



Review

Portable sensors equipped with smartphones for organophosphorus pesticides detection

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ARTICLE INFO

Keywords:

Organophosphorus pesticide

Smartphone

Sensor

Optical

Electrochemistry

ABSTRACT

Organophosphorus pesticides (OPs) play an important role in agricultural production and the accurate detection of OP residues is essential to ensure food safety. Portable sensors are expected to be a potential device due to their high detection efficiency, easy-to-use processes and low cost. Due to the widespread popularity and powerful capabilities of smartphones, smartphone-based sensing systems have rapidly developed into ideal tools for portable detection, however, a systematic review on the detection of OPs is still lacking. Therefore, a comprehensive overview of sensors equipped with smartphones for OP detection in recent year is provided; this overview includes their sensing signals (colorimetric, fluorescent, chemiluminescent and electrochemical signals), detection mechanism, analysis applications, advantages/disadvantages and perspectives. Moreover, the progress of sensors equipped with smartphones for the detection of OPs in food is thoroughly summarized. This review contributes to food safety and the development of efficient and reliable methods for smartphone-based OPs detection.

1. Introduction

The rapid growth of the world population, which will reach 10 billion in 2050 (Umaphathi et al., 2021), has led to higher demands on agricultural productivity. To increase food production, pesticides (insecticides, herbicides, rodenticides, fungicides, etc.) have become essential tools in modern agriculture to prevent diseases, kill pests and remove weeds (Fang et al., 2020). In recent decades, the use of pesticides has been effective in ensuring sustained food production; thus, pesticide use has significantly increased (Li et al., 2022; Umaphathi et al., 2022a). However, some pesticides are toxic and carcinogenic and can cause serious health problems once they enter the human body; these problems include liver damage, cancer, blindness, reproductive problems, neurological disorders, endocrine disorders, and respiratory diseases (Umaphathi et al., 2022b). Moreover, some pesticide residues are persistent, and they can enter the food chain through air, water and soil, thus causing bioaccumulation and biomagnification. Therefore, pesticide pollution has attracted widespread global attention as a public health problem that cannot be disregarded (Bedair et al., 2022; Dar et al., 2022; Schleiffer & Speiser, 2022; Tudi et al., 2021).

Among many pesticides, organophosphorus pesticides (OPs) are currently the most widely used pesticides in agricultural production worldwide and account for 45% of the pesticide market due to their high toxicity and wide range of targets; the common OPs include chlorpyrifos, paraoxon, glyphosate, malathion, and dichlorvos (Fu et al., 2022; Mali et al., 2023; Mdeni et al., 2022). In actual agricultural production, the effective utilization of OPs is relatively low, and due to continuous and excessive use, OP residues have been detected in crops, air, soil, and various water bodies, causing concerns regarding food safety and health risks (Pathiraja et al., 2023; Umaphathi et al., 2022a). OPs that enter the body can recognize the active site of acetylcholinesterase (AChE, a serine hydrolase) and covalently bind to the -OH group of serine, thereby irreversibly inhibiting the activity of AChE (Khosropour et al., 2022; Lazarević-Pašti, 2023). In turn, this leads to a massive accumulation of acetylcholine (ACh, a neurotransmitter of the nervous system) in the body and causes sustained nerve impulses that can lead to abnormalities in the nervous, respiratory, cardiovascular, and digestive systems (Mali et al., 2022; Umaphathi et al., 2022a). Considering the severe impacts of OPs, accurate and rapid detection of OP contamination and timely response are necessary to ensure food safety and protect

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<https://doi.org/10.1016/j.foodchem.2023.137456>

Received 2 July 2023; Received in revised form 3 September 2023; Accepted 10 September 2023

Available online 12 September 2023

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public health.

Although conventional methods for OP detection, such as gas chromatography-mass spectrometry (GC-MS) and liquid chromatography-mass spectrometry (LC-MS), are sensitive, reproducible and reliable, they are based on laboratory techniques and require experienced technicians and expensive large-scale instruments (Issaka et al., 2023; Umaphathi et al., 2021). Moreover, the sample preparation and pre-treatment processes are tedious and time-consuming. These inherent limitations render traditional methods of pesticide detection and screening unsuitable for on-site detection of OPs, especially in remote rural areas where resources are scarce. Therefore, to address these limitations, there is an urgent need to develop portable detection equipment that is simple to operate, cost-effective and sensitive.

Compared to laboratory-based analytical technologies, the sensors developed in recent years tend to be miniaturized and integrated, and many portable sensors for pesticide detection have been developed (S. Huang et al., 2019; Poudyal et al., 2023). However, to practically apply these portable sensors for on-site detection, most need to be integrated with other accessory devices to implement the detection process, analyse the response signal, and display the results. Currently, smartphones have become indispensable mobile devices in the daily lives of most people (Sun and Hall). A modern smartphone resembles a microcomputer equipped with a user-friendly operating system, a multicore processing system, customized applications, a high-capacity battery, large memory, audio and USB interfaces, wireless communication systems (Bluetooth, Wi-Fi, near-field communication), a global positioning system (GPS), and high-resolution cameras. They are ideal to use as a data analysers, image processors, monitors, and controllers in portable sensing strategies for detection outside the laboratory (Issaka et al., 2023). Moreover, the detection data and signals of target analytes can be transmitted to the smartphone or to the cloud via wired communication or wireless communication for further analysis. Due to the rapid development of smartphones in recent years and their popularity, portability, reliability, and wide applicability, an increasing number of studies have been conducted to effectively integrate smartphones with portable sensors (W. Chen et al., 2021; Cheng et al., 2021; Choi & Seokheun, 2016; Choi et al., 2021; Ding et al., 2022; Guan et al., 2019; Kayani & Omer, 2022; Madrid et al., 2022; Zeng et al., 2022; Zhang & Shi, 2022; L. Zhang et al., 2022). In addition, combining smartphones with portable sensors helps to reduce the size of the detection device compared to laboratory-based instruments. Also, the production, operation and maintenance costs of smartphones are much lower, causing them to be cost effective. Moreover, smartphones equipped with high-resolution cameras and smart chips simplify data collection and processing, and nonprofessional consumers can access target analyte detection results in a short period. Therefore, sensors equipped with smartphones are expected to be widely used in less developed or remote areas.

In summary, due to the severity of OP hazards, the necessity of portable on-site detection, and the prospect of smartphone applications, the recent progress in portable sensors equipped with smartphones for OP detection is reviewed; these sensors mainly include optical sensors and electrochemical sensors. An in-depth review of the OP detection mechanisms is provided based on colorimetric, fluorescent, chemiluminescent and electrochemical methods and the advantages and limitations of each sensing strategy is presented. Finally, the prospects and challenges of these sensors for future practical applications are discussed. It is expected that this review paper will contribute to ensuring food safety and provide a theoretical reference for the design of sensors equipped with smartphones for OP detection.

2. Optical sensors equipped with smartphones for OP detection

Currently, the main optical sensors equipped with smartphones for OP detection include fluorescent, colorimetric, chemiluminescent and colorimetric and fluorescent dual-mode sensors (Umaphathi et al., 2022b;

Umaphathi et al., 2021). They are simple, highly sensitive and selective, and they can rapidly detect and screen target analytes on site; particularly, these can be applied in remote or rural areas where resources are scarce. Functional materials are critical components of optical sensors and play an essential role in signal recognition, transduction, and amplification. Common functional materials include noble metal nanomaterials, quantum dots, graphene and metal organic frameworks (Q. Chen et al., 2021; Umaphathi et al., 2022b; Yan et al., 2018). The combination of different optical systems and functional materials enables a single optical sensor to output multiple responses, which improves the accuracy of the results. Furthermore, smartphones contain complementary metal oxide semiconductor (CMOS) image sensors, which convert optical images into electronic signals (Huang et al., 2018; Ramirez-Coronel et al., 2023). Specifically, in smartphone-based optical strategies, images are initially captured by the smartphone, and then analysis applications (ImageJ, Color Recognizer, Color Picker, etc.) on the smartphone are used to convert the colour information of the images into digital signals under a certain colour space (Red-Green-Blue (RGB), Cyan-Magenta-Yellow-black (CMYK), Hue-Saturation-Brightness (HSB)/Hue-Saturation-Lightness (HSL), CIE tristimulus values (CIE XYZ) model, etc. (Fan et al., 2021)); finally, a relationship between the digital signals and the concentration of the analyte is constructed, thus achieving the quantitative analysis of the analyte. Information on the applications of optical sensors equipped with smartphones for detecting OPs is summarized in Table 1.

