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Comparative Assessment of Pesticide Residues in Groundnuts Sold in Selected Markets in Port Harcourt Metropolis, Rivers State, Nigeria

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Abstract
In this study, pesticide residues [pyrimethanil, Diazinon, Malathion, Chlorpyrifos, Dichlorodiphenyldichloroethane (DDD), Dichlorodiphenyltrichloroethane (DDT), and Dichlorodiphenyldichloroethylene (DDE)] were investigated in groundnut (*Arachis hypogaea* L.) samples from two prominent markets [the Mile three (3) Market, and Sagana Market] in Port Harcourt metropolis, Rivers State, Nigeria. Fresh and roasted samples were bought and analysed from both markets. The same parameters were also analysed in "homemade" samples. Samples were transported to the laboratory in well tagged sterile polythene bags for analysis. Characterisation was done using the MassHunter quantitative analysis. Thereafter, the GC-MS machine was used for detection of pesticide residues. Findings from this study revealed the presence of pesticide residues in all the groundnut samples analysed. Homemade samples had relatively low amounts forDiazinon(0.0966mg/kg), Malathion (1.0636 mg/kg) and Chlorpyrifos (0.0095 mg/kg) that were within the available FAO/WHO maximum residues limits (MRLs) for groundnut (legume vegetables) unlike the samples from both Sangana and Mile 3 Markets. The concentration of investigated pesticide residues followed the order, DDE>DDT >DDD> Malathion > Diazinon > Pyrimethanil, with DDE measuring as high as 1127.785mg/kg in fresh groundnut sample from a location in Mile 3 Market. Results revealed pesticides in various samples had their unique peculiarities. Further studies to investigate the causes behind these peculiarities could reveal how pesticides interact within distinct environments and/or market conditions. The detection of some pesticides above permissible limits call for more studies to forestall the incidence of toxicity from the ingestion of pesticides contaminated food substances, including groundnuts.

Keywords: Diazinon, Groundnut, Malathion, Pesticides, Pyrimethanil.

Introduction

Groundnut (*Arachis hypogaea* L.) is the thirteenth most important food crop of the world; fourth most important source of edible oil and the third most important source of vegetable protein (Taru et al., 2008). *Arachis hypogaea* L. belongs to class, magnoliopsida; order, fabales; family, fabaceae; subfamily, faboideae; tribe, aeschynomeneae; and genus *Arachis*. The World production figure of groundnut in 2019 was 48.8 million tonnes from 29.6 million hectares with average production of 1647 kg ha-1 (FAO, 2021). The production of groundnut is concentrated in Asia and Africa, where the crop is grown mostly by smallholder farmers under rain-fed conditions with limited inputs. In Africa, about 11.7 million hectares of land is utilized for groundnut production and 10.9 million tons of yearly generation (FAO, 2013). Globally, Nigeria was the third largest producer of groundnut as at 2019, with an annual production of 4.4 million tonnes, after China 17.1 million tonnes and India 6.7 million tonnes (FAO, 2021). Nigeria's groundnut production represents 10 % of total global production, 39% of Africa's production and 51 % of groundnut production in the West Africa region (Nahanga, 2017; Ajeigbe et al.,2014).

