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Recent progress in the development of recognition bioelements for polychlorinated biphenyls detection: Antibodies and aptamers

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ABSTRACT

Polychlorinated biphenyls (PCBs) are persistent pollutants, which have expanded in foods and the environment. Detection of PCBs is considered essential due to recognized side-effects of PCBs on health and the public concerns in this regard. On the other hand, due to the trace levels of these organic chlorine compounds, reliable and sensitive assays must be developed. Recognition elements are essential parts of analytical detection assays and sensors of PCBs since these elements are involved in the selective identification of the analytes of interest. Understanding the fundamentals of the recognition elements of PCBs and the benefits of the sensor strategies result in the development of next-generation recognition devices. This review aimed to highlight the recent progress in the recognition elements as key parts of biosensors. We initially, focused on the developed antibody-based biosensors for the detection of PCBs, followed by discussing the aptamers as novel recognition elements. Furthermore, the recent advancement in the development of aptamer-based solid phase extractions has been evaluated. These findings could contribute to improving the design of commercial PCB-kits in the future.

1. Introduction

Polychlorinated biphenyls (PCBs) are infamous classes of man-made by-products of persistent organic pollutants (POPs), which are resistant to biological, chemical, and radiation degradation. PCBs threaten the ecosystem and food safety due to their high toxicity, long-term stability, poor degradation, and bioaccumulation [1–3]. Before 1977, PCBs were often used as coolants, heat carriers, insulating oils, and lubricant oils, as well as in electrical equipment in industries [4,5]. PCBs are colorless with no smell or taste, which were banned in industrial processes in 1977 due to their suspected adverse health and environmental effects associated with the exposure [6]. However, PCBs are still produced and applied in developing countries and also found in the products procured before 1979. PCBs are 209 homologs, and PCB77, PCB169, and PCB126 are coplanar and considered to be the most toxic dioxin-like PCBs [7]. PCBs could remain cycling between air, water, and soil for long periods of time. They bioaccumulate in fats, and their levels increase in the upper food chains. Food is the most important source of exposure to PCB. Some parts of these compounds enter the body through swallowing dust, while small parts enter the body through breathing the air contaminated with these compounds. The World Health Organization (WHO) and the Food and Agriculture Organization (FAO) authorize a daily PCB intake of 6 μg/kg per day [8].

PCBs have some adverse health effects, including carcinogenic, mutagenic, teratogenic, and immunotoxic effects [9,10]. PCBs have been classified as carcinogens by the International Agency for Research on Cancer (IARC) [11]. Evidence suggests that the most common type of PCB-induced malignant melanoma is a rare, severe form of skin and brain cancer [12,13]. Furthermore, PCBs are considered to be a serious threat to food safety and human health. Environmental Protection Agency (EPA) has regulated maximum contaminant level of 0.0005 ppm of PCBs in the drinking [14]. Food and Drug Administration (FDA) mandates the range of 0.2–3.0 ppm of PCBs for all foods and 2 ppm in fish. FDA limits PCBs in paper food-packaging to 10 ppm [15]. Sensitive and low-cost measuring systems for the monitoring of PCBs in food and environmental samples are essential to minimizing human health and environmental risks. Monitoring of dioxins and PCBs in human and animal foods is the most efficient technique to evaluate the risk of human and animal exposure. In addition, determining the level of pollution could lead to the reduction of pollution sources in the food chain, which in turn results in the identification and elimination of pollution sources. In general, the most frequently used techniques for PCB analysis are divided into three categories of chemical analysis, bioassay, and sensor technology. Suspected samples are often tested by gas chromatography, along with various other methods [16–18]. However, these methods are time-consuming, laborious, and costly,

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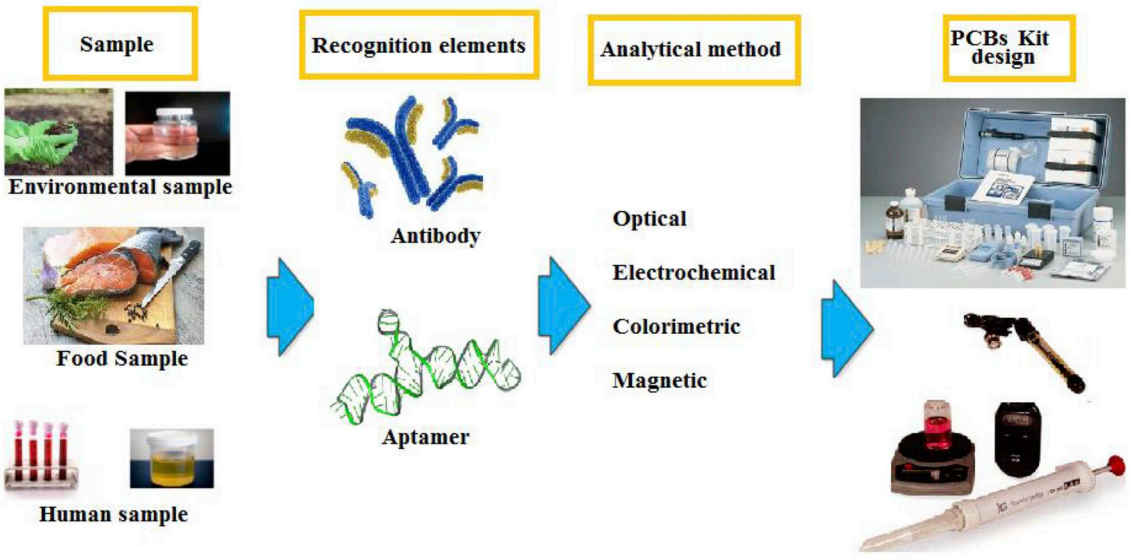

Scheme 1. Schematic illustration of recognition bioelements development for polychlorinated biphenyls detection.

while their other limitations include the complex pre-treatment process and need for specialized personnel. Therefore, it is vital to access simple, rapid, cost-effective, portable, and on-site screening systems for the evaluation of the risk of PCB exposure through food and environmental samples. The biosensor technology is considered promising in this regard, which could be an alternative solution to the mentioned problems. Application of these rapid techniques is expanding since they are easy to use and affordable, providing real-time, on-site inspection. Biosensors could be used to transform the biophysical changes in the binding target and recognition elements into selective and measurable signals [19–23]. The present review aimed to provide an overview of the most recent findings on antibody and aptamer-based biosensors for the detection of PCBs. Biosensors are composed of a bioprobe as the recognition element and a transducer. In addition, we have discussed two important categories of transducers (optical and electrochemical) and their potential applications in the screening of PCBs by antibodies and aptamers as the recognition elements, as well as their advantages and drawbacks (Scheme 1). Finally, the present study has assessed the application of aptamers as novel bioprobes in solid-phase extraction.

2. Antibodies as recognition probes for PCBs detection

Antibodies are the most well-known bioprobes used in various assays and techniques for different purposes, such as clinical diagnosis, food safety, environmental monitoring, and veterinary medicine [24,25]. Antibodies are produced by the immunization of mammals (e.g., mice, rabbits, and goats) against foreign substances (antigens), along with the use of antibodies as highly specific reagents to recognize the intended analyte [26]. The principal advantage of antibody-based methods is the specificity of detection, which is derived from the selective instinct of antibody-antigen binding interactions. With the development of biotechnology, several antibodies have been obtained to target various congeners of PCBs. Moreover, several antibody-based approaches, such as radioactive tracer (radioimmunoassay) [27], enzyme-linked immunosorbent assay (ELISA) [9,28–30], other multi-analyte immunoassays and immunosensors [31–33], have been utilized as alternatives to the routine techniques based on gas chromatography and mass spectroscopy for the analysis of dioxin-like compounds in food and environmental samples.

