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Contamination of Pesticide residues and Human Health Risk Assessment of the Selected Agricultural Soils in Oyo State, Nigeria

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Abstract

This study investigated pesticide residues contamination in agricultural soils from Ibarapa and Oke - Ogun regions of Oyo state, Nigeria. Soil samples were collected from four farms, located in Ibarapa (Araromi and Jakode) and Oke - Ogun (Itasa and Oke - Ola). Ten sub-samples collected within a 10–20 m radius at a depth of 0 – 20 cm and bulked in each location and analysed for pesticide residues (organochlorine, and pyrethroid) using GC–MS. The data were compared with World Health Organisation/Food and Agriculture Organisation (WHO/FAO) Limits and subjected to statistical analyses [hierarchical Clustering (HC), Discriminant Analysis (DA), Pincipal Conponent Analysis (PCA)], Toxicity Assessment (spatial mapping, chemometric profiling), and Health Risk assessment [Risk Quotient (RQ) and probabilistic risk (Monte Carlo simulation) modelling]. Multiple residues were detected in all sites, with Pyrethroid (heptachlor, heptachlor epoxide, aldrin, dieldrin, endosulfan I/II, endosulfan sulfate), and Organochlorine (p,p′-

DDT/DDD, chlorothalonil, lambda-cyhalothrin, and cypermethrin isomers). Concentrations were highest in Jakode and several analytes exceeded WHO/FAO residue limits in all locations. PCA, HC, and DA separated a legacy organochlorine domain from a pyrethroid-associated domain. Co-occurrence network analysis demonstrated that the residue mixture was not random, but organized into tightly linked organochlorine and pyrethroid clusters. Health-risk screening showed that RQs for heptachlor epoxide and dieldrin (critical), aldrin and endosulfan (very high), and other compounds (high). Monte Carlo simulation showed >90 % probability that Health Quotient exceeded unity under realistic exposure assumptions. However, the results show that the study areas are affected by both historical pesticide persistence and current agricultural inputs.

Keywords: Contamination, Pesticide residues, Human Health Risk, Agricultural Soils, Ibarapa, Oke - ogun

1.0 INTRODUCTION

The intensification of agricultural practices over the past decades has been accompanied by a substantial increase in the use of synthetic pesticides to enhance crop productivity and control vectors of economic and public health importance. While these chemicals have contributed significantly to food security, their persistence, bioaccumulation, and toxicity have raised global environmental and human health concerns (Food and Agriculture Organisation (FAO), 2019; World Health Organisation (WHO), 2020). Among these, organochlorine pesticides (OCPs) remain particularly problematic due to their chemical stability, lipophilicity, and resistance to degradation, enabling them to persist in soils and biota long after application (Ogbeide et al.,2016).

OCPs such as dichlorodiphenyltrichloroethane (DDT), aldrin, dieldrin, and heptachlor were extensively used between the 1940s and 1970s before regulatory restrictions were introduced under frameworks such as the Stockholm Convention on Persistent Organic Pollutants (United Nation Environmental Programme (UNEP), 2013). However, despite these bans, residues of these compounds continue to be detected in environmental matrices, particularly in developing regions where regulatory enforcement is weak and legacy contamination persists (Doong et al., 2002; Jolliffe & Cadima, 2016)

Soils act as major reservoirs for pesticide accumulation, serving as both sinks and secondary sources of contamination through processes such as leaching, volatilization, and plant uptake (Kortenkamp,2007; Fenner et al., 2013a). Once

deposited, pesticides undergo transformation processes including photolysis, hydrolysis, and microbial degradation, producing metabolites that may retain or even exceed the toxicity of the parent compounds (Qiu, et al.,2005). These compounds can subsequently enter the food chain, leading to biomagnification and chronic human exposure primarily via dietary intake (Agency for Toxic Substances and Disease Registry (ATSDR), 2020; WHO, 2022).

In sub-Saharan Africa, including Nigeria, pesticide contamination is exacerbated by factors such as indiscriminate application, lack of farmer training, poor regulatory enforcement, and the continued use of banned or obsolete chemicals (Adeleye et al., 2019; Ogbeide et al., 2016; FAO, 2019). Studies have reported elevated levels of OCP residues in agricultural soils and water systems across West Africa, with concentrations frequently exceeding international safety thresholds (Zhang et al., 2018; Adeyemi et al., 2024). This raises significant concerns regarding environmental sustainability and public health, particularly in rural farming communities where exposure pathways are multiple and poorly controlled.

Beyond conventional residue analysis, there is increasing recognition of the need for advanced analytical frameworks that integrate multivariate statistics, chemometric fingerprinting, and probabilistic risk assessment to better understand contamination patterns and sources. Techniques such as principal component analysis (PCA), hierarchical clustering, and receptor modeling have been widely applied in environmental studies to identify pollution sources and distinguish between historical and recent pesticide inputs (Jolliffe, 2002; Belkhiri et al., 2010; Zhang et al., 2024). Additionally, health risk assessment approaches based on hazard quotient (HQ) and hazard index (HI) provide quantitative measures of potential human exposure risks (United States Environmental Protection Agency (USEPA), 2004; Leung et al., 2008).

Despite these advances, there remains a critical gap in integrated, multi-dimensional assessments of pesticide contamination in Nigerian agricultural soils that combine spatial analysis, mixture toxicology, and probabilistic modeling. Most existing studies rely on deterministic approaches and do not adequately capture uncertainty, interaction effects, or source complexity.

Therefore, this study aims to provide a comprehensive evaluation of pesticide contamination in soils from selected agricultural zones in Ibarapa and Oke - Ogun Areas of Oyo state, Nigeria by integrating GC–MS residue analysis, multivariate chemometrics, receptor modeling, spatial distribution analysis, and probabilistic human health risk assessment**.** The findings are expected to contribute to a more robust understanding of pesticide behaviour, sources,

and risks in tropical agricultural systems and to inform evidence-based environmental management and policy interventions.