In Nigeria, groundnuts are processed into different dishes and food substances. Abdulrahaman et al.(2014) identified some food substances prepared from groundnut in different tribes in Nigeria to include: groundnut oil, kulili, yaji, sisi pelebe, donkwa, kunu geda, chin-chin, groundnut soup, and roasted groundnut. In the northern parts of Nigeria, groundnuts are also cooked or boiled, processed into groundnut paste (commonly known as groundnut butter), groundnut cake, salted groundnut and groundnut soup (Mustapha et al.,2015). Boiled and roasted groundnuts are common street side food in Port Harcourt metropolis, Rivers State, Nigeria. Whether freshly boiled or fried, groundnut is a good traditional snacking item with its unique taste. Fried groundnuts are enjoyed by many people who combine them with bread, cucumber, banana, roasted or boiled maize, garri (soaked garri), boiled rice etc. The oil cake or groundnuts cake, a by-product of groundnut oil production, forms one of the most sort after livestock cakes of commercial product. Intensive agricultural practice and industrial processes has been known to pollute the environment and contaminate food and agricultural products. Environmental toxicants include pesticides, and threaten entire ecosystems, seriously damaging its components, function and structure, in some cases. Pesticides are considered as efficient, economical, and effective weapons in integrated pest management systems. They include insecticides, herbicides, nematicides, fungicides, molluscicides, rodenticides, plant growth regulators, and other compounds (Zhang et al., 2021). A pesticide has been defined as any component or mixture of compounds intended for use as a plant regulator, defoliant, or desiccant (United States Environmental Protection Agency, 2004). The global pesticide consumption in 2019 was approximately 4.19 million metric tons (Fernández, 2021).Though pesticides are useful chemical substances to farmers in controlling pests, diseases, and weeds, ensuring optimal crop yield and quality, their indiscriminate and excessive use can lead to their persistence in the environment and subsequent accumulation in crops, including groundnuts (Garcia et al., 2020). Pesticide residues in groundnuts are a cause for concern due to their potential adverse effects on human health, such as acute and chronic toxicity, developmental abnormalities, and carcinogenicity (Garcia et al., 2020; Gupta et al., 2019). From the foregoing, it is evident that effective assessment of groundnut is necessary to ensure best practices and consumer protection. Also, recently, the social media has been awash with videos showing the use of chemicals in the industrial processing of groundnut which if true, could predispose innocent consumers to diseases related to the toxicity of such chemicals; hence the need for this study. This study is aimed at ascertaining pesticide residues in groundnut seeds from Sangana Street market and Mile 3 market both in Port Harcourt, Rivers State, Nigeria.

Materials and Methods

Due to its strategic location in Nigeria and the economic and commercial activities, the Port Harcourt metropolis serves as an excellent market for locally produced commodities from the agricultural and commercial sectors from other parts of Nigeria, including the vast majority of rural communities. The markets sampled are the Sangana Street market and Mile 3 Market, both situated in Port Harcourt metropolis, Rivers State, Nigeria. These markets were selected because they are major markets in the City. The Sangana Street market and Mile 3 markets both have a large influx of people, who throng the markets to buy various commodities including groundnuts.

Groundnut samples were bought from Sangana Street market and Mile 3 market separately. Samples were put in well tagged sterile polythene bags accordingly, and transported to the laboratory for analysis. Groundnut seeds were analysed for the presence of pesticide residues. Sampling was carried out three times in three different months of the year. Homemade groundnuts samples were also analysed for pesticide residues. In the laboratory, groundnut seed samples were washed with tap water, and rinsed with deionised water to remove likely sources of impurities like soil particles and debris. The samples were then pounded in mortar with pestle. The mortar and pestle were thoroughly washed to remove any possible dirt and other contaminants before use. Pounding was separately done for the different samples from the different markets, and for the homemade sample, until a homogeneous mixture was achieved. Pesticide residue samples characterisation was done using the Mass Hunter qualitative/quantitative analysis, where the EPA 8015 method was used for the sample extraction. Thereafter, the GC-MS machine was used for the characterisation and detection of the presence of chemical pesticides residue in the groundnut samples according to standards. The following key was used to distinguish the samples used for this work: SF-1, represents Sangana market fresh groundnut sample for location 1; SF-2, represents Sangana market fresh groundnut sample for location 2; SR-1, represents Sangana market roasted groundnut sample for location 1; SR-2, represents Sangana market roasted groundnut sample for location 2; MF-1, represents Mile three fresh groundnut sample for location 1; MF-2, represents Mile three fresh groundnut sample for location 2; MR-1, represents Mile three roasted groundnut sample for location 1; MR-2, Mile three roasted groundnut sample for location 2; H, represents homemade groundnut sample. Relevant statistical analytical tools such as the mean, standard deviation, charts (graphs) and analysis of variance (ANOVA) at 0.05 confidence level was utilized to arrive at a conclusion.

Results

The concentrations of Pyrimethanil vary moderately across the samples ranging between 0.0902mg/kg in SF-2 to 0.4315mg/kg in MF-2 as shown in Table 1..

