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Legacy Pesticides in Aquatic Systems: Mechanistic Persistence, Regulatory Lag, and Catchment-Scale Remediation Imperatives

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ABSTRACT

Legacy pesticides remain a persistent environmental challenge despite regulatory bans and reductions in agricultural pesticide use. Continued detections in surface water and groundwater indicate that historical residues stored in soils and sediments contribute to long-term contamination. This review evaluates the mechanistic processes governing legacy pesticide persistence, including adsorption–desorption dynamics, sediment storage, preferential flow pathways, and groundwater recharge behavior. A structured literature synthesis of peer-reviewed studies published between 2000 and 2025 was conducted to assess monitoring trends, physicochemical determinants of persistence, and remediation technologies applicable at the catchment scale. Evidence indicates that exceedances of the 0.1 µg L^{-1} regulatory threshold remain frequent in vulnerable agricultural regions because of delayed release from environmental reservoirs rather than ongoing application alone. Adsorption-based technologies, particularly activated carbon systems, show potential for practical mitigation, while biochar amendments and advanced oxidation processes offer complementary treatment pathways. The analysis indicates that regulatory restrictions should be coupled with targeted remediation and soil risk assessment to achieve measurable improvements in water quality. An integrated catchment-level management framework is therefore proposed to address legacy contamination and support sustainable water resource protection.

1. INTRODUCTION

The intensification of agricultural production over recent decades has been accompanied by a substantial increase in pesticide use worldwide, supporting global food security but also contributing to diffuse environmental contamination (Carvalho 2017, Silva et al. 2019, Ramchandar et al. 2025). Although regulatory frameworks across the European Union and other regions have progressively restricted or banned numerous active substances, measurable improvements in water quality have not consistently followed at the anticipated rate (European Commission 2020, European Environment Agency 2022). This apparent regulatory–environmental disconnect has drawn attention to the persistence of historically applied compounds, commonly referred to as legacy pesticides.

Legacy pesticides are compounds whose environmental presence continues despite regulatory withdrawal or substantial reduction in use. Their persistence is increasingly attributed to strong soil–organic matter interactions, slow degradation kinetics, and delayed release mechanisms rather than ongoing application alone (Fenner et al. 2013, Geissen et al. 2015). Several monitoring programs continue to report exceedances of the 0.1 μg L^{-1} drinking-water threshold in agricultural catchments, particularly for herbicides such as atrazine, 2,4-D, and MCPA (Gilliom et al. 2006, Lapworth et al. 2012). These findings suggest that soil and sediment compartments function as long-term contaminant reservoirs capable of sustaining chronic exposure (Gzyl et al. 2014).

Environmental persistence is not solely governed by degradation half-life. Instead, it emerges from coupled processes involving adsorption–desorption dynamics, preferential flow pathways, sediment remobilization, and groundwater recharge (Jarvis 2007, Schwarzenbach et al. 2006). While some studies emphasize natural attenuation as a sufficient recovery mechanism, others argue that active remediation strategies may be required to accelerate environmental restoration (Gavrilescu 2005, Sharma et al. 2018). This divergence highlights an ongoing debate regarding the relative roles of regulatory reduction and targeted intervention.

Although substantial research has examined pesticide transport and treatment technologies independently, fewer studies have integrated mechanistic persistence with catchment-scale mitigation planning. In particular, the translation of soil-based risk assessment into practical remediation strategies remains limited. Addressing legacy contamination therefore requires bridging laboratory-scale understanding with policy implementation and watershed management.

Despite increasingly stringent regulatory frameworks, significant regional differences persist in pesticide governance, contributing to regulatory lag. In the European Union, a precautionary approach has established a strict drinking-water limit of 0.1 μg L^{-1} for individual pesticides, yet exceedances continue to be reported because of legacy residues and delayed environmental release. In contrast, regulatory thresholds in countries such as the United States are generally compound-specific and often higher, reflecting a risk-based approach. In countries such as India, regulatory enforcement and monitoring capacity remain variable, further complicating the effective management of pesticide contamination. These disparities indicate that reductions in pesticide use alone are insufficient and that legacy contamination requires integrated remediation strategies alongside regulatory controls (European Union 2009, European Union 2021, Knauer 2016, Harmon O'Driscoll et al. 2022, Lewis et al. 2016).

