A Systematic Review of Photolysis and Hydrolysis Degradation Modes, Degradation Mechanisms, and Identification Methods of Pesticides

1. Introduction

Pesticide, as a chemical synthetic substance or a natural substance from the other bionts, used to prevent, destroy or control diseases, insects, grasses, and other harmful organisms that endanger agriculture, forestry plants and their products, and purposefully regulate the growth and development of plants and insects. As an important agricultural means of production, the primarily function of pesticides is to ensure crop yield, quality, and safety. They are divided into several major types depending on their control objects, such as insecticide, fungicide, acaricide, herbicide, nematicide, and plant growth regulator. The development and application of pesticides had played a very important role in promoting the harvest of agricultural food crops. However, excessive dependence on pesticides and irrational use of pesticides had a certain negative impact on agricultural production. With the mass production and widespread use of pesticides, the application of pesticides inevitably causes residues and pollution in the environment and has caused a widespread concern in countries around the world [1–4]. Pesticides must be residual when used, these residues will have incalculable implications for food, the environment, biomes, and even humans. The ecological effects of pesticide residues and the fate of these residues in the environment are urgently needed to be solved for both the scientific community and the general public. Therefore, it is necessary for us to explore the photolysis, hydrolysis mechanism, and identification method of pesticide degradation mode to provide a theoretical basis and technical support for multi-titudinous pesticide detection technology and residue solution in the future.

There are many reasons for causing and affecting pesticide residues. The properties of pesticides, environmental factors, and the application methods of pesticides are the main factors affecting pesticide residues. The pollution of pesticides to the water environment mainly come from (I) direct application to the water environment; (II) migration of pesticides applied to the farmland with rainwater or irrigation water; (III) discharge of wastewater from pesticide...
production and processing enterprises; (IV) during the use of pesticides, the droplets, or dust particles migrating with the wind and settle into the water body and the cleaning of the application tools and instruments. For example, some pesticides were found in the water environment with extensive agricultural activity [5, 6]. Furthermore, many pesticides were found in urban streams [7], lakes [8–11], underground water [12], and rivers [13–16]. After the pesticides were applied, some of them adhered to the plant body, or part infiltrated into the body of the plant, and the other part was scattered on the soil or evaporates, dissipates into the air, or flows into the lake with rainwater and farmland drainage, thereby polluting the water body. Pesticide residues mainly enter the human body through the atmosphere, water, soil and food, and cause various chronic or acute diseases.

Both photolysis and hydrolysis are important ways in the degradation of pesticides in the environment. Hydrolysis is a hydrolysis reaction of pesticides because there are chemical structures in the pesticide molecules that can be hydrolyzed, such as ester bonds, ether bonds, amide bonds, cyano group, and acyl chloride group. Photolysis of pesticides is a process in which a pesticide molecule gets light radiation energy, and light energy is converted into a molecular bond of the compound to break the bond and generate an internal reaction. Pesticide molecules must absorb a certain wavelength of light energy in an excited state in order to carry out a photochemical reaction. Due to the structural properties of the pesticide itself, most pesticides are very sensitive to photolysis. In the past few decades, a large number of studies have been conducted on the photolysis of pesticides and the effects of organic and inorganic constituents in natural world on the degradation of pesticides in water [17–33].

In this paper, the literature on pesticide degradation in recent years is reviewed, the degradation methods of photolysis and hydrolysis are introduced, the degradation mechanism of pesticides is discussed, and the identification methods of degradation products are summarized. Finally, this review provides some useful data and recommendations for future research that will be urgently needed to inform pesticide users, developers, and governmental regulators as well as will have a more thorough reference point from which the future of these widely applied pesticides can be determined.

