## **REVIEW**

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# Efficiency of combined techniques in pesticide residues removal from food: a systematic review



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## Abstract

The global use of pesticides has steadily increased in recent years to enhance food production and protection and prolonge its shelf life. Nevertheless, pesticide residues and their association with various human diseases have raised concerns among the population, as the primary route of human exposure to pesticides is through food consumption. Consequently, removing pesticide residues from agricultural products and food is crucial for mitigating associated risks. Various treatments are employed to eliminate and degrade pesticide residues, with commonly used conventional techniques such as washing, peeling, and cooking being used. Additionally, emerging techniques such as ultrasound, ozone, electric current, plasma, and ultraviolet light have been applied to remove these residues. In this study, we systematically reviewed 38 articles to assess the efficacy of combined techniques for pesticide residue removal in food. The findings revealed that using combined techniques resulted in significantly higher levels of residue removal. Furthermore, combining emerging techniques with other treatments has demonstrated increased removal efficiency, significantly reducing the variability in the percentage range of residue removal. The synergistic use of ultrasound, ozone, and ultraviolet light techniques demonstrated a notably enhanced efficacy in removing pesticides, resulting in a higher elimination percentage. Among the 38 studies, 12 exhibited substantially lower variability. Moreover, ultrasound emerged as the technique with the most significant synergistic effect when combined with other techniques, enhancing the overall efficiency of pesticide residue removal. Among the 12 studies with lower variability, 9 incorporated ultrasound, 4 ozone, and 3 ultraviolet light as part of the combined treatment. However, it is essential to note that conventional techniques also achieved considerable residue removal, even with more significant variability. This information can serve as valuable guidance for managers, decision-makers, and the public in effectively selecting appropriate techniques to eliminate pesticide residues from food before consumption or sale.

Keywords Pesticide degradation, Pesticide removal, Combined techniques, Multiple techniques

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#### Introduction

The potential health risks associated with pesticide residues in food have raised significant global concern (UNEP, 2022). Despite the establishment of Maximum Residue Levels (MRLs) for pesticides by the Codex Alimentarius, exceeding these established limits has been identified in various food and agricultural products in several countries (Boudebbouz et al., 2022; de Andrade et al., 2023; Elgueta et al., 2017; Kuang et al., 2023; Liu et al., 2019; Mert et al., 2022). Furthermore, multiple studies have demonstrated that exposure to pesticides can have adverse effects on human health, including carcinogenic (Melanda et al., 2022; VoPham et al., 2017; Wallace & Buha Djordjevic, 2020), hepatotoxic (Alarcan et al., 2020; Chang et al., 2017; Lozano-Paniagua et al., 2021), teratogenic (Sharma et al., 2020), pulmonary (Benka-Coker et al., 2020; Ratanachina et al., 2020; Zhang et al., 2019), and reproductive (Cremonese et al., 2017; El-Nahhal, 2020; Landeros et al., 2022), among others (Kalyabina et al., 2021; Mostafalou & Abdollahi, 2017; Salazar-Flores et al., 2022). Therefore, exposure to pesticide residues through food represents a significant threat to human health, highlighting the importance of developing methods for removing these residues and, in turn, minimizing the associated risks.

Agricultural products are commonly subjected to preconsumption treatments by different industries to lower harmful residues, including conventional methods such as washing, peeling, cooking, sterilization, and chemical treatments involving detergents, bleach, and electrolyzed water (Rawn et al., 2008; Shiroodi & Ovissipour, 2018; Wu et al., 2019). Nevertheless, these methods have demonstrated limited effectiveness in removing pesticide residues from food (Khan & Rahman, 2017). Furthermore, some of these treatments may have adverse effects on the sensory qualities of the products, such as color, texture, and taste, particularly in cases of chemical or thermal treatments (Kontou et al., 2004; Łozowicka & Jankowska, 2016). Additionally, specific

treatments may result in partial product loss, as in the peeling procedure (Chung, 2018; Yang et al., 2017). In contrast, emerging methods such as cold plasma, irradiation, highpressure processing, ozone, electric current, and ultrasound have shown promising results in removing pesticide residues without compromising the quality of the product (Mir et al., 2022). Combining conventional and modern treatments has demonstrated higher pesticide residue removal than when applied individually. For instance, the effectiveness of captan residue removal in apples improved when washing and peeling treatments were used in combination, compared to washing alone (Rawn et al., 2008). Furthermore, a decrease in pesticide residues in different fruits was observed when a combination of electrolyzed water, chlorine dioxide, and photocatalysis was applied (Calvo et al., 2019). Similarly, the combined use of ultrasound, washing, ozone, and electric current has demonstrated higher efficacy in removing pesticide residues in tomatoes and cherry juice (Akdemir Evrendilek et al., 2020; Cengiz et al., 2018).

However, an in-depth analysis of combining techniques to remove pesticide residues in food effectively has received limited attention (Akdemir Evrendilek et al., 2020; Calvo et al., 2019; Cengiz et al., 2021; Vasseghian et al., 2020). Therefore, this systematic review aimed at identifying the most effective combined techniques for removing pesticide residues (PRs) in fruits, vegetables, and processed food.

## Methodology

## Systematic search methods

The Preferred Report Items for Systematic Review and Meta-Analysis (PRISMA) guidelines were used to document the results of the article selection process (Shamseer et al., 2015) to obtain the information required. Thus, the subsequent steps were followed: 1) identification of research questions, 2) identification of relevant studies, 3) selection of studies, 4) selection and data extraction, and 5) reporting the results.

## **Research question**

The PICO (Population, Intervention, Comparator, Outcome) strategy was used to formulate the following research question: What combined treatments or techniques are most effective for removing pesticide residues from food? Where P = food, I = application of combined treatments or techniques, <math>C = application of single treatments, and O = effective remotion of PRs.

#### Data sources and search strategy

The information search was conducted in April 2023 in two databases: Web of Science (https://www.webofscien ce.com) and Scopus (https://www.scopus.com). These databases were selected due to their extensive coverage of scientific research studies relevant to the objective. The initial selection of scientific articles was performed by analyzing the titles, abstracts, and keywords. Articles that did not address the removal or degradation of pesticides in food were excluded from the analysis.

Articles published in the past 15 years, specifically from January 2008 to April 2023, were selected because there has been an increased number of publications focusing on removing pesticide residues on different matrices since 2008. This choice ensures that our analysis encompasses the most relevant and up-to-date research in the field. The search terms used included:

## Web of science

("Pesticide Residues" OR pesticide OR herbicide OR insecticide OR agrochemical) AND (remov\* OR degrad\* OR elimin\* OR degener\* OR reduc\* OR dissipa\*) AND (combin\* OR multiple OR "multi-objective" OR "multiobjective") AND (techni\* OR proce\* OR treatment\* OR method\*) AND (food\* OR "agricultur\* product\*" OR product OR fruit OR vegetable).