Among numerous immunoassay techniques, ELISA is most widely used in commercial kits. The commercially available PCB immunoassay kit has been validated by the U.S. Environmental Protection Agency (EPA) for particular purposes and used increasingly for regulatory objects [7]. Table 1 presents some of immunoassays commercial test kits for PCB determination and their specifications, as reported by the manufactures.

Within the past decade, remarkable efforts have been made to develop antibody-based biosensors. Immunosensors present one of the most useful applications of immunoassays in PCB detection. Immunosensors are based on the biological interactions between the antibodies and targets, followed by transduction, amplification, and detection. The transduction mechanism converts the biophysical interaction into detectable signals. The type of transduction defines the optical immunosensors and electrochemical immunosensors. The recent achievements regarding antibody-based immunosensors for PCB quantification have been presented in the following section, as well as the advantages and disadvantages of the investigated immunosensors.

2.1. Optical antibody-based PCB biosensors

Endo et al. [33] developed a simple micro-flow immunosensor chip for the measurement of coplanar-PCB (Co-PCB) using anti-Co-PCB monoclonal antibody through fluorescence detection. In this competitive ELISA assay, the Co-PCB antibodies have been immobilized on polystyrene beads and introduced into the flow channel. After the competitive reaction between the target and horseradish peroxidase (HRP), the conjugated antigen, hydrogen peroxide and fluorogenic substrate were injected into the microchip. As a result, a fluorescent dye was produced and monitored in terms of changes in the fluorescence intensity. The integration of ELISA into a biochip could reduce the consumption of the probes and samples, providing a rapid detection of approximately 30 s. This rapid, cost-efficient system could be practical as an automated system.

In recent years, there has been considerable interest in the development of surface plasmon resonance (SPR) immunosensors in different target detections [34,35]. SPR analysis has particular characteristics including sensitivity, selectivity, does not require tagging agents or carriers, portability, miniaturization, and on-site analysis [36,37]. Thus, SPR immunosensors have great promise for biosensors for a wide variety of analyte detections in complex matrices. In this fact, Shimomura et al. presented a novel IgG-modified biosensor surface for the rapid detection of PCBs by SPR analysis [38]. They used SPR sensor chips to the screening of three endocrine disrupting chemicals (EDCs); 2,3,7,8-TCDD, PCB and atrazine. With this method, it was possible to

Table 1
Some of the commercial kits for PCB detection.

Commercial Name	Manufacturer	Format	sample	LOD (ppm)	Action Levels (ppm)	Analysis Time (min)	Source
RaPID assay PCB Test kit	SDI	ELISA	water	0.0002[a]	0.0005–0.01[a]	–	www.sdix.com
Ensys PCB Soil	SDI	enzyme immunoassay	soil	0.5[a]	0.5–10[a]	–	www.sdix.com
HAH Pocket col II test kit (PCB Soil)	HACH	colorimeter/Immunoassay	soil	1[c]	1–50[c]	20	www.hach.com
L2000DX	Dexsil	electrochemical	s[d]: 2 w[e]: 0.01	s: 2–2000 w: 0.01–2000	10	www.dexsil.com
Clor-N-Oil	Dexsil	fixed endpoint colorimetric titration	transformer oil	20	20–500	5	www.dexsil.com
Clor-N-Soil kit	Dexsil	fixed endpoint colorimetric titration	soil	50	–	10	www.dexsil.com
Envirogard PCB test kit	Millipore, TM Inc.)	colorimeter/immunoassay	soil	0.5	0.5–500	30	www.modernwater.com
PCB ELISA- Abraxis Kits	Abraxis LLC	ELISA	water, soil, sediment, fish tissue	0.00025	0.00025–0.025[a]	45	www.abraxiskits.com

[b]PCB concentration expressed as Aroclors 1260, 1254, 1248, 1242, 1232 and 1016.
[a] PCB concentration expressed as Aroclor 1254.
[c] Concentration expressed as Aroclor 1248.
[d] Soil.
[e] Water.

detect three kinds of EDCs at a ppb (part-per-billion) level within 15 min. Additionally, these biosensor requires small sample volume (20 µl) and it is cost-effective. On the other hand, Bender et al. [39] applied a direct electrochemical immunosensor for the detection of PCBs. The biosensing system was developed through the immobilization of the anti-PCB antibody on a conducting pyrrole polymer electrode. The interaction between the PCB and anti-PCB antibody produced a measurable current. The electrochemical immunosensor was used for the detection of PCBs in environmental water and soil samples.

2.2. Electrochemical antibody-based PCB biosensors

Electrochemical biosensors are promising in terms of the point-of-care detection due to the ability to miniaturize for portable application [40]. For instance, Laschi et al. [41], have developed an immunosensor based on screen-printed, disposable electrodes for the identification of PCBs. In this amperometric platform, HRP conjugated with the antibody was used as the enzyme label, and ferrocenemonocarboxylic acid (FCA) was employed as an electron mediator. A competitive reaction between a mixture of polychlorinated biphenyls (PCBs) and the optimization concentration of an enzyme-labeled PCB led to the reduction of the cathodic current with increasing concentration of the target (Fig. 1). Additionally, Laschi et al. established other similar disposable electrochemical immunosensor for the detection of PCBs [42]. In this competitive assay, alkaline phosphatase was used as the enzyme label, along with differential pulse voltammetry (DPV) assay (Fig. 1).

Magnetic separation is a practical technique that is widely used in sensing platforms owing to its easy manipulation and remarkable convenience in coupling with detection methods [43–45]. In this regard, Centi et al. [46] introduced a disposable immunomagnetic electrochemical sensor for the detection of PCBs based on antibody-coated magnetic beads (MBs) similar to the design proposed by Silva et al. [47]. In this biosensing approach, magnetic beads were used to optimize the immunoassay and enhance the sensitivity of the system. The biosensing system was fabricated through the immobilization of the antibody onto the surface of the MBs in order to construct the capture probe. After magnetic separation, a direct, competitive immunoassay was observed between the analyte and PCB-labeled alkaline phosphatase (AP). Following that, the beads solution from the second step of the magnetic separation was dropped onto the working electrode and incubated using an enzymatic substrate (Fig. 2). The performance of the immunosensor was considered successful on real samples with DPV. In addition, the researchers compared the electrochemical magneto-immunosensor (EMI) using an electrochemical immunosensor (EI) for the detection of PCBs in terms of sensitivity, reproducibility, and response frequency [48]. The obtained results confirmed the improvement of the sensing performances of the immunosensor based on the EMI strategy. The use of magnetic beads in EMI strategy can greatly improve the performance of the antibody-based biosensor, owing to the increase of the surface area, also the faster kinetics of reaction achieved due to the beads are in suspension so the analyte cannot migrate very far [49].

In another research, Centi et al. [50] developed other similar disposable, low-density, array-based techniques on the competitive assay and DPV signals. The modified MBs were captured on the surface of the graphite, screen-printed, low-density electrode, leading to the development of the immunosensor combined with solid-phase extraction, which were used for the analysis of PCBs in milk samples. Similarly, Lin et al. [51] designed an electrochemical immunosensor to detect PCBs using the sensing scheme similar to the one employed by Centi [46] and Silva [47]. The scheme of this biosensor was based on a direct, competitive ELISA using PCB antibody-coated MBs and HRP-labeled PCB (RP–PCB). The performance of the biosensor was validated using the commercial PCB-ELISA kit.