2.1. Colorimetric sensors equipped with smartphones

Colorimetric sensing techniques enable qualitative or semi-quantitative analysis of target analytes based on changes in their colour and intensity. Although the sensitivity and accuracy of colorimetric strategies need further improvement compared to other optical methods, they are most readily available and low-cost; thus, they are the most common optical sensing strategy for using smartphones as detectors. Therefore, their use as auxiliary analytical tools for preliminary detection to screen large quantities of pesticides is highly attractive. In addition, 3,3',5,5'-tetramethylbenzidine (TMB), indoxyl acetate (IDA), and 5,5'-dithio-bis-2-nitrobenzoate (DTNB) are commonly used chromogenic reagents in OP colorimetric detection. They can be converted after reactions into TMB_{OX}, indigo dye and 2-nitro-5-thiobenzoate (TNB), respectively, with a distinct colour change. The extent of these colour changes induced by the detection reactions is correlated with the OP amounts (Gong et al., 2022). The following is a detailed discussion of colorimetric sensors equipped with smartphones for OP detection in food; these sensors are classified according to the different chromogenic reagents.

Colourless TMB can be oxidized to blue TMB_{OX} by compounds with strong oxidizing properties, such as H₂O₂ or dissolved oxygen, in the presence of nanozymes, and the chromogenic reaction will be inhibited in the presence of OPs (Y. Huang et al., 2019). Based on the above principles, Wei et al reported a test paper for the detection of dichlorvos (DDVP) in vegetables and fruits (Wei et al., 2020). They used AChE and choline oxidase (ChOx) to catalyse the generation of H₂O₂ from acetylcholine chloride (AChCl). Moreover, the nickel ferrite/nickel oxide nanoparticles co-loaded with three-dimensional reduced graphene oxide (3DRGO-NiFe₂O₄/NiO NPs) had mimetic peroxidase activity and catalysed the production of OH radicals from H₂O₂, which in turn oxidized TMB. When OPs were present, AChE and ChOx activity was inhibited, and the blue colour of the test paper became lighter. The images of the test papers were converted into saturation (S) and lightness (L) through a smartphone, enabling quantitative analysis of the OPs. The logarithmic concentration of DDVP showed a favourable linear relationship ($R^2 = 0.99$) with the S/L value, and the limit of detection (LOD) was 10 µg/mL.

While most research in the past has reported on nanozymes with mimetic peroxidase activity, the development of nanozymes with

Table 1
Application information of optical sensors equipped with smartphones for OP detection.

Analytes	Matrix	Optical signal	Smartphone app	Colour model	LOD	Linear range	Ref.
Dichlorvos	Fruit and vegetable	Colorimetry	/	HSL	10 µg/mL	50–2.4 × 10 ⁴ µg/mL	(Wei et al., 2020)
Malathion	/	Colorimetry	Color Picker	RGB	7.5 ng/mL	0.0083–5.333 µg/mL	(P. Liu et al., 2021)
Paraoxon	Juice	Colorimetry	SAMPS	Gray	0.5 ng/mL	5 × 10 ⁻⁴ –1.5 µg/mL	(Jin et al., 2021)
Malathion	Lake water and apple	Colorimetry	Color Assist	RGB	1 nM	1–10 nM	(Ge et al., 2022)
Malathion	Water	Colorimetry	Sentinel Reports	RGB	10 nM	1 × 10 ⁻⁸ –1 × 10 ⁻⁶ M	(Sicard et al., 2015)
Paraoxon							
Malathion	Apple, cucumber, and tomato	Colorimetry	/	RGB	/	10–500 nM	(Jang et al., 2020)
Malathion	Domestic water	Colorimetry	Adobe Capture CC and Desmos	RGB, CMYK	51.9 ng/mL	/	(Wu & Wang, 2018)
Methyl parathion	Apple	Colorimetry	QuickMark	CMYK	0.2 µg/mL	20–100 µg/mL	(Guo et al., 2015)
Malathion	Water, plant, and apple	Colorimetry	ImageJ	Gray	/	0.1–50 mM	(Li et al., 2021)
Methyl paraoxon	Edible plants	Colorimetry	iDropper	CMYK	0.42 µM	2.1–21 µM	(Wei et al., 2019)
Paraoxon	/	Fluorescence	Color recognizer	RGB	0.73 ng/mL	/	(J. Chen et al., 2020)
Paraoxon	Soil and Water	Fluorescence	Color recognizer	RGB	0.03 ng/mL	0.1–250 ng/mL	(F. Liu et al., 2021)
GLP	/	Fluorescence	Color recognizer	RGB	0.93 nM	0–5 µM	(Wei et al., 2023)
DDVP	/	Fluorescence	Color Picker	RGB	4.1 ng/mL	0.005–0.04 µg/mL	(Liu et al., 2020)
Chlorpyrifos	Vegetable and fruit	Fluorescence	ImageJ	Gray	0.73 ng/mL	/	(Cheng et al., 2018)
Diiazinon					6.7 ng/mL		
Malathion					0.74 ng/mL		
OPs	Vegetables	Fluorescence	Detect	Gray	0–20 µg/mL	0.01–0.5 mg/kg	(Jiao et al., 2021)
Paraoxon	Environmental and agricultural Samples	Ratiometric fluorescence	ImageJ	HSL	<10 ng/mL	10–500 ng/mL	(Jin et al., 2019)
DDVP	Water and foods	Ratiometric fluorescence	ImageJ	RGB	0.38 ppb	1–40 ppb	(Huang et al., 2021)
OPs	Buffered aqueous solutions and apple juice	Ratiometric fluorescence	Color recognizer	RGB	1.8 ng/mL	5–1000 ng/mL	(Li et al., 2023)
Methyl parathion	Rice and tap water	Ratiometric fluorescence	/	HSL	0.06 µg/mL	0.1–7 µg/mL	(Fahimi-Kashani & Hormozi-Nezhad, 2020)
OPs	Foods and water	Colorimetry-fluorescence	Sensor Photo	Gray	0.67 µg/L	/	(Cai et al., 2021)
Trich	Foods	Colorimetry-fluorescence	Color Picker	RGB	0.4 µg/L		
					0.083 ng/mL	0.28–50.0 ng/mL	(Shen et al., 2023)
					0.11–50.0 ng/mL		
					0.033 ng/mL		
GLP	/	Colorimetry-fluorescence	Color recognizer	RGB	3.31 nM	0–10 µM	(Wei et al., 2021)
Paraoxon	/	Colorimetry-fluorescence	Color recognizer	RGB	2.81 nM		
					0.01 µg/L	0.01–1000 µg/L	(Pei et al., 2021)
					0.20 µg/L	5–100 µg/L	
						100–1000 µg/L	
GLP	water and cereal samples	Colorimetry-fluorescence	Color recognizer	RGB	9.06 µg/L	0.01–6.67 mg/L	(Luo et al., 2022)
					141.39 µg/L	0.20–6.67 mg/L	
					45.0 µM	/	(Montali et al., 2020)
Chlorpyrifos methyl	/	CL	ImageJ	RGB			

oxidase-mimicking activity is more attractive because H₂O₂ with high toxicity and poor stability is not required to be involved in the reactions they catalyse (Q. Chen et al., 2020). As shown in Fig. 1A, the Ag₃PO₄/UIO-66 composite proposed by Liu et al. catalysed the conversion of dissolved oxygen to superoxide radicals, which in turn oxidized TMB (P. Liu et al., 2021). When OPs were present, the OPs rapidly adsorbed to the surface of the composite, masking its active site and thus inhibiting the chromogenic reaction. This system responded to malathion in the range of 0.0083 to 5.333 µg/mL, and the LOD was 7.5 ng/mL. Notably, this method was only applicable to pesticides containing phosphothionate groups (malathion, chlorpyrifos, fenthion, etc.) since it was based on the formation of Ag-S bonds between the target analyte and Ag₃PO₄ to inhibit the activity of Ag₃PO₄/UIO-66.

In addition, nanozymes with oxidase-mimicking activity can directly

catalyse the oxidation of TMB into TMB_{OX} without the involvement of dissolved oxygen. For example, Jin et al used MnO₂ nanoflakes (NFs) to directly catalyse the oxidation of TMB into TMB_{OX} under acidic conditions, and the grey value of the images showed good linearity (R² = 0.9923) with the logarithmic concentration of paraoxon in the juice over the range of 5 × 10⁻⁴ to 1.5 µg/mL with an LOD of 0.5 ng/mL. Interestingly, this sensing device could be applied to the detection of other pesticides by simply changing the corresponding enzymes (Jin et al., 2021).