Diazinon concentrations show a wider range, with the highest concentration found in MT-1(1.5818mg/kg) and the lowest in H (0.0966 mg/kg).

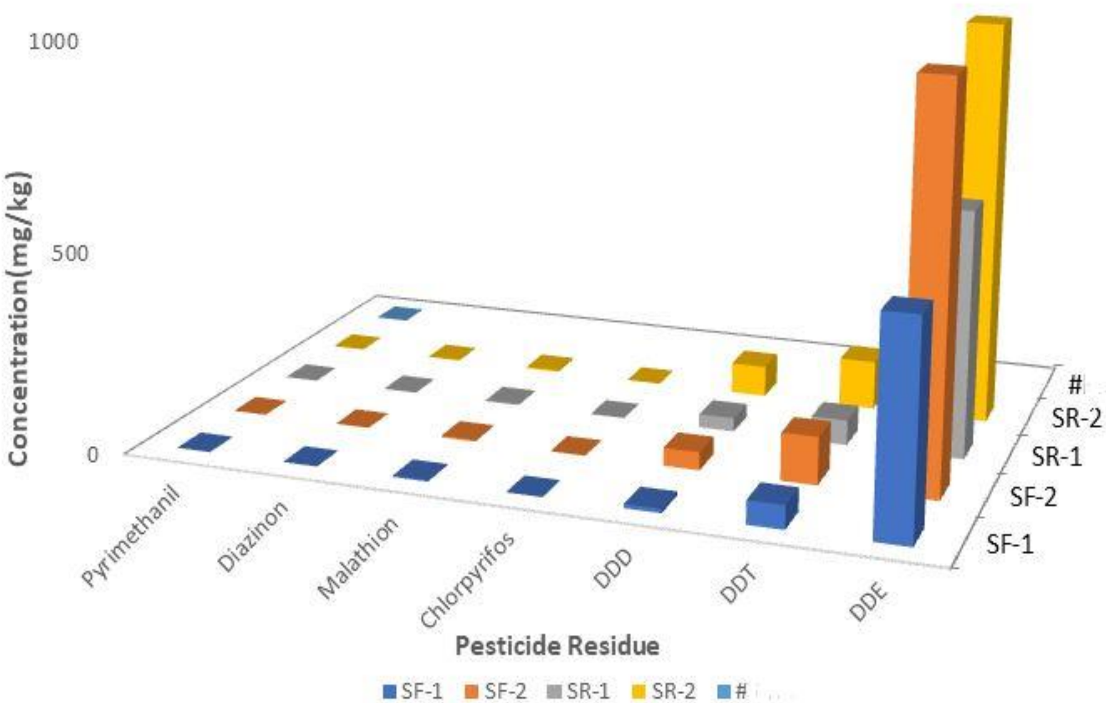
Malathion concentrations were highest in SF-2 (4.1350mg/kg) and lowest in H (1.0636 mg/kg).
Chlorpyrifos concentrations were relatively low across all samples, with the highest concentration in SF-2 (0.1323 mg/kg) and the lowest in H (0.0095 mg/kg).
DDD concentrations varied considerably, with SR-2having the highest value (77.99 mg/kg) and SF-1 the lowest (10.33 mg/kg).
DDT concentrations fluctuated considerably with SR-2 having the highest concentration (122.73mg/kg) and H the lowest (23.58 mg/kg)
DDE concentrations exhibited a relative wide range, with MR-2 having the highest value (1154.73 mg/kg) and SF-1 the lowest (523.14 mg/kg).

- The results highlight significant variability in pesticide concentrations across the different samples, reflecting potential differences in agricultural practices and/or environmental exposure.
- SR-2 consistently showed higher concentrations across multiple pesticides, indicating a potential hotspot for pesticide residue.
- H consistently exhibits the lowest concentrations, suggesting lower levels of pesticide contamination, possibly due to being homemade or locally sourced.

Table 1: Mean concentrations of pesticide residues in groundnut samples in milligrams per kilogram (mg/kg) from the various groundnut seed samples showing variations in the residues concentrations across different groundnut seed samples.