#### Scopus

("Pesticide Residues" OR pesticide OR herbicide OR insecticide OR agrochemical) AND (remov\* OR degrad\* OR elimin\* OR degener\* OR reduc\* OR dissipa\*) AND (combin\* OR multiple OR "multi-objective" OR "multiobjective") AND (techni\* OR proce\* OR treatment\* OR method\*) AND (food\* OR "agricultur\* product\*" OR product OR fruit OR vegetable) AND NOT ("wastewater" OR "geograph\*" OR residential OR "behavio\*" OR "soil remediation" OR "water remediation" OR wood OR seawater OR freshwater OR gis OR network OR "surface water" OR "groundwater" OR "metal" OR "synthesis" OR "biological" OR "urine" OR "blood" OR "injury" OR "mechanism\*" OR "algorithm\*" OR "genetic" OR "air" OR "water management" OR "soil pollution" OR "air pollution" OR "sediment" OR "circular economy" OR effluent OR "drinking water" OR remediation OR "medicinal plants" OR "forest management" OR "river" OR "basin" OR "medicine" OR "ecosystem\* service\*" OR suicide OR wetland OR gender OR "global warming" OR biodiversity OR microplastic\* OR "development goals" OR allergy OR "forest management" OR communit\* OR "machine learning" OR "heavy metal\*" OR plastic OR polymer OR "economic benefit\*") AND ( LIMIT-TO ( PUBYEAR,2023) OR LIMIT-TO ( PUBYEAR, 2022) OR LIMIT-TO ( PUBYEAR,2021) OR LIMIT-TO ( PUBYEAR,2020) OR LIMIT-TO ( PUBYEAR, 2019) OR LIMIT-TO ( PUBYEAR,2018) OR LIMIT-TO (PUBYEAR,2017) OR LIMIT-TO ( PUBYEAR, 2016) OR LIMIT-TO ( PUBYEAR,2015) OR LIMIT-TO (PUBYEAR,2014) OR LIMIT-TO ( PUBYEAR, 2013) OR LIMIT-TO ( PUBYEAR,2012) OR LIMIT-TO (PUBYEAR,2011) OR LIMIT-TO (PUBYEAR,2010) OR LIMIT-TO (PUB-YEAR,2009) OR LIMIT-TO (PUBYEAR,2008) ) AND (LIMIT-TO (PUBSTAGE,"final")) AND (LIMIT-TO (LANGUAGE,"English")) AND (LIMIT-TO ( SRCTYPE,"j"))

#### Inclusion and exclusion criteria

The essential information from articles discovered in databases, including author names, abstracts, titles, publication years, journals, and DOIs, was exported to Excel for organization. Then, all the citations were imported into a web-based systematic review software, Covidence (https://www.covidence.org/). The software eliminated duplicate references and facilitated the screening, selection, and data extraction process. That process was done based on pre-specified criteria. Subsequently, articles were selected based on the following inclusion and exclusion criteria:

- a) Only original articles written in English and published in peer-reviewed journals between January 2008 and April 2023, containing relevant information on techniques for removing PRs from food, were included.
- b) Articles that evaluated the efficacy of PRs removal by applying combined techniques were included.
- c) Books, conference papers, abstracts, letters, presentations, reviews, meta-analyses, posters, and theses were excluded.
- d) Articles irrelevant to the study objective, duplicate publications, and those lacking sufficient data for analysis were excluded.

The flow chart of the study selection process is shown in Fig. 1.

## Selection and data extraction

The selection and extraction of data from articles was carried out using a structured Excel spreadsheet. The extracted information was categorized into the following domains: a) Author and publication year; b) Food material; c) type of pesticide; d) applied method; e) processing conditions; and f) removal yield of PRs. Table 1S summarizes these data.

## **Results and discussion**

#### Descriptive analysis of the studies

The search strategy yielded a total of 6020 articles, with 3129 studies identified in Scopus and 2891 in Web of Science. Subsequently, 1802 articles were removed automatically due to duplicates, resulting in 4218 articles for further screening. Following a comprehensive assessment

of titles and abstracts, 183 studies were deemed suitable for full-text analysis. After carefully examining these articles, it was determined that 145 did not meet the predefined inclusion criteria. Consequently, a total of 38 articles were selected for further data extraction, as depicted in Fig. 1.

This study identified the application of 46 combined techniques and the presence of 60 pesticide residue types (Tables 1 and 2, respectively). The pesticides that were analyzed in more than three studies were: Acetamiprid (5,04%),  $\lambda$ -cyhalothrin (5,04%), Boscalid (4,32%), Chlorpyrifos (4,32%), Pyraclostrobin (4,32%), Cyprodinil (2,88%), Imidacloprid (2,88%), Iprodione (2,88%), Thiamethoxam (2,88%), Azoxystrobin (2,16%), Bifenthrin (2,16%), Captan (2,16%), Cypermethrin (2,16%) and Procymidone (2,16%).

## Techniques definition for pesticide degradation *Main conventional techniques*

Domestic and commercial treatments encompass a range of methods for food processing. These methods include washing, peeling, canning, and thermal processing such as cooking, boiling, pasteurization, blanching, and drying.

#### Washing

This procedure has as a primary objective the removal of dust, contaminants, and microorganisms from an object, employing a combination of mechanical actions, solvents, and the addition of chemical substances such as detergents and disinfectants (Andrade et al., 2015).

#### Peeling

The process involves the removal of the outer layer or skin of a fruit or vegetable. It can be achieved through manual peeling, blanching, scoring, or steaming (Rawn et al., 2008). These methods facilitate the separation of the outer layer, which could contain dirt, contaminants, or pesticide residues, from the product.

#### Canning

It is a preservation method that involves placing food in a container followed by a heat treatment to eliminate contaminants and inactivate microorganisms. The food product is typically prepared by washing, peeling, and sometimes cooking before canning (Jankowska & Łozowicka, 2022).

#### Thermal processes

Cooking, boiling, pasteurization, and blanching are part of these processes. Each method involves the application of heat to food products, resulting in the enhancement of their organoleptic properties, reduction in contaminant



Fig. 1 PRISMA Flowchart of the literature search and strategy for selecting the relevant studies

levels, and inactivation of microorganisms. The heating process induces structural and molecular alterations in different chemical substances, facilitating their removal and reduction. Elevated temperatures effectively eliminate and inactivate microorganisms, ensuring the product's safety and quality. The boiling method is based on applying heat to a liquid to raise its temperature to the boiling point. Typically, this method changes the physical

Utilized techniques						
Air (air)	Different aqueous solutions (Aq. Sol)	Microbubble-water (MCB)	Rehydration (ReH)			
Air Drying (AirD)	Different chemical solutions (Ch. Sol)	Microwave Owen (Mw.Ck.)	Salt Dipping (S.Dipp.)			
Alkaline electrolyzed water (AIEW)	Dioxido de titanio (TiO <sub>2</sub> )	Ozone (Oz)	Saponin camelia seed cake (CSE)			
Aqueous Ozone (AqOz)	Dipping (Dipp.)	Ozone microbubbles (OzMB)	Saponin soybean extract (SBE)			
Baking Soda (Bak. Soda)	Dry (Drying)	Peeling (PEL)	Steaming (Stm)			
Blanching (Bl)	Dry pasteurization (Dry.Past.)	Photocatalysis (PhotoC)	Stir-frying (SFry)			
Boiling (Boi)	Electrical current (EC)	Photosensitizers (Photo.Sent.)	Sunlight (S.light)			
Canning (Cann.)	Electrolyzed Water (EW)	Plasma-activated buffer solution (PABS)	Ultrasound (US)			
Chemical Boiling (Ch.Boi.)	Frying (Fry)	Plasma-activated liquid (PAL)	Ultraviolet (UV)			
Chlorine dioxide (ClO <sub>2</sub> )	Hydrogen peroxide (Hidro.Perox.)	Plasma-activated water (PAW)	Washing (WS)			
Cooking (Ck)	In-pack sterilization (IP.ST.)	Preliminary treatment (Pre.Treat.)				
Detergente (DTR)	Lactic Acid (Lact.Ac.)	Pulsed electric field (PEF)				