Electrochemical impedance spectroscopy (EIS) is considered to be an effective, non-destructive electrochemical technique since the inverse correlation of the impedance with the current enables the

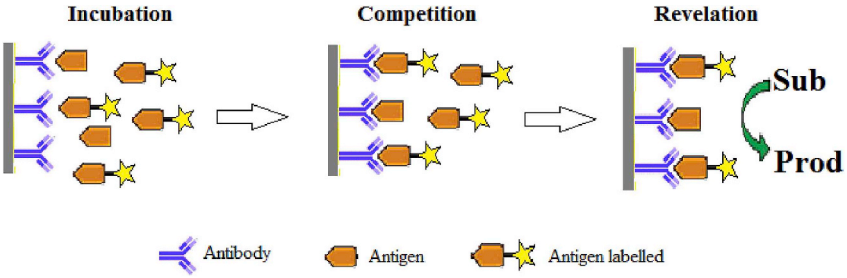

Fig. 1. Scheme of the three steps of the electrochemical immunosensor mechanism. The working electrode was modified by the immobilization of antibodies against PCBs, and a competition was generated between the free antigen (PCBs) and the same antigen labeled with an enzyme. An affinity complex antigen-antibody is formed during the incubation, and the concentration of the analyte is determined by electrochemical measurement of a redox substrate which produced by the reaction with the enzyme-label. Reprinted with permission [42].

measurement of the low concentration of the target with high precision [52,53]. Moreover, Date et al. [54] designed a label-free immunosensor using the EIS method to detect highly sensitive PCBs. In this biosensor, insulating oil samples were prepared through a simple cleanup procedure. A micro gold electrode was modified using the novel monoclonal antibody (RU6F9) for the identification of PCBs. The binding of PCBs to antibodies on the surface of the electrode altered the charge transfer resistance and was recorded using electrochemical impedance spectroscopy.

Khesuoe and colleagues [31] fabricated an improved electrochemical immunosensor for PCB detection using a modified working electrode composed of polyaniline (PANI) doped by silver nanoparticles (Ag-NPs). The polyclonal anti-PCB antibody (Ab) was immobilized on the Ag NPs-doped PANI, modifying the GCE (GC/PANI/Ag NPs) surface through a covalent linkage of glutaraldehyde with Ab, which reinforced the stability of the biosensor. Moreover, the capturing of PCB by Ab resulted in an electrochemical peak, which was recorded as square wave voltammetry signals. This biosensor could also detect PCB contamination in tap water and guava juice samples. A summary of the reports on PCB immunosensors based on their specifications are listed in Table 2.

In summary, antibodies are the most applicable recognition elements in commercial kits. Although antibodies are extremely sensitive and selective, they have some limitations in terms of stability and production time. Moreover, the screening of antibodies is costly and time-consuming, which requires biological organisms. The limitations of these bioreceptors in analytes detection have stimulated extensive research in order to achieve persistent, economic alternative bioprobes (e.g., aptamers) [55].

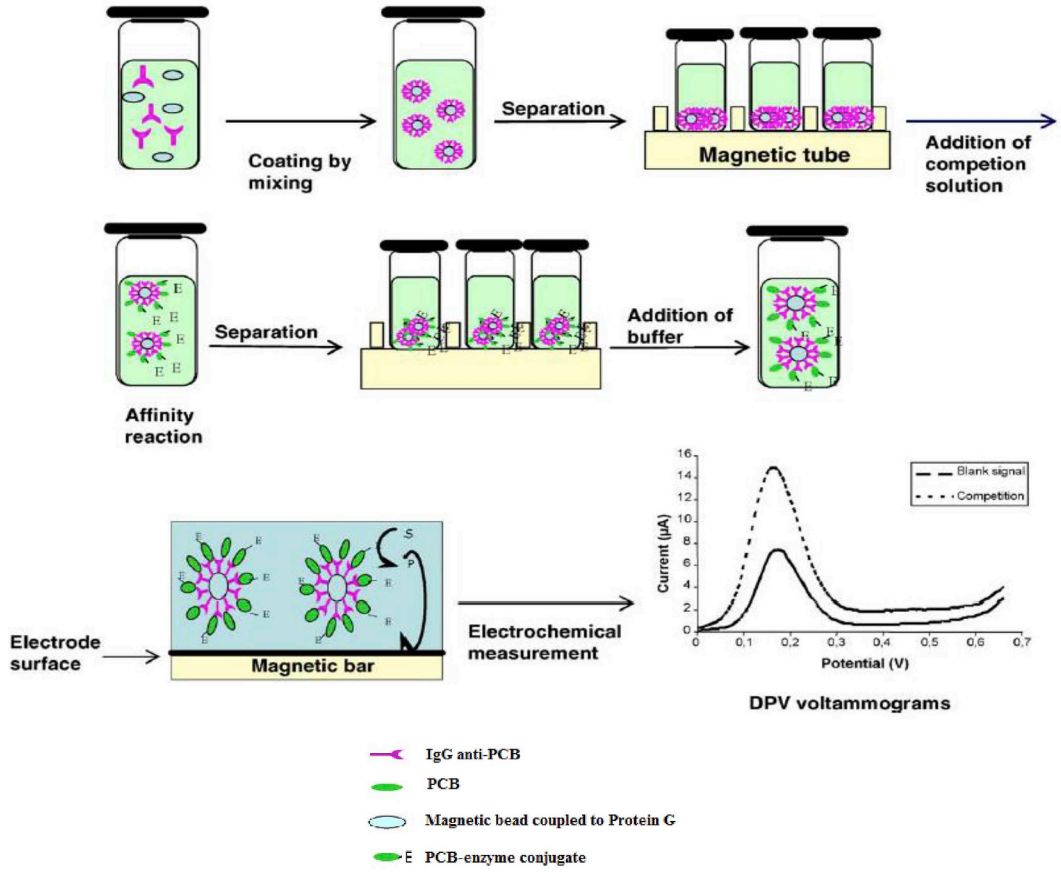

Fig. 2. Schematic showing of the immunochemical binding, magnetic separation, electrochemical reaction and voltammograms. The magnetic beads coupled with protein G as the capture probe. After magnetic separation, a competitive immunoassay was observed between the analyte and PCB-labeled an enzyme. The beads solution from the second step of the magnetic separation was dropped onto the working electrode and incubated using an enzymatic substrate. Reprinted with permission [46].

Table 2
Available PCB immunosensors.

Target	Analytical method	Biosensing principle	Real sample	LOD (nmol L^{-1})	linear range (nmol L^{-1})	Reference
Co-PCB	fluorescence	ELISA	water and soil extract	[a]3.4×10^{-4}	[a]3.4×10^{-4} to 3.4×10^{2}	[33]
PCB	SPR	indirect inhibition immunoassay		7.65×10^{1}	–	[38]
PCBs	electrochemical	antibody-immobilize on conducting polymer	water and soil	[b]1.3×10^{1}, [c]5.3, [d]1.2, and [e]6.4	[b]1.1 to 3.8×10^{2}	[39]
PCBs	electrochemical	competitive format	–	7×10^{-3}	0.1 to 10	[41]
PCBs	electrochemical	competitive format	food samples (sheep milk, bovine adipose tissue, and bovine muscle)	[f]3.6×10^{1}	[f]3.6×10^{1} to 1.8×10^{5}	[42]
PCBs	electrochemical	electrochemical magneto-immunosensor	marine sediment and soil	[b]1.1, [c]1.4, [d]1.5 and [e]3.1	–	[46]
PCBs	electrochemical	antibody coated magnetic beads	milk	[f]3.6×10^{1} and [g]3.9	–	[47]
PCBs	electrochemical	direct competitive enzyme-linked immunosorbent assay	river water	[d]3×10^{-2}	[d]3.1×10^{-2} to 4.6	[51]
PCBs	electrochemical	label-free impedimetric immunoassay	insulating oils	[h]3.3×10^{-3}	[h]3.3×10^{-2} 3.3×10^{1}	[54]
PCBs	electrochemical	silver nanoparticles doped polyaniline modified glassy carbon electrode	tap water and guava juice	[g]2.4×10^{-1}	[g]7.8×10^{-1} and 4.6	[31]

[a] For 4-methoxy-3, 3,4-trichlorobiphenyl.
[b] PCB concentration expressed as Aroclor 1242.
[c] PCB concentration expressed as Aroclor 1248.
[d] PCB concentration expressed as Aroclor 1254.
[e] PCB concentration expressed as Aroclor 1016.
[f] PCB concentration expressed as Aroclors 1242 and 1248.
[g] PCB concentration expressed as PCB28.
[h] PCB concentration expressed as Aroclors of 1242, 1248, 1254, and 1260; 1:1:1:1.

