC-MS. The presence of DDT, dieldrin, methoxychlor, heptachlor, endosulfan and aldrin were recorded in all the samples analyzed Cemile, (2016).

Evaluation of pesticides residues in 350 samples of vegetable and fruit parched from six (6) different markets in Kumasi using GC-ECD for organochlorine (aldrin, gamma-HCH, , dieldrin, methoxychlor, DDT, endrin, and DDE,) and pyrethroid (deltamethrin, permethrin, , cypermethrin, cyfluthrin, fenvalerate,) residues. Hence 37.5% of the samples analyzed recorded no contaminant, 19.0% exceeded the permissible limits, whereas 43.5% recorded below the limit Crentsil et al.,(2011). Deigna et al., (2021), used gas chromatography in conjunction with an electron-capture detector to measure the levels of organochlorin residue in kola nuts in Cote d'Ivoire. Results confirmed the presence of quitozene, chlorthal dimethyl, hexachlorobenzene, endrin ketone, methoxychlor, hexachlorobenzene, and DDE (LOQ) at a low concentration limit. Aldrin and lindane concentrations were found to range from 5.19 to 92.93 ug/kg. According to the findings of the modified QuECHERS extraction procedure with gas chromatography flame ionization detector (GC-FTD), seven (7) different organophosphorus pesticide residues were found in vegetables and eggplants. Chlorpyrifo, dimethoate, and diazinon were detected in 20%, 6%, and 10% of the samples, respectively, with concentration ranges of 0.540 to 0.980 mg/kg, 0.052 to 0.132 mg/kg, and 0.033 to 0.42 mg/kg for diazinon and 0.035-0.42 mg/kg for chlorpyrifo, respectively. Twenty one percent (21%) of the samples had contamination levels above the EU-MRLS safety limit (Habib *et al.,* 2021). Pesticide residues in vegetables (carrots, spinach, cucumbers, turnips, eggplant, and peppers) grown in Lomé city gardens were evaluated qualitatively and quantitatively using gas chromatography-mass spectrometry and liquid chromatography-mass spectrometry. Chlorpyriphos ethyl, dithiocarbamates, chlorothalonil, carbendazime, prothioconazole, pendimethalin and cypermethrin presence were detected above the permissible limits Housséni, (2018). Ibotom and Mohammade (2016) identified the presence of residues organophosphorus organochlorine pesticide in vegetable (cucumber, cabbage as well as lettuce) and fruits (lime, apple, and orange) samples in selected markets in Kaduna, using GC-MS. Pesticide residues of organochlorine (aldrin, endrin, dieldrin, endosulfane, toxaphene, chlordane, and hexachlorobenzene) and organophosphorus (parathion, malathion, dichlorvos, and chlorvos) were detected. The results revealed that lime had 0.002 mg/kg of malathion, orange was contaminated at 0.002 mg/kg of malathion and lettuce 0.008 mg/kg of chlordane and dieldrin. Cabbage contained chlordane and aldrin at 0.005 and 0.015mg/kg below the EU's maximum residue limit. Using GC-MS, it was discovered that sorrel, spinach, and pumpkin (vegetable leaves) taken from farms in Nasarawa State, Nigeria, contained pesticide residues above the allowable limit. The results showed that pumpkin contained 0.75 mg/kg of DDT, spinach contained 0.319 mg/kg, and sorrel contained 0.219 mg/kg. -BHC (0.359 mg/kg) as well as alpha-BHC (0.647 mg/kg) were only found in pumpkin leaves. Dieldrin concentrations in spinach and sorrel were 0.124 mg/kg and 0.053 mg/kg, respectively. According to Ibrahim et al., Endrin and Aldrin were found in pumpkin and sorrel at concentrations of 0.732 and 0.095 mg/kg, respectively Ibrahim et al., (2018). Isaac et al.,(2021), assessed organophosphate and carbamates in Kenyan vegetables. Twenty-one percent (21%) of the samples recorded profenofos, omethoate, acephate, methamidophos, and chlorpyrifos at 10 to 1343 ug/kg concentrations. Twenty one percent (21%) of French beans, 10% of kales, 8% of spinach, and 22% of tomatoes exceeded the EU-MRL. The amount of residue of organochlorin in dried beans samples from Kano,