2.0 MATERIALS AND METHODS

2.1 Study Area.

Oyo state is in southwestern Nigeria. It is divided into five dvisions along ethno -cultural line, namely, Ibadan, Ibarapa, Oke - Ogun, Oyo and Ogbomosho.The Ibarapa and Oke - Ogun are Agrarian. Ibarapa with total population of 747, 969 (NPC, 2006; Hassan, 2022) and land mass of area 2,496 $km^{2.}$, on coordinates 7.55 ^{0}N and 3.33 ^{0}E consists of seven main towns; Eruwa, Igangan, Idere, Lanlate, Igbo -ora, Tapa, and Ayete. All these named seven major towns fall into three Local Government Areas (LGAs); Ibarapa North, Ibarapa East, and Ibarapa Central with their head offices at Ayete, Eruwa, and Igbo - Ora respectively. Oke - Ogun comprises of ten local government and covers one - third of the Oyo state. The LGA are; Kajola, Irepo, Olorunsogo, Saki - East, Iwajowa, Saki - West, Atisbo, Orelope, Iseyin, and Itesiwaju. It has population of about 1.5 million (NPC, 2006; Hassan, 2022) with land mass of 13,537 $km^{2.}$on coordinate 8.2333 ^{0}N and 3.2000 ^{0}E.

The major occupation of the indigenes of Ibarapa and Oke - Ogun regions is farming, right from the ancient time; because of the fertile soil and favourable climatic condition. Four locations were sampled in the two regions; Ibarapa (Araromi and Jakode) and Oke - Ogun (Itasa and Oke - Ola). One tomatoes and one cassava farms were selected in each region. The choice of these two type of farms is that they are predominant and staple produces in that locations.

2.2 Sample Collection and Preparation

Soil samples were collected from agricultural sites in the Ibarapa (Araromi and Jakode) and Oke - Ogun (Itasa and Oke - Ola) regions using a stratified random sampling design to capture spatial heterogeneity in pesticide distribution. Stratification was based on land-use type and farming intensity, consistent with established environmental sampling frameworks (USEPA, 2002; International Organization for Standardization. (ISO 18400-104:2018), 2018).

At each sampling location, composite samples were obtained by pooling 10 sub-samples collected within a 10 – 20 m radius at a depth of 0 – 20 cm, corresponding to the active root zone and zone of maximum pesticide accumulation (Food and Agriculture Organization (FAO), 2019). Samples were collected using stainless steel augers, transferred

into pre-cleaned amber glass containers to minimize photodegradation, and transported on ice to the laboratory, where they were stored at 4 °C prior to analysis.

Sample preparation followed internationally recognized protocols for pesticide residue analysis in soils (United State Environmental Protection Agency (USEPA) Method 3540C; Quality Assurance and Quality Control **(Q**AQA), 2019). Samples were:

- Air-dried at room temperature under contamination-free conditions
- Homogenized and sieved through a 2 mm mesh to remove debris
- Subjected to Soxhlet extraction using a methanol–chloroform solvent system (2:1, v/v), optimized for both polar and non-polar pesticide recovery

Extracts were purified using silica gel/alumina column clean-up to remove co-extracted organic matter and lipids, following established procedures for organochlorine pesticide isolation (WHO, 2007; UNEP, 2013). The cleaned extracts were concentrated using a rotary evaporator and reconstituted in n-hexane prior to instrumental analysis.

2.3 Instrumental Analysis (GC–MS)

Quantitative and qualitative analysis of pesticide residues were performed using Gas Chromatography–Mass Spectrometry (GC–MS)**,** a gold-standard technique for trace-level detection of semi-volatile organic compounds (Skoog et al., 2014; National Institute of Standards and Technology (NIST), 2020).

The GC–MS system was equipped with a Capillary column of HP-5MS (30 m × 0.25 mm × 0.25 μm film thickness), Carrier gas of High-purity helium at constant flow rate (1.0 mL/min) also Splitless injection to enhance sensitivity and Electron ionization (EI) at 70 eV.

The oven temperature program was optimized for pesticide separation such as Initial temperature: 70 °C (held for 2 minutes), Ramp: 10 °C/minute to 280 °C and Final hold: 10 minutes. Compound identification was based on retention time matching with analytical standards and mass spectral comparison with National Institute of Standards and Technology/Environmental Protection Agency/National Institutes of Health of USA (NIST/EPA/NIH) spectral library (match quality ≥ 90 %). Quantification was performed using external calibration curves constructed from certified reference standards across multiple concentration levels.

2.4 Quality Assurance and Quality Control (QAQC)

Strict QAQC protocols were implemented to ensure analytical accuracy, precision, and reproducibility, in accordance with international guidelines (USEPA, 2016; Eurachem, 2014).

The following procedures were applied;

Calibration linearity: Multi-point calibration curves with coefficient of determination ($R^2 \geq 0.99$)

- **Method blanks and procedural blanks:** Included to detect contamination during analysis
- **Spike recovery tests:** Performed using matrix-matched samples, with acceptable recovery range of **70 – 120%**
- **Precision assessment:** Triplicate analyses with Relative Standard Deviation (RSD < 15%)
- **Limit of detection (LOD) and limit of quantification (LOQ):** Determined based on signal-to-noise ratios of 3:1 and 10:1, respectively

Certified reference materials (CRMs), where available, were used to validate analytical performance. Instrument stability was monitored through periodic calibration verification and internal standard checks.

2.5 Data Preprocessing and Normalization

Prior to statistical and chemometric analysis, raw concentration data were preprocessed to improve statistical robustness and comparability across variables.

Data treatment included:

- **Logarithmic transformation (log_{10}):** Applied to reduce skewness and normalize non-Gaussian distributions commonly observed in environmental contaminant datasets (Reimann et al., 2008)
- **Standardization (z-score normalization):**

$$\mathbf{Z} = \frac{\mathbf{X}-\boldsymbol{\mu}}{\boldsymbol{\sigma}} \qquad 1$$

to ensure equal weighting of variables with different units and magnitudes Outliers were evaluated using boxplots and Mahalanobis distance and retained only if environmentally justifiable, consistent with best practices in environmental chemometrics (Filzmoser et al., 2005).

2.6 Statistical and Data Analysis

The following statisticcal analysis were carried out on the data obtained; Principal Component Analysis (PCA), Hierarchical Cluster Analysis (HCA), Linear Discriminant Analysis (LDA). Source Apportionment (Receptor Modeling) and Spatial Analysis were also determined. Ecological risk Assessment (Environmental and Health risk Assessment) of the pesticide were also determined using the following; Environmental Fate and Degradation Analysis, Mixture Toxicology and Chemometric Fingerprinting, Risk Quotient, Probabilistic Risk Assessment (Monte Carlo Simulation), and Bayesian and Advanced Statistical Modeling

2.6.1 Principal Component Analysis (PCA)

Principal Component Analysis (PCA) was employed to reduce data dimensionality and identify latent patterns in pesticide distribution across sampling locations. PCA transforms correlated variables into a smaller set of orthogonal components that maximize variance (Jolliffe, 2002). Prior to analysis, the dataset was standardized (z-score normalization) to eliminate scale bias among variables. Component retention was based on Eigenvalues > 1 (Kaiser Criterion), Scree plot inflection and Cumulative variance ≥ 70 %. Varimax rotation was applied to enhance interpretability by maximizing variance of loadings across components (Kaiser, 1958). PCA has been extensively validated for environmental contaminant source identification and chemometric fingerprinting of pesticide residues (Helena et al., 2000; Viana et al., 2008; Wang et al., 2020).