3. Aptamers as recognition probes of PCBs

3.1. Selection of PCB-Binding DNA aptamers

Aptamers are chemical antibodies with adequate affinity, which have been featured as promising alternatives since 1990 [56]. These short, nucleic acid-based sequences could recognize a wide range of food and environmental contaminants, such as heavy metal ions, pesticides and insecticides, antibiotics, drugs and drug residues, microbial cells, and toxins, owing to their unique three-dimensional structure [19,57–60]. Aptamers have numerous advantages, including high affinity and specificity, better target versatility, low immunogenicity, cost-effectiveness, reproducibility, and faster synthesis compared to antibodies. As such, they are proper candidates for therapeutic and diagnostic applications. *In-vitro* selection of aptamers is inexpensive and could be carried out within a few weeks [61,62]. Among the other benefits of aptamers are simplicity, wide variety of chemical modifications, and unlimited shelf-life, which facilitate their application in the measurement of targets in complex matrices with various transducers.

Therefore, the applications of aptamers remain very widespread, with increasing explorations in the several scientific fields of biosensing, diagnostics, therapeutics, food safety and environmental monitoring [63–65]. There have been a numbers of excellent reviews in recent years with different emphases [17,66,67].

Recent reports have denoted the selection of single-stranded DNA aptamer for the recognition of some proof-of-concept PCBs [68,69]. The specific aptamers that tightly bind to PCBs have been isolated from 10^{15} random sequences using the systematic evolution of ligands through exponential enrichment (SELEX). SELEX consists of several binding and elution steps of oligonucleotides in the vicinity of the intended target until reaching an enriched pool of the sequence with high affinity for the target. Table 3 lists several aptamers that have been developed to detect some PCB congeners.

3.2. Aptasensors for PCBs detection

Aptasensors are a novel class of biosensors with aptamers as the bioreceptors. Single-strand DNA could fold into distinct conformational structures, so that they would become dedicated to binding to their target with high affinity [71,72]. In general, the developed aptasensors for PCBs could be divided into optical and electrochemical aptasensors.

3.2.1. Optical aptamer-based PCB biosensors

Optical aptasensors are a subject of great interest owing to their inherent advantages, such as simplicity, fast operation, high sensitivity, and low cost. The assays that are based on colorimetric, fluorescent, and surface-enhanced Raman spectroscopy are the three subclasses of PCB optical aptasensors.

3.2.1.1. Colorimetric-based aptasensors. As the most practical of optical sensors, colorimetric biosensors have attracted remarkable attention. Colorimetric biosensors are used for the rapid and on-site detection of a target with the naked eye. Therefore, they are more likely to be considered in the manufacturing of commercial kits than other sensors. Colorimetric biosensors often rely on the size-dependent optical properties of gold nanoparticles (AuNPs), which could easily change from red to blue due to the variations in the particle size.

AuNPs have unique optical properties, such as large surface area, ease of synthesis, high chemical stability, proper biocompatibility, and peroxidase-like activity [19,73]. Colorimetric aptasensors are classified into semi-quantitative methods, while the signal could be observed spectrally to obtain qualitative results. In this regard, Cheng et al. [70] presented the first colorimetric aptasensor for the detection of PCB77 using AuNPs. The truncated aptamer (35-mer) could serve as a stabilizer, protecting AuNPs from salt-induced aggregation (Fig. 3). Upon

Table 3
PCBs aptamer Sequences obtained by SELEX.

Target	Aptamer Sequence	Size(mer)	K_d (nmol L^{-1})	Reference
PCB77	GGCGGGGCTACGAAGTAGTGATTTTTTCCGATGGCCCGTG	40	400	[69]
PCB77	GGCTACGAAGTAGTGATTTTTTCCGATGGCCCGTG	35	–	[70]
PCB72	AGCAGCACAGAGGTCAGATGCACTCGGACCCCATTCTCCTTCCATCCCTCATCCGTCCACCCTATGCGTGCTACCGTGAA	80	63	[68]
PCB106	AGCAGCACAGAGGTCAGATGCACTCGGACCCCATTCTCCTTCCATCCCTCATCCGTCCACCCTATGCGTGCTACCGTGAA	80	99	[68]

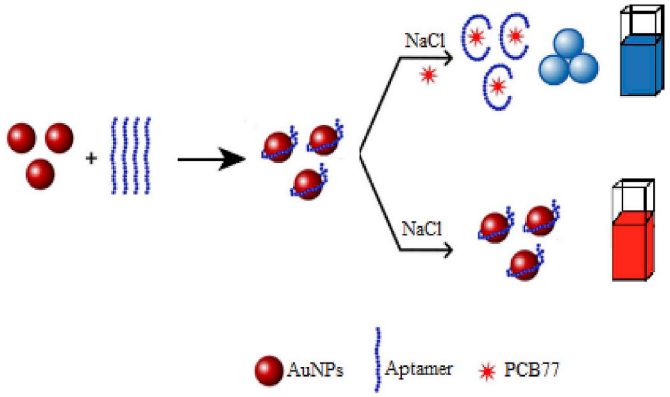

Fig. 3. Principle of colorimetric aptasensor for the detection of PCB77. The aptamers were directly adsorbed on the surface of AuNPs, forming aptamer-functionalized gold nanoparticles. Upon the addition of the target, the binding between PCB77 and aptamer induced aggregation of AuNPs that changed the color of the solution from red to blue. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

the addition of PCB 77, the aptamers switched from random coil structures to an advanced structure, thereby leaving AuNPs and forming a purple-blue color. The practical application of this assay has been confirmed for the detection of PCB 77 in the water samples collected from Huangpu River, showing proper consistency with the results of gas chromatography tandem mass spectrometry (GC-MS).

3.2.1.2. Fluorescent aptasensor. As a strong diagnostic method with high sensitivity, fluorescence has been considered extensively in the designing of aptamer-based biosensors owing to easy operation, rapid detection, good reproducibility, and potential for multi-analyte analysis [74–77].

Upconversion nanoparticles (UCNPs) are a promising new class of imaging agents with unique properties, including long lifetime, absence of autofluorescence, high photo-stability, and low toxicity [78]. In a study, Wang et al. [79] presented an ultrasensitive PCB aptasensor through recovering the quenched fluorescence emission of UCNPs. As is illustrated in Fig. 4, the specific PCB aptamer was fixed onto the surface of magnetic microspheres (MMPs) via avidin-biotin linking, followed by hybridization with cDNA. In the presence of PCB, the aptamers bound to the target, and the released cDNA was transferred into the solution of two designed hairpins (H_1/H_2) to further improve the detection sensitivity. As the initiator strands, the cDNA could trigger an enzyme-free nucleic acid polymerization, as well as the hybridization chain reaction (HCR). HCR opened the hairpins, causing the fluorescence of the connected UCNPs to be restored. In the second step (amplification signal), nicking endonuclease were added so as to cut the DNA strands at a specific site. The UCNPs were released, distancing further from the quencher tags, so that the recovery of fluorescence could reach the maximum. This advanced, complex aptasensor with dual amplification strategy could be used for the detection of PCB72/106 with high sensitivity, wide linear range, and high specificity.

3.2.1.3. SERS-based aptasensors. Surface-enhanced Raman scattering (SERS) is a surface-sensitive technique, in which an analyte is adsorbed onto an active SERS substrate with nanoscale roughness. The substrate is irradiated by monochromatic radiation (usually laser), and the resultant scattering is analyzed using a Raman spectrometer [81]. Typically, gold and silver metal surfaces are used in this technique, and the preparation of the surface could be performed using the metallic coating of a nanostructured substrate or through the deposition of metallic nanoparticles [82].