Table 1 Identified techniques for pesticide residue removal and their abbreviations

#### Table 2 Pesticide residues identified

Pesticides identified in all the studies						
Acetamiprid	Cyfluthrin	Ethion	Indoxacarb	Propamocarb		
Azoxystrobin	Cyhalothrin	Fenarimol	Iprodione	Pyraclostrobin		
Bifenthrin	Cypermethrin	Fenbuconazole	Lufenuron	Pyridaben		
Boscalid	Cyprodinil	Fenpropathrin	Malathion	Quinalphos		
Bupirimate	Deltamethrin	Fenvalerate	Mancozeb	Spinetoram		
Captan	Dichlofluanid	Fipronil	Metalaxyl	Tebuconazole		
Carbaryl	Dichlorvos	Fludioxonil	Methamidophos	Thiabendazole		
Carbendazim	Difenoconazole	Flusilazole	Methomyl	Thiacloprid		
Carbosulfan	Diflubenzuron	Fluvalinate	Myclobutanil	Thiamethoxam		
Chlorfenapyr	Dimethoate	Folpet	Oxamyl	Thiodicarb		
Chlorothalonil	Dimethomorph	Glyphosate	Phoxim	Triadimefon		
Chlorpyrifos	Esfenvalerate	Imidacloprid	Procymidone	Trichlorfon		

and sensorial properties of the food, enhancing its quality. The pasteurization process requires the application of specific heat treatment conditions to remove contaminants and microorganisms without damaging the quality of the product. The blanching involves a brief product exposure to boiling water or steam to achieve partial cooking. This process also aims to remove contaminants and reduce the microbial load (Xiao et al., 2017).

### Drying

This process involves the removal of moisture from food products through evaporation, decreasing the water content. This reduction inhibits microbial growth, enzymatic activity, and chemical reactions, enhancing the product's preservation and removing contaminants (Lee & Jung, 2009).

#### **Emerging techniques**

Advanced techniques are being employed for the removal of pesticide residues. These methods include cold plasma, electrical current, pulse electric field, photocatalysis, electrolyzed water, ultrasonication, and ozone.

#### Cold plasma and plasma-activated water

Cold plasma or non-thermal plasma is a form of matter that has been electrically energized and is composed of gas molecules, positively and negatively charged particles, free radicals, electrons, and photons (Misra et al., 2014). These highly reactive species are produced at nearly room temperature (30 - 60 °C). When a product is exposed to a cold plasma source, it is irradiated with ionizing irradiation and reactive species, which catalyze oxidation reactions to degrade pesticides (Mir et al., 2022). Additionally, when non-thermal plasma based on air gaseous source is irradiated into the water, it forms a solution rich in reactive oxygen and nitrogen species, generating plasma-activated water (PAW). It has been reported that PAW exhibits similar disinfection and sterilization properties as non-thermal plasma due to free radicals with high half-life, acidification, and higher redox potential, which enhance its oxidative capacity. Furthermore, PAW allows the reactive species to flow in the medium freely, increasing their reach and contact with food residues containing pesticides, thus promoting their degradation (Zheng et al., 2019).

#### Electrical current and pulsed electric field

These techniques are part of advanced electrochemical oxidation processes that enable the degradation of contaminants and microorganisms in food by applying an electric current. The continuous application of electric current generates reactive species such as hydroxyl radicals, active chlorine, ozone, and hydrogen peroxide within the medium. These reactive species interact with pesticide residues, initiating the degradation process. (Moreira et al., 2017). On the other hand, applying an electric pulse requires using an electric pulse generator. This generator supplies a high voltage directed towards a chamber containing the food and its load of contaminants and microbes. The contaminants and microbes undergo degradation via oxidation reactions driven by the presence of hydroxyl radicals (Arshad et al., 2020).

#### Photocatalysis

This process involves accelerating the reaction rates between two or more substances by using a catalyst and exposing them to a light source. Substances that can absorb light and act as catalysts are called photocatalysts. The process occurs when a semiconductor material is exposed to a specific wavelength of light, where the photons possess enough energy to be absorbed by the valence electrons, causing them to become excited and move to a higher energy level or conduction band. This simultaneous excitation results in the formation of holes in the valence band. Consequently, a photoexcited state is formed, generating a negatively charged electron and a positively charged hole (lack of negative charge), enabling the electron to react with electron acceptor molecules through reduction processes. In contrast, the hole reacts with electron donor molecules through oxidation processes. As a result, an environment of simultaneous oxidation and reduction is created, facilitating the degradation of contaminants and microorganisms (Ameta & Ameta, 2018).

## Electrolyzed water

Electrolyzed water is produced by the electrolysis of diluted NaCl in water, with the electrodes separated by

a membrane. At the anode, oxygen gas  $(O_2)$ , chlorine gas  $(Cl_2)$ , hypochlorite ion  $(OCl^-)$ , hypochlorous acid (HOCl), and hydrochloric acid (HCl) are generated, while at the cathode, hydrogen gas  $(H_2)$  and sodium hydroxide (NaOH) are produced. Consequently, the solution at the anode is referred to as acidic electrolyzed water, while the solution at the cathode is known as alkaline electrolyzed water (AIEW). Due to the generation of reactive species during this process, electrolyzed water exhibits disinfecting, degreasing, and antimicrobial properties. As a result, it becomes a highly effective solution for removing and disintegrating contaminants (Ding et al., 2019).

#### Ultrasonication

The application of ultrasound for the degradation of pesticide residues and microorganisms present in food has significantly increased in recent years due to its environmental friendliness, high effectiveness, and synergistic potential when combined with other techniques. Ultrasound application in an aqueous medium fosters the occurrence of acoustic cavitation, a phenomenon characterized by the formation, growth, and subsequent collapse of bubbles. These bubbles generate elevated temperatures and pressures upon implosion, facilitating reactive oxidizing species production. These highly reactive species actively participate in chemical reactions, effectively disintegrating contaminants and microorganisms within the food matrix. This sustainable approach represents a powerful strategy for mitigating foodborne risks while minimizing environmental impact (Pollet & Ashokkumar, 2019).

#### Ozone

Ozone is extensively employed for the disinfection of food and crops. Its remarkable oxidative capacity enables the degradation of molecular structures, making it a highly effective tool. Ozone finds application in storing and preserving fruits and vegetables, owing to its exceptional stability in atmospheric conditions. However, its aqueous form is typically employed in situ due to its decomposition into harmless oxygen ( $O_2$ ). When synergistically combined with hydroxy radicals, ozone intensifies organic compounds' oxidation and degradation process, including pesticide residues (Pandiselvam et al., 2020). This versatile and eco-friendly approach presents significant potential for enhancing food safety and quality.