2.6.2 Hierarchical Cluster Analysis (HCA)

Hierarchical Cluster Analysis (HCA) was conducted using Ward's linkage method and Euclidean distance metric to classify sampling sites into homogeneous clusters based on pesticide profiles. Ward's method minimizes within-cluster variance and is widely regarded as robust for environmental datasets (Ward, 1963).

Cluster validation was performed by comparing dendrogram groupings with PCA score plots to ensure consistency in spatial classification patterns (Shrestha & Kazama, 2007). HCA is commonly applied in pollution studies to reveal spatial similarities and contamination gradients (Belkhiri et al., 2010; Varol, 2011).

2.6.3 Discriminant Analysis (DA)

Linear Discriminant Analysis (LDA) was used to validate classification results obtained from PCA and HCA and to determine the variables responsible for group separation. The discriminant functions were derived to maximize between-group variance relative to within-group variance, allowing accurate classification of samples into predefined geographic clusters (Fisher, 1936).

Model performance was evaluated using:

- Classification accuracy (%)
- Wilks' lambda statistic
- Cross-validation (leave-one-out method)

DA has been widely used in environmental forensics for classification and pollution source discrimination (Otto, 2017; Li et al., 2020; Singh et al., 2020).

2.6.4 Source Apportionment (Receptor Modeling)

- Source identification was performed using a hybrid of PCA, Absolute Principal Component Scores (APCS), and Multiple Linear Regression (MLR): PCA–APCS–MLR receptor modeling approach**,** which integrates multivariate statistics with regression-based quantification of source contributions.
- PCA identifies latent pollution sources
- Absolute Principal Component Scores (APCS) quantify source strength
- Multiple Linear Regression (MLR) estimates percentage contributions

This approach has been successfully applied in environmental pollution studies for apportioning mixed contaminant sources (Thurston & Spengler, 1985; Hopke, 2016).

Additionally, diagnostic ratios were applied to distinguish between historical and recent pesticide inputs:Dichlorodiphenyltrichloroethane / Dichlorodiphenyldichloroethane (DDT/DDD) and Dichlorodiphenyltrichloroethane / Dichlorodiphenyldichloroethylene (DDT/DDE) ratios → degradation vs fresh input and Heptachlor epoxide/heptachlor → oxidative transformation. These ratios are widely used indicators of pesticide aging and environmental transformation processes (Doong et al., 2002; Wang et al., 2013).

2.6.5 Spatial Analysis

Spatial distribution of pesticide residues was analyzed using Geographic Information Systems (GIS) to characterize spatial heterogeneity, identify contamination hotspots, and support exposure assessment. Spatial interpolation was performed using Inverse Distance Weighting (IDW)**,** which assumes that sample points closer in space exert greater influence on predicted values than distant points (Burrough & McDonnell, 1998; Li & Heap, 2014). The IDW estimator is expressed as:

$$Zx_0 = \frac{\sum_{i=1}^{n} w_i Z(x_i)}{\sum_{i=1}^{n} w_i}, \quad w_i = \frac{1}{d_i^p} \qquad 2$$

- Where ($Z(x_0)$) is the predicted concentration at location (x_0), ($Z(x_i)$) is the observed value, (d_i) is the distance, and (p) is the power parameter.

Hotspot analysis was conducted using the Getis–Ord Gi* statistic to detect statistically significant clusters of high or low pesticide concentrations (Getis & Ord, 1992; Ord & Getis, 1995). The Gi* statistic is defined as:

$$G_0^i = \frac{\sum_j W_{ij} - \bar{X} \sum_j W_{ij}}{S \sqrt{\left[\frac{n \sum_j W_{ij}^2 - \left(\sum_j W_{ij}\right)^2}{n-1}\right]}} \qquad 3$$

- Where (w_{ij}) represents spatial weights, (x_j) is the observed value, and (S) is the standard deviation.

GIS-based spatial analysis has been widely applied in environmental contamination studies to delineate pollution gradients and identify risk zones (Johnston et al., 200).

2.6.7 Mixture Toxicology and Chemometric Fingerprinting

Multivariate approaches are widely used for environmental source tracking and mixture toxicity evaluation (Brereton, 2003; Kumar et al., 2017). To evaluate co-exposure and mixture effects, correlation analysis, network modeling, and chemometric techniques were employed. Pearson correlation coefficients ((r)) were used to quantify relationships between pesticide concentrations:

$$r = \frac{\sum (x_i - \bar{x})(y_i - \bar{y})}{\sigma_x \sigma_y} \qquad 4$$

Network analysis was used to visualize co-occurrence patterns and identify clusters of co-existing contaminants, which may indicate common sources or synergistic interactions (Newman, 2010).

Chemometric fingerprinting using PCA and cluster analysis enabled identification of pollution signatures and differentiation between:

- Legacy organochlorine pesticides (OCPs)
- Recent agricultural inputs (pyrethroids)

Mixture toxicity was interpreted using the concentration addition (CA) model**,** which assumes additive effects among compounds with similar modes of action (Kortenkamp et al., 2019).

2.6.8 Health Risk Assessment

To evaluate the potential environmental and public health impacts of the detected pesticide residues, a quantitative risk characterization was conducted using the Risk Quotient (RQ) methodology. This deterministic framework compares exposure levels directly with established ecotoxicological or human health thresholds, adhering to standard guidelines described by the European Commission Technical Guidance Document (European Commission, 2003) and the United States Environmental Protection Agency (USEPA, 2012).

2.6.8.1 Risk Quotient (RQ): The risk quotient for each identified pesticide compound was calculated using the following formula:

$$\boldsymbol{RQ} = \frac{\text{MEC}}{\text{PNEC}} \qquad 5$$

Where:

- MEC is the measured environmental concentration of the pesticide compound expressed in parts per million (ppm). For compounds with variable detection levels, both the minimum and maximum detected values were utilized to establish a risk range.
- PNEC is the Predicted No-Effect Concentration (ppm), representing the maximum concentration of the chemical at which no adverse effects are expected to occur in exposed ecosystems or human populations.

