A review of spectroscopic probes constructed from aptamer-binding gold/silver nanoparticles or their dimers in environmental pollutants’ detection

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Abstract
The issue of environmental pollutant residues has gained wide public attention all along. Therefore, it is necessary to develop simple, rapid, economical, portable, and sensitive detection techniques, which have become the focus of research in the pollutants detection field. Spectroscopy is one of the most convenient, simple, rapid, and intuitive analytical tools that can provide accurate information, such as ultraviolet spectroscopy, fluorescence spectroscopy, Raman spectroscopy, plasmon resonance spectroscopy, etc. Gold nanoparticles, silver nanoparticles, and their dimers with unique optical properties are commonly used in the construction of spectroscopic probes. As a class of oligonucleotides that can recognize specific target molecules, aptamers also have a strong ability to recognize small-molecule pollutants. The application of aptamer-binding metal nanoparticles in biosensing detection presents significant advantages for instance high sensitivity, good selectivity, and rapid analysis. And many spectroscopic probes constructed by aptamer-binding gold nanoparticles, silver nanoparticles, or their dimers have been successfully demonstrated for detecting pollutants. This review summarizes the progress, advantages, and disadvantages of aptamer sensing techniques constructed by visual colorimetric, fluorescence, Raman, and plasmon resonance spectroscopic probes combining gold/silver nanoparticles or their dimers in the field of pollutants detection, and discusses the prospects and challenges for their future.

Keywords Aptamer · Gold/silver nanoparticles · Spectroscopic probes · Environmental pollutants

Introduction
With the increasing level of industrialization in the world, the migration, enrichment, transformation, and metabolism of trace pollutants remaining in the environment (such as soil and water bodies) pose a potential threat to the ecological environment and human health. The main standard methods currently used for the detection of environmental pollutants are GC (Gas Chromatography), HPLC (High-Performance Liquid Chromatography), LC–MS (Liquid Chromatography–Mass Spectrometry), AAS (Atomic Absorption Spectrometry), ICP (Inductively Coupled Plasma-Atomic Emission Spectrometry), and AFS (Atomic Fluorescence Spectrometry), etc. [1–3]. These assays have great sensitivity and selectivity, but they frequently need complex equipment, costly sample pre-treatment processes, and are challenging to obtain quickly in situ detection [4]. Therefore, in recent years, many researchers have worked on the application of miniaturized biosensing devices with the same high sensitivity and specificity for the detection of environmental pollutants, combined with portable instruments for real-time monitoring in the field.

The operating basis of a biosensor consisting of a recognition element and a signal transmission element is the signal changes (optical signals, electrical signals, etc.) generated by the reaction between the biomolecule [5]. Based on the recognition elements, biosensors are classed as enzyme-based biosensors [6], immunobiosensors [7], aptasensors [8], and so on. They can be classified as electrochemical
biosensors, optical biosensors, and thermal and piezoelectric biosensors, depending on the signal transmission method [9, 10]. Compared with traditional contaminant detection technologies, biosensors have the advantages of moderate cost, fast analysis, and environmental adaptability. And they have been widely studied in the fields of food safety, clinical testing, and environmental detection.

With the emergence of aptamers called "artificial antibodies", aptasensors have become a hot topic of research in the field of environmental detection due to their excellent properties. Aptamer, used as a recognition element in biosensors, is a single-stranded oligonucleotide screened by the systematic evolution of ligands by exponential enrichment (SELEX) technique. It can specifically identify single-stranded oligonucleotides that capture target molecules [11, 12]. Compared with antibodies, aptamers have the advantages of simple preparation, lower cost, easy modification stability, and better recognition ability. Researchers have screened aptamers for a variety of environmental pollutants and used them to construct aptasensors for the detection of environmental pollutants [13, 14].

Metal nanoparticles are widely used in the construction of biosensors because of their unique physicochemical properties such as small particle size, ease of preparation, and high specific surface area [15]. To improve biosensors' sensitivity, detection speed, specificity, and other performances, researchers often change the size, shape, and structure of nanoparticles to construct new metal nanoparticles. Existing studies have studied many common forms of metal nanoparticles, including metal nanospheres, nanorods, nanocore–shell structures, nanoclusters, and nanocages [16]. In particular, metal nanoparticle dimers composed of two identical or different metals are simple to prepare and have a strong "hot spot" effect [17], especially gold and silver nanoparticle dimers. Their effects are so outstanding that become one of the most popular materials in biosensors' research in recent years.