2. Types and Characteristics of Pesticides

Pesticides are divided into several major types depending on their control objects, such as insecticides, fungicides, acaricides, herbicides, nematicides, and plant growth regulators. Insecticides were agents that control the chemical or biological sources of insects. Insecticides could control insects which may be due to killing the insect or otherwise preventing it from engaging in a considered destructive behavior. Historically, humans have had long experience using pesticides [34, 35]. Compared with insecticides, fungicides have a short development history. It was not until 1807 that the first practical chemical used for disease control, the first fungicide, was discovered [36, 37]. In recent years, the development of Strobilurin fungicides was remarkable. The mechanism of action was to block electron transmission, inhibit mitochondrial respiration, and inhibit fungal growth [38]. Ammoniacides have been developed due to the economic losses caused by the rampant reproduction of herbivorous mites affecting fruits, cotton, and vegetables. Today, to achieve the goal of drug resistance, most of the new acaricides can be classified as mitochondrial respiratory inhibitors, growth inhibitors, and neurotoxins [39, 40]. Herbicides act on large numbers of metabolic functions and energy transfer sites in plant cells [41]. The first commercial synthetic herbicide was created in the 1940s [42]. Herbicides are divided into three categories: the first, biochemical pathways and physiological processes related to photosynthesis; the second, inhibit bioconstruction or assemble into biopolymers; and the third, other modes of action [43]. Plant parasitic nematodes cause huge economic losses to agriculture around the world annually [44]. Compounds controlling nematodes began synthesis only in the 19th century [45, 46]. Plant hormones play regulatory roles in growth and development, while synthetic chemicals with similar physiological activities, or compounds capable of otherwise modifying plant growth, are called plant growth regulators [47]. Plant growth regulators play a mainly active regulatory role in plant development and affect the balance of plant body hormones generally [48].

3. Pesticide Degradation Mode

The degradation mode of pesticides in the environment is divided into biological degradation and nonbiological degradation. Nonbiological degradation is also divided into hydrolysis and photolysis.

3.1. Hydrolysis. Hydrolysis reactions are important processes when many pesticides are degraded. Because of adsorption catalytic reactions, pesticide hydrolysis is faster in soil than in soil-free systems. This is more significant for the degradation of chlorotriazine herbicides and organophosphate insecticides [49]. We take organophosphorus insecticides as an example. Its hydrolysis reaction can occur by homogenization machine production. Water and hydroxide ions participate as nucleophiles in bimolecular nucleophilic replacement reactions. Iron and aluminum oxides as well as different clays can increase the hydrolysis rates by providing the surface positions of the nucleophiles and the hydrolysis reactions. Despite much speculation about the hydrolysis mechanisms, there is still uncertainty [50, 51].

3.2. Photolysis. The light-induced chemical reactions of pesticides on the surface of the atmosphere, water bodies, or objects (e.g., plants and soil) are an important nonbiodegradation pathway of pesticides and has a significant impact on pesticide residues, efficacy, toxicity, and the environment. Photocatalytic degradation is a relatively cheap and effective degradation method and has good potential [52–55]. Photodegradation requires absorption of light energy and only pesticides that absorbed light above 285 nm.
could be decomposed by natural sunlight. Therefore, photocatalytic degradation experiments are usually carried out under high intensity light. Pesticides absorbed light radiation and produced hydroxyl, superoxide, and ozone radicals, which ultimately lead to degradation products. Photocatalytic degradation reactions might be isomerization, substitution, or oxidation. The reaction type is affected by the physical properties of pesticides, environmental factors, reagents, and so on [56, 57]. Photocatalytic degradation reactions generally required photocatalysts. The ideal material for photocatalysts should have high photoactivity, photocorrosion resistance, chemical inertia, low cost, and low environmental toxicity in the near ultraviolet and visible regions of the electromagnetic spectrum [58]. Titanium dioxide and zinc oxide were the main catalysts in photocatalytic reaction experiments [59–62]. However, recent research using semiconductors as catalysts has emerged [63].

3.3. Biodegradation. Biodegradation has the advantages of efficiency, economy, flexibility, wide range of degradation objects, stable degradation ability, and no secondary pollution to the environment [64]. The main reactions involved in microbial degradation include hydrolysis, oxidation, alkylation, and dealkylation [65]. Biodegradation might be thought of as the transformation of a complex substance into one or more simpler substances by biological machinery, through the production of enzymes that broke a chemical bond, and the degradation of large compounds into small ones, rendering them inactive. In addition to degrading enzymes, biodegradation could also be influenced by environmental factors, including the soil type, water content, temperature, and pH [66–68]. The role of oxygen in pesticide biodegradation was complex; for example, as an oxygenase substrate involved in biodegradation reactions. If oxygen was sufficient, the end products of degradation will be carbon dioxide, water, sulfate, nitrate, phosphate, chloride ions, etc. If oxygen was insufficient, anoxic conditions can stimulate the activity of potential anaerobic microorganisms, which may directly or indirectly affect the transformation of pesticides [69].