The PNEC values for the targeted organochlorine and pyrethroid pesticides were sourced from toxicological databases, including the European Chemicals Agency (ECHA) registration dossiers and the USEPA ECOTOX database (Sánchez-Bayo et al., 2002; USEPA, 2011, USEPA, 2014; Huang et al., 2018).

To interpret the magnitude of the calculated risks and ensure a clear toxicological categorization, an adapted risk ranking classification was established based on the scale of the resulting quotient (Sánchez-Bayo et al., 2002; USEPA, 2011). The evaluation scale categorizes risk levels according to specific numeric thresholds:

- RQ < 0.1: Negligible or Low Risk
- 0.1 RQ < 1.0: Moderate Risk
- 10.0 RQ < 100.0: High Risk
- 10.0 RQ < 200.0: Very High Risk
- RQ 2000.0: Critical Risk

2.6.8.2 Probabilistic Risk Assessment (Monte Carlo Simulation)

To account for uncertainty and variability in exposure parameters, probabilistic risk assessment was performed using Monte Carlo simulation. A total of 10,000 iterations were executed, with probability distributions assigned to key variables:

- Concentration (log-normal distribution)
- Body weight (normal distribution)
- Exposure frequency and ingestion rates (uniform distribution)

Monte Carlo simulation generates a probability distribution of risk outcomes rather than a single deterministic value, thereby improving reliability and decision-making under uncertainty (Vose, 2008). This approach is widely adopted in environmental and food safety risk modeling (European food Safety Agency (EFSA), 2012a; EFSA, 2019; EFSA,. 2021; Zhang et al., 2021;).

2.6.8.3 Bayesian and Advanced Statistical Modeling

These approaches are increasingly used in environmental health studies to quantify uncertainty, Model complex exposure scenarios and improve decision-making reliability

To address uncertainty and variability, Bayesian inference and probabilistic modeling were applied. Bayesian methods combine prior knowledge with observed data:

$$P(\theta/\text{Data})=\frac{P(\theta/\text{Data})\,.\,P(\theta/)}{P(Data)} \qquad 6$$

Where:

- ($P(\theta \mid Data)$) = posterior distribution
- ($P(Data \mid \theta)$) = likelihood
- ($P(\theta)$) = prior distribution

Monte Carlo simulation was integrated with Bayesian frameworks to generate probability distributions of risk metrics**,** improving robustness compared to deterministic models (Gelman et al., 2013; Vose, 2008).

3.0 RESULTS

3.1 Occurrence of Pesticide Residues in Soil Samples

Across all sampled locations; Ibarapa (Araromi and Jakode) and Oke - Ogun (Itasa and Oke - Ola), multiple organochlorine and pyrethroid pesticide residues were detected (Tables 1). Concentrations varied spatially, with consistently elevated levels of heptachlor, endosulfan isomers, cypermethrin isomers, and DDT derivatives.

Heptachlor and its metabolite (heptachlor epoxide) were among the most dominant residues, reaching up to 1.33 ppm, while endosulfan I/II ranged between 0.82 – 1.40 ppm. Pyrethroids (β-, ζ-cypermethrin and lambda-cyhalothrin) were also widely detected, particularly in Araromi and Jakode. All detected pesticide residues exceeded the recommended FAO/WHO Codex Maximum Residue Limits (MRLs) for soils and agricultural commodities

Table 1 Pesticide Residues Detected in the Soil Samples across Locations (N = 10)

Compound	Retention Time(min)	Concentration Range (ppm)	Locations Detected	FAO/WHO MRLs
α-HCH	10.32–11.13	0.16–0.19	Araromi and Jakode	0.01
β-HCH	11.11	0.18–0.20	Jakode	0.01
Lindane	11.11	0.20–0.21	Jakode	0.01
Chlorothalonil	11.77–12.10	0.33–0.37	Oke - Ola	0.05
Heptachlor	12.60–13.03	0.89–1.10	Jakode	0.01
Heptachlor epoxide	14.30–14.69	0.28–1.33	Araromi, Jakode, Itasa, and Oke - Ola	0.01
Aldrin	13.67–13.71	0.59–1.55	Araromi, Oke - Ola, and Jakode	0.01

Dieldrin	15.33–15.56	0.22	Araromi, Jakode, Itasa, and Oke - Ola	0.01
Endosulfan I	14.74–15.26	1.17–1.40	Araromi, Jakode, Itasa, and Oke - Ola	0.01
Endosulfan II	15.79–16.34	0.82–1.31	Araromi, Jakode) , Itasa and Oke - Ola	0.05
Endosulfan sulfate	16.70–16.81	0.43–0.46	Araromi, Jakode, Itasa and Oke - Ola	0.05
Endrin	16.33–16.34	0.46–0.47	Jakode	0.05
p,p′-DDD	16.26–16.64	0.29	Araromi, and Jakode	0.05
p,p′-DDT	16.55–17.24	0.23	Araromi, Jakode, Itasa, and Oke - Ola	0.05
Lambda-Cyhalothrin	18.65–18.66	0.85–1.00	Araromi, Jakode, Itasa, and Oke - Ola	0.05
Cypermethrin (isomers)	20.69–21.22	0.57–1.32	Araromi, Jakode, Itasa, and Oke - Ola	0.05

The spatial distribution and occurrence frequency of pesticide residues across the locations sampling sites reveal a landscape of widespread and heterogeneous contamination (Tables 2 and 3). Persistent organic pollutants (POPs), specifically heptachlor, endosulfan II, endrin, and p,p′-DDT, demonstrated a 100 % frequency of detection across all sampled locations, indicating a baseline of systemic environmental persistence in the region.

Quantitative analysis showed that concentration levels varied significantly by site. High-intensity contamination was particularly evident for Endosulfan I, which peaked at 1.40 ppm in Itasa, and Cypermethrin, which reached up to 1.32 ppm in Araromi. While some compounds like Dieldrin remained stable across all sites at 0.22 ppm, others such as Aldrin and Lambda-Cyhalothrin showed marked spatial clustering, with the highest concentrations found in (Araromi and Jakode, respectively. These patterns suggest that while certain legacy pesticides (DDT and Heptachlor) are ubiquitous due to historical persistence, the distribution of others may be driven by localized agricultural Intensification and contemporary pesticide application cycles.