Similar to the TMB, colourless IDA was converted to blue products when catalysed by AChE. However, the chromogenic reaction was inhibited when OPs were present. As shown in Fig. 1B, Sicard et al. proposed a paper-based analytical device (µPAD) for OP detection in water (Sicard et al., 2015). Based on the inhibition of AChE by OPs, the

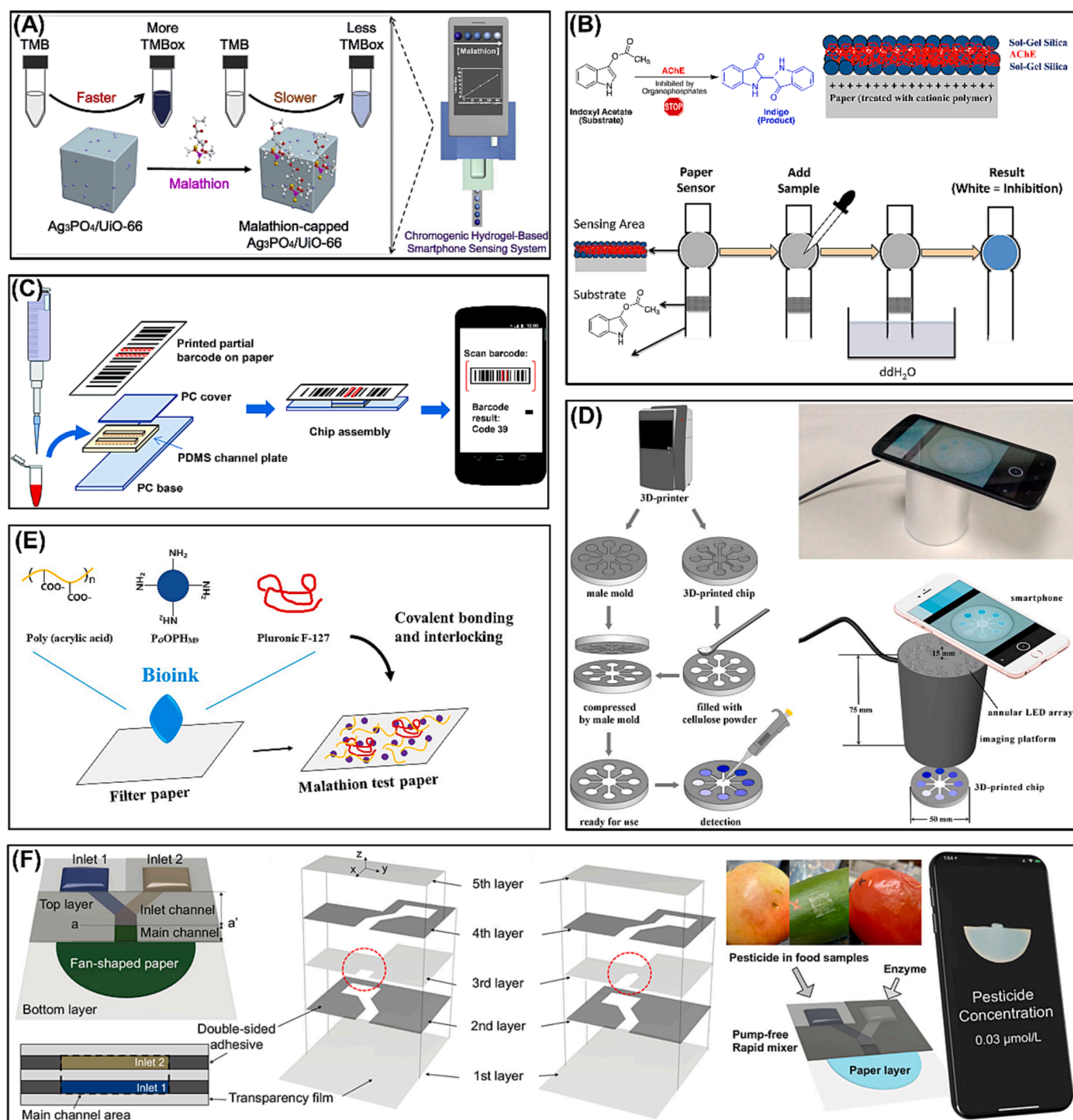


Fig. 1. Colorimetric sensors equipped with smartphones for OP detection. (A) Rapid detection of malathion by $\text{Ag}_3\text{PO}_4/\text{UIO-66}$ nanozyme-based hydrogel spheres (P. Liu et al., 2021); (B) Smartphone-based μPAD for OP detection in water (Sicard et al., 2015); (C) Smartphone-readable barcode for methyl parathion (Guo et al., 2015); (D) 3D printed chip for OP detection (Wu & Wang, 2018); (E) Paper-based sensor for malathion (Li et al., 2021); (F) Pump-free microfluidic device for OP detection in food (Jang et al., 2020).

μPAD could generate a colorimetric signal associated with the concentration of OPs. Furthermore, the quantitative analysis of OPs was achieved by simultaneously capturing the μPAD images of the water sample and pure water sample and analysing the ratio between the different pixel values of the two μPAD images in the RGB colour model. The response range of this device was 1×10^{-8} to 1×10^{-6} M for malathion and parathion, and the LOD was 10 nM. Additionally, to achieve rapid mixing and stable flow of the sample in the paper-based sensor, Jang et al. designed a low-cost pump-free microfluidic device combining

laminar and paper-based channels for the detection of OPs in food (Jang et al., 2020). As shown in Fig. 1F, the overlapping inlets assembled by a composite transparent film enabled multiple fluids to simultaneously enter the downstream channels and rapidly mix. The outlet was integrated into a fan paper that not only acted as a capillary pump but also provided the reaction zone. More specifically, the food samples and AChE were mixed in the overlapping inlet channels and then reacted with the IDA on the fan paper. Although solid-phase carrier paper plays a crucial role in colorimetric detection, it still suffers from several

shortcomings, including the lack of clear hydrophobic boundaries and poor stability of the chromogenic signal (Jin et al., 2021). To improve the above problems, Wu et al. designed a 3D printed chip (Wu & Wang, 2018). As shown in Fig. 1D, the centre of the chip was a flower-shaped hollow network structure filled with cellulose powder which was used to avoid flow interruptions in the channels, these channels included one central zone, eight linear channels and eight detection zones. The quantitative detection of OPs was achieved by customized applications that analysed the chip images captured by a smartphone on a dedicated imaging platform. Notably, the sensing system could not only select the most effective colour channel for the signal analysis for different targets but also directly set the calibration equation. The system achieved malathion detection within a few minutes with an LOD of 51.9 ng/mL.

Ellman's reagent DTNB reacts with thiocholine (TCh) containing thiol groups (-SH) to produce yellow TNB; however, the presence of OPs inhibits the production of TNB. For example, Guo et al. (Guo et al., 2015) designed a smartphone-readable barcode for the qualitative detection of methyl parathion (Fig. 1C). The barcode selected for this research was Code 39, and the results were read by directly scanning the barcode through the QuickMark app on a smartphone. The barcode was scanned and showed a "+" when OPs were found and a "-" when OPs were not found or in low concentration. In contrast to the principle based on the inhibition of enzyme activity by OPs, Li et al. (Li et al., 2021) designed a paper-based sensor by immobilizing the malathion hydrolase *PoOPH_{M9}* on filter paper through an interlocking network of Pluronic F127 (PF127)-poly(acrylic acid)-*PoOPH_{M9}* conjugates (Fig. 1E). Specifically, mercaptan, which is the hydrolysed product of malathion, reacted with DTNB to produce a bright yellow or dark yellow colour, and the intensity of the colour increased as the concentration of malathion increased. The sensor enabled the malathion (0.1–50 mM) detection in less than 5 min. Notably, this sensor achieved OP detection on site by direct immersion in water samples or by wiping food surfaces with it.

Moreover, several colorimetric strategies can produce detectable colour changes without the involvement of chromogenic agents. Wei et al. used nanoceria to directly catalyse the hydrolysis of OPs to bright yellow *para*-nitrophenol (p-NP) with colour intensity proportional to the OP concentration (Wei et al., 2019). Their research results showed a favourable linear relationship between colour intensity and methyl parathion concentration in the range of 2.1 to 21 μ M ($R^2 = 0.9942$) and detection limits of 0.42 μ M for both the smartphone-based colorimetric and spectrophotometric methods; thus, this sensing strategy has promising practicality.

In general, smartphone-based colorimetric strategies have demonstrated unique advantages in OPs analysis, such as user-friendliness, low cost, practicality and naked-eye detectability. Furthermore, the targets were identified based on colour changes even under relatively low concentration conditions. However, their detection sensitivity and accuracy were prone to interference by the background colour of the samples. Therefore, the selectivity of the chromogenic reaction and the effectiveness of the colour signal were significant factors in improving the colorimetric detection accuracy of smartphones (Kumaran et al., 2022). In addition, some measures were needed to obtain a more reliable response when constructing a colorimetric sensing strategy. For example, the interference of extraneous light sources during image capturing could be reduced by integrating the smartphone with suitable light sources and external accessories (shade boxes, light boxes, etc.). Additionally, colour calibration (white balance, luminance correction, saturation correction, etc.) could be used to reduce image errors caused by the lighting conditions, shooting angle/distance, shooting parameter settings and different brands of smartphones (Huang et al., 2018; Kap et al., 2021). Moreover, a suitable colour model for image analysis according to different needs is an essential way to improve the accuracy and efficiency of detection (Jliden et al., 2020).