3.4. Factors Affecting the Degradation. The factors affecting pesticide degradation in the water environment and soil were generally related to the nature of soil and water itself, such as water soil pH, temperature, soil water content, soil organic matter content, and different soil types (the actual essence was related to the organic matter content) [70, 71]. Some influencing factors were related to the nature of the pesticide itself; for example, the effectiveness of pesticides on soil pests was also the main factor affecting the pesticide degradation in the soil. There were other environmental factors, such as other soil phenomena (adsorption) and environmental exposure of nontarget organisms [72].

4. Pesticide Degradation Mechanism

4.1. Insecticide. In the study of nonbiodegradation of insecticides in nature (soil or water), the experimenter will explore the consistency of hydrolyzed photolysis products through comparative experiments and will compare the effects of natural light and man-made radiation sources on photolysis. The main photolysis mechanism of insecticide was ester group breaking (Figure 1(a)) [73, 74]. However, the photolysis pathway of pesticides was not only the breaking of the ester group. There were many kinds of pesticides with different components. Some pesticides have only one degradation pathway, some have two, and some even have a variety of degradation pathways. For example, the photolysis of the organophosphorus insecticides-ethyl parathion, methyl parathion, and phenylisopropylphosphorothioate in water and soil was found to include oxidation, hydrolysis, isomerization, and reduction (Figures 1(b)–1(d)) [75]. Pesticide hydrolysis was also the same. In the experiment of hydrolysis of chlorpyrifos (O,O-diethyl-O-(3,5,6-trichloro-2-pyridyl) phosphorothioate), nucleophilic attack was done by water of ethoxy carbon degradation products (Figures 1(e) and 1(f)) [76]. The effect of environmental factors on the photolysis and hydrolysis of pesticides was also a subject worthy of study. It was found that three insecticides were stable under acidic conditions. Dealkylation occurred in a neutral medium. In the alkaline medium, the ether bond broke to form phenol derivatives and dialkyl phosphoric acid (Figures 1(g)–1(j)) [77].

4.2. Fungicide. Like insecticides, the degradation pathways of fungicides were diverse. When the main degradation reaction occurs, it will be accompanied by parallel reactions. For example, the main photoproducts of azoxystrobin fungicides were produced by ether bond breaking. The secondary photolysis products might be due to demethylation. Photoisomerization and cleavage of acrylate double bonds were parallel pathways of degradation (Figures 2(a)–2(e)) [78]. For the degradation of fungicides, the researchers also considered the influence of environmental factors on the reaction. For example, the common humus in soil, acid-base environment, and the effect of micellar medium on degradation. The research showed that humus as the oxidant extracts electrons or hydrogen atoms from bactericides to form oxidation products. In the acidic environment, the disulfide bond and carbon-nitrogen bond in bactericide were destroyed, and there will be byproducts in the absence of metal tin ions. In nonionic micelles, the loss of the ethyl ester group and the opening of a dihydrocarbon ring will be inhibited. In the alkaline hydrolysis of benzoic acid and benzyl fungicides, the degradation mechanism was inferred to be a carboxyl fracture (Figures 2(f)–2(k)) [79–82].

4.3. Acaricide. For acaricides with a relatively complex structure, its degradation pathway also has many routes. For example, the photolysis pathway of abamectin, an acaricide composed of two colorless homologs with the same macrolide structure included oxygenation, demethylation, and isomerization (Figures 3(a) and 3(b)) [83]. In order to understand the degradation of acaricides under different conditions, the experimenters will design the degradation of acaricide under different light conditions in different solutions to master the possible different degradation
mechanisms. For example, in the study of photoysis of non-naphthoquine, it was found that the degradation mechanism was the cleavage of ether bond between quinazoline and the phenyl ring system, the oxidation of a tert-butyl substituent and the oxidation of the heterocyclic part of a quinazoline ring (Figures 3(c)–3(e)) [84]. Even if the possible degradation mechanism was mastered, the renewal of acaricides was changing with each passing day. This means that it is very important to study the degradation of the new mechanism. In the study of a new benzoyl acetamide acaricide cyflumetofen (CYF), it was found that the electrophilic group carbon atoms on the CYF were found to be easily damaged by a nucleophilic attack, producing a hydrolysate 1. When the carbon-carbon single bond breaks, it binds with an amino group to produce hydrolysate 2 (Figures 3(f) and 3(g)) [85].