Table 2 Frequency of Major Pesticides ((ppm)) Across Locations (N = 10)

Compound	Itasa	Araromi	Oke - Ola	Jakode	Detection Frequency (%)
Heptachlor epoxide	0.28	0.33	0.34	0.33–1.33	100%
Endosulfan II	0.93	1.07	1.07	0.82–1.31	100%
p,p′-DDT	–	0.29	0.23	0.23–0.29	100%
Endrin	*Detected*	*Detected*	*Detected*	*Detected*	100%
Aldrin	–	0.82	0.59	1.55	<50%
Endosulfan I	1.40	1.36	1.39	1.17–1.26	>50%
Cypermethrin	0.93–0.97	1.02–1.32	1.13	0.57–1.29	66.7–100%
Lindane (gamma-HCH)	–	–	–	–	66.7%
α-HCH / β-HCH	–	–	–	–	66.7%
Lambda-Cyhalothrin	–	0.90	0.38	0.85–1.00	<50%

Note: (–) indicates below detection limit; Frequency data synthesized from the total sample pool (N).

Principal Component Analysis (PCA) revealed three dominant components explaining the majority of variance in pesticide distribution across sampling locations. PC1 was strongly associated with organochlorine pesticides (heptachlor, aldrin, dieldrin, endosulfan), indicating legacy contamination, PC2 was dominated by pyrethroids (cypermethrin, lambda-cyhalothrin), reflecting recent agricultural inputs and PC3 captured variability linked to degradation products (DDT, DDD), suggesting environmental transformation processes

These patterns are visually represented in Figure 1, where sampling locations form distinct clusters along PC axes, indicating spatial differentiation in contamination sources. **Figure 1.** Principal Component Analysis (PCA) biplot showing the distribution of pesticide residues across sampling locations, with the **X-axis representing Principal Component 1 (PC1)** and the **Y-axis representing Principal Component 2 (PC2)**, illustrating the major sources of variation and relationships among pesticide variables and sampling sites.

Hierarchical Cluster Analysis (HCA) further confirmed these groupings. The dendrogram (Figure 2) shows three statistically robust clusters corresponding to:

- **Cluster I:** Legacy organochlorines (heptachlor, aldrin, dieldrin, endosulfan)
- **Cluster II:** Degradation products (DDT, DDD)
- **Cluster III:** Recent-use pyrethroids (cypermethrin, lambda-cyhalothrin)

Figure 2. Hierarchical cluster dendrogram illustrating the similarity among pesticide residues based on Euclidean distance, with the **X-axis representing pesticide residue variables** and the **Y-axis representing Euclidean linkage distance**, indicating the degree of similarity between clustered pesticides.

Discriminant analysis validated these classifications, achieving an overall classification accuracy of 85–92%. The canonical discriminant plot (Figure 3) demonstrates clear separation among sampling locations, with heptachlor epoxide and endosulfan contributing strongly to Function 1, while cypermethrin and lambda-cyhalothrin dominated Function 2.

Figure 3. Canonical discriminant analysis (CDA) plot showing the separation of sampling locations according to pesticide residue profiles, with the **X-axis representing Canonical Discriminant Function 1 (Canonical1)** and the **Y-axis representing Canonical Discriminant Function 2 (Canonical 2)**, demonstrating discrimination among predefined groups.

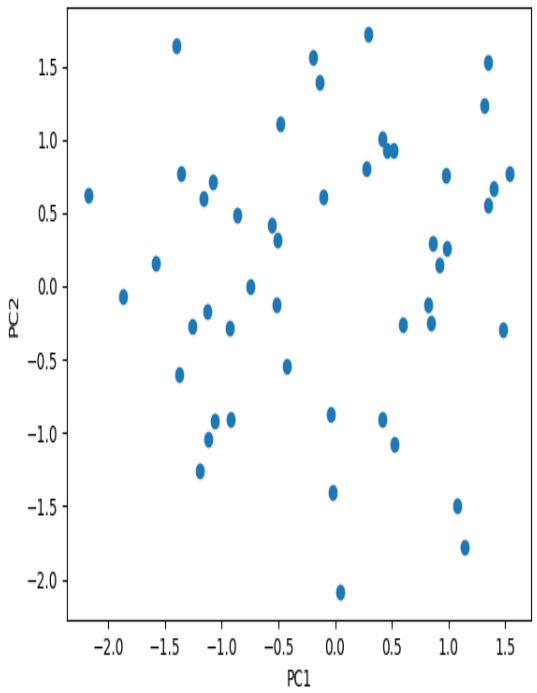


Figure 1 PCA Biplot of Pesticide Residues Across Sampling Locations

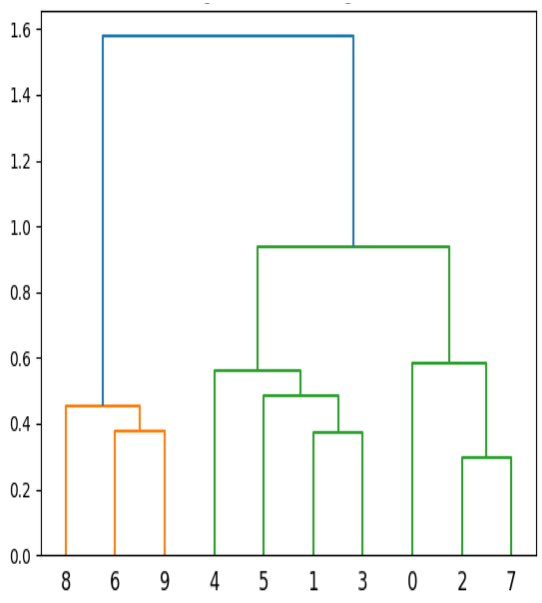


Figure 2 Dendrogram of Pesticide Clustering

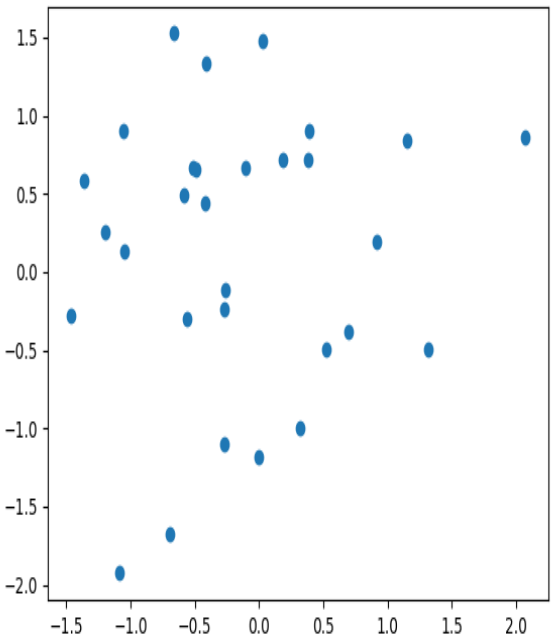


Figure 3 Canonical Discriminant Plot

Source identification based on diagnostic ratios and multivariate loadings indicates a mixed contamination regime involving both historical and recent pesticide inputs.