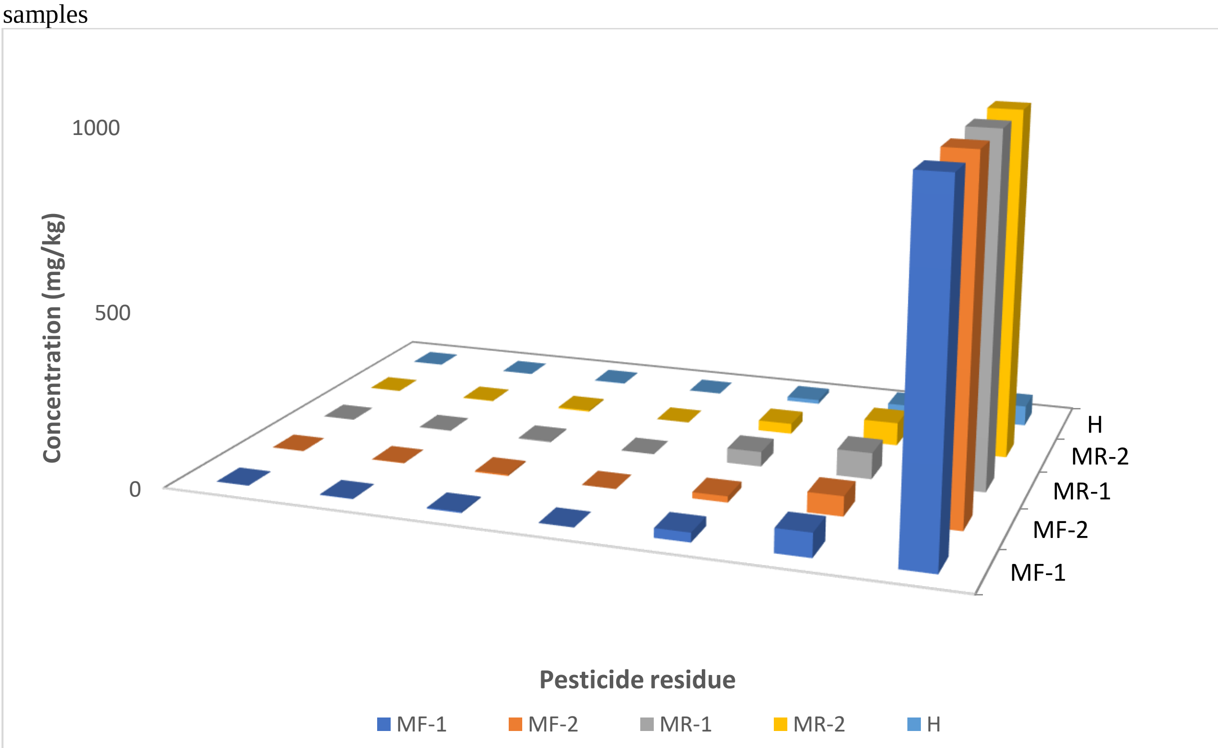
	Pyrimethanil	Diazinon	Malathion	Chlorpyrifos	DDD	DDT	DDE	TOTAL
SF-1	0.1464	0.5697	3.0232	0.0819	10.33	58.32	523.14	595.60
SF-2	0.0902	0.7867	4.1350	0.1323	44.81	118.33	980.86	1149.15
SR-1	0.1083	0.9637	2.8903	0.0344	33.62	62.90	609.05	709.57
SR-2	0.0869	1.3307	3.9849	0.0952	77.99	122.73	1116.81	1323.03
MF-1	0.2758	1.3478	3.4560	0.0564	26.75	67.59	1127.52	1227.00
MF-2	0.4315	1.1334	3.6791	0.0446	17.60	56.84	1133.55	1213.27
MT-1	0.2232	1.5818	2.8686	0.0227	42.67	75.77	1137.11	1260.24
MR-2	0.3951	1.4061	3.7972	0.0113	30.20	66.88	1154.73	1257.42
H	0.1789	0.0966	1.0636	0.0095	11.89	23.58	58.56	95.37
FAO/WHO, 2008	NA	0.2	2	0.05	NA	NA	NA	NA

Key: SF-1, represents Sangana market fresh groundnut sample for location 1; SF-2, represents Sangana market fresh groundnut sample for location 2; SR-1, represents Sangana market roasted groundnut sample for location 1; SR-2, represents Sangana market roasted groundnut sample for location 2; MF-1, represents Mile three fresh groundnut sample for location 1; MF-2, represents Mile three fresh groundnut sample for location 2; MR-1, represents Mile three roasted groundnut sample for location 1; MR-2, Mile three roasted groundnut sample for location 2; H, represents homemade groundnut sample.