#### Combined techniques for pesticide residues removal

In this study, we analyzed the effectiveness of combined techniques for removing pesticide residues in food based on a systematic review of 38 studies, as shown in Table 1S. The results revealed a wide range of pesticide residue removal, from 0 - 100% effectiveness (Fig. 2). However, it is noteworthy that only 12 studies (31,6%) exhibited lower variability, demonstrating a percentage of pesticide residue removal between 50 and 80% (Table 3).

Of these 12 studies, nine (75%) used US combined with other techniques. Additionally, Oz, UV, and EC were employed in four (33%), three (25%), and two (17%) studies, respectively. Furthermore, other techniques were employed in at least one study, including EW, Bak. Soda, Hidro. Perox., Ch. Sol, and WS.

#### Ultrasound combined with other techniques

The notable prevalence of ultrasound in combination with other techniques can be attributed to its remarkable synergistic potential, which can be attributed to the capacity of such combinations to generate additional reactive oxygen species and hydroxy radicals, thereby expediting the degradation of contaminants. For instance, Siddique et al. (2021) determined that the combined application of US and Oz achieved a remarkable removal efficiency of 99 – 100% for carbendazim and carbosulfan in spinach without compromising the quality of the vegetable. They carefully assessed the impact of these combined techniques on the product by analyzing various quality parameters, including vitamin C content (VC), total dissolved solids (TSS), titratable acidity (TA), and weight loss (WL). They found no significant alterations resulting from the application of US and Oz. The synergy between US and Oz arises from the inherent instability of ozone in aqueous media, leading to the formation of hydroxyl radicals, peroxide, and superoxide. Under the influence of US, the concentration of hydroxyl ions in the aqueous medium is enhanced, thereby facilitating the degradation of pesticide residues (Malakootian et al., 2020). Furthermore, Maryam et al. (2021), Fan et al. (2015), and Whangchai et al. (2017) employed combined US and Oz treatments to effectively remove PRs from strawberry fruits, lettuce, and tangerines, resulting in reduction percentages ranging from 98% to 99,99%, 68.02% to 82.16%, and 42% and 73%, respectively.

The variation observed in the reduction percentages might be attributed to factors such as: 1) the ozone medium (aqueous or gaseous), 2) ozone concentration, 3) physicochemical properties of the pesticide, 4) exposure



Fig. 2 Pesticide residue removal ranges by combined techniques

Techniques	Minor reduction (%)	Major reduction (%)	Pesticides	References
US+Oz	99.75	100.00	Thiamethoxam Imidacloprid Acetamiprid Thiacloprid Carbendazim	(Siddique et al., 2021)
AqOz+US	98.00	99.99	Thiamethoxam Imidacloprid Acetamiprid Thiacloprid Carbendazim	(Maryam et al., 2021)
US + UV	90.00	97.00	Chlorpyrifos	(Yuan et al., 2022)
UV + Hid.Perox	92.63	100.00	Boscalid Pyraclostrobin Fenbuconazole Glyphosate	(Skanes et al., 2021)
PAL+US	80.23	89.28	Chlorothalonil	(Ali et al., 2022)
US+CE	81.99	93.99	Captan Thiamethoxam Metalaxyl	(Cengiz et al., 2021)
US+Oz	68.02	82.16	Methamidophos (MDP) Dichlorvos (DDVP)	(Fan et al., 2015)
US + Bak.Soda	70.61	92.15	Fenpropathrin Cypermethrin Deltamethrin	(Yu et al., 2020)
WS+OMB+MCB+Aq.Sol	77.00	100.00	Carbosulfan Trichlorfon	(Li et al., 2021)
US+CE	69.80	95.06	Captan Thiamethoxam Metalaxyl	(Cengiz et al., 2018)
EW+US	50.00	89.00	Cyprodinil	(Zhao et al., 2018)
UV+TiO <sub>2</sub>	53.00	94.00	Pyraclostrobin Boscalid Fludioxonil Azoxystrobin	(Choi et al., 2020)

Table 3 Studies with lower variability and high pesticide residue removal by combining different techniques

time, 5) pesticide concentration, 6) medium temperature, 7) ultrasound frequency and energy input (Ameta & Ameta, 2018).

Akdemir Evrendilek et al. (2020) conducted a study to evaluate the effectiveness of US, Oz, and PEF in the removal of chlorpyrifos, fluvalinate, cyprodinil, pyraclostrobin and malathion from cherry juice. The results showed a wide range of residue reduction, varying from 8 to 97%. Due to the application of PEF done on microseconds, it appears that this technique contributes primarily to the degradation of PRs by enhancing the vibrational and rotational motion of the molecules, facilitating their interaction with reactive species in the medium. However, it is essential to note that the primary mechanism responsible for the residue degradation is likely attributed to oxidation reactions involving hydroxyl radicals (•OH) and reactive oxygen species (ROS) generated through the combined use of US and Oz. Additionally, Cengiz et al. (2018, 2021) combined US and EC to remove captan, thiamethoxam, and metalaxyl from tomatoes and lettuce. The results demonstrated significant removal ranging from 69,80% to 95,06% for tomatoes and 81,99% to 93,99% for lettuce. In these studies, EC was continuously applied for 1 to 10 min in the presence of a NaCl solution to achieve optimal removal. The degradation mechanism of the residues in these cases can be attributed to several factors, including free chlorine, hydrogen peroxide, ROS, and <sup>•</sup>OH. Additionally, dynamic and physical factors such as the generation of hotspots, acoustic shocks, microflows, high hydrodynamic tensions, and intense localized shear forces contribute to the degradation process. The chlorine compounds and hydrogen peroxide generated through electrochemical reactions in the aqueous medium, along with the hydrodynamic and acoustic cavitation effects induced by US, facilitate the oxidation reactions that lead to the degradation of pesticide residues (Calvo et al., 2019; Kilicli et al., 2019).

Yuan et al. (2022) achieved a removal efficiency of 97% of chlorpyrifos residues in milk by applying US and UV combined. Their findings revealed a synergistic effect

between these techniques. The high degradation percentage can be attributed to the amplifying effect of UV irradiation, which enhances the production of  $\bullet$ OH and reactive species generated by US. The mass transfer processes and the efficient transport of oxidizing agents throughout the sample also contribute to the overall system efficiency, promoting pesticide residue removal.