4.4. Herbicide. In the study of herbicide degradation, photoysis and hydrolysis were also studied by changing the light. It was found that Sulfuron could cause the sulfonyle group to fall off under neutral and alkaline conditions. Photooxidation and photoisomerization were the main reactions of triketone herbicides (Figure 4(a)) [86, 87]. Of course, herbicides were mostly organic, and their degradation reactions were complex and changeable, and there were many degradation products. There were nine photolysis products of ketene herbicides. It was conceivable that there were many photolysis pathways, including isomerization and cracking of the oxime ether bond, cracking of dechlorinated isopropyl group, sulfoxide, and oxidative cracking of the dechlorinated epoxy group (Figures 4(b) and 4(c)) [88]. Water molecules play an indispensable role in the hydrolysis of herbicides. They often attack compounds as nucleophiles, as do other types of pesticides. For example, the hydrolysis of sulfonyle herbicides. The hydrolysis mechanism was similar to the nucleophilic substitution reaction in which water molecules attack carbon groups from the aryl or heterocyclic side (Figure 4(d)) [89].

4.5. Nematicide. The most of degradation products detected in the degradation of nematicides were sulfoxides and sulfones because sulfur ions or methyl groups would undergo oxidative desulfurization and sulfur oxidation. The degradation of amine (4-methylisopropyl phosphoramid) nematicide and thiazole phosphorus were taken as an example (Figure 5) [90, 91]. It has also been shown above that titanium dioxide often participates in photolysis as a photocatalyst. The photolysis of fenamiphos nematicide has two steps under the action of catalyst, the first was oxidation and the second was mineralization [92].

4.6. Plant Growth Regulator. Photoisomerization often occurs under photolysis and plant growth regulators are no exception. The photolysis mechanism of gibberellin A3 derivatives was photoinduced aromatization of a ring (Figures 6(a) and 6(b)) [93]. Generally, the degradation of plant growth regulators is also needed to be considered as the influence of environmental factors. In the study of the degradation of methyl phosphonate (MPN), the effects of
different environmental factors on its degradation were compared. It was found that the degradation mechanism was that electron transfer produces free radicals, reacts with oxygen to form peroxy radicals, and finally decomposes into hydroxyl radicals to attack methyl in MPN (Figure 6(c)) [94]. The degradation of plant growth regulators also involved many ways. For example, the photolysis pathway of malehydrazine included carbon-carbon double bond

Figure 2: Degradation pathways of fungicides.
Figure 3: Degradation pathways of acaricides.

Figure 4: Main photolysis mechanism of herbicides.

Figure 5: Degradation mechanism of nematicides.
transfer site, ketone group, and amino group cleavage (Figure 6(d)) [95].

5. Identification Methods

In recent years, the methods and techniques for quantitative and qualitative analysis of pesticides were constantly updated. The critical methods and techniques were very useful for the residue analysis of pesticides and the identification of degradation products, degradation mechanism, and reaction pathway of the pesticides. Many technologies, such as gas chromatography-mass spectrometry (GC-MS/MS) [96–98], liquid chromatography-mass spectrometry (HPLC-MS/MS) [99–102], ultraperformance liquid chromatography-quadrupole-time-of-flight mass spectrometry (UPLC-Q-TOF-MS) [103–109], ultraperformance liquid chromatography-orbitrap mass spectrometry (UPLC-Orbitrap MS/MS) [110–114], and high-resolution mass spectrometry (UPLC-HRMS) [115–117] were used to separate the degradation products and identify the structure of products. So far, the high-resolution mass spectrometry combined with UPLC is increasingly used in the qualitative screening and degradation mechanism of pesticide metabolites, for its high-resolution ensures high sensitivity, accuracy, and high specificity required for complex sample analysis. Through these techniques, the degradation mechanisms and reaction pathways of many pesticides in the environment were identified. Pesticide may undergo physical and chemical processes in the soil. Therefore, a combination of physical and chemical unit processes was required and actually employed to ensure the removal of pesticide residues and byproducts from the environment. The mode and mechanism of chemical degradation were closely related to the molecular structure of pesticides. Generally, pesticides with functional groups such as halooalkyl, amide, amine, carbamate, epoxy, cyano, phosphate, and sulfate were easily hydrolyzed.

5.1. High-Performance Liquid Chromatography.

High-performance liquid chromatography (HPLC) used a liquid as the mobile phase and high pressure infusion system. Single solvent or mixed solvent with different polarities and buffer were pumped into the chromatographic column. After being separated in the column, the sample was detected by the detector. High-performance liquid chromatography was used to study the degradation rate of fenpyroximate in apples, oranges, and grapes. The classical QuEChERS method was used for pretreatment. The C18 column was used for separation and HPLC-PDA was used for detection [118].