A comparative overview of regulatory frameworks and associated challenges across regions is presented in Table 1.

The primary aim of this review is to critically evaluate the mechanistic drivers of legacy pesticide persistence in aquatic systems and to assess the effectiveness of available remediation strategies within a catchment-scale management framework. By synthesizing evidence from monitoring data, soil transport studies, and adsorption-based treatment research, this review seeks to clarify the factors sustaining long-term contamination and to propose integrated approaches capable of supporting measurable improvements in water quality.

2. MATERIALS AND METHODS

2.1. Review Design and Scope

This study employed a structured narrative review approach, integrating mechanistic laboratory investigations, field monitoring evidence, and policy analyses addressing legacy pesticide persistence. The review aimed to synthesize current scientific understanding of environmental reservoirs, transport pathways, persistence drivers, and remediation strategies applicable at the catchment scale.

This study adopted a structured narrative review approach informed by PRISMA reporting principles without conducting a full systematic review. Although a meta-analysis was not conducted, systematic principles were followed to ensure transparency, reproducibility, and methodological rigor.

2.2. Literature Search Strategy

A systematic literature search was conducted using the Scopus, Web of Science, and Google Scholar databases. The search period covered publications from January 2000 to March 2025 to capture contemporary developments in environmental persistence research and remediation technologies.

Search strings included combinations of the following keywords: "legacy pesticides", "groundwater contamination", "surface water monitoring", "adsorption–desorption", "activated carbon remediation", "soil transport risk", and "catchment management".

Table 1: Comparative overview of pesticide regulatory frameworks and associated challenges.

Region	Regulatory Approach	Drinking Water Limit	Key Challenge
European Union	Precautionary	0.1 μg L^{-1} (individual)	Persistent legacy residues despite regulatory bans
United States	Risk-based	Compound-specific (generally higher)	Variability across compounds and standards
India	Developing/variable	Variable/not uniformly enforced	Limited monitoring and enforcement capacity

Boolean operators (AND/OR) were applied to refine the search results. Reference lists of selected articles were also screened to identify relevant studies not retrieved during the primary database search.

The initial database search yielded 628 records. After removal of duplicate entries (n = 80), 548 records were retained for title and abstract screening.

2.3. Screening and Study Selection

Titles and abstracts of 548 records were screened for relevance. A total of 360 records were excluded because they were irrelevant, focused on toxicity without environmental context, or were not peer reviewed. The remaining 188 studies were assessed for full-text eligibility. Of these, 110 studies were excluded because of insufficient data or lack of relevance. Finally, 78 studies were included in the qualitative synthesis. The study-selection process is summarized in Fig. 1.

2.4. Inclusion and Exclusion Criteria

2.4.1. Studies were Included if They

- Reported field-based monitoring data of pesticide residues in surface or groundwater;
- Investigated adsorption–desorption mechanisms in soil or sediment systems;
- Examined physicochemical parameters influencing persistence (e.g., DT_{50}, K_oc, log K_ow);
- Evaluated remediation technologies under laboratory, pilot-scale, or field conditions;
- Addressed regulatory or policy implications related to pesticide persistence.

2.4.2. Studies were Excluded if They:

- Focused exclusively on current-use pesticides without documented persistence relevance;