The combined application of US and PAL on tomatoes removed chlorothalonil residues from 80,23% to 89,28% (Ali et al., 2022). The degradation of these residues by PAL can be attributed to the acidity of the medium, generation of metastable reactive oxygen and nitrogen species (RONS), such as O<sub>3</sub>,NO<sub>2</sub>,NO<sub>3</sub>, as well as the high oxidation-reduction potential and electrical conductivity exhibited by PAL (Ekezie et al., 2019; Sarangapani et al., 2020; Zheng et al., 2019). Different studies have highlighted that an increase in acidity can enhance the activity of PAL (Bai et al., 2020; Qi et al., 2018). However, recent studies have indicated that prolonged PAL usage may lead to the formation of nitrites  $(NO_2^-)$  and nitrates  $(NO_3^-)$ , potentially compromising the final product quality (Ekezie et al., 2019; Johnson Esua et al., 2021). Therefore, the combined application of PAL and US is recommended, as it effectively mitigates prolonged exposure of the product to these species while capitalizing on the synergistic effects of US to amplify pesticide residue reduction. These effects can be summarized as follows: 1) increase in the production of oxidizing agents, including hydroxyl radicals, atomic oxygen (O), and hydroperoxyl radicals  $(HO_2^{\bullet})$ ; 2) dynamic mass transport phenomena and efficient dispersion of reactive species across the entire product surface; and 3) formation, expansion, and implosion of bubbles during the phenomenon of acoustic cavitation.

Zhao et al. (2018) conducted a study examining the synergistic effect of combining US and EW on removing cyprodinil residues in spinach. The findings revealed a notable removal rate ranging from 50 - 89%. The use of EW to eliminate microorganisms and contaminants has been extensively investigated and implemented in many studies (Calvo et al., 2019; Ding et al., 2019; Hao et al., 2011; Hricova et al., 2008; Shiroodi & Ovissipour, 2018; W. Zhang et al., 2021a, 2021b). In this study, the degradation mechanism of cyprodinil by EW can be attributed to the formation of specific chemical species at the anode  $(O_2, Cl_2, OCl^-, HOCl, HCl)$  and the cathode  $(H_2, NaOH)$ . The species actively react with the pesticide molecules, leading to their decomposition. The application of ultrasound catalysts facilitates the transportation of reactive species from the EW throughout the system via hydrodynamic phenomena. Consequently, the probability of interaction between these reactive and the pesticide molecules increases, thereby promoting their degradation. Furthermore, it is worth noting that ultrasound enhances transport and contributes to residue decomposition through acoustic cavitation and the generation of reactive oxygen species (ROS).

Yu et al. (2020) assessed the combined application of US and Bak. Soda on cabbage to facilitate the removal of fenpropathrin, cypermethrin, and deltamethrin residues. The study demonstrated an average removal ranging from 70.61 - to 92.15%. Bak. Soda was employed in an aqueous solution due to previous evidence highlighting the instability of pyrethroids in alkaline environments (Yang et al., 2017; Zuo et al., 2012). Similar to prior investigations, US generated ROS and OH that effectively reacted with the target pesticides. Moreover, this study meticulously explored the optimal parameters encompassing frequency, power intensity, exposure time, and temperature, which are crucial for achieving efficient pesticide reduction. The optimal values were determined: 28 kHz for frequency, 240 W for power intensity, 10 min of exposure time, and 20 °C for temperature. It was also observed that a lower frequency of US resulted in enhanced residue removal, owing to the formation of larger bubbles, thereby promoting more energetic bubble collapse during cavitation. Furthermore, it was noted that excessively high power intensity could impede cavitation by obstructing bubble formation, consequently impeding interaction with the residues. Additionally, the investigation showed a positive correlation between increased exposure time and more significant pesticide reduction. However, beyond a certain threshold, prolonged exposure led to a rise in residue levels, attributed to system saturation and counterproductive acoustic cavitation, wherein contaminants were compelled to re-interact with the product, causing a rebound effect. Temperature was another critical factor affecting the efficacy of US treatment. Elevated temperatures facilitated the escape of smaller bubbles from the system, impeding cavitation and consequently hindering residue degradation. Ultimately, the combined utilization of US and Bak. Soda exhibited notable potential in reducing residue levels within agricultural products, primarily due to the synergistic effect of US in facilitating the distribution of baking soda in less accessible regions, where conventional washing methods may fall short. This finding was supported by Li et al. (2022), who employed a combination of US and WS to eliminate triadimefon and boscalid residues in colza, resulting in a reduction from 0 - 61%. The lower removal percentage observed was attributed to several factors, including the lower concentration of oxidizing agents generated by applying a single technique, the physical structure of the product, the initial concentration, and the physicochemical characteristics of the pesticides. Notably, boscalid demonstrated higher hydrophilicity

than triadimefon, facilitating its removal through waterbased washing methods. Moreover, boscalid exhibited a greater affinity for the physical structure of the vegetable, leading to its accumulation in the product and, consequently, elevated residue levels. Additionally, the study highlighted the influence of pH on triadimefon removal, emphasizing its pH-dependent behavior and confirming the impact of its physicochemical properties on its removal efficiency. Finally, this research provided compelling evidence that achieving higher removal requires an extended washing duration time, elucidating the relatively lower residue removal efficiency of the combined technique compared to the approach employed by Yu et al. (2020).

Hsu et al. (2022) investigated the combined effects of US, air, SBE, and CSE on removing pesticide residues from fruits and vegetables. The most effective combination was the application of aeration with SBE and CSE, resulting in a remarkable removal from 25.58 - 100%. Ultrasound was used in this study due to its ability to generate ROS and free radicals, which acted in conjunction with acoustic cavitation. However, in this study, the application of US for 30 min was less effective than other techniques. This finding aligns with the observations and discussions presented by Yu et al. (2020), which emphasized that prolonged exposure to ultrasound can lead to the reintroduction of previously removed residues due to the counterproductive effects of acoustic cavitation. Thus, it is plausible that the adverse effects of ultrasound from the combined treatment in this study were influenced by the extended exposure time of the products to this specific technique. Furthermore, the study demonstrated that saponins, such as SBE and CSE, exhibit remarkable efficacy in removing pesticide residues. Saponins can form micelles, which encapsulate and sequester contaminants and pesticides. This capability can be attributed to the unique molecular structure of saponins, characterized by both hydrophilic and hydrophobic regions, giving them surfactant-like properties such as foaming, emulsification, and antibacterial activity (Manahan, 2017; Zhou et al., 2018). The positive synergistic effect observed between SBE and CSE and the aeration technique can be attributed to the continuous application of airflow, which optimizes the interaction between saponins and pesticides, enhancing the encapsulation process. Finally, the considerable variability observed in the range of residue removal can be attributed to several factors, including the concentration and physicochemical characteristics of the pesticides, particularly their persistence. For instance, this study indicated that removing pesticides such as imidacloprid and chlorfenapyr was more feasible through aeration combined with SBE than dinotefuran. This difference can be attributed to the higher persistence of dinotefuran despite its higher water solubility.

#### Ultraviolet irradiation combined with other techniques

We found four investigations that utilize UV in conjunction with other techniques, apart from US, to remove PRs. Skanes et al. (2021) evaluated the degradation of boscalid, pyraclostrobin, fenbuconazole, and glyphosate in apples by applying UV irradiation combined with Hidro.Perox. The study findings revealed a remarkably high degree of pesticide residue removal, ranging from 92.63 - 100%. Similarly, Ho et al. (2020) achieved 30 -80% removal efficiency for chlorpyrifos in apples. The remarkable synergistic effect observed between these techniques, which belong to the group of advanced oxidation processes, can be attributed to the optimal assistance provided by UV irradiation in decomposing H<sub>2</sub>O<sub>2</sub> into OH. As previously mentioned, hydroxyl radicals possess a high oxidative capacity, facilitating the degradation of pesticides. Furthermore, it is essential to note that the degradation of pesticides by UV radiation requires the presence of chromophores within their molecular structures, capable of absorbing light energy and promoting their degradation or decomposition. For instance, aromatic benzene rings, which are prevalent in the studied pesticides and many other pesticide compounds, can absorb UV light. Additionally, it is plausible that the primary variation observed in the removal efficiency between the two studies is influenced by the specific properties of the pesticide molecules and the experimental conditions employed (details in Table 1S).