SERS has gained remarkable attention for the development of selective and sensitive food and environmental biosensors [83–85] owing to its significant benefits, such as ultra-high sensitivity (as high as a single molecule) [86,87], narrow spectral bandwidths [88], real-time detection, well-established instrumentation, and slight or no need for sample preparation [81]. Lu et al. introduced a SERS aptamer-based biosensor for the detection of PCB77 using SiO_2 core and Au shell nanostructures (SiO_2@Au) [89]. In the mentioned research, the PCB77 binding aptamers were covalently attached to the gold shell of SiO_2@Au. In the absence of PCB77, SiO_2@Au nanostructures exhibited strong SERS signal (I [660 cm^{-1}]/I[736 cm^{-1}]), while in the presence of the target, the aptamer conformations changed, thereby leading to the reduction of the SERS signals. The aptasensor shows the limit of detection (LOD) to be as low as 1 μM.

In a similar, recent study, Sun et al. illustrated another SERS label-free aptasensor for PCB77 detection [90]. In the mentioned research, the aptamer was immobilized on the Ag-nanorod arrays with S—Ag bonds. In the presence of PCB-77, the aptamer changed into a hairpin loop structure, and the SERS signals of the aptamer guanines increased. The high selectivity of this novel SERS aptasensor for PCB-77 detection has also been verified by other PCB congeners. Moreover, the developed SERS aptasensor has been reported to accurately detect PCB77 in the lake water samples with the LOD of 0.33 μM.

Similarly, Fu et al. constructed a SERS microfluidic aptasensor for the detection of PCB77 [91]. To construct this microfluidic chip, two patterned polydimethylsiloxane samples were prepared with two input channel grooves form a close chip (Fig. 5). In addition, the Ag nano-crown array was used for the SERS substrate and detection zone. According to the obtained results, the PCB77 could not chemically adsorb on the surface of the Ag nano-crown array in the SERS measurements. However, the mercapto-aptamers that was injected from another channel was used as the capturing agent for PCB77 and immobilized on the detection zone via S—Ag bonds. With a small analyte volume, this rapid SERS aptasensor could detect the trace amount of PCB77 with the LOD of 10 nmol L^{-1}.

3.2.2. Electrochemical aptasensors

Electrochemical aptasensors have emerged as a promising tool for food and environmental analysis owing to their inherent advantages, such as high sensitivity and selectivity, rapid response, simplicity, low cost, and high stability [92,93]. Biophysical interactions are often detected in the close proximity of the electrode surface based on the type of the output signal. Electrochemical aptasensors could be classified as amperometric, voltammetric, potentiometric, and conductometric [57]. In an interesting approach, Pilehvar et al. [94], introduced an electrochemical aptasensor for hydroxylated polychlorinated biphenyl (OH-

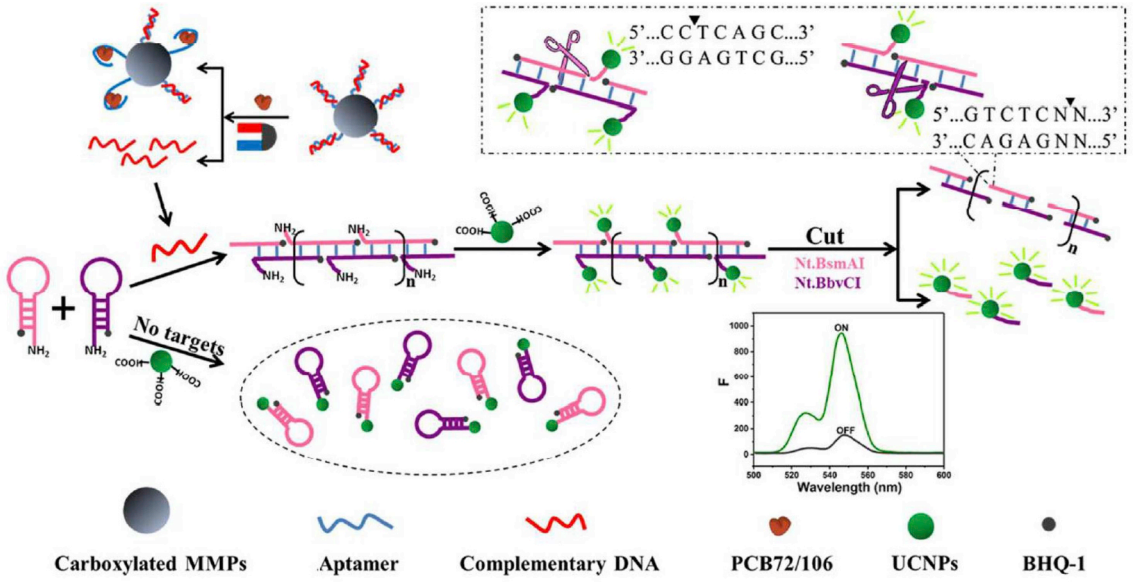

Fig. 4. Representation of the principle of the fluorescent aptasensor for PCB72/106 detection based on the dual-amplification strategy. In the absence of target, the HCR process cannot be triggered and the UCNPs were connected to the hairpin. A fluorescence quenching occurred as a result of the close distance between the UCNPs and BHQ-1. In the present of target, the released cDNA initiated HCR as the initiator, and formed a long double-stranded DNA. Then it connected to the UCNPs. The fluorescence recovered as a result of the hairpin structure opening and increase of the distance between the UCNPs and BHQ-1. Reprinted with permission [80].

PCB) using carboxyl functionalized multi-walled carbon nanotubes (MWCNT-COOH). Sensing platform consist of a glassy carbon electrode modified by MWCNT-COOH, which acts as a capture probe for aminated-aptamer immobilization via amide linkage. In addition, the desired specifications of functionalized MWCNTs (high surface area, abundant functional groups, and unique electronic and mechanical properties) have attracted the attention of researchers for the application of this nanomaterial for sensing platforms [95,96]. The addition of OH-PCB to this biosensor has resulted in aptamer conformation switching, and the changes in the interfacial properties have been characterized by EIS. The LOD in this impedimetric aptasensor has been estimated at 10 nmol L^{-1}. Aptasensor has been employed for the detection of OH-PCB in blood serum samples.

In order to enhance PCB detection systems, Wu et al. [97] developed a sensitive aptasensor with the Au electrode, which was modified by AuNP dotted reduction graphene oxide (RGO-AuNP) as a platform for aptamer immobilization. The unique synergistic properties of RGO and AuNP could develop multi-active sites, which are hydrophilic to graft more aptamers, so that the sensitivity of the designed aptasensor would be almost twice higher than the common Au-based biosensors. The aptamers modified by ferrocene (Fc) have also been considered as the redox probe from 3′ terminals. In the presence of PCB77, the immobilized aptamer is switched into a folded structure, forcing the redox probe to be in the proximity of the electrode surface and increasing the corresponding signal. This aptasensor has exhibited the LOD of as low as 0.1 pgL^{-1} and has been used in the analysis of tap water samples.

Black phosphorus (BP) is a novel discovered 2-D layered system, which has recently triggered an extensive interest owing to its terrific properties [98]. However, the intrinsic instability of BP toward oxygen and humidity has restricted its practical applications [99]. In a very recent attempt, Liang et al. [100] was passive BP by coating with hexamethylenediamine (HMA). The resultant BP-HMA nanocomposite showed the stability of more than one month and has been dotted with AuNPs before binding to PCB77 aptamers. Upon PCB77 addition, the target reacted with the aptamer, thereby inducing structural switching and causing the redox probe to become adjacent to the electrode surface. This electrochemical aptasensor is based on gold nanoparticle-dotted BP and has shown excellent analytical performance as a novel biosensing platform.