Spectroscopy is the most common analytical tool in aptasensors. The principle of applying aptamer-bound metal nanoparticles to construct spectroscopic probes for detection is based on the interaction between the aptamer and the target pollutant. According to measuring the optical signals generated or amplified by nanomaterials and analyzing the corresponding spectral information, we can achieve the aim of detecting pollutants. Spectroscopic methods include colorimetric (using UV spectroscopy), fluorescence (using fluorescence spectroscopy), surface-enhanced Raman scattering (using Raman spectroscopy), surface plasmon resonance (using surface plasmon spectroscopy), etc. [18]. We classify spectroscopic probes constructed from different means of spectroscopic analysis combined with aptamers and gold/silver or dimeric nanoparticles, and then discuss their advantages, limitations, and research perspectives for their application in the detection of environmental pollutants in recent years.

**Spectral probes**

**Aptamer**

As a class of oligonucleotide sequences consisting of a DNA or RNA fragment of 20–80 bases, nucleic acid aptamers have been isolated through the systematic evolution of ligands by exponential enrichment (SELEX) [19]. Aptamers recognize specific target molecules with high affinity and high specificity comparable to antibodies [20, 21] (Table 1). Many types of molecules can be recognized by aptamers, including bacteria, cells, organisms, proteins, metal ions, and so on.

Specific recognition between aptamers and these target molecules is achieved through different binding modes and structures, such as folding, specific hydrogen bonding, and conformational complementation [22]. With the continuous development of SELEX technology, researchers have come up with many more effective aptamer screening methods for different application routes, such as immunoprecipitation coupled SELEX, cellular SELEX, capillary electrophoresis SELEX, capture SELEX [23], etc. The screening of more aptamers that specifically recognize different target molecules will lead to a wide range of applications in clinical medicine, food safety, environmental testing, and other fields.

**Metal nanoparticles and their dimers**

Metal nanoparticles are used to be functionalized by aptamers and nanoprobes providing signal amplification, thanks to their unique optical properties, biocompatibility, and high specific surface area [24]. In recent years, the combination of metal nanoparticles with UV absorption spectroscopy, fluorescence spectroscopy, Raman spectroscopy, and other spectroscopic analysis techniques to construct spectroscopic probes for pollutants detection has gradually become a hot research topic in the field of environmental detection. Gold nanoparticles (AuNPs) and silver nanoparticles (AgNPs) are two of the monometallic nanoparticles that are widely studied in aptasensors. AuNPs can be prepared by hydrothermal reactions using the trisodium citrate reduction method. It is uniformly dispersed under electrostatic conditions. Gold nanoparticles of different particle sizes appear burgundy or purplish-red. However, due to the high surface energy of metal nanoparticle colloids, the particles are prone to collision and aggregation, making metal nanoparticles unsuitable for long-term storage [25]. Thus, the surface of aptamer-functionalized AuNPs can be modified by sulphydryl groups,
disulfide bonds, etc. forming Au–S bonds and enhancing the stability of AuNPs [26]. AgNPs have better optical properties such as Raman enhancement and fluorescence enhancement. However, their size homogeneity and dispersion are not easily controlled in the preparation process [27], which limits their application and development to a certain extent. The researchers have studied and synthesized dimers combining the advantages of different metals.

Metal nanoparticles’ dimers are typically composed of two metals. It is the presence of two metals that give the dimer its more unique flexibility, more powerful surface plasmon resonance, surface-enhanced Raman scattering activity, electromagnetic coupling enhancement effects, and other properties [28–30]. They offer more options for metal nanoparticle applications in pollutants detection. Homogeneous or heterogeneous dimers of gold and silver are the most common metal dimer materials used in spectroscopic probes. Many scholars have studied the optical properties of different forms of gold and silver dimers, demonstrating that the coupling effect that exists when two metal nanoparticles come into contact gives rise to many new properties [31–34]. For example, M. Jose et al. prepared gold core silver shell nanoparticles (Au@AgNPs). And by studying the extinction coefficient, reflectivity, refractive index, and other parameters, it is proved that this core–shell type metal nanoparticle exhibits optical properties several times higher than that of single metal gold or silver nanoparticles [35] (Fig. 1).

**Typical spectral probe sensing applications**

Spectroscopic probes of aptamer-bound metal nanoparticles are widely studied for the detection of environmental pollutants residues. They can be classified as visualization spectroscopic probes, fluorescence spectroscopic probes, surface-enhanced Raman spectroscopic probes, surface plasmon resonance spectroscopic probes, etc. The following section will focus on the construction of aptasensors based on spectroscopic probes for the analytical detection of pollutants by various spectroscopic means. We list some examples of pollutants detected by these methods in Table 2.