Choi et al. (2020) achieved in their study a pesticide residue removal from 53 – 94% for pyraclostrobin, boscalid, fludioxonil, and azoxystrobin in carrots using a combined approach of UVC, VUV, and TiO<sub>2</sub> PhotoC. The degradation mechanism during photolysis involves the specific cleavage and formation of the substances' chemical bonds, rearrangements, and oxidation/reduction process (Katagi, 2018). UV-catalyzed reactions commonly target functional groups such as double bonds, aromatic rings, and carboxyl groups. Oxidation occurs through reactions with oxidizing species and radicals that form upon exposure to UV light or when photocatalysts are employed. For instance, UV radiation can degrade pesticide molecules through direct interaction with light or through reactions with  $\bullet$ OH and  $H_2O_2$  generated during light exposure in the medium.  $\mathrm{TiO}_2$  acts as a photocatalyst, increasing the effectiveness of UV radiation and promoting the production of ROS, such as <sup>•</sup>OH and superoxide radicals  $(O_2^{\bullet-})$ . However, this study demonstrated that  $TiO_2$  did not effectively enhance the performance of UV in terms of residue removal. This phenomenon may be attributed to the overproduction of intermediate species,

which hinders the continuous degradation of residues. Consequently, the authors recommend the implementation of a continuous flow of water or air to induce turbulence, potentially boosting residue degradation. The range of reduction in pesticide residues can be attributed to various factors, including the substances' molecular structure and physicochemical properties, their stability in the degradation medium, and the energy of the photons involved. For example, the degradation of boscalid in water and soil poses challenges due to its high stability (EPA, 2003). Furthermore, different studies have shown that the reduction percentage of this particular residue diminishes as the energy of the photons decreases (Food Safety Commission, 2004).

In their study, Baig et al. (2021) investigated the effectiveness of mint and riboflavin extracts in conjunction with light and UV irradiation for removing deltamethrin and  $\lambda$ -cyhalothrin residues. The results demonstrated a removal from 30.12% to 78.51%. This study revealed that their specific physicochemical characteristics hindered the degradation of both pesticides by UV radiation alone. Photoinitiators such as mint and riboflavin were employed to facilitate and expedite the UV-catalyzed photodegradation process and to overcome this result. In the context of the combined use of these techniques, the degradation mechanism involves the reaction of photoinitiators with UV light, leading to the generation of ROS and free radicals. These species subsequently react with the pesticides, synergistically enhancing the degradation process alongside the previously discussed actions resulting from UV exposure.

#### Ozone combined with other techniques

We found three studies that used ozone in the form of microbubbles and solutions, in conjunction with other treatments, to remove pesticide residues. Zhang et al. (2021a, 2021b) achieved removal from 36.60 - 46.37% for phoxim and chlorothalonil residues in baby cabbage through washing with air microbubbles and Oz. Similarly, Li et al. (2021) employed WS, OzMB, and Ch.Sol to remove carbosulfan and trichlorfon residues, resulting in a reduction percentage ranging from 77 - 100%. The pesticide degradation mechanism by combining these techniques can be attributed to 1) the oxidative properties exhibited by ozone, 2) the physical effects during the formation, growth, and collapse of the bubbles, and 3) the production of free radicals. During the bubble collapse, vortices and liquid jets are formed, facilitating the removal of pesticides from the surface of the food through physical mechanisms. The mechanisms of action of ozone and hydroxyl radicals have been previously discussed.

Pounraj et al. (2021) investigated the combined utilization of Oz and Lact.Ac. to remove chlorpyrifos and  $\lambda$ -cyhalothrin residues from tomatoes, cucumbers, carrots, and lettuce. The study yielded residue removal efficiencies from 26 – 100%, achieved through ozone oxidation reactions facilitated by the presence of free radicals in the medium. Furthermore, the observed synergistic effect between Lact.Ac. and Oz can be attributed to the gas stability under acidic conditions, enabling an extended lifespan within the medium and enhancing its reactivity with pesticides (Wang et al., 2019; Yuk et al., 2007).

#### Electrolyzed water combined with other techniques

Wu et al. (2019) assessed the pesticide residue removal from kumquats, spinach, and cucumbers using various treatments, including AIEW, activated oxygen, calcium solutions, sodium solutions, Oz, and sodium bicarbonate. The study achieved residue removal efficiencies ranging from 4 – 85%. The wide variability in the removal percentages can be attributed to several factors: 1) the physicochemical characteristics of the pesticides, such as their octanol-water partition coefficient (Kow) and acid dissociation constant (pKa), as well as the properties of the solutions used, including solute concentration and temperature, and 2) the parameters associated with the emerging techniques employed, such as current intensity, power level, and ozone concentration. The degradation mechanism of pesticides by ozone action has been previously discussed. The study selected AIEW due to its proven effectiveness in removing pesticide residues from fruits and vegetables (Hao et al., 2011). AIEW, generated at the cathode during the electrolysis of a saline solution, possesses alkaline properties. The observed synergy between AIEW and the use of calcium and sodium bicarbonate solutions can be attributed to the elevation of pH in the solution, facilitated by these substances, which enhances residue removal through basic hydrolysis reaction mechanisms. However, it should be noted that certain substances may not readily undergo degradation via such reactions. Hence, activated oxygen, which encompasses a range of ROS ( $O_3, O_2, O_2^{\bullet-}, HO^{\bullet}, O_2^{\bullet-2}$ ), was employed to degrade the pesticides through oxidation-reduction reactions.

Furthermore, Calvo et al. (2019) removed the residue from 3 - 75% for cyprodinil, tebuconazole, and iprodione using EW, ClO<sub>2</sub>, and PhotoC. The utilization of photocatalysts such as TiO<sub>2</sub> has been previously discussed in this study. These treatments enable the degradation of pesticides through the interaction of UV light, PhotoC, and Aq.Sol, resulting in the generation of highly reactive •OH. Moreover, in this particular study, neutral EW was employed, leading to a limited removal capacity. This result can primarily be attributed to the relatively low disinfection efficacy of neutral EW (Ding et al., 2019; Hricova et al., 2008). Similarly, like Oz,  $ClO_2$  degrades pesticides through oxidation mechanisms due to its potent oxidizing properties. However, the formation of chlorite  $(ClO_2^-)$  and chlorate  $(ClO_3^-)$  as byproducts when reacting with other substances necessitate removal due to potential environmental and human health implications (EFSA, 2015; Hebert et al., 2010; USEPA, 2022).