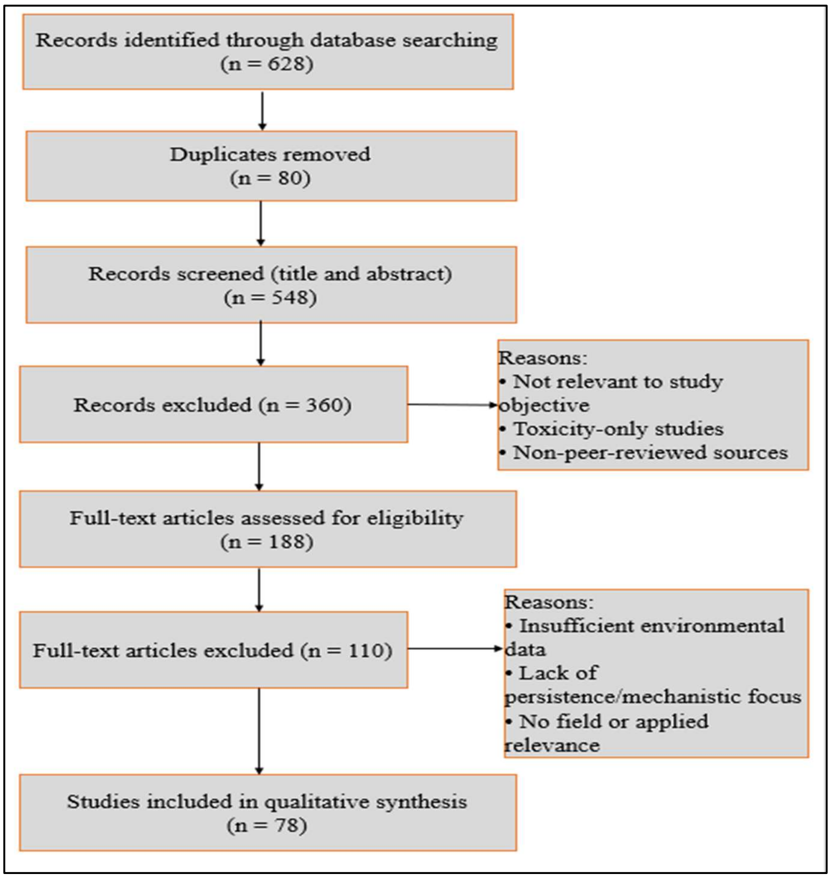

Fig. 1: PRISMA-style flow diagram illustrating the literature search and study selection process. A total of 628 records were identified through database searches, of which 548 were screened after duplicate removal. Following eligibility assessment, 78 studies were included in the structured narrative synthesis.

- Reported acute toxicity studies without environmental transport or fate analysis;
- Were not peer-reviewed publications.

2.5. Quality Assessment of Included Studies

The quality of included studies was assessed based on study design, analytical reliability, and reporting completeness. Studies were categorized as high, moderate, or low confidence. High-confidence studies included field-based investigations with quantitative data, while moderate-confidence studies were laboratory-based with clear methodology. Low-confidence studies were excluded from synthesis.

A summary of the quality appraisal of the included studies (n = 78), based on study design and methodological reliability, is presented in Table 2.

2.6. Data Extraction and Synthesis

From each eligible study, the following information was extracted:

- Compound characteristics (e.g., half-life, sorption coefficient, aqueous solubility);
- Environmental compartment investigated (soil, sediment, surface water, groundwater);
- Reported concentration ranges;
- Remediation method evaluated and associated removal efficiency;
- Geographic and climatic context.

Due to heterogeneity in monitoring methodologies, analytical techniques, and reporting units, a quantitative meta-analysis was not performed. Instead, a structured qualitative synthesis was conducted, supported by semi-quantitative comparison of reported parameter ranges (e.g., DT_{50}, Koc, and concentration levels), to identify recurring mechanistic patterns, dominant drivers of persistence, and consistent policy implications.

2.7. Comparative Assessment of Remediation Technologies

Remediation technologies were comparatively evaluated based on the following criteria:

- Dominant mechanism of contaminant removal (adsorption, degradation, immobilization);
- Demonstrated field applicability (laboratory, pilot, or full-scale);
- Regeneration potential and long-term sustainability;
- Scalability and operational feasibility at the catchment scale;
- Reported removal efficiency ranges where available.

This evaluation framework enabled systematic comparison among adsorption-based systems, soil amendments, and advanced oxidation processes.

Where possible, comparative interpretation was supported by reported performance ranges and field validation evidence to distinguish between laboratory efficiency and real-world applicability.

2.8. Data Availability and Transparency

All data analyzed in this review were derived from published peer-reviewed sources. No new experimental data were generated. The study selection process, synthesis approach, and extracted parameters are described in detail to ensure transparency and reproducibility. In accordance with journal policy, the reference list and extracted synthesis tables will be made fully accessible upon publication. There are no restrictions on data availability.

2.9. Ethical Considerations

This study is based exclusively on previously published data and does not involve human participants, animal experimentation, or intervention-based trials. Therefore, ethical approval was not required.