In an electrochemical assay, based on aptamer, Au-nanoparticles/poly dimethyl diallyl ammonium chloride (PDDA)-graphene composite was established for the specific detection of OH-PCB in water samples

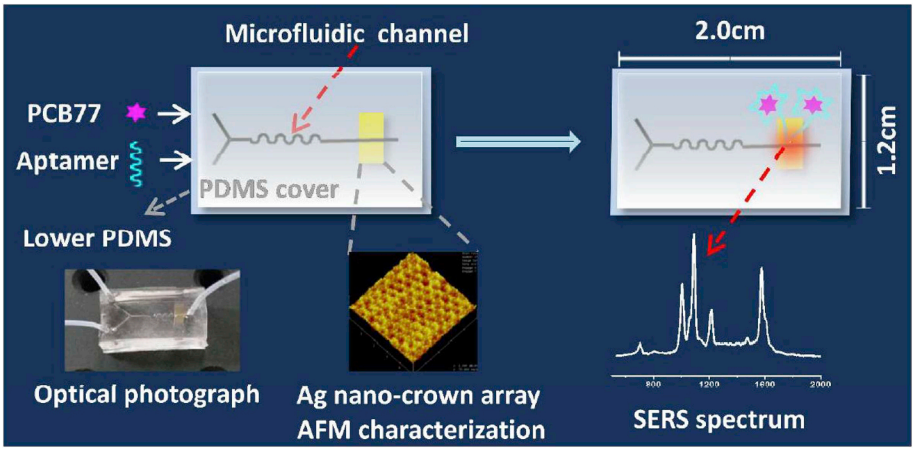

Fig. 5. Scheme of depicting the SERS microfluidic PCB77 aptasensor design. The PCB77 could not chemically adsorb on the surface of the detection zone. However, the mercapto-aptamers that was injected from another channel was used as the capturing agent for PCB77 and immobilized on the detection zone via S—Ag bonds. Reprinted with permission [91].

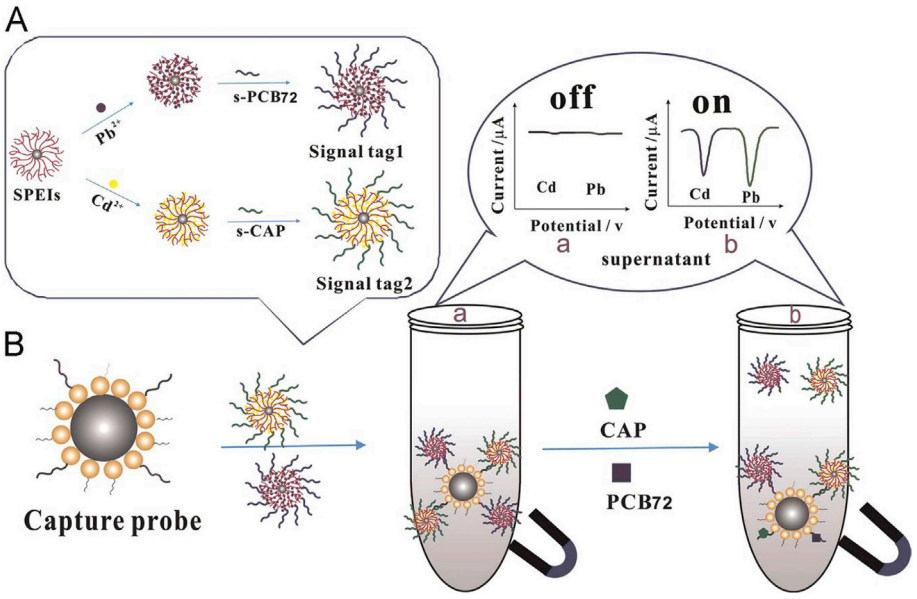

Fig. 6. Simultaneous electrochemical detection of PCB72 and CAP based on metal ions encoded nanospherical brushes spherical structures. The dispersion of Pb^{2+} and Cd^{2+} ions onto the prepared SPEIs obtained the MSPEIs. Then MSPEIs were incubated with s-PCB and s-CAP that generated the tracers; (B) Schematic representation of simultaneous detection of chloramphenicol and PCB72 based on a voltammetric aptasensing platform. The capture probe, including the magnetic AuNPs labeled with the selective aptamers of chloramphenicol and PCB72, was exposed to the tracers through the hybridization between the s-DNAs and the aptamers. The presence of the chloramphenicol and PCB72 caused the target attachment to the aptamer-MGPs that released the s-DNA-MSPEIs tracers from the capture probe. Therefore, an increased amount of the tracers could be detected through the SWV measurement that switched the signals of the aptasensor to "on" state. Reprinted with permission [104].

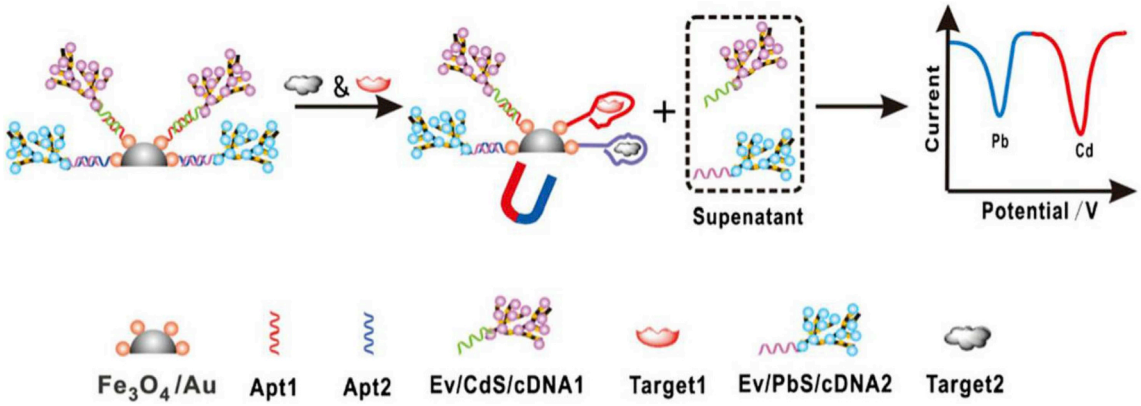

Fig. 7. The simultaneous detection of two target based on aptamer-QDs encoded dendritic probes. The encoded nanotracers were linked to the capture probes by the base pairing between cDNA and aptamer. In the presence of the targets, the nanotracer was released into the supernatant. After magnetic separation, PCB72 and CAP were detected simultaneously through the monitoring of the voltammogram peaks of the metal ion tags Cd(II) and Pb(II) from the supernatant solution. Reprinted with permission [105].

[101]. In this aptamer-based sensor, the OH-PCB aptamer was firstly fixed on the surface of the modified glassy carbon electrode (AuNPs/PDDA-GN/GCE). In the presence of OH-PCB, the electron transfer resistance (R_{et}) increased due to the formation of the aptamer/target complex, which was quantified by EIS.