Similarly, (Zheng et al., 2019) successfully removed phoxim residues from grapes, achieving removal percentages ranging from 10 – 73,60% through the combined application of PAW, WS, and AirD. The observed synergistic effects among these techniques in pesticide residue removal can be attributed to the physical actions involved, such as washing and airflow, and the chemical reactions facilitated by RONS by PAW and AirD, which effectively degrade the pesticide compounds. <sup>•</sup>OH.

#### Conventional techniques combined with other techniques

Here, we presented a comprehensive review of 15 studies that employ combined conventional treatments to remove various PRs from diverse food samples. The observed percentage of pesticide residue removal ranges from 0 - 100% (Table 1S). The substantial variability in removal efficacy can be attributed to multiple factors, including 1) the effectiveness of each treatment technique, 2) the physicochemical properties of the pesticides, 3) specific physical parameters such as temperature, pressure, and cooking time, 4) the type of agricultural products under investigation, and 5) the experimental design employed. For instance, WS and Bl methods were more efficient in removing residues with lower octanol-water partition coefficients (Kow) compared to Fry and S.fry techniques (Huan et al., 2015; T. Liu et al., 2020). The Kow, a measure of the hydrophilicity and hydrophobicity of a chemical compound, indicates the tendency of a substance to partition between octanol (representing a hydrophobic environment) and water (representing a hydrophilic environment). A lower kow value denotes higher hydrophilicity, whereas a higher value suggests greater hydrophobicity (Cumming & Rücker, 2017). Hence, methods involving water-based solutions or polar chemical agents, such as WS and Bl, facilitate the removal of pesticides with lower Kow values due to their increased hydrophilicity (Ratnamma et al., 2021). In contrast, Fry and SFry techniques employ oil as a cooking medium, effectively removing pesticide residues with higher Kow values or greater hydrophobicity. Accordingly, more polar molecules interact with water, while less polar molecules exhibit greater affinity for oil (Łozowicka & Jankowska, 2016). Nevertheless, the efficacy of these techniques can vary depending on the specific mode of action of the pesticide in plants. For instance, Lee and Jung (2009) investigated the removal of dichlofluanid, flusilazole, folpet, iprodione,  $\lambda$ -cyhalothrin, and lufenuron from hot peppers leaves using the WS, Bl, and dry method. Interestingly, despite its greater lipophilicity,  $\lambda$ -cyhalothrin demonstrated superior removal efficiency through WS compared to lufenuron. This disparity can be attributed to the systemic properties of each pesticide. While  $\lambda$ -cyhalothrin functions as a contact pesticide, lufenuron exhibits systemic characteristics, facilitating its penetration and translocation within the plant. This effect leads to a diminished efficacy of the washing method in removing systemic pesticides compared to contact pesticides. Consequently, in this case, the efficacy of WS as a removal method is primarily governed by the physical and dynamic forces involved rather than intermolecular interactions between water and the pesticide. Moreover, similar findings have been reported by Kim et al. (2015) and Hanafi et al. 2016, who advocated for a greater focus on the transfer mechanisms and modes of action of pesticides rather than solely considering their hydrophobic or lipophilic properties.

Furthermore, it is crucial to consider that certain pesticides can undergo degradation through physical processes, such as volatilization and leaching, as well as chemical processes, including hydrolysis reactions, which are strongly influenced by the physicochemical properties of each molecule. For instance, Aktar et al. (2010) proposed a basic hydrolysis reaction as the degradation mechanism for quinalphos, resulting in the formation of quinoxalinol and quinoxalinyl-thiophosphate as degradation products. Additionally, (Tomer et al., 2014) demonstrated that the pH of the solution plays a significant role in facilitating acidic or basic hydrolysis reactions of pesticide residues. In their study, cypermethrin was readily observed to degrade in highly acidic or alkaline solutions. Furthermore, pH variation affects the ionizable functional groups within the molecules, influencing their adherence to the cuticle of the product and subsequently facilitating their removal. Consequently, certain studies have employed substances such as sodium carbonate, bicarbonate, calcium hydroxide, and acetic acid to modify the pH conditions (Andrade et al., 2015; Rasolonjatovo et al., 2017; Tomer et al., 2014). Therefore, it is essential to consider parameters such as the pKa and electrostatic potential map of each molecule when evaluating the degradation mechanisms of pesticides, as they provide valuable insights into the most reactive sites. Finally, regarding pesticide residue removal, consecutive applications of conventional techniques mimicking household practices have shown promising results, particularly with frying and blanching methods. This result is attributed to the increased solubility of pesticide residues in water

or oil, depending on their affinities. Furthermore, hightemperature processes like boiling, pasteurization, frying, and stir-frying have proven effective in removing pesticides with lower vapor pressure through volatilization and thermal degradation (Bonnechère et al., 2012; Hanafi et al., 2016; Jankowska & Łozowicka, 2022; T. Liu et al., 2020; Łozowicka & Jankowska, 2016).

## Effects of applying combined techniques on product quality

In this review, studies employing conventional combined methods for PR removal exhibited negligible effects on the quality of agricultural produce. In contrast, the application of emerging combined techniques manifested variable impacts on food quality. These effects were assessed by analyzing various parameters, including vitamin content, TSS, TA, WL, alterations in physical appearance, and textural changes in food products. Notably, Yuan et al. (2022) explored the synergistic application of US and UV irradiation for the degradation of chlorpyrifos in milk. Their findings indicated that these advanced techniques had a minimal influence on the milk quality, affecting nutrient composition, fatty acid, lipid oxidation, physicochemical properties, and color to a negligible extent. Among these parameters, a slight increase in fatty acids and lipid oxidation was observed, alongside a nuanced alteration in coloration, most notably when US exceeded a duration of 30 min. However, no significant alterations were noted when US was applied for less than 20 min. Zheng et al. (2019) employed PAW, alongside WS and AirD techniques, to remove phoxim residues from grapes. Additionally, the study assessed the influence of PAW on various quality parameters, including color, firmness, sugar content, VC, and superoxide dismutase activity. The outcomes revealed no significant alterations in these metrics, indicating that PAW application does not adversely affect the grapes' bioactive compounds or their antioxidant capacity. In a separate investigation, Ali et al. (2022) analyzed the impact of PAL combined with US on tomato quality, particularly focusing on color, texture, pH, TSS, ascorbic acid (AscAc), and lycopene during the evaluation of chlorothalonil residue removal. The results demonstrated negligible effects on theses essential quality parameters. Furthermore, Maryam et al. (2021) successfully reduced bacterial load and PRs, and delayed fungal decomposition in strawberries, thereby enhancing their shelf life by applying AqOz and US. This treatment not only maintained the strawberries' quality regarding TSS, TA, pH, AscAc content, and anthocyanins but also suggested an extended preservation of fruit quality beyond that of the control group. Whangchai et al. (2017) applied OzMB and US to mandarins for ethion residue removal, noting that this combined treatment did not significantly alter the quality of fruits. The evaluated parameters included peel color, WL, TSS, TA, and AscAc, concluding that these were unaffected by the treatment.