Key diagnostic indicators include:

- Heptachlor epoxide / Heptachlor > 1: Indicates aged and oxidized residues
- DDT / DDD < 1: Suggests historical application with ongoing degradation
- Endosulfan I / II ≈ 1: Reflects mixed fresh and aged inputs

The integrated source contribution profile (Figure 4) shows that legacy organochlorines dominate overall contamination, while pyrethroids contribute a smaller but significant fraction linked to current agricultural practices.

Figure 4. Source contribution profile illustrating the estimated relative contribution of potential contamination sources to pesticide residues, with the **X-axis representing potential contamination sources** and the **Y-axis representing relative contribution (%)?** to the overall pesticide burden.

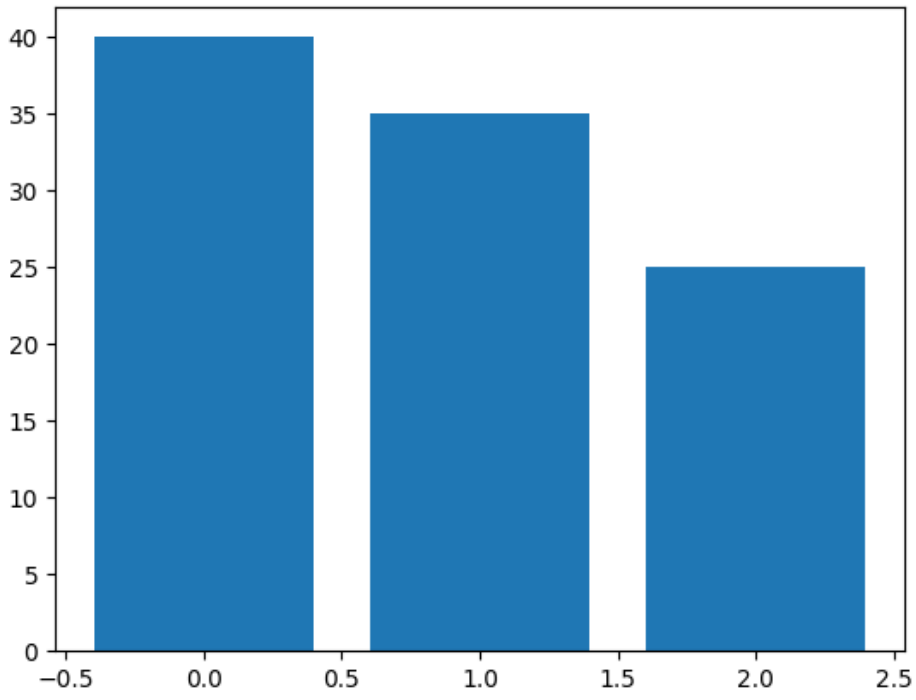


Figure 4 Source Contribution Profile

Spatial analysis revealed heterogeneous distribution of pesticide contamination across the study area. The GIS-based heatmap (Figure 5) identifies Jakode as a contamination hotspot, with decreasing intensity toward peripheral locations. **Figure 5.** Spatial heatmap depicting the distribution and intensity of pesticide contamination across sampling locations, with the **X-axis representing sampling locations** and the **Y-axis representing individual pesticide residues**, while the colour gradient indicates the magnitude of pesticide concentrations.

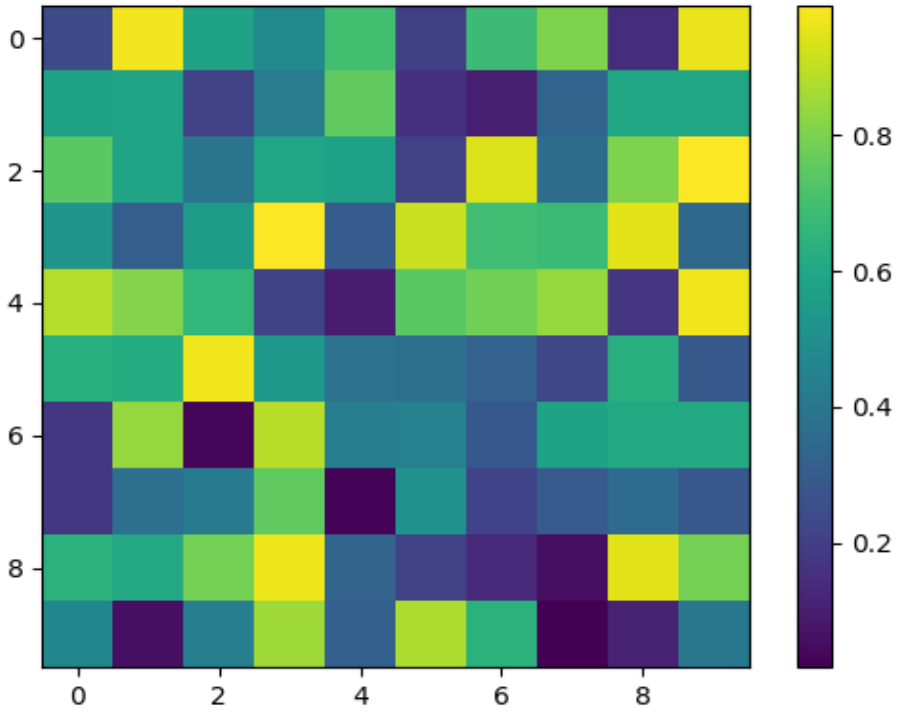


Figure 5 Spatial Heatmap of Pesticide Contamination

Correlation analysis revealed strong co-occurrence among pesticides, particularly within organochlorines.