3. RESULTS AND DISCUSSION

3.1. Environmental Persistence Mechanisms

3.1.1. Physicochemical Controls on Long-Term Storage

While sorption affinity (Koc) is widely recognized as a primary control on pesticide retention in soils, evidence across field-scale studies indicates that its influence is highly context-dependent. High Koc values promote strong adsorption to soil organic matter, reducing immediate mobility; however, this effect can be overridden under conditions of high hydrological flux or preferential flow pathways, particularly in coarse-textured or structured soils (Gondar et al. 2013, Kodešová et al. 2011, McManus et al. 2017).

Table 2: Quality appraisal summary of included studies (n = 78).

Study Type	Number of Studies	Key Characteristics	Confidence Level
Field monitoring	34	Long-term environmental monitoring, real-world datasets	High
Laboratory studies	26	Controlled adsorption/desorption experiments	Moderate
Modeling/Combined	18	Risk assessment, modeling, and integrated analysis	Moderate

Similarly, although degradation half-life (DT_{50}) is commonly used as an indicator of persistence, several studies demonstrate that compounds with moderate DT_{50} values may still exhibit long-term environmental presence when strong sorption delays degradation or when residues are protected within soil matrices. Conversely, pesticides with relatively short DT_{50} values can be rapidly transported to groundwater systems under high rainfall or irrigation conditions, indicating that transport processes may dominate over degradation (Aguiar Jr. et al. 2017, Hakoun et al. 2017, Jørgensen & Spliid 2016).

These contrasting findings suggest that persistence cannot be attributed to a single controlling parameter. Instead, it emerges from the dynamic interaction between sorption, degradation kinetics, and hydrological connectivity, with the dominant control varying across environmental conditions. The principal physicochemical controls are summarized in Table 3.

These parameters collectively determine persistence through their interaction, with the dominant controlling mechanism varying across environmental conditions such as soil texture, rainfall intensity, and hydrological connectivity, rather than following a fixed mathematical relationship.

In addition to qualitative interpretation, several studies provide indicative threshold ranges that help explain variability in environmental persistence. Pesticides with soil half-lives (DT_{50}) greater than 100 days are generally classified as persistent, while those exceeding 365 days are considered highly persistent, with potential for long-term environmental storage. However, persistence risk increases significantly when moderate DT_{50} values (30–100 days) are combined with high sorption affinity ($Koc > 1000$), as delayed degradation may occur due to strong binding within soil matrices.

Conversely, compounds with low sorption coefficients ($Koc < 300$) and high water solubility exhibit increased leaching potential, particularly in sandy or permeable soils where preferential flow pathways dominate. Under such conditions, transport processes may outweigh degradation effects, leading to rapid groundwater contamination even for compounds with relatively short DT_{50} values.

These findings reinforce that persistence emerges from interacting parameter ranges rather than isolated physicochemical properties.

A hierarchical interpretation of controlling mechanisms further clarifies the variability in persistence behavior across environmental settings. Under low rainfall and fine-textured soils (e.g., clay-rich systems), sorption processes (Koc) tend to dominate, limiting mobility and promoting long-term storage within soil matrices. In contrast, in coarse-textured or structured soils with high permeability, hydrological connectivity becomes the primary controlling factor, facilitating rapid transport through preferential flow pathways regardless of sorption strength.

Degradation processes, represented by DT_{50}, exert a secondary control that is strongly influenced by environmental conditions. In well-aerated soils with active microbial communities, degradation may significantly reduce persistence; however, in saturated or subsurface environments, degradation rates are often limited, allowing stored residues to persist over extended periods.

This hierarchy indicates that hydrology and soil structure frequently override physicochemical retention under dynamic field conditions, while sorption dominates under stable conditions. Consequently, persistence should be interpreted as a condition-dependent outcome rather than a fixed property of individual compounds.

3.1.2. Adsorption-Desorption and Delayed Release

Adsorption–desorption dynamics play a critical role in sustaining long-term pesticide persistence beyond initial application periods. Several studies report nonlinear and hysteretic behavior in adsorption–desorption processes, indicating that desorption is often slower and less complete than adsorption. This asymmetry results in the formation of bound residues within soil matrices, which are not immediately bioavailable but can act as long-term contaminant reservoirs (Barriuso et al. 2008, Boivin et al. 2005, Foo and Hameed 2010).