On the other hand, Baig et al. (2021) removed PRs from cauliflower using a combination of photosensitizers and UV light, further determining that these treatments did not significantly affect the antioxidant activity of natural antioxidants present in the product. Siddique et al. (2021) utilized Oz and US con spinach leaves, successfully removing PRs without adversely affecting quality parameters such as VC, TSS, TA, and WL over a week of storage under refrigerated conditions. However, they recommend limiting US exposure to less than 15 min. Li et al. (2021) achieved pesticide residue removal in apples using OzMB, tap water, hypochlorous acid, AqOz, and MCB, noting that OzMB had a minimal impact on the superficial color of the apples. Also, significant color differences were observed after 10 min of Oz exposure, highlighting the importance of assessing these washing methods on the effects of fruit quality. Fan et al. (2015) employed US and Oz to treat lettuce contaminated with PRs, finding that this combination effectively degrades them without negatively affecting the lettuce quality. Quality parameters evaluated, such as TA, soluble sugars, VC, chlorophyll, carotenoids, and nitrates, showed no significant changes. Nevertheless, a decrease in TA and VC was observed after 60 min of treatment, attributable to the reaction of VC with the Oz and radicals generated by US. Akdemir Evrendilek et al. (2020) applied PEF, Oz, and US in combination on cherry juice, demonstrating its effectiveness in pesticide removal and microbial inactivation without adversely affecting the physical, bioactive, and sensory properties of the juice except when Oz was used alone or in combination with other techniques. This combined treatment did not adversely affect the juice's pH, TSS, and electrical conductivity. Nonetheless, treatments with Oz alone for more than 10 min and in combination with PEF and US significantly affected the initial color values (L\*, a\*, b\*, C\*), turbidity, Total Antioxidant Capacity (TAC), Total Monomeric Anthocyanin Content (TMAC), Total Phenolic Compounds (TPC), and AscAc concentration. However, treatments with US for less than 30 min did not affect product quality. Calvo et al. (2019) used EW, ClO<sub>2</sub>, and PhotoC in combination to remove PRs in various stone fruits, determining that these treatments did not significantly affect fruit quality indicators such as TSS, TA, pH, and firmness, with a maximum exposure to  $\text{ClO}_2$  of 25 min. Pounraj et al. (2021) found that the combined application of Oz and Lact.Ac. on fresh vegetables was more effective in reducing PRs and bacterial load than individual treatments without observing significant differences in sensory attributes compared

to untreated vegetables, suggesting that sensory quality remained intact. Hsu et al. (2022) applied saponins extracted from natural products along with US for 30 min to remove PRs in fresh fruits and vegetables without affecting the color of the products. Yu et al. (2020) used US and sodium bicarbonate to remove pyrethroid residues in cabbage and observed no significant changes in VC, total soluble sugar, texture, and sensory evaluations, indicating that the treatments did not compromise product quality. In summary, the application of emerging combined techniques for pesticide residue removal has a varied impact on the quality of agricultural products, with most studies reporting that these effects are insignificant. The observed impacts on quality are typically associated with prolonged exposure to these techniques.

#### **Future perspectives**

The intersection between the effectiveness of PRs elimination and the maintenance of quality in agricultural products underscores the potential of novel, combined decontamination techniques. These are recognized for their efficacy and safety and their capacity to conserve the nutritional integrity and overall quality of treated foods. Despite these advances, there remains a pivotal need for further optimization of these elimination technologies. Therefore, there is a need for a comprehensive evaluation encompassing physicochemical parameters, precise dosing of oxidizing agents, and meticulous calibration of exposure durations to these technologies. Progress in these domains is expected to catalyze the development of innovative technological solutions that leverage these techniques, facilitating their adoption in commercial applications that benefit consumers, producers, and the agricultural sector at large. The integration of these practices within the industry is motivated by an escalating demand for products free of pesticide residues and by international regulatory mandates aimed at minimizing PR presence in exported goods.

Moreover, the market presently offers consumer-grade devices, predominantly originating from China, that utilize US, Oz, and EC treatment for food purification. However, the scientific foundation underpinning these devices, along with the efficacy of their operational settings in mitigating PRs and other contaminants, remains largely opaque. This finding highlights an imperative for developing and validating such devices through stringent scientific scrutiny.

Additionally, this review reveals a paucity of research dedicated to exploring the degradation byproducts resulting from the application of combined decontamination techniques. Yuan et al. (2022) have identified five degradation byproducts of chlorpyrifos with reduced toxicity compared to the original pesticide, as determined through density functional theory (DFT) analyses. Similarly, Ali et al. (2022) have delineated the degradation pathway of chlorothalonil following PAL-U treatment, recognizing various byproducts without assessing their toxicity. Baig et al. (2021) elucidated the degradation mechanisms of the pesticides under study without examining the toxicity of the resultant byproducts. Furthermore, Calvo et al. (2019) were unable to identify specific degradation byproducts, although they noted instances where such byproducts may present greater toxicity than their precursor compounds. Therefore, it is imperative for future investigations to prioritize the identification and toxicological assessment of these degradation byproducts, ensuring the comprehensive safety and efficacy of these emergent decontamination technologies.

### Conclusions

The analysis of multiple studies reveals that combining different techniques increases the efficacy of pesticide residue elimination from food. Furthermore, combining emerging techniques such as ultrasound, electrical current, electrolyzed water, ultraviolet, and plasma irradiation with other treatments demonstrated enhanced removal efficiency and decreased variability in residue elimination. Also, ultrasound appears to be the optimal technique to pair with others in achieving enhanced results, given its significant synergy with the other methods examined in this review.

The primary degradation mechanism is related to the interaction between pesticide residues and the reactive species generated by the combined techniques, such as reactive oxygen species, hydroxyl radicals, and physical factors such as hotspots, acoustic shocks, mass transfer, and intense shear forces.

Ozone combined with microbubbles, lactic acid, and other techniques, such as UV irradiation and electrolyzed water, showed significant removal percentages for various pesticide residues in baby cabbage, tomatoes, cucumbers, and lettuce. The oxidative properties of ozone, physical effects of bubble formation and collapse, and generation of free radicals contributed to the degradation of pesticide residues.

Conventional techniques, including washing, blanching, frying, and stir-frying, demonstrated variable but generally effective removal of pesticide residues when combined with other treatments. The efficacy depended on factors such as the physicochemical properties of the pesticides, the type of agricultural products, and specific parameters of the techniques employed.

In summary, combining ultrasound, electrical current, ozone, plasma and ultraviolet irradiation, and conventional techniques has shown great potential in removing pesticide residues from food samples. These combined techniques enhance degradation efficiency, improve mass transport phenomena, generate reactive species, and facilitate physical removal. However, the specific combination and parameters should be carefully selected based on the targeted pesticides, food products, and desired residue removal efficiency.

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Supplementary Material 1.

#### Authors' contributions

Conceptualization, JSFT, and DS; methodology, JSFT, and DS; validation, JSFT, and DS; formal analysis, JSFT, and DS.; investigation, JSFT; resources, XX; data curation, XX; writing—original draft preparation, JSFT; writing—review and editing, JSFT; visualization, JSFT, and DS; supervision, DS All authors have read and agreed to the published version of the manuscript.

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## Declarations

#### **Consent for publication**

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The authors declare no conflict of interest.

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