Recently, several electrochemical biosensors have emerged as efficient sensing tools for the simultaneous detection of the ultra-trace level of pollutants [102,103]. For instance, Yan et al. [104] have reported the development of a sensitive assay to simultaneously detect PCB72 and chloramphenicol (CAP). In an intelligent layout, the nanostructures with amino groups (nanospherical hyperbranched polyethylene imine brushes [SPEIs]) adsorbed the Pb^{+2} and Cd^{+2} ions as the redox probes in their structure. Owing to the superior properties of SPEIs (e.g., three-dimensional spherical structure with a larger specific area and ability to load metal ions by maintaining their electrochemical activities), they could act as ideal trappers in the design of multi-target biosensors. As is depicted in Fig. 6, two SPEIs loaded the Pb^{+2} and Cd^{+2} ions as the tags for PCB72 and CAP, respectively, conjugating with their aptamer complementary strands (s-CAP and s-PCB) as the tracers (signal tags 1 and 2). On the other vial, the PCB72 and CAP aptamers were simultaneously immobilized on magnetic gold nanoparticles, forming the capture probe. Afterwards, the capture probe and tracers could form a complex (vial a) through the hybridization of sDNA with the aptamers. In the presence of PCB72 and CAP, the targets could react with the aptamers on the capture probes, thereby releasing the SPEIs. The released SPEIs in the supernatant could be simultaneously detected based on their ion peaks. It is notable that these aptasensors have been reported to have extremely low LOD (0.3 $pgmL^{-1}$) for CAP and PCB72. This analytical technique has been able to detect CAP and PCB72 in three types of fish samples.

Using a similar approach, Chen et al. [105] developed an electrochemical aptasensor for the simultaneous detection of PCB72 and CAP. The adopted strategy was based on the EnVision complex (EV), which loaded quantum dots (CdS or PbS), while linking to complementary strands (EV/CdS or PbS/cDNA), and was used as the encoded nanotracers and magnetic gold nanoparticles (Fe_3O_4@Au) modified with PCB72 and CAP aptamers as capture probes (Fig. 7). The EV complex is a water-soluble, dendric complex consisting of HRP and secondary antibodies, and a large number of its enzyme sites are used for the immobilization of CdS and PbS QDs [106,107].

Initially, the encoded nanotracers were linked to the capture probes by the Watson-Crick base pairing between cDNA and aptamer. In the presence of the targets, the nanotracer (EV/CdS or PbS/cDNA) was released into the supernatant. After magnetic separation, PCB72 and CAP were detected simultaneously through the monitoring of the voltammogram peaks of the metal ion tags Cd(II) and Pb(II) from the

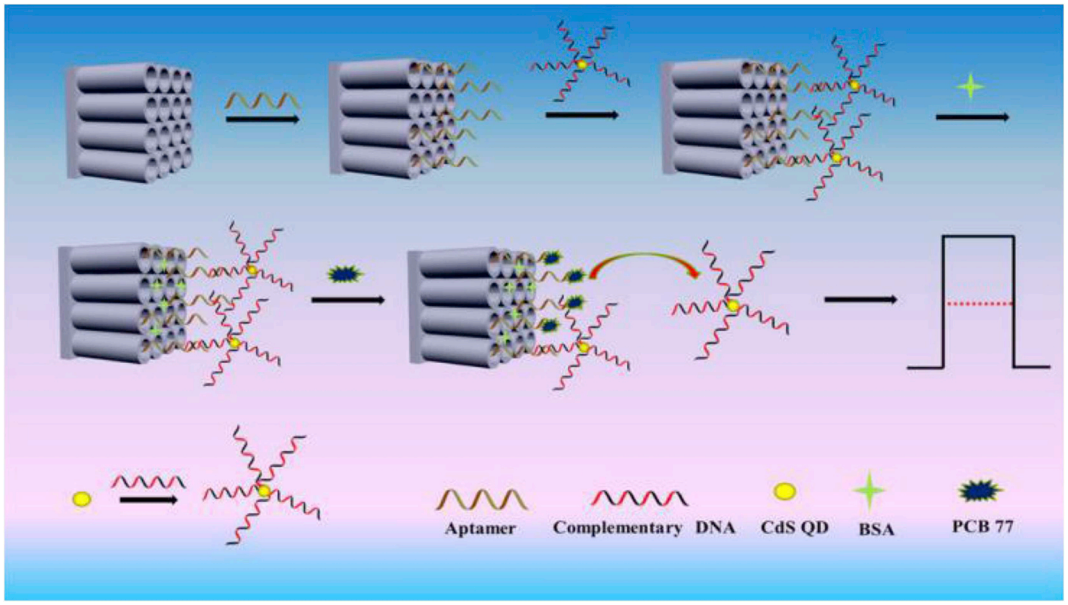

Fig. 8. Illustration of the PEC aptasensor construction and the detection mechanism of PCB77. After anchoring the aptamer on the ordered N-doped TiO_2 NTs surface, DNA-CdS QDs were introduced on the sensing interface by hybridizing. In the presence of PCB77, the target reacts with the aptamer, thereby inducing the release of DNA-CdS QDs from the aptasensor surface and leading to the reduction of the photocurrent. Reprinted with permission [109].

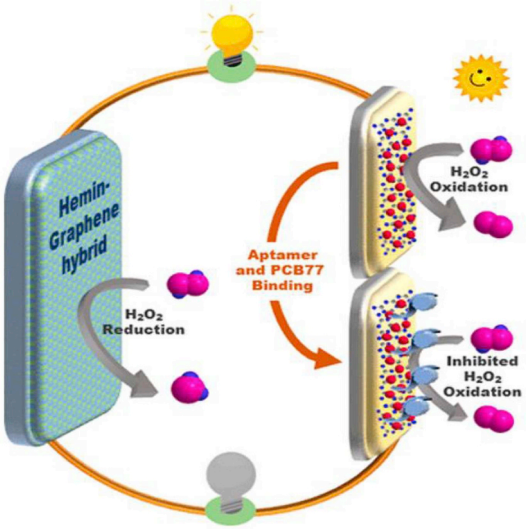

Fig. 9. Schematic representation of the of PFC aptasensor for PCB77 determination. In presented green fuel with photoelectrocatalytic oxidization at the aptamer-modified anode, hydrogen peroxide (H_2O_2) electrocatalytically reduces at the hybrid cathode, thereby inducing electric power generation in the external circuit. The incubation of PCB77 into the system and complex formation on the photoanode inhibit electron transfer. Reprinted with permission [111].

supernatant solution. The LOD for CAP and PCB72 was estimated at 0.33 and 0.35 pg mL^{-1}, respectively, which is 2–3 times lower than the commercially available ELISA.

3.2.2.1. Photoelectrochemical-based aptasensors. Recently, PEC-based biosensors have attracted the attention of researchers for sensitive detection purposes owing to advantages such as high sensitivity, simple and inexpensive equipment, and ease of miniaturization [108]. In PEC sensors, the excitation source is light, and the resultant current is the detectable signal. Photon-to-electricity conversion on photoactive electrodes could be coupled with a biophysical binding between a recognition element and an analyte. In a research in this regard, Fan et al. exploited N-doped TiO_2 nanotubes as an appropriate photoactive surface for the anchoring of the PCB77 aptamer (Fig. 8) [109]. After immobilization on the ordered N-doped TiO_2 nanotube, the aptamer hybrids with the complementary strand modified with CdS quantum dots (DNA-CdS QDs). CdS QD is a well-known PEC label with its unique electrical and optical properties. In the presence of PCB77, the target could react with the aptamer, thereby inducing the release of DNA-CdS QDs from the aptasensor surface and leading to the reduction of the photocurrent. This PEC aptasensor has exhibited a wide linear range and a low LOD for PCB77.

Recently, there has been growing interest regarding photocatalytic fuel cells (PFCs) as sensors for the utilization of permanent, natural solar energy [110]. These self-powered sensors often consist of two electrodes and generated electrical signals proportional to the target levels. An appealing PFC-based aptasensor has also been proposed by Ref. [111] which consists of a gold nanoparticle-decorated graphitic C_3N_4 nanosheet as the photoanode and hemin-graphene nanocomposites as the cathode, Fig. 9. As a green fuel with photoelectrocatalytic oxidization at the aptamer-modified anode, hydrogen peroxide (H_2O_2) electrocatalytically reduces at the hybrid cathode, thereby inducing electric power generation in the external circuit. The incubation of PCB77 into the system and complex formation on the photoanode inhibit electron transfer, leading to the reduction of the signals. Highly sensitive and selective detection of the results could be applied in PCB77 monitoring.