2.2. Fluorescent sensors equipped with smartphones

The fluorescent sensing strategy enables analysis by direct or indirect detection of the fluorescence signal of the target analyte. Compared to colorimetric strategies, fluorescent strategies are more likely to achieve high specificity and sensitivity, thus, they are a more attractive option when the amount of target analytes is too small or too complex (Umapathi et al., 2022b; Xie et al., 2021). In fluorescence detection, an external light source (e.g., LED or laser diode) is usually required to excite the fluorescence to be detected. Moreover, analyte detection can be indirectly achieved by introducing a fluorescent material when the analyte is not fluorescent. Commonly used fluorescent materials include fluorescent dyes, semiconductor nanomaterials, rare earth materials, metal nanomaterials and carbon nanomaterials (Gong et al., 2022; Kalyani et al., 2020). In addition, to obtain high detection accuracy and low background signals, housings, filters and lenses are commonly integrated (Di Nonno & Ulber, 2021). Furthermore, the design of the optical path is essential to improve the detection sensitivity in smartphone-based fluorescence strategies (Ding et al., 2019). The fluorescence response mechanisms commonly used include fluorescence resonance energy transfer (FRET), photoinduced electron transfer (PET), the inner filter effect (IFE), aggregation-caused quenching (ACQ) and aggregation-induced emission (AIE) (Shen et al., 2022). FRET is usually accomplished via two fluorescent molecules. When the excitation spectra of the donor fluorescent molecule and the acceptor fluorescent molecule overlap, the donor fluorescent molecule induces the acceptor fluorescent molecule to fluoresce, while the fluorescence intensity of the donor fluorescent molecule decreases or is even quenched. During the PET process, electrons transfer from the highest occupied molecular orbital (HOMO) of the receptor to the energy-closed HOMO of the fluorophore. As the HOMO of the fluorophore is filled, the excited electrons in the lowest unoccupied molecular orbital (LUMO) cannot return to their HOMO upon photoexcitation, resulting in fluorescence quenching. IFE relies on the quencher to absorb the excitation or emission light of the fluorescent molecule, resulting in diminished fluorescence of the fluorescent molecule. ACQ refers to conventional fluorescent molecules whose fluorescence is diminished or quenched at high concentrations. However, fluorescent molecules with AIE properties can emit intense fluorescence when present as agglomerates or clusters (Chowdhury et al., 2018; Shen et al., 2022). The main response modes of fluorescence include turn-off (fluorescence quenching), turn-on (fluorescence enhancement) and ratiometric fluorescence (Li et al., 2022). The following is a detailed discussion and comparison of fluorescent sensors equipped with smartphones for OPs in food samples.

In turn-off fluorescent sensors, the enzyme-catalysed hydrolysis products can react specifically with quenchers, such as metal cations and nanomaterials (Yan et al., 2018), enabling fluorescence recovery. The catalytic activity of the enzyme is inhibited when OPs are present, leading to fluorescence quenching. For example, Chen et al. designed a fluorescent paper-based analysis device (FPAD) for OP detection (J. Chen et al., 2020). As shown in Fig. 2A, AIE nanoparticles (PTDNPs-0.10) adsorbed onto the surface of two-dimensional MnO_2 nanoparticles (2D-MnNFs) due to the electrostatic attraction, forming composite PTDNP-MnNFs and quenching the blue-green fluorescence of PTDNPs-0.10 through FRET. Furthermore, the AChE-catalysed hydrolysis of ATCh produced TCh that could reduce MnO_2 to Mn^{2+} , thus blocking FRET, and the brightness of FPAD was enhanced with increasing AChE doses. Nevertheless, due to the irreversible inhibition of AChE by OPs, the brightness of FPAD gradually diminished with increasing OP concentration. The detection limit of this sensor for paraoxon in real samples (kale extract, etc.) was 0.73 ng/mL. Furthermore, Wei et al. developed a hydrogel sensor loaded with GSH-gold nanoclusters (Au NCs)@ZIF-8 for the sensitive and selective detection of Cu^{2+} and OPs (Fig. 2C) (Wei et al., 2023). Specifically, TCh could inhibit the collapse of ZIF-8 through sulfhydryl group (-SH) coordination with Cu^{2+} , enabling the fluorescence of Au NCs to recover. Nevertheless, TCh

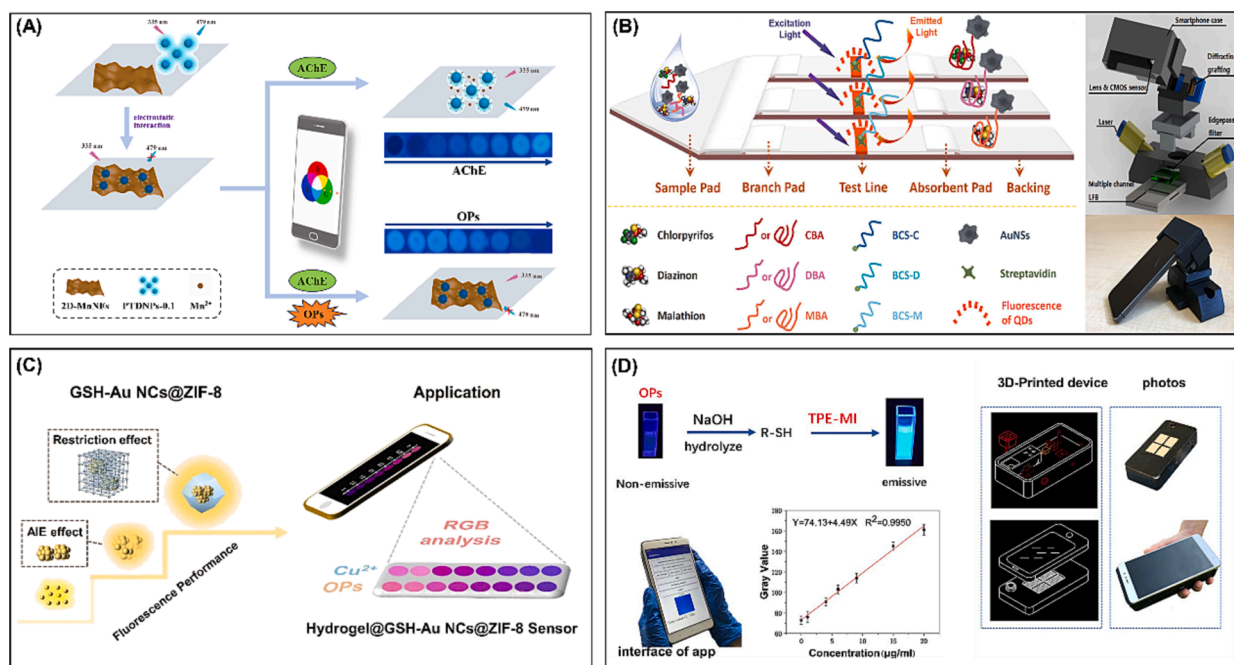


Fig. 2. Turn-off and turn-on fluorescent sensors equipped with smartphones for OP detection. (A) FPAD for OP detection (J. Chen et al., 2020); (B) Aptamer-based three-channel lateral flow biosensor and smartphone fluorescence spectrum reader (Cheng et al., 2018); (C) GSH-Au NC@ZIF-8 based hydrogel sensor for Cu^{2+} and GLP (Wei et al., 2023); (D) OP detection by TPE-MI (Jiao et al., 2021).

generation was inhibited when OPs were present. Moreover, the RGB values demonstrated a good linear relationship with the concentration of Cu^{2+} or glyphosate (GLP) in the range of 0 to 5 μM , with LODs of 0.05 μM and 0.93 nM for Cu^{2+} and GLP, respectively.