Over time, gradual desorption from soil organic matter and sediment compartments leads to sustained low-level release of pesticide residues into pore water and connected aquatic systems. This delayed release mechanism explains

Table 3: Physicochemical properties governing environmental persistence of legacy pesticides.

Parameter	Definition	Influence on Persistence
log Kow	Hydrophobicity indicator	Promotes soil/sediment accumulation
Koc	Soil organic carbon affinity	Enhances long-term storage
DT_{50}	Degradation half-life	Determines degradation rate
Water solubility	Mobility indicator	Controls runoff and leaching potential
pKa	Ionization behavior	Influences adsorption–desorption

the continued detection of legacy pesticides in surface water and groundwater long after their regulatory withdrawal.

Importantly, the rate and extent of desorption are strongly influenced by environmental conditions. In fine-textured soils with high organic matter content, strong sorption can prolong retention and delay release. In contrast, changes in hydrological conditions—such as rainfall events, fluctuations in groundwater levels, or disturbance of sediments—can enhance remobilization of previously stored residues.

These findings indicate that persistence is not only a function of initial sorption but also of long-term desorption kinetics and environmental perturbations. As a result, legacy pesticide contamination should be understood as a time-dependent release process from environmental reservoirs rather than a static outcome of past applications.

3.1.3. Sediment Reservoirs and Episodic Mobilization

Sediments function as important secondary reservoirs for legacy pesticides, where particle-bound residues accumulate over extended periods due to strong sorption to organic matter and suspended solids. While this process reduces immediate bioavailability, it does not result in permanent immobilization, as stored contaminants remain susceptible to remobilization.

Under stable hydrological conditions, sediments predominantly act as sinks, facilitating long-term accumulation of pesticide residues. However, during high-flow events such as intense rainfall or seasonal runoff, these sediments can be resuspended, releasing previously stored contaminants into the water column. This episodic mobilization results in transient concentration spikes and explains continued detections even in catchments where pesticide application has declined or ceased (Andrade et al. 2021, Halbach et al. 2021, Ivanova et al. 2021).

The relative importance of sediment-mediated transport is strongly influenced by catchment characteristics and hydrological variability. In low-energy systems, storage processes dominate, whereas in dynamic catchments with frequent disturbance, sediments transition from sinks to active sources, significantly contributing to contaminant transport.

These findings demonstrate that sediment-associated persistence is governed by a dynamic balance between storage and remobilization processes. Therefore, effective management of legacy pesticide contamination should explicitly incorporate sediment dynamics within catchment-scale monitoring and mitigation strategies. The principal persistence pathways are illustrated in Fig. 2.

3.2. Monitoring Evidence and Regulatory Disconnect

Long-term monitoring datasets demonstrate persistent exceedances of the 0.1 µg L^{-1} threshold in vulnerable agricultural regions (European Commission 2020). Surface-water monitoring often indicates gradual improvement, whereas groundwater recovery lags because of longer residence times (Lapworth et al. 2012, Chow et al. 2020, McKnight et al. 2015, Melin et al. 2020, Papadakis et al. 2018, Schreiner et al. 2016, Stehle and Schulz 2015).

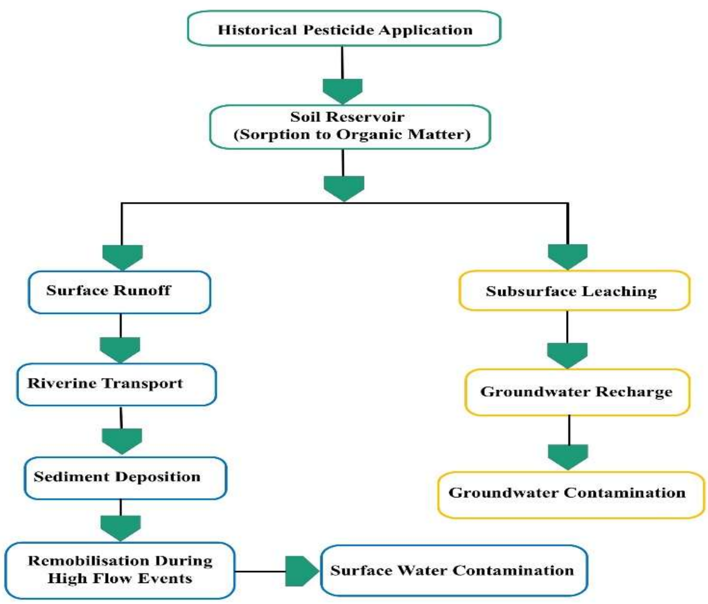

Fig. 2: Conceptual pathways governing legacy pesticide persistence across soil, sediment, and groundwater compartments.