We summarized the progress that has been made in aptamer-based detection of PCBs. These aptasensores have been listed in Table 4.

3.3. The role of aptamers as recognition elements in extraction

PCBs are observed nearly in all matrixes such as food, water, soil and air in trace quantities. Therefore, it is an urgent need to develop an effective pretreatment method to enrich the trace level of PCBs. The extraction of PCBs from various contaminated sites is the next challenging stage after sampling and pretreatment procedures. Some literature have demonstrated sampling and pretreatment methods for PCBs analysis from different sources such as air, water, soil, sediments and other biological contaminated sources [112,113].

There is the several sample extraction methods including solid

Table 4
Available PCB aptasensors.

Target	Analytical method	Biosensing principle	Nanoparticles	Real sample	LOD (nmol L^{-1})	linear range (nmol L^{-1})	Reference
PCB77	colorimetric	salt-induced aggregation of AuNPs	AuNPs	real water	5×10^{-2}	5×10^{-1} to 9×10^{2}	[70]
PCB72 and PCB106	fluorescence	hybridization chain reaction and nicking endonuclease	NaYF4:Yb,Er UCNPs	water and soil	0.01	1.1×10^{-2} to 2.3×10^{3}	[80]
PCB77	SERS	aptamer modified silica Au/core-shell nanoparticles	SiO_2@Au	–	1×10^{3}	1×10^{3} to 2×10^{4}	[89]
PCB77	SERS	aptamer modified Ag-nanorod arrays	Ag-nanorod Arrays	real lake water	3.3×10^{2}	3.3×10^{2} to 1×10^{3}	[90]
PCB77	SERS	aptamer capturing in a microfluidic device	Ag nano-crown	–	1×10^{1}	10^{1} to 1×10^{5}	[91]
OH-PCB	electrochemical	MWCNT-COOH modified glassy carbon electrode	MWCNT	blood serum	1×10^{1}	1.6×10^{2} to 7.5×10^{3}	[94]
PCB77	electrochemical	target-induced conformational changes of aptamer	AuNP -reduction graphene oxide	–	3.4×10^{-7}	3.4×10^{-7} to 3.4×10^{3}	[97]
PCB77	electrochemical	target-induced conformational changes of aptamer	AuNPs	tap water	1×10^{-4}	3.4×10^{-2} to 3.4×10^{1}	[100]
OH-PCB	electrochemical	target-induced conformational changes of aptamer	Graphene and AuNPs	lake water	5.3×10^{-3}	2.9×10^{-2} to 2.9×10^{2}	[101]
PCB77	electrochemical	multi-metal ions encoded nanospherical brushes	MSPEIs[a]	fish	1×10^{-3}	3.4×10^{-3} to 3.4×10^{1}	[104]
PCB72	electrochemical	displacement of nanotracer	Fe_3O_4@Au	fish	0.9×10^{-5}	2.7×10^{-3} to 2.7×10^{2}	[105]
PCB77	photoelectrochemical	target-induced release of the DNA-CdS QDs probe	CdS quantum dots and N-doped TiO_2 nanotubes	tap water and domestic sewage	3.4×10^{-4}	3.4×10^{-4} to 3.4×10^{-1}	[109]
PCB77	photoelectrochemical	target-induced complex formation and inhibited the electron transfer	gold nanoparticles-decorated graphitic C_3N_4 nanosheet and hemin-graphene nanocomposites	environmental water sample	1.5×10^{1}	3.4×10^{1} to 3.4×10^{3}	[111]

[a] MSPEIs: metal ions encoded nanospherical hyperbranched polyethylene imine brushes.

phase extraction (SPE), supercritical fluid extraction (SFE), soxhlet extraction, solvent extraction and other latest microextraction methods [112].

Solid phase extraction (SPE) is a sample preparation process for the extraction of the trace levels of contaminants in food and environmental samples. SPE is partitioning of solutes between two phases, liquid (testing sample) and a solid (sorbent) phase. An SPE method often inclusive three to four sequential steps. First, the solid sorbent must be conditioned using an appropriate solvent, followed by the same solvent as the sample solvent. The next step is the percolation of the sample via the solid sorbent. The third step (optional) may be the washing of the solid sorbent with an appropriate solvent, to delete matrix components of sample that have been maintained by the solid sorbent. The final step consists in the elution of the targets by an appropriate solvent, without removing maintained matrix components [114,115].

Recently, aptamer has been loaded as an SPE sorbent on the coating of SPE in order to improve the limitations of conventional SPE [116–118]. In this regard, Lin et al. [119] developed a novel dispersive SPE containing a specific aptamer for PCB72 and PCB106 coupled with GC-MS, which provided high selectivity, stability, repeatability, good binding capacity, and reproducibility for the extraction of PCBs from soil samples. In this process, Fe_3O_4 nanoparticles were functionalized with UiO-66-NH_2 membrane using polydopamine (PDA) shell as the covalent linker (Fe_3O_4@PDA@UiO-66-NH_2). At the next stage, the aminated aptamer was immobilized via amide linkage On UiO-66-NH_2. The aptamer-functionalized adsorbent (Fe_3O_4@PDA@UiO-66-Apt) specifically captured PCBs from a complex matrix, and the adsorbent was easily isolated from the samples through magnetic separation. Finally, the detection of PCB72 and PCB106 was performed through GC-MS. In another study, stir bars were functionalized using aptamers and used to develop SPE. Metal-organic framework (MOF-5), which is a novel porous material, was also deposited on stainless steel wires using electrochemical methods [120]. Following that, poly (diallyldimethylammonium chloride) (PDDA) with a positive charge was modified on the MOF substrate, where it could electrostatically bind to the negative-charge aptamer. Finally, the developed Apt-MOF fiber was used for the extraction of PCBs from fish samples, and desorption solution was analyzed using the GC-MS for the detection of PCBs. The LOD of the designed Apt-SBSE/GC-MS assay was estimated at 0.003–0.004 ng kg^{-1} for the PCBs in the fish samples. The key advantages of this new sorbent are high selectivity and enrichment capability. In a study in this regard, Jiang et al. [121] employed aptamer-functionalized magnetic conjugated organic frameworks (COF) abundant in carboxylic groups and constructed a selective extraction of the trace level of 2-OH-CB 124 in human serum. The amino-modified aptamers were also immobilized onto the surface of the magnetic COFs and used as SPE material for the selective detection of OH-PCBs through liquid chromatography-mass spectrometry.

4. Conclusions and perspectives

According to the legal regulation for PCBs and their side-effects on health, sensitive and on-site screening systems for PCBs are urgently needed to minimize their risk. The selection of bioreceptors is the most important issue in the detection of PCBs. Although antibodies are considered to be the first bioreceptors for the detection of PCBs, aptamers have recently become the new bioreceptors for such purposes. The present study provided an overview of the aptamers employed to detect PCBs. The range of the examples presented in this review can provide a comprehensive outlook toward the development of novel biosensors for PCB monitoring. Moreover, we discussed the advantages and limitations of antibodies and aptamers as bioreceptors and transducers. Currently, there are no available aptamer-based commercial kits for PCB detection. However, as complementary schemes to the existing high-resolution mass spectrometry and antibody-based screening methods, aptamer-based assays are highly expected and will be developed for the determination of POPs in food matrices in the near future. In conclusion, the recent advancement in the development of aptamer-based SPE has been highlighted, and it is recommended that further investigation be conducted regarding the construction of commercial aptamer-based kits.

Novelty statement

Herein, we provide an overview of the most recent findings on antibody and aptamer-based biosensors for the detection of PCBs considering the latest research articles as case studies. Finally, the recent advancement in the development of aptamer-based solid phase extractions has been evaluated. These findings could contribute to improving the design of commercial PCB-kits in the future.

Declaration of interests

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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