Lagos, and Abuja, Nigeria, were investigated by Isegbe et al., (2016) using EPA method 8081B. Aldehyde, endrin, heptachlor, endrin- ketone, a-chlordane, a-BHC, endosulfan I, and epoxide was detected. Assessment of residues carbamate, Organochlorines, pyrethroids and Organophosphates in 250 samples of wheat, pulses and rice from selected markets of the Bidar district of north Karnataka using HPLC and GC (gas chromatography and high performance liquid chromatography). Varieties of pesticides analyzed were found present in 80 samples. Fifty-eight (58) samples were below the permissible limits whereas 22 samples exceeded permissible pesticide residues limits in grain samples. The presence of organophosphates (most chlorpyrifos, ethion parathion, triazofos and profenofos) dominated Organochlorines and pyrethroids in all the samples evaluated Jagadish, (2015). Assessment of pesticides residue in Lagos State, Nigeria was carried out in apple, watermelon, and sweet orange fruits with GC-MS. The pesticide residue concentrations in apple and orange fruits were below WHO/FAO MRLs. The concentrations of oxamyloxime, omethoate, and atrazine in watermelon were all above the WHO/FAO MRLs at 0.088 to 0.125 mg/kg, 0.094 to 0.205 mg/kg, and 0.088 to 0.133 mg/kg, respectively Koleayo et al.,(2017). Qualitative and quantitative determination of twenty (20) pesticide residues of organochlorin in 150 samples of vegetables (lettuce , tomato and cabbage) collected from four (4) agro-ecological regions of Togo using GC-ECD. All samples (100%) that were analysed revealed the presence of DDT, drins , heptachlor, HCH and chlordane at levels below the allowable limits of FAO/WHO Lankondjua et al., (2016). Using the QuEChERS multi-residue extraction method and gas chromatography-mass spectrometry (GC-MS) for both qualitative and quantitative pesticide determination, Mustapha et al., (2017) evaluated the presence of 34 pesticide residues in 150 fruit and vegetable samples in Kuwait. Twenty-two percent (21%) of the samples had residues that were above the permitted residue limits (MRL). Fifty-six 56 (79%) of the samples whereas 62 (42%) samples were free from pesticide contamination. Ndidi et al.,(2019) assessed the levels of the residue of organophosphorus, organochlorine, and Pyrethroid pesticides in 20 cowpeas (*vigna inguiculata*) Using gas chromatography GC-MS. Organochlorine (lindane, endrine ketone, and endosulfan ii), organophosphorus (carbophenothion, malathion, and parathion, and pyrathion), and pyrethroide (B-cypermethrin, Alfer – cypermethrin, and cyfluthrin) were all found in all of the samples tested. The presence of parathionm, endosulfan ii, and baythroide was confirmed, with levels of 6578.64, 745.20, and 24.79 mg/kg, respectively, exceeding the Maximum Residue Limits set for food. One hundred-twenty seven (127) samples of fruits and vegetables from Qatar were examined for the presence of organochlorine pesticides (OCPs) using Dionex-Accelerated Solvent Extractor with GCECD/GC-MS. Ninety-nine percent (99%) of the samples contained concentrations above the MRLs. Heptachlor was frequent OCP detected in 75 samples Noora et al., (2016). Evaluation of organochlorine, rganophosphorus, and pyrethroides in 21 samples of beans using GC-SM. As a result, the majority of pesticide residues were discovered below the harmful HRI. In all of the samples, aldrin was discovered. Some organochlorine pesticides had Health Risk Index (HRI) values is greater than one, (Olawale and Confidence, 2021). Aldrin, endrin, dieldrin, heptachlor, chlordane, and endosulfan residues in cowpea and yam chips were found in some Ile-Ife markets using gas chromatography and an electron capture detector.
The study's findings revealed that all sample analyses were contaminated with heptachlor (0.402 or 0.072) in cowpea and dried yam chips, as well as aldrin (0.580 + or – 0.456 mg cowpea) exceeded maximum residue limit of European Union, Olufade et al.,(2014). Standard QuEChERS extraction method was used followed by the use of GC-HRT/MS for quantitative evaluation of residues of organochlorin in vegetables (cabbage, swiss chard and potato) from selected markets in the city of Bloemfontein, South Africa. The detection of the organochlorine residues was below the recommended maximum Nthabiseng et al.,(2021). Residues organophosphate were recorded in vegetables as well as fruits from wet and supermarket sources in China, Mar province, Northern Thailand, using gas chromatography coupled with flame photometry detector. Chlorpyrifos was detected in 31.10 percent of vegetable samples and 21.43 percent of fruit samples that did not test positive for the pesticide. Profennfos at concentration 0.531 mg/kg and triazophos at concentration.188 mg/kg in orange fruit exceeded Codex WHO/FAO MRLs, according to Thiphavanh et al., (2017). Srivastava et al., (2011) evaluated 48 pesticide residues of pyrethroids, organochlorines and organophosphates in 20 different vegetables samples from a selected market. In all of the samples (60), there were 23 pesticide residues found, with concentrations ranging from 0.05 to 12.35 mg/kg. Expect for cumber, okra, carrot, capsicum, onion, tomato, bottle ground, beetroot, cauliflower, colocassia, cabbage, radish, French beans, fenugreek seeds and other vegetable samples analyzed recorded pesticide residue below the permissible limit.