Key observations include:

- Continued detection of atrazine despite its ban in the EU;
- Recurring exceedances of MCPA and 2,4-D in intensive catchments;
- Higher transport risk in sandy soils than in clay-dominated systems.

These findings suggest that regulatory restrictions alone cannot eliminate contamination sustained by historical soil reservoirs. Frequently detected legacy pesticides and selected persistence indicators are summarized in Table 5.

3.3. Comparative Evaluation of Remediation Technologies

Efficiency ranges are indicative and vary depending on experimental conditions, contaminant type, and environmental settings.

The reviewed remediation technologies exhibit significant variability in performance, scalability, and field applicability, indicating that no single approach is universally applicable across all environmental settings. Adsorption-based systems, particularly granular and powdered activated carbon, have been shown to consistently achieve high removal efficiencies (typically 70–95%) under both laboratory and field conditions, making them among the most reliable and widely implemented options for water treatment (Cosgrove et al. 2019, Cosgrove et al. 2022, Ighalo et al. 2021, Mojiri et al. 2020, Saleh et al. 2020).

However, their long-term effectiveness is constrained by media saturation and the need for regeneration or replacement, which introduces operational and cost considerations. In contrast, biochar-based amendments offer a more sustainable and cost-effective alternative for in situ soil remediation, but their performance is highly variable (30–80%) depending on feedstock properties, soil type, and environmental conditions.

Advanced oxidation processes provide efficient chemical degradation of pesticide residues under controlled conditions, often achieving high removal rates. Nevertheless, their application at the catchment scale is limited by energy requirements, operational complexity, and reduced efficiency in heterogeneous environmental matrices.

Emerging technologies such as magnetic nanocomposites show promising adsorption and recovery potential; however, their application remains largely experimental due to scalability constraints and limited field validation (Intisar et al. 2022, Shahid et al. 2021).

These comparisons indicate that adsorption-based technologies are most suitable for immediate water quality improvement, while soil-based amendments and advanced oxidation processes serve complementary roles depending on site-specific conditions. Therefore, effective remediation strategies should adopt a combined, site-adaptive approach, integrating multiple technologies based on contaminant characteristics, environmental conditions, and management objectives (Butkovskyi et al. 2021, Jing et al. 2021, Lorenz et al. 2022, Prosser et al. 2020). The comparative performance

Table 4: Comparative assessment of remediation strategies for legacy pesticide mitigation.

Technology	Mechanism	Efficiency (%)	Field Applicability	Key Limitation
Granular Activated Carbon	Adsorption	80–95%	High	Media saturation
Powdered Activated Carbon	Adsorption	70–90%	High	Sludge management
Biochar Amendments	Immobilization	30–80%	Moderate	Performance variability
Magnetic Nanocomposites	Adsorption + recovery	60–85%*	Experimental	Scalability
Advanced Oxidation	Chemical degradation	70–95%	Moderate	Energy demand

* Performance based primarily on laboratory or pilot-scale studies; limited field validation available.

Table 5: Frequently detected legacy pesticides in aquatic monitoring studies and selected persistence indicators.

Compound	Chemical Class	Typical Soil Half-life (DT_{50}, days)	log K_ow	Primary Detection Matrix	EU Regulatory Status
Atrazine	Triazine herbicide	60–100	2.6	Groundwater	Banned
MCPA	Phenoxy herbicide	15–50	2.8	Surface water	Restricted
2,4-D	Phenoxy herbicide	7–30	2.7	Surface and groundwater	Approved with limits
Chlorpyrifos	Organophosphate insecticide	30–120	4.7	Sediment	Withdrawn
Imidacloprid	Neonicotinoid insecticide	40–200	0.6	Surface water	Restricted

Sources synthesized from Gilliom et al. 2006, Silva et al. 2019, Szöcs et al. 2017, and Urseler et al. 2022. Values represent typical ranges reported across monitoring studies and may vary depending on environmental conditions.

of the reviewed remediation technologies is summarized in Table 4.