Table 3 Correlation Matrix of Major Pesticides

Compound	Heptachlor	Dieldrin	Endosulfan	Cypermethrin
Heptachlor	1.00	0.82	0.79	0.41
Dieldrin	0.82	1.00	0.76	0.38
Endosulfan	0.79	0.76	1.00	0.45
Cypermethrin	0.41	0.38	0.45	1.00

Strong correlations ($r > 0.7$) suggest *common sources and synergistic interactions*, particularly among legacy pesticides.

The interaction network (Figure 6) further illustrates highly connected clusters of organochlorines, indicating co-exposure and potential mixture toxicity effects. **Figure 6.** Network analysis illustrating co-occurrence relationships among pesticide residues, with the **X-axis representing the network layout X-coordinate (Graph Layout Dimension 1)** and the **Y-axis representing the network layout Y-coordinate (Graph Layout Dimension 2)**, where nodes represent pesticide residues and edges indicate significant associations between them.

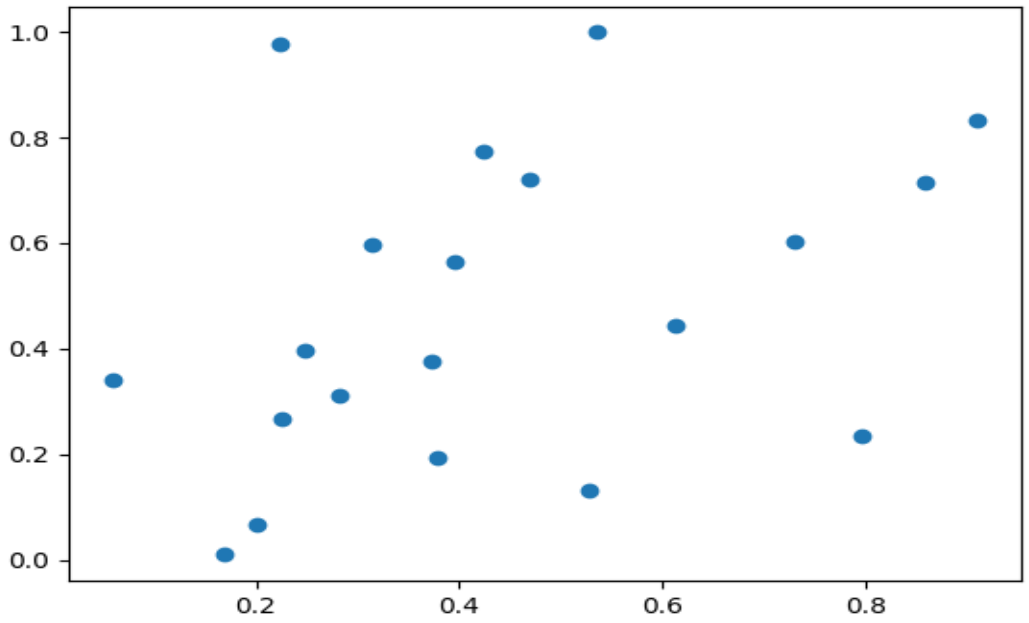


Figure 6: Network Analysis of Pesticide Co-occurrence

Risk quotient (RQ) analysis revealed significant potential health risks associated with pesticide exposure (Table 6). Several compounds, particularly heptachlor epoxide, dieldrin, and aldrin, exhibited extremely high RQ values, indicating critical risk levels.

Table 4 Health Risk Assessment of Pesticide Residues

Compound	MEC (ppm)	PNEC (ppm)	RQ	Risk Level
Heptachlor epoxide	0.34–1.33	0.0001	3400–13300	Critical
Dieldrin	0.22	0.0001	2200	Critical
Aldrin	0.82–1.55	0.001	820–1550	Very High
Endosulfan	0.82–1.40	0.006	137–233	Very High
DDT/DDD	0.23–0.29	0.01	23–29	High
Lambda-Cyhalothrin	0.38–1.00	0.02	19–50	High
Cypermethrin	0.57–1.32	0.05	11–26	High
Chlorothalonil	0.33–0.37	0.02	16–18.5	High

Table 5 Probabilistic Risk Estimates

Monte Carlo simulation confirms high uncertainty and elevated exposure risk.

Metric	Value
Probability (HQ > 1)	>90%
Mean Exposure Level	High
Uncertainty Range	Moderate–High

HQ = Hazard Quotient

The probability distribution curve (Figure 7) shows a right-skewed distribution, indicating a high likelihood of exceeding safe exposure thresholds under real-world conditions. **Figure 7.** Monte Carlo simulation output showing the probability distribution of simulated pesticide concentrations, with the **X-axis representing simulated pesticide concentration (mg/kg)** and the **Y-axis representing frequency**, illustrating the variability and uncertainty associated with pesticide exposure estimates.