Key: SF-1 represents Sangana fresh groundnut samples for location 1; SF-2 represents Sangana fresh groundnut samples for location 2; SR-1 represents Sangana roasted groundnut samples for location 1; SR-2 represents Sangana roasted groundnut samples for location 2;and H represents homemade groundnut samples.

Figure 1: Pesticide residue concentrations of fresh and roasted groundnut from Sangana Market and Homemade samples



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Key: MF-1 represents Mile 3 fresh groundnut samples for location 1; MF-2 represents Mile 3 roasted groundnut samples for location 2; MR-1 represents Mile 3 roasted groundnut samples for location 1; MR-2 represents Mile 3 roasted groundnut samples for location 2; and H represents homemade groundnut samples.

Figure 2: Pesticide residue concentrations of fresh and roasted groundnut from Mile 3 Market and Homemade samples

Table 2: Statistical significance of pesticide residue concentrations in groundnut seeds across different locations.

	Pyrimethanil	Diazinon	Malathion	Chlorpyrifos	DDD	DDT	DDE
SF-12	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
SR-12	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
MF-12	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
MR-12	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
S-FR	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
M-FR	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
SM-F	$P < 0.05$	$P < 0.05$	$P > 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
SM-D	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
HSR1	$P < 0.05$	$P < 0.05$	$P > 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
HSR2	$P < 0.05$	$P < 0.05$	$P > 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
HMR1	$P < 0.05$	$P < 0.05$	$P > 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$
HMR2	$P < 0.05$	$P < 0.05$	$P > 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$	$P < 0.05$

Key: SF-12 represents Sangana Fresh groundnut samples for location 1 to 2; SR-12 represents Sangana Roasted groundnut samples for location 1 to 2; MF-12 represents Mile 3 Fresh groundnut samples for location 1 to 2; MR-12 Mile 3 Roasted groundnut samples for location 1 to 2; S-FR represents Sangana Fresh to roasted groundnut samples; M-FR Mile 3 Fresh to Roasted groundnut samples; SM-F represents Sangana to Mile 3 Fresh groundnut samples; SM-R represents Sangana to Mile 3 Roasted groundnut samples; HSR1 represents Homemade to Sangana for Roasted in location 1; HSR2 represents Homemade to Sangana for Roasted in location 2; HMR1 represents Homemade to Mile 3 for Roasted groundnut samples in location 1; and HMR2 represents Homemade to Mile 3 for Roasted groundnut samples in location 2.

Discussion

The findings from this study revealed all the investigated pesticide residues were present in all the groundnut samples analyzed. Pyrimethanil, characterized by a range from 0.0902(mg/kg) to 0.4315(mg/kg) demonstrates moderate variability, with the highest concentration recorded in sample MF-2. Diazinon, on the other hand, displays a broader range from 0.0966(mg/kg) to 1.5818(mg/kg) with its highest concentration observed in MT-1. Malathion, exhibiting concentrations from 1.0636(mg/kg) to 4.1350(mg/kg) showed substantial variability, with SF-2 presenting the highest concentration. Chlorpyrifos, with concentrations ranging from 0.0095(mg/kg) to 0.1323(mg/kg) reflects relatively low levels across all samples. DDD and DDT reveal considerable variability, with SR-2 recording the highest concentrations. DDE, had its highest concentrations 1154.73(mg/kg) again sees MR-2. The cumulative total of pesticides ranged from 95.37(mg/kg) in sample H, to 1323.03(mg/kg) in sample SR-2.These findings underscore the significant variability in pesticide concentrations, suggesting potential disparities in agricultural practices or environmental exposure which agrees with some earlier studies (Stackpoole et al., 2021; Curl et al., 2015). Results also reveal SR-2 consistently emerges as a hotspot for pesticide residue, exhibiting higher concentrations across multiple pesticides. Conversely, the homemade samples, represented by H, consistently demonstrate the lowest concentrations, underscoring the influence of sourcing and/or preservation practices on contamination levels.