5.2. Liquid Chromatography-Mass Spectrometry.

Liquid chromatography-mass spectrometry (LC-MS). Pesticide residues were mainly detected by liquid chromatography tandem mass spectrometry, high performance liquid chromatography tandem mass spectrometry, and ultrahigh liquid chromatography tandem mass spectrometry.

Of course, in the experiment, the experimental method was not single and unchanging. There will be different methods in sample extraction, such as ultrasonic extraction,
solid phase extraction, and so on. For example, ultrasonic extraction and HPLC-APCI-MS were used to determine antifouling pesticides and their degradation products in the sea. Solid phase extraction combined with liquid chromatography-mass spectrometry was used to determine benzophenone, carbazine, and their degradation products in water samples [119, 120]. However, overly complex experimental methods will consume a lot of human, material, and financial resources. Liquid chromatography tandem mass spectrometry was used to determine pesticides and their degradation products [121, 122]. With the development of science and technology, the identification and determination experiments of pesticides were constantly innovated. In the experiment of studying the photo-hydrolysis and degradation products of neonicotine insecticides, the absorbance of each neonicotine was determined by spectrophotometer, and the reaction products were determined by UPLC-MS/MS. There are also technologies such as ultrahigh liquid chromatography time of flight mass spectrometry (UPLC-Q-TOF-MS) and high-resolution mass spectrometry (UPLC-HRMS) to separate degradation products and identify the structure of products [123–125].

5.3. Gas Chromatography and Gas Chromatography-Mass Spectrometry. Gas chromatography was a chromatographic separation and analysis method using a gas as mobile phase. The vaporized sample was carried into the column by the carrier gas. The molecular forces of each component were different and the outflow time was different, so that the components were separated from each other. In pesticide degradation experiments, solid phase microextraction was often used to treat samples, regardless of the final detection method. This was the case in the experiment of studying the degradation residues of amitraz, which was treated by solid-phase microextraction and detected by gas chromatography ion trap detector (GC-ITD) [126].

In terms of gas chromatography-mass spectrometry, mass spectrometry has a unique ability to identify unknown compounds with extremely high sensitivity, making GC-MS one of the most powerful tools for separating and detecting complex compounds. Sometimes, the toxicity of degradation products will also be determined in the experiment because it is impossible to distinguish whether the degradation products of pesticides are still toxic. For example, the disappearance of methyl organophosphorus, toxic phosphorus, and malathion in aqueous solution and the formation of photodegradation products were detected by gas chromatography-mass spectrometry (GC-MS). The toxicity was determined by FIA-ache-TLS bioassay [127].

5.4. Other Methods. Isotope labeling was a method to understand the detailed process of chemical reactions by tracing compounds labeled by tracer elements. Carbon elements were used as tracer elements in pesticide degradation product recognition. Pesticide degradation research methods using carbon 13 and carbon 14 isotope markers have emerged and have gradually been widely used. This technique could be readily seen in studies of the degradation pathways and the identification of the degradation products of multiple pesticides [128, 129].

6. Concluding Remarks

Mentioned in this article are the different types of pesticide and its effects on human health and biological environment. Not only that, but also for pesticide degradation model, different kinds of pesticide degradation mechanisms and methods to identify degraded products are mentioned.

In today's world, the widespread use of pesticides and pesticide residues did become a hot topic. For the health of human life and better biological environment, degradation of pesticide in nature should be familiar with the cognitive method and understand the photolysis hydrolysis and influencing factors (temperature, pH, light, etc.). Through the degradation mechanism and influencing factors, the degradation time could be accelerated, and the theoretical basis and basic support for pesticide residues could be provided in the future. In this paper, the biodegradation of pesticides was not described too much, mainly focusing on the photolysis and hydrolysis of pesticides. However, in recent years, there were more and more research studies on the biodegradation of pesticides. In the future development, the biodegradation of pesticides is also crucial to the development of pesticide degradation residues.

The identification method of pesticide degradation products was also mentioned in this paper. Through consulting data, the author found that high-performance liquid chromatography-mass spectrometry was mostly used in research experiments, mainly because of its advantages of high efficiency, high sensitivity, and wide application. With the development of technology, identification methods would be gradually updated, including UPLC-Q-TOF-MS and ultraperformance liquid chromatography-orbitrap mass spectrometry (UPLC-Orbitrap MS/MS).

Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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