In contrast to the turn-off fluorescent sensors, the intensity of fluorescence can be recovered or significantly enhanced when OPs are present in turn-on fluorescent sensors. For instance, Liu et al. designed a paper-based sensor using CuS nanoparticles (CuS_{NPs}) and Amplex red (AR) for DDVP detection under enzyme-free and hydrogen peroxide-free conditions (Liu et al., 2020). Specifically, the synergistic function of CuS_{NPs} and DDVP caused AR to be oxidized to resorufin, emitting red fluorescence. The sensing system responded to DDVP in the range of 0.005 to 0.04 $\mu\text{g}/\text{mL}$ with a detection limit of 4.1 ng/mL. Notably, although the strategy provided high detection accuracy, the incubation time for DDVP detection was long (2.5 h); therefore, this needs to be considered when it is used for on-site detection. In addition, to achieve the simultaneous detection of multiple pesticides on site, Cheng et al. designed a fluorescent aptamer-based three-channel lateral flow biosensor (apta-LFB) integrated with a smartphone spectrum reader (Cheng et al., 2018). As shown in Fig. 2B, the aptamers were used to alternately recognize elements in the LFB, and quantum dot nanobeads and gold nanoparticles (AuNPs) were used as fluorescent emitters and quenchers, respectively. When the AuNPs-aptamers were mixed with the sample containing the target, the AuNPs-aptamers were not captured by the complementary sequences (BCS-C, BCS-D, BCS-M); thus, no quenching effect of AuNPs was observed, and the normal fluorescence emission of the quantum dot nanobeads on the test line occurred. Furthermore, the 3D printed reader enabled zero background readings and measurement of the full emission spectrum on the apta-LFB. This sensing system was optimized to achieve detection limits of 0.73 ng/mL for chlorpyrifos, 6.7 ng/mL for diazinon and 0.74 ng/mL for malathion. Moreover, Jiao et al. demonstrated that organophosphorus compounds containing P—S—C bonds (e.g., dimethoate, malathion, demeton, phorate, disulfoton) were prone to hydrolysis to thiols under alkaline conditions (Jiao et al., 2021). Therefore, the concentration level of OPs was assessed by detecting thiols. As shown in Fig. 2D, the AIE luminogen maleimide-functionalized tetraphenylethylene (TPE-MI) could

selectively react with the thiols to produce a fluorescent thiolation product (TPE-BSP). The fluorescence images were captured via smartphone on a dedicated 3D printing device, which in turn enabled the quantitative detection of OPs. The amounts of dimethoate in peas and celery were 0.47 and 0.62 $\mu\text{g}/\text{mL}$, respectively, and the amount of methomyl in cabbage was 0.32 $\mu\text{g}/\text{mL}$. These results were generally consistent with the LC-MS/MS analysis.

However, in the process of using a single-emission fluorescence sensing system to detect a target analyte, the fluorescence signal is susceptible to extraneous factors, such as environmental fluctuations, probe photobleaching, excitation light source fluctuations, sample matrix, and instrument drift (Sun et al., 2020; Y. Zhang et al., 2021). Ratiometric fluorescent sensors with two or more emissions provide a self-calibration function that effectively eliminates interfering signals, thus improving the specificity, accuracy and reproducibility of fluorescence strategies (Fahimi-Kashani & Hormozi-Nezhad, 2020; Liu et al., 2022; Luo et al., 2022). Ratiometric fluorescence-based sensing strategies commonly integrate two fluorophores into a single matrix, and the ratio of the two fluorescence intensities is used as the response of the target analyte (Shen et al., 2022; Zhang et al., 2023). Moreover, the reported smartphone-based ratiometric fluorescent strategies for OP detection have been mainly divided into two types: the single-signal change type and the dual-signal reverse change type.

In the single-signal change model, two fluorescent signals act independently; one signal shows a specific response to the target and serves as a sensing signal, while the other fluorescent signal is insensitive to the target and used as a reference signal. For example, Huang et al. prepared a hydrogel kit for DDVP detection in tap water and food (Huang et al., 2021). As shown in Fig. 3A, the fluorescence of carbon dots (CDs) as a reference signal remained constant, while the presence of DDVP led to the Cu^{2+} release, this resulted in the quenching of the fluorescence of the CdTe quantum dots (QDs) by Cu^{2+} through electron transfer. The sensing system responded to DDVP in the range of 1 to 40 ppb with a detection limit of 0.38 ppb and a detection time of 50 min. Similarly, a shorter detection time sensing strategy was proposed by Fahimi-Kashani et al, who combined CDs and cetyltrimethylammonium bromide (CTAB)-CdTe QDs to prepare an enzyme-free ratiometric fluorescent

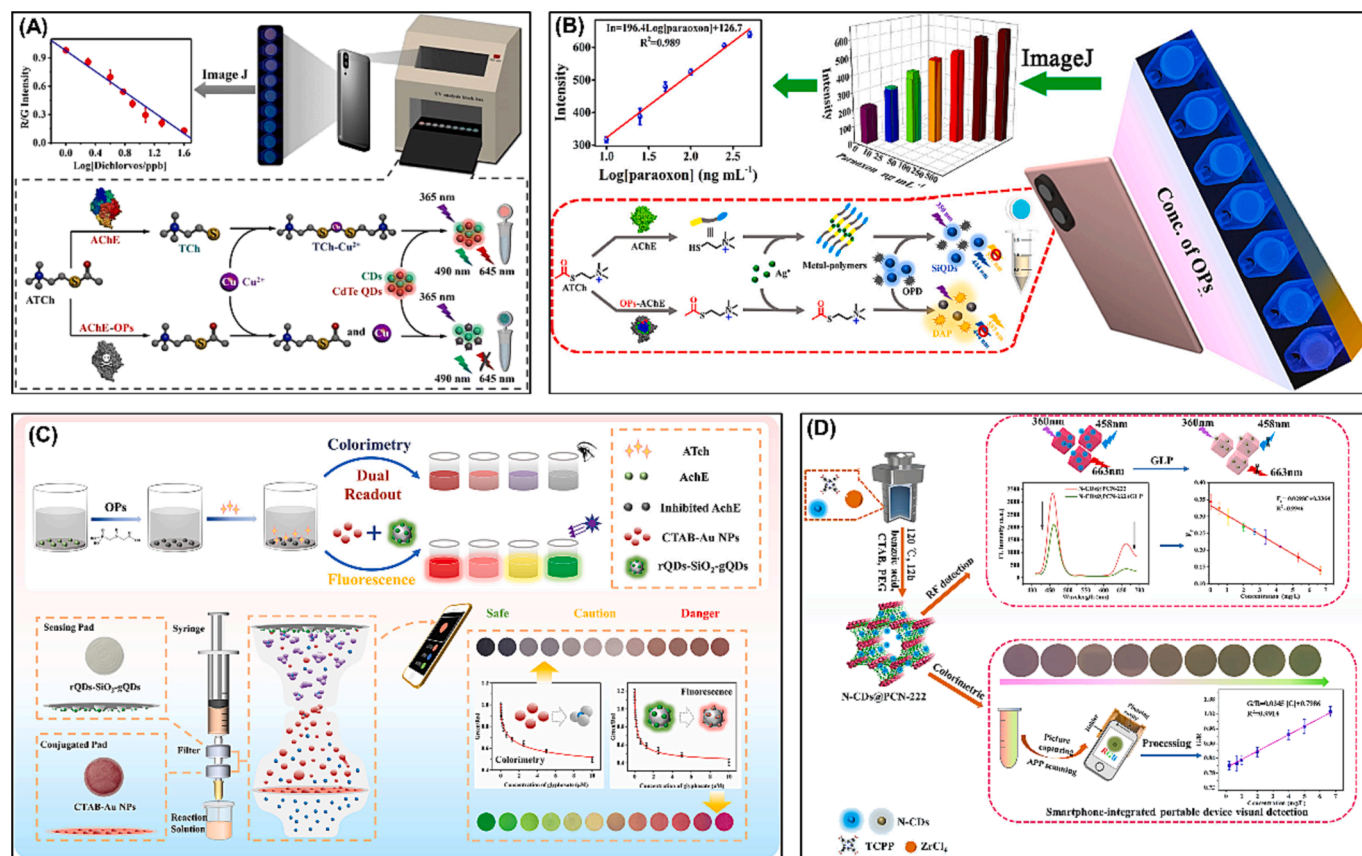


Fig. 3. (A) Target-responsive hydrogels based on CDs and CdTe QDs for DDVP detection in tap water and food (Huang et al., 2021); (B) Ag⁺-responsive hydrogels based on OPDs and SiQDs for paraoxon detection in pear juice (Jin et al., 2019). (C) Portable syringe equipped with smartphones for OP detection (Wei et al., 2021). (D) Colorimetric and fluorescent dual-mode sensing strategy based on N-CDs@PCN-222 for the detection of GLPs in water samples and cereals (Luo et al., 2022).

probe for methyl parathion (MP) detection in rice and tap water (Fahimi-Kashani & Hormozi-Nezhad, 2020). Specifically, the presence of MP induced CTAB to self-assemble on the surface of CdTe QDs, causing the fluorescence of CdTe QDs to be quenched, while the fluorescence of CDs remained constant. The detection limit was 0.06 µg/mL and the detection time was only 10 min.

In the dual-signal reverse change mode, the two fluorescent signals are correlated with each other and demonstrate sensitive and opposite changes to the target. One of the fluorescence signals shows fluorescence enhancement, while the other fluorescence signal exhibits fluorescence weakening. For example, Jin et al. integrated silicon quantum dots (SiQDs) and the organic molecule o-phenylenediamine (OPD) to design a Ag⁺-responsive hydrogel for paraoxon detection (Jin et al., 2019). As shown in Fig. 3B, the presence of OPs prevented the coordination of Ag⁺ to TCh, enabling the free Ag⁺ to oxidize the OPD to yellow DAP, which emitted yellow-green fluorescence, while DAP quenched the fluorescence of SiQDs through IFE. There was a favourable linear relationship (R² = 0.989) between the paraoxon concentration and the hue intensity, and the detection limit of this sensing system was less than 10 ng/mL.