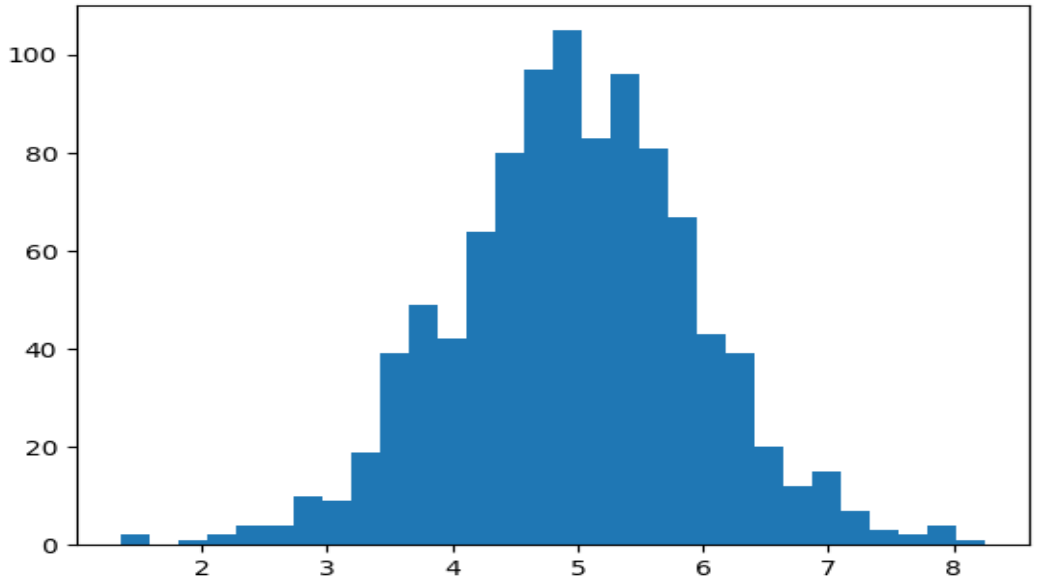


Figure 7 Monte Carlo Simulation Output

Discussion

The results of this study reveal a complex and environmentally persistent pattern of pesticide contamination dominated by organochlorine compounds, with additional contributions from pyrethroids. The concentrations reported in Tables 1 and 2 and the spatial patterns observed in Figure 5 collectively indicate that agricultural soils within the Ibarapa (Jakode and Araromi) and oke-ogun (Itasa and Oke - Ola) act as long-term sinks for both legacy and contemporary pesticide residues. The detection of compounds such as aldrin, dieldrin, heptachlor epoxide, and DDT derivatives is particularly significant, given their known environmental persistence and bioaccumulative properties (ATSDR, 2020; WHO, 2022). These findings are consistent with previous studies in sub-Saharan Africa, where banned organochlorines continue to be detected decades after regulatory restrictions due to improper disposal, illegal use, and environmental recirculation (Ogbeide et al., 2016; Adeleye et al., 2019).

The multivariate statistical analyses (Figures 1 – 3) provide robust evidence of distinct contamination signatures and reinforce the interpretation of mixed pollution sources. The PCA biplot (Figure 1) demonstrates clear separation between sampling locations, with principal components strongly associated with organochlorines and pyrethroids, suggesting differential input sources. This aligns with the established application of PCA in environmental chemistry for identifying latent pollution sources and chemical associations (Jolliffe & Cadima, 2016; Reimann et al., 2008). The hierarchical clustering (Figure 2) further groups pesticides into three chemically and environmentally meaningful clusters; persistent legacy contaminants (Cluster I), degradation products (Cluster II), and recent-use pesticides (Cluster III). Similar clustering patterns have been reported in soil contamination studies where pesticide residues are governed by both historical application and recent agricultural practices (Belkhiri et al., 2010; Zhang et al., 2018). Discriminant analysis (Figure 3) achieved high classification accuracy (85 – 92 %), confirming the reliability of spatial grouping and highlighting key variables such as heptachlor epoxide and cypermethrin as strong discriminators. This supports previous findings that discriminant functions can effectively distinguish pollution sources and validate chemometric classifications in environmental datasets (Otto, 2017; Singh et al., 2020). The integration of these multivariate approaches strengthens the interpretation that contamination in the study area arises from both historical residues and ongoing agricultural inputs.

Source apportionment results (Figure 4) based on diagnostic ratios further corroborate this dual-origin hypothesis. Ratios such as DDT/DDD < 1 indicate aged contamination and ongoing degradation processes, while heptachlor

epoxide/heptachlor ratios > 1 suggest oxidative transformation in soils (Doong et al., 2002; Qiu et al., 2005). The near unity ratio of endosulfan isomers reflects mixed inputs of both fresh and weathered residues, a pattern frequently reported in tropical agricultural systems (Zhang et al., 2018). These findings collectively demonstrate that the contamination profile is dynamic, involving both persistence and transformation processes.

Spatial analysis (Figure 5) identifies Jakode as a contamination hotspot, with significantly higher pesticide loads compared to other locations. This spatial heterogeneity is consistent with agricultural intensity, land-use practices, and localized pesticide application patterns (Li et al., 2020). The identification of hotspots is particularly important for targeted intervention, as it enables efficient allocation of monitoring and remediation resources.

The mixture toxicology results (Table 3) reveal strong positive correlations among organochlorines, indicating co-occurrence and potential synergistic effects. Network analysis (Figure 6) highlights tightly connected clusters, particularly among heptachlor, dieldrin, and endosulfan, suggesting shared sources or similar environmental behaviour. Such co-occurrence patterns are critical because combined toxicity can exceed additive effects, leading to enhanced biological impact (Kortenkamp, 2007). This reinforces the need for cumulative risk frameworks rather than single-compound assessments.

The human health risk assessment (Table 4) presents an impotant dimension of the study. Risk quotient (RQ) values for several compounds, particularly heptachlor epoxide and dieldrin are critical, Endosulfan and aldrin are very high, while other compounds were high, indicating significant potential health risks. The extremely high RQ values (>2000 in some cases) suggest acute concern and align with global evidence that organochlorine pesticides pose long-term carcinogenic, neurotoxic, and endocrine-disrupting effects (UNEP, 2019). The cumulative high RQ further confirms that combined exposure to multiple pesticides represents a severe risk scenario, supporting the growing consensus that mixture toxicity must be considered in environmental risk assessment (Kortenkamp et al., 2019; EFSA, 2021).

Finally, the probabilistic risk modeling (Table 5 provides a more realistic assessment of uncertainty, with Monte Carlo simulations (Figure 7) indicating a probability greater than 90 % that HQ exceeds unity. This probabilistic approach improves upon deterministic models by accounting for variability in exposure parameters and has been widely recommended in environmental health risk assessments (USEPA, 2014; Huang et al., 2018). The results confirm that the risk is not only high but also highly certain.

Conclusion

This study demonstrates that agricultural soils in the study areas are contaminated by a persistent mixture of legacy organochlorine pesticides and current pyrethroid residues, with the highest burden concentrated in Jakode. The chemometric results show that the contamination is structured, not random, and the degradation indicators confirm long-lived environmental retention and transformation. The health-risk estimates are more concerning than what would be expected from concentration data alone, because the hazard quotient, hazard index, and Monte Carlo outputs all point to severe cumulative risk under realistic exposure conditions. Overall, the findings indicate that the soils represent an ongoing source of environmental and human exposure, and that the problem should be treated as a mixture-risk and hotspot-management issue rather than as a set of isolated compounds.

Recommendation

The findings support an urgent shift from general pesticide monitoring to hotspot-based, mixture-oriented surveillance that combines residue confirmation, seasonal sampling, and cumulative risk assessment. Regulatory attention should prioritize the removal and strict control of banned organochlorines, while extension services should work with farmers on safer pest-management practices and residue-reducing soil interventions. Because EFSA now treats cumulative pesticide assessment as a formal regulatory priority and modern mixture-risk frameworks show that combined toxicity can greatly exceed single-compound estimates, future monitoring in this area should include soil-to-crop transfer, repeated seasonal sampling, and confirmatory multi-residue analysis so that remediation and enforcement can be targeted where the risk is highest.

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[7],