3.4. Integrated Catchment-Level Interpretation

The synthesis suggests that effective mitigation requires integration of soil risk screening, monitoring, and targeted intervention, as illustrated in Fig. 3.

The environmental benefit of remediation should be assessed using:

1. Reduction in exceedance frequency;
2. Decline in mean annual concentration;
3. Improvement in groundwater trend slopes.

Such metrics provide more meaningful indicators of recovery than pesticide sales statistics alone.

4. CONCLUSIONS

Legacy pesticides continue to influence aquatic environments long after regulatory withdrawal because of persistent soil storage, delayed desorption, and hydrological redistribution processes. This review demonstrates that environmental persistence is governed by interacting physicochemical properties and catchment-scale transport dynamics rather than degradation half-life alone. Monitoring evidence indicates that regulatory reductions in pesticide use do not automatically translate into immediate water quality recovery, particularly in groundwater systems with long residence times.

The synthesis highlights the importance of integrating soil risk screening, continuous monitoring, and targeted remediation strategies to address historical contamination reservoirs. Among available technologies, adsorption-based systems such as activated carbon remain the most technically mature and scalable options, while soil amendments and advanced oxidation processes offer complementary mitigation pathways under site-specific conditions.

The findings contribute to existing knowledge by bridging mechanistic understanding of pesticide persistence with practical catchment-scale management approaches. Future research should prioritize long-term field validation of remediation systems, development of predictive soil transport indices, and incorporation of adaptive policy feedback mechanisms. Sustainable water quality restoration will depend not only on reducing current pesticide inputs but also on actively managing legacy contamination sources.

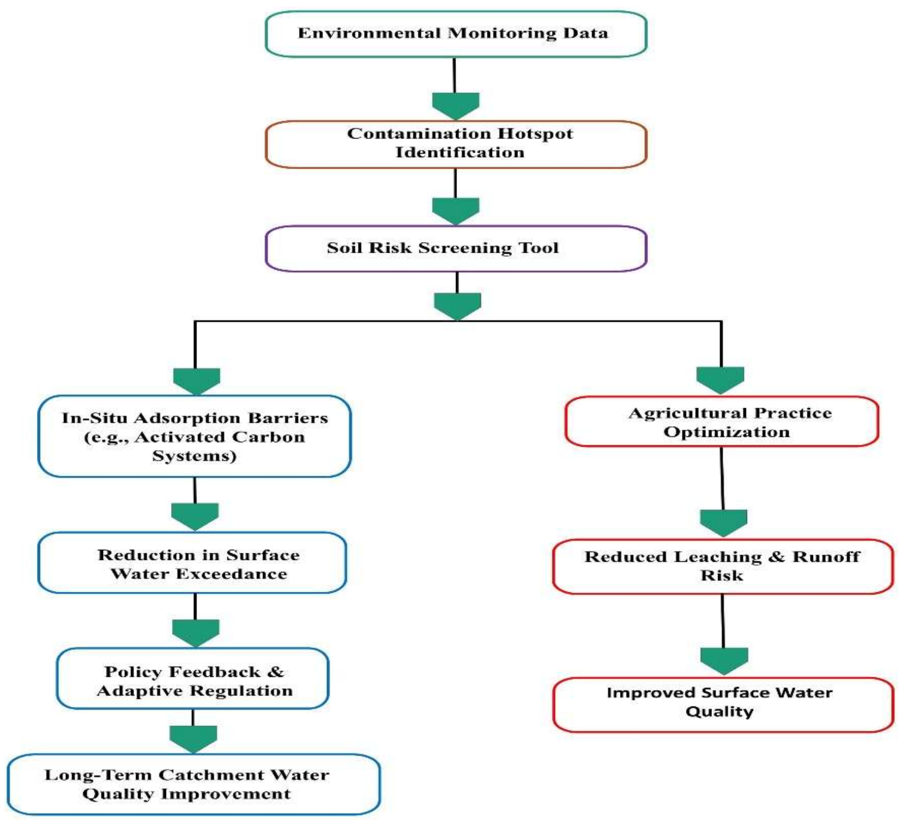

Fig. 3: Integrated catchment-scale mitigation framework combining monitoring, soil risk assessment, and adsorption-based remediation.

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