In general, smartphone-based fluorescence sensing strategies feature high sensitivity, accuracy, specificity and ease of operation. Among them, the turn-off fluorescence strategy exhibits a higher background signal compared to the turn-on strategy because the turn-off fluorescence strategy needs at least one initial fluorescence reference signal (Gong et al., 2022; Shen et al., 2022; Y. Zhang et al., 2021). Furthermore, the single fluorescence signal sensing strategy tends to be susceptible to target-independent factors, such as changes in detection temperature, probe photobleaching, background light scattering, excitation light source fluctuation and instrument drift. The ratiometric fluorescent strategy with self-calibration can effectively eliminate false-

positive signals, especially in complex matrices, and can further improve the repeatability, specificity and reliability of the fluorescence sensing strategy. However, similar to turn-off fluorescence strategies, ratiometric fluorescent strategies also require at least one initial fluorescent reference signal, which may result in inferior detection sensitivity compared to low-background turn-on fluorescence sensing strategies (Shen et al., 2022). Notably, the dual-emission turn-on ratiometric fluorescent strategy reported in several studies is expected to achieve low background noise and high detection sensitivity (Geng et al., 2018; Shen et al., 2019). In addition, indispensable accessories, such as excitation light sources, filters and housings used to block interfering light sources cause difficulty in the integration of fluorescent systems with smartphones. At present, smartphone-based fluorescent strategies are still in the early stage for OP detection, and it is anticipated that with the development of materials science, nanotechnology and instrument manufacturing, smartphone-based fluorescence strategies will attract more attention in the field of organophosphorus detection.

2.3. Colorimetric and fluorescent dual-mode sensors equipped with smartphones

Similar to the ratiometric fluorescent strategy, the dual-mode analysis strategy also provides self-calibration that can reduce or even avoid the influence of the detection environment, thus facilitating a higher detection accuracy than the single analysis mode. Currently, the smartphone-based dual-mode optical strategies used for OPs mainly rely on colorimetric and fluorescent signals. For example, Cai et al. packaged AuNCs into ZIF-8 and designed a dual-mode biosensor for OP detection in food and water samples (Cai et al., 2021). Specifically, both the disruption of the ZIF-8 structures and the chromogenic reaction of TMB

were inhibited when OPs were present, which in turn caused significant fluorescence changes and colour changes. The detection limits of this sensing system for fluorescent and colorimetric detection of OPs were $0.67 \mu\text{g/L}$ and $0.4 \mu\text{g/L}$, respectively.

Pei et al. broadened the application of Ti_3C_2 MXene quantum dots (Ti_3C_2 MQDs) in the field of sensing. They prepared Ti_3C_2 MQDs by a fluorine-free method for OP detection (Pei et al., 2021). Specifically, the IFE between Ti_3C_2 MQDs and TNB resulted in the fluorescence quenching of Ti_3C_2 MQDs. However, as the OP concentration increased, the production of TNB was inhibited, which led to the recovery of the fluorescence, moreover, the colour of the reaction system changed from yellow to colourless. The fluorescence and colorimetric detection limits of the system were $0.20 \mu\text{g/L}$ and $0.01 \mu\text{g/L}$, respectively. However, the incubation time for this sensing strategy was too long and should be optimized and improved in the future.

Furthermore, in addition to combining the colorimetric signal with a single fluorescent signal, integrating the colorimetric signal with the ratiometric fluorescent signal is a more reliable and accurate option. For instance, Wei et al. (Wei et al., 2021) designed a portable syringe for OP detection (Fig. 3C). Specifically, the hydrolysis product of ATCh, TCh, caused the aggregation of CTAB-encapsulated gold nanoparticles (CTAB-Au NPs); this resulted in a colour change from red to grey. Moreover, the aggregated CTAB-Au NPs moved upwards and bound to the rQDs@ SiO_2 @gQDs, which reduced the FRET effect between CTAB-Au NPs and gQDs; thus, the green fluorescence was recovered; additionally, the red fluorescence of rQDs@ SiO_2 @gQDs remained constant. The production of TCh was inhibited when OPs were present in the reaction solution, and the colour change of CTAB-Au NPs and the fluorescence change of QDs@ SiO_2 @gQDs were used to quantify the OPs. The response range of the sensing system was 0 to $10 \mu\text{M}$ for glyphosate, and the LODs were 2.81 nM and 3.31 nM for the fluorescence and colorimetric strategies, respectively. Notably, this is the first portable

device proposed using a dual-mode strategy to detect OPs.

In contrast to the previously proposed ratiometric fluorescent strategy, Luo et al. (Luo et al., 2022) combined a colorimetric signal with dual-quenching ratiometric fluorescence for GLP detection in food samples (Fig. 3D). More specifically, the *N*-CDs@zirconium porphyrin metal-organic framework (PCN-222) formed compounds with GLP, and the fluorescence of PCN-222 was quenched through the PET process, while the occurrence of IFE quenched the fluorescence of *N*-CDs. Furthermore, the solution colour changed from pinkish purple to green upon the addition of GLP. The fluorescent and colorimetric detection limits of this strategy were $9.06 \mu\text{g/L}$ and $141.39 \mu\text{g/L}$, respectively.

2.4. Other optical sensors equipped with smartphones

In addition to colorimetric and fluorescent signals, some other optical signals, such as chemiluminescence (CL) signals, are also often used to develop optical sensors equipped with smartphones (Khalafazad-Kordasht et al., 2021; Li et al., 2020). The CL process refers to the excited intermediate formed after a chemical reaction of a luminescent substance returns to the ground state and emits photons to produce an optical signal (Q. Zhang et al., 2021). Compared to fluorescence-based analytical methods, CL-based strategies do not require the addition of an extra light source, which reduces the interference of scattering and decreases the background noise. Therefore, the CL sensors are ideal for the development of portable sensors equipped with smartphones.

Montali et al. (Montali et al., 2020) designed a foldable paper-based biosensor for AChE inhibitors (e.g., OPs, warfare agents, etc.) (Fig S1). Briefly, the sensor was integrated with AChE, ChOx and horseradish peroxidase (HRP). After the sensor was completely folded, the CL signal was obtained by adding a luminol solution to trigger the CL reaction catalysed via HRP. When OPs were present, the light emission was reduced. Moreover, the 3D-printed miniature box used to capture CL