Results show a consistent trend of statistical importance in SF-12, SR-12, MF-12, MR-12, S-FR, M-FR, SM-F, SM-D, HSR1, HSR2, HMR1, and HMR2, which suggests that specific locations or/and conditions significantly impacts on pesticide concentrations in groundnut seeds as reported by Thelin and Stone (2013). In the case of SM-F and HSR1 samples, an interesting finding emerged the p-value for Malathion exceeding 0.05. This occurrence implies that within these specific scenarios, there might not be a statistically significant disparity in Malathion concentrations between the groups being compared. Investigating the causes behind these exceptions could reveal how pesticides interact within distinct environment or market conditions. An Analysis of Variance (ANOVA) examining the pesticide residues concentrations in roasted groundnuts involving “Total Pesticides” and “Individual Pesticides” was carried out. The evaluation of Total Pesticides showcases two sources of variation—Between Groups and Within Groups. The Between Groups Variation doesn't reveal statistical significance, indicating that the observed differences in total pesticide concentrations among different categories are likely due to random variability rather than substantive distinctions. Conversely, the examination of Individual Pesticides highlights a stark contrast; the Between Groups Variation indicates a statistically significant difference among specific pesticides present in the roasted groundnuts. This suggests that while the total concentrations might not differ significantly, individual pesticides exhibit notable variations in their levels within the sampled roasted groundnuts. A variation in individual pesticides was also reported by Onwujiogu et al. (2022) in their study on a legume vegetable (Bambara nut). H (homemade) had the lowest concentrations of all samples, and was within the FAO/WHO maximum residues limits (MRL) for Diazinon, Malathion and Chlorpyrifos. This was however not the case for the other samples, whose concentrations of Diazinon, Malathion all exceeded the set limits. Galani et al. (2020) also reported very high pesticide residues in groundnut and other vegetables that exceeded the maximum residue limits. The relative low amounts of pesticides in the homemade samples (though statistically not significant) suggests preparation and/or storage practices may have played a role in the amount of pesticides found in groundnut samples from Sangana and Mile 3 Markets in Port Harcourt. The concentration of investigated pesticide residues followed the order, DDE >DDT > DDD > Malathion > Diazinon > Pyrimethanil.

Conclusion

Pesticide residues are amongst the priority pollutants globally. They are of great importance because of their toxicity even at low levels, and a tendency to accumulate and biomagnify in living tissues. Pesticides toxicity in food grains and vegetables have been implicated in numerous human health issues, including damage to the central and peripheral nervous systems, cancer, allergies and hypersensitivities, reproductive disorders, and disruption of the immune system. Findings from this study revealed the presence of pesticide all the groundnut samples analyzed. Homemade samples had the lowest amount of residues and were within the available FAO/WHO maximum residues limits (MRLs) for legume vegetables, unlike the samples from Sangana and Mile 3 Markets. Results also revealed the concentrations of various pesticides had their unique peculiarities. DDE measuring as high as 1127.785 mg/kg in fresh groundnut sample from Mile 3 Market (location 1). The concentration of investigated pesticide residues followed the order, DDE >DDT > DDD > Malathion > Diazinon > Pyrimethanil. Homemade samples had the lowest amount of residues and were within the available FAO/WHO maximum residues limits (MRLs) for legume vegetables, unlike samples from Sangana and Mile 3 Markets. The findings from this work do not reveal any statistical significance in the concentrations of pesticide residues between roasted, fresh and homemade groundnuts. Hence the observed differences in total pesticide concentrations among different categories are likely due to random variability.

Recommendations

1. Pesticides can cause harm by contaminating food crops and the environment, thereby causing several health issues. Their use in groundnut production, preservation and storage should be minimized.
2. Care should be taken to wash groundnut properly before processing and consumption.
3. More studies should be carried out on pesticide residues in groundnut and other food crops in our markets on a regular basis. This will forestall the incidence of pesticide toxicity from the ingestion of contaminated food substances.

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