Using soxhlet extraction with GC- ECD the presence of 17 OPCs analyzed in the maize samples were above the permissible limit. Endrin aldehyde, endosulfan ii and gamma-HCH and Heptachlor epoxide 1.039, 0.592, 0.118 and 18.642 mg/kg respectively in maize, Sosan et al., (2018). The quantities of pesticides with organochlorines left over in kola nuts in Osun State, Nigeria, were reported by Susan and Onyekunle (2017). GC-ECD was used to examine 17 OCPs pesticide residue at which all the samples recorded pesticide contamination above the recommended residual limits. Lindane (0.243 mg/kg), p, p-DDE (0.480 mg/kg), endosulfan (0.243 mg/kg), and dieldrin (0.310 mg/kg) were among the concentrations that were measured.

Tahir et al., (2011) examined organophosphate (OP), pyrethroid and organochlorine (OC) pesticide residues in eight fruit samples (pear, orange, banana, apple, guava, persimmon guava and grapes) selected from local markets in Naw. Only the apple exceeded the Codex Alimentarius Commission's MRL among the fruits tested. The assessment of 26 pesticide levels in some of the most popular fruits in Hyderabad, Pakistan was conducted in GC MS. Only 3 (2%) out of the 131 samples assessed recorded the presence of pesticide residues below the limit, whereas 53 (40%) fruits recorded above the permissive pesticide residue limits. Almost all of the samples tested positive for chlorpyrifos and dieldrin. In orange and apple samples, chlorpyrifos (1256ug/kg) and endosulfansulfate (1236 ug/kg) residues recorded the highest concentration levels Yawar et al., (2001.)

CONCLUSION

Fruits and grains are sources of nutritional components in human food that, when contaminated with synthetic chemicals, put human lives at risk. The data gathered so far for this review confirmed different levels of organochlorin and organophosphorus in grains, fruits, and vegetables, implying that these chemicals should be monitored in food regularly. As a result, natural pesticides (bio-pesticides) are highly recommended as a pest control alternative to synthetic pesticides for food security and environmental sustainability.

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