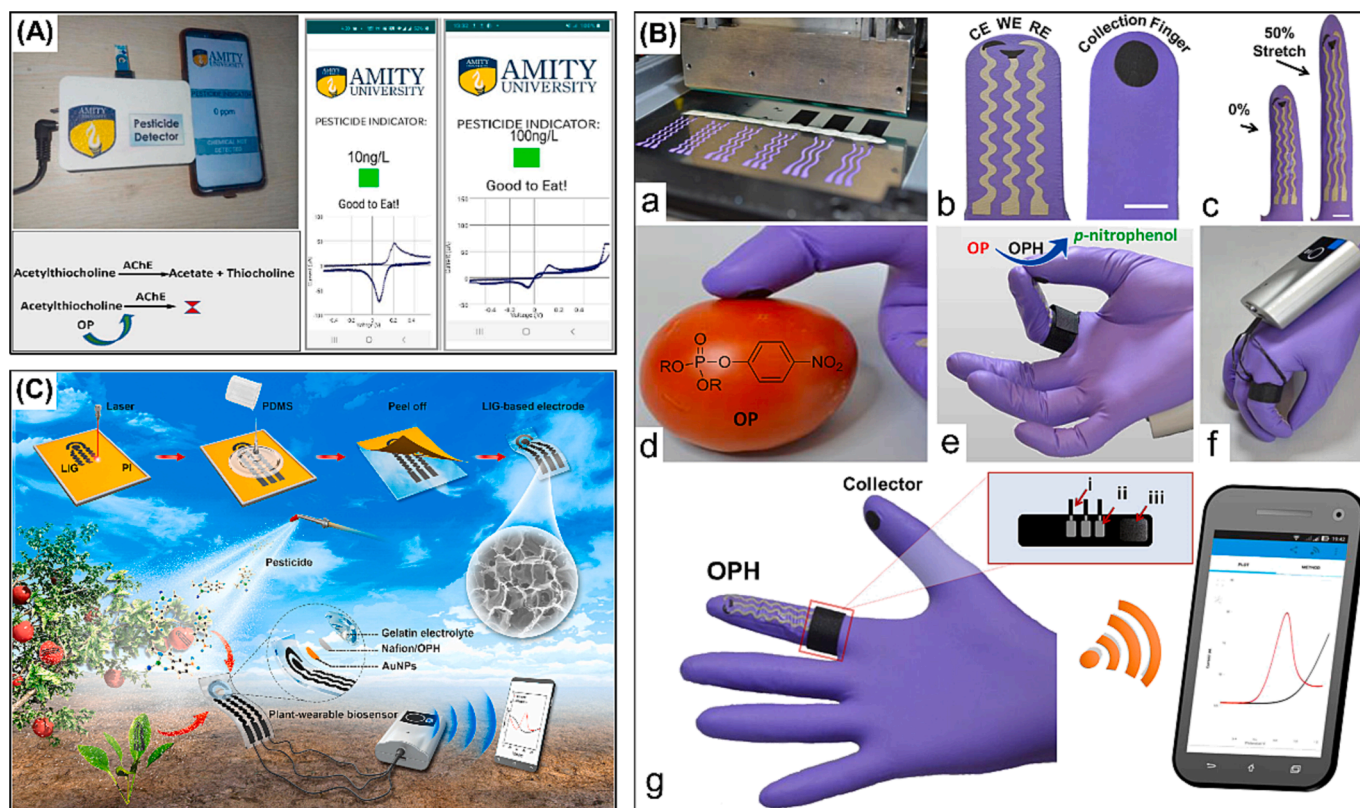


Fig. 4. Electrochemical sensors equipped with smartphones for OP detection. (A) OP bioprobe for rapid detection of OPs in vegetable extracts (Chansi et al., 2022; He et al., 2023); (B) Flexible glove-based electrochemical biosensor for rapid on-site detection of OPs (Mishra et al., 2017); (C) Plant wearable electrochemical sensor for methyl parathion detection on crop surfaces (Zhao et al., 2020)).

images is shown in Fig. 4B. The LOD of this sensor for chlorpyrifos-methyl was 45.0 μM , and the total detection time was 25 min.

Optical sensors equipped with smartphones have shown promising applications in detecting OPs. Among them, colorimetric sensors exhibit the advantages of simple operation, naked-eye detectability, rapid response, and low cost; however, the detection sensitivity and accuracy are easily affected by the background signal. These sensors can be used as auxiliary analysis tools for preliminary detection of OPs. Compared with colorimetric strategies, fluorescence sensors provide higher sensitivity and are more applicable for the detection of OPs in complex environmental matrices. Nevertheless, fluorescence sensors need additional excitation light sources, and the lifetime of fluorescence and background noise impact the accuracy and stability of fluorescence sensors (Kumaran et al., 2022). In addition, the single optical sensing signals based on smartphones are susceptible to extraneous factors, such as the detection environment and instrument settings, causing false positives and unreliable data. Ratiometric fluorescent sensors and dual-mode sensors with self-calibration improve this limitation, can effectively eliminate the interfering signals and provide higher detection specificity and reliability. Furthermore, in comparison with the fluorescence strategy, the CL strategy is simpler, does not require the addition of an extra light source, and CL features a low background signal and high sensitivity. However, the luminescence intensity of CL is relatively low and often requires more sophisticated and sensitive detectors to improve the signal-to-noise ratio (Gong et al., 2022; M. Zhang et al., 2022). In addition, smartphones are mainly used to capture and process images in these optical strategies, and the processing algorithm of the analysis app in smartphones can potentially be affected by interfering light sources (Beduk et al., 2022). Moreover, with the development of 3D printing technology, the integration of smartphones with external optical accessories used to improve the capturing conditions can effectively improve the sensitivity, accuracy and repeatability of optical sensing sensors. Optical sensors equipped with smartphones are anticipated to play an important role in ensuring food safety in the future.

3. Electrochemical sensors equipped with smartphones for OP detection

Electrochemical detection tends to be more sensitive and selective than optical assays; however, conventional electrochemical sensing devices are expensive, bulky, rigid, and not conducive to the on-site detection of analytes. In contrast, portable electrochemical sensors have been developed that not only provide simple operation, short response time, low sample consumption and low detection limits but also have the advantage of being available for real-time monitoring and can respond to poor conditions in real time (Liu et al., 2016). Currently, electrochemical sensors have been widely used in various fields (Xu et al., 2022). In agriculture, electrochemical methods commonly used for pesticide residue detection mainly include potentiometry, impedance, amperometry, and voltammetry (cyclic voltammetry, square wave voltammetry and differential pulse voltammetry) (Kumaran et al., 2022; Lazarević-Pašti, 2023; Malvano et al., 2017). However, there are few

examples of electrochemical sensors equipped with smartphones for OP detection. In the present smartphone-based electrochemical sensing strategies used to detect OPs, smartphones are usually used as controllers, data receivers and processors, and displays. The portable potentiostat is a commonly used electronic device in electrochemical on-site analysis; it applies stimulation voltages or currents between electrodes, records the currents and voltages generated at the same electrodes and transmits these signals to the smartphone in real time via wired communication (USB interface, audio interface, etc.) or wireless communication (Bluetooth, NFC, etc.) for further quantitative analysis (Chansi et al., 2022; Mishra et al., 2017; Shi et al., 2018; Umapathi et al., 2022a; Zhao et al., 2020). In Table 2, information on the recent applications of electrochemical sensors equipped with smartphones for OP detection is summarized. The following is a discussion of recent research on the detection of OPs via electrochemical sensors equipped with smartphones, classified according to the electrochemical methods used to detect OPs.

Chronoamperometry (CA) quantifies the steady-state current as a function of time by applying a potential to the working electrode. During the application of a potential, electroactive analytes present in the electrolyte are oxidized or reduced, resulting in a current associated with the amount of the target (Pérez-Fernández et al., 2020). Shi et al. designed a dual-channel screen-printed electrode (DSPE) modified with Prussian blue (PB) and AChE; this electrode was combined with a smartphone-controlled potentiostat to achieve rapid OP detection (Shi et al., 2018). Specifically, when OPs were present on one of the channels, the inhibitory effect of OPs on AChE led to a decrease in the response current of that channel, while the current of the channel without OPs remained constant. The current signal on both channels was simultaneously measured using the potentiostat, and the enzyme inhibition rate was obtained by the amount of change in current. The sensing system was used for trichlorfon, oxamyl, and isocarbophos and showed a good linear relationship between the negative logarithm of the OP concentrations and enzyme inhibition ratios in the range of 1×10^{-7} to 5×10^{-6} g/mL with an LOD of 10^{-7} g/mL under the optimum experimental conditions.

Cyclic voltammetry (CV) is a continuous wave technique that obtains a response current distribution mainly by applying a potential between the working electrode and the reference electrode, causing the redox reaction of the electroactive substances on the electrode. In cyclic voltammograms, the peak current is proportional to the concentration of the target analyte (Umapathi et al., 2022a). Chansi et al. (Chansi et al., 2022) designed a bioprobe for OP detection in vegetable extracts (Fig. 4A). The bioprobe was based on the principle of inhibition of AChE by OPs, the current signal produced by TCh was recorded by the potentiostat, and the signal was output to the smartphone app in real time in the form of a CV response and OP concentration. The sensing system responded to OPs in the range of 10 to 100 ng/L with an LOD of 9.8 ng/L and showed stability for 85 days.

Square wave voltammetry (SWV) is a sensitive, large-amplitude differential technique based on the principle of applying a square-wave potential waveform to a working electrode; this causes a redox reaction of the electroactive substance at the electrode, and the current

Table 2
Application information of electrochemical sensors equipped with smartphones for OP detection.

Analytes	Matrix	Method	Electrode	Smartphone use	LOD	Linear range	Ref.
Trichlorfon Oxamyl Isocarbophos	Cowpea cabbage	CA	Dual-channel SPE	Controlling the detector	1.0×10^{-7} g/mL	1.0×10^{-7} – 5.0×10^{-6} g/mL	(Shi et al., 2018)
Chlorpyrifos	Vegetable	CV	Carbon-based SPE	Signal processing and display responses	9.8 ng/L	10–100 ng/L	(Chansi et al., 2022)
Methyl parathion Methyl paraoxon	Fruits and vegetables	SWV	The serpentine structures SPE	Displaying the results	/	/	(Mishra et al., 2017)
Methyl parathion	Apple and spinach	SWV	3D porous laser-induced graphene-based serpentine three-electrode	Visualizing results in real-time	0.01 μM	20–500 μM	(Zhao et al., 2020)

response of the electrode in the potential sweep cycle is then measured (Pérez-Fernández et al., 2020). The peak current profile obtained by SWV is proportional to the concentration of the analyte. Moreover, the background currents generated during the detection process can be effectively suppressed using SWV; thus, the signal-to-noise ratio is enhanced, providing higher peak currents and lower detection limits (Umapathi et al., 2022a).

Mishra et al (Mishra et al., 2017) developed a wearable electrochemical biosensor for OP detection. As shown in Fig. 4B, the design of the serpentine structure and the use of pressure-enduring inks enabled the glove to provide excellent resilience to mechanical deformation. Moreover, the thumb with a printed carbon pad was used for collecting samples, and the index finger with an electrode containing immobilized organophosphate hydrolase (OPH) was used for detection. Furthermore, the potentiostat on the back of the hand was used to record SWV signals and wirelessly communicate with a smartphone for rapid voltammetric results. Methyl parathion and methyl paraoxon on the surface of foods were detected and their relative standard deviations (RSDs) were 2.10% and 3.05%, respectively, indicating that the sensor provided promising practicality and reproducibility. Based on wearable sensors, Zhao et al. designed a plant-wearable electrochemical sensor for the in situ detection of OPs (Zhao et al., 2020). As shown in Fig. 4C, the electrode system was integrated with OPH, gelatine electrolyte, and AuNPs and could maintain a favourable electrochemical response on irregular crop surfaces to selectively capture and detect OPs in real time. The sensing system responded to methyl parathion in the range of 20 to 500 μM with an LOD of 0.01 μM . Notably, these flexible and wearable electrochemical sensors featured noninvasiveness, low cost, simple operation, rapid response, and in situ recognition and avoided a tedious sample pre-treatment process. Furthermore, the development of this sensor solved the limitations (e.g., rigidity and bulkiness) of traditional electrochemical devices, providing great application prospects for the portable detection of pesticides in food safety.

Among the reported smartphone-based electrochemical sensing strategies for detecting OPs, voltammetry has received more attention due to its advantages of rapid response, high sensitivity and simultaneous detection of multiple targets. In particular, SWV provides higher sensitivity, selectivity and lower background current than CV (Pérez-Fernández et al., 2020), and it is one of the most commonly used sensing strategies in voltammetry. In addition, compared to voltammetry, CA is a strategy with simple operation, low hardware requirements and high applicability. The current integration interval of CA is longer, leading to a higher signal-to-noise ratio. Moreover, both voltammetry and CA can be effectively applied for real-time monitoring.

Smartphones have been extensively integrated with portable electrochemical sensors for portable detection, analysis and screening of various targets (Umapathi et al., 2022a). Overall, electrochemical sensors equipped with smartphones provide the advantages of low cost, short response time, high accuracy and sensitivity, simple and easy sample pretreatment and preparation, easy integration with electrochemical devices, and real-time information acquisition of target analytes. In addition, the detection performance of electrochemical sensors relies heavily on the recognition element, and the combination of advanced materials and nanomaterials with unique structures and high conductivity with electrodes can enhance the electron transfer and electrochemical signal strength in the sensing device (Du et al., 2021). Enzymes, such as AChE and OPH, provide excellent selectivity and reproducibility and are commonly used as biorecognition elements in electrochemical sensors for the detection of OPs. Nevertheless, overloaded enzymes can block electron transfer at the electrode (Mishra et al., 2017; Shi et al., 2018; Umapathi et al., 2022a; Zhao et al., 2020). Moreover, in recent years, with the development of 3D printing and screen-printing technologies and depending on the different analytical purposes and carrier materials, various types of portable electrode systems (paper-based, plastic, gloves, etc.) have been exploited (Lu et al., 2019; Mishra et al., 2017), thus enabling the miniaturization and

portability of electrochemical sensors. However, there are still relatively few studies on electrochemical sensors equipped with smartphones for OP detection compared to optical sensors equipped with smartphones. Therefore, several challenges, such as the compatibility of smartphones with potentiostats, interference between metals, oxidation of samples, high background currents and stability of electrodes, remain to be addressed through further studies to utilize the enhanced performance of electrochemical sensors (Kumaran et al., 2022; Umapathi et al., 2022a; Yan et al., 2022).

4. Conclusions, challenges and perspectives

OP residues in food pose a severe risk to public health. Optical and electrochemical sensors equipped with smartphones provide great potential for addressing the demand for rapid OP detection due to their efficient sensing performance and low cost. This review focuses on the recent research progress in smartphone-based colorimetric, fluorescent, chemiluminescent and electrochemical sensing technologies for OP residue detection in food samples, the sensing principles and methods are detailed, and their advantages and disadvantages are compared.

In general, the popularity of smartphones improves the application value of organophosphorus sensors and effectively reduces the cost; furthermore, nonprofessionals or consumers can easily operate these portable sensors. Optical sensors equipped with smartphones provide the advantages of easy operation, naked-eye detectability, low cost and practicality for OP detection. In particular, ratiometric fluorescent sensors and the dual-mode sensors with self-calibration functions can effectively solve the problems of false positives and unreliable detection data; these problems are caused by the single optical sensing signal being easily affected by extraneous factors, such as the detection environment and instrument settings. The integration of optical sensors with advanced materials, nanomaterials and external optical accessories can effectively improve the overall performance of optical sensors equipped with smartphones. In addition, electrochemical sensors equipped with smartphones have a wide range of analyses, low sample volume requirements, simple sample preparation, and the advantage of real-time monitoring (Kumaran et al., 2022). Moreover, in contrast to the smartphone-based optical sensing strategies, the smartphone-based electrochemical sensing strategies are largely independent of smartphone functions, are less susceptible to smartphone configurations, matrix colours and environmental lighting conditions, and they further provide better quantification and interference resistance (Sun and Hall).

Despite the promising prospects of sensors equipped with smartphones for OP detection in food, the applicability and portability of current sensors in the field is still in its infancy, and the functions of smartphones are not presently fully utilized. For example, as a medium between the cloud and sensors, smartphones with wired and wireless communication functions are anticipated to achieve an internet-based detection method for pesticide residues, facilitating the traceability of pollution sources in food. In addition, due to their powerful communication and computing capabilities, smartphones are anticipated to become ideal tools for controlling, analysing and sharing information in the Internet of Things (IoT), showing great potential in facilitating the development of the IoT. In short, the smartphone will play a crucial role in future food analysis as an essential link between people, sensors and the internet.

To fully exploit and broaden the applications of sensors equipped with smartphones in food analysis, much research needs to be performed in the future; these include the following: (1) Developing novel, high-performance and highly stable recognition elements, chromogenic substrates, chemiluminescent materials and fluorescent materials to minimize nonspecific reactions and enhance the signal-to-noise ratio to improve the sensitivity, reliability and reproducibility of sensors equipped with smartphones. However, the high cost of preparation and environmental toxicity of nanomaterials and advanced materials need to be addressed in future food analysis applications. (2) Developing

smartphones equipped with higher pixel intensities and more powerful processors as well as more effective optical accessories to ensure the stability of optical image capturing conditions; the development of a unified and feasible image analysis app for optical sensors equipped with smartphones is needed to avoid biased output results due to the functional differences between different apps. (3) Designing sensors equipped with smartphones as high-throughput sensing devices that can detect a wide range of pesticides and interfering substances in food with low detection limits. In addition, automated sample pretreatment microsystems integrated with smartphones need to be developed to ensure more rapid pesticide residue analysis. Accurate automation of smartphone-based sensing devices and the development of smartphone applications that can reliably analyse sensing signals are both needed. (4) Developing sensors equipped with smartphones that further combine advanced technologies, such as big data, cloud computing, IoT and artificial intelligence, to better achieve simple, efficient, cost-effective, and real-time monitoring of pesticides in food. Moreover, future research needs to be devoted to verifying and optimizing the applicability and accuracy of these sensing strategies and regularly upgrading the sensing devices such that new detection devices for other hazardous chemicals in food can be developed.

With the continuous advancement of materials science, nanoscience, optical science, electronic science, artificial intelligence and instrument manufacturing, it is anticipated that these challenges will be effectively addressed and that the detection technology for OP residues in food will become more powerful and reliable. Due to the high penetration rate of smartphones, smartphone-based sensing strategies are expected to play a vital role in the future detection of pesticides, with each consumer having the opportunity to participate in ensuring food safety.

CRedit authorship contribution statement

Jiaqi Xue: Conceptualization, Writing – original draft, Writing – review & editing. **Kang Mao:** Conceptualization, Writing – review & editing, Funding acquisition, Supervision. **Haorui Cao:** Writing – review & editing. **Rida Feng:** Writing – review & editing. **Zhuo Chen:** Writing – review & editing. **Wei Du:** Writing – review & editing. **Hua Zhang:** Conceptualization, Writing – review & editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgment

The authors acknowledge the support from the National Natural Science Foundation of China (42107486, 42377456), Guizhou Provincial Science and Technology Projects (Qiankehe Jichu-ZK [2022] Yiban 565), Youth Cross Team Project of CAS (JCTD-2021-17), and the Youth Innovation Promotion Association CAS (2023415).

Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.foodchem.2023.137456>.

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