**Visualization spectroscopic probes: simple, intuitive, and instantly qualitative**

As a class of commonly used analytical tests, the colorimetric methods have the advantages of fast response time, ease of operation, and high sensitivity. These methods are based on the color changes of the system, and then achieve the purpose of analysis by visual recognition or relying on the ultraviolet spectrums.

Gold nanoparticles are used as one of the most common nanomaterials for visualization spectroscopic probes because of their unique optical properties, electronic properties, good biocompatibility, colloidal stability, and ease of synthesis [58]. The local surface plasmon resonance (LSPR) is produced when the oscillation of free electrons on the AuNPs’ surface coincides with the vibration frequency of the incident light. In the macroscopic manifestation as a specific wavelength of light in the UV–visible absorption spectrum in the local surface plasmon resonance absorption peak. The LSPR absorption peak of AuNPs in the uniformly dispersed state is around 520 nm. When NaCl or other electrostatic salt solutions are added to the system, the AuNPs aggregate, and the distance between the particles changes, causing an obvious red shift of LSPR. And the color change was visible to the naked eye from burgundy to blue–violet or blue–gray (Fig. 2). Researchers often use this color change in combination with aptamers for visual colorimetric analysis of pollutants residues.

Bai et al. [59] developed a rapid assay for the simultaneous detection of six organophosphorus pesticides based on the principle of competitive binding of AuNPs and target molecules to nucleic acid aptamers. In this method, the organophosphorus pesticide aptamer was adsorbed directly onto the surface of the AuNPs, which enabled the AuNPs to remain stable and homogeneous under high salt conditions.
When the target molecule was added, it competed with the AuNPs to bind the aptamer, the AuNPs were aggregated and the color of the solution changed from red to purple or blue. The method allowed direct visual observation of the presence of pollutants in the sample and achieved higher detection sensitivity by employing UV spectroscopy. In addition to the traditional colorimetric method of NaCl-induced aggregation of AuNPs, some studies have used the interaction of cationic polymers or surfactants with colorimetric spectroscopic probes to construct colorimetric sensors. For example, a group of researchers used the interaction between the cationic polymer poly (dimethylallylammonium chloride) (PDDA), the aptamer, AuNPs, and the target to construct colorimetric aptasensors for Bisphenol A (BPA) detection with a detection limit of 1.50 nM, and successfully applied it to the detection of BPA in tap water and river water [60]. More simply, Qi et al. [61] devised a colorimetric aptasensor that does not require NaCl or other substances to mediate the color change. In this study, positively charged gold nanoparticles were synthesized (The AuNPs made by the commonly used trisodium citrate reduction method were coated with citrate and thus negatively charged.) Normally, negatively charged aptamers could be easily wrapped around the surface of (+)AuNPs, resulting in the aggregation of AuNPs and the blue color of the solution; when the aptamer was bound to the target, the (+)AuNPs remained dispersed (Fig. 3). The team used this principle to achieve sensitive detection of aminopyralid in the environment with a detection limit of $5.6 \times 10^{-10}$ M. This is an order of magnitude lower than the colorimetric method based on AuNPs with a negative charge.

Because AuNPs’ monometallic nanoparticles themselves have very distinct color change characteristics and are easily analyzed using UV spectroscopy, but silver nanoparticles lack good color change characteristics, there are fewer visualized spectroscopic probes constructed by applying metallic silver nanoparticles and metal dimers. Huang et al. [62] prepared Au/Fe$_3$O$_4$ nanodimer hybrid materials, which were combined with Ochratoxin A (OTA) aptamers for the colorimetric detection of OTA with a detection limit of 1.15 ng mL$^{-1}$.

In the studies of classical visualization for spectral probe sensing applications, researchers have often focused on the attachment of AuNPs to the aptamer and the binding of the aptamer to the target, but have neglected the effect of AuNPs on the adsorption of certain targets themselves. LIU’s team [63] recently conducted a more in-depth study of this issue, using dopamine, melamine, and potassium ions as comparative subjects (Fig. 4). Surface-enhanced Raman spectroscopy and other means revealed that AuNPs adsorbed very weakly on potassium ions, but both dopamine and melamine could adsorb directly on the AuNPs surface and even inhibited the adsorption of aptamer on the AuNPs. The reason for the change in color of the system results from the adsorption of dopamine by AuNPs. The team also carried out colorimetric control experiments with chloramphenicol on the same substance. The aptamer of chloramphenicol was used as the experimental group and the non-aptamer sequence as the control group. It was found that similar color changes could be observed in both the experimental and control groups. This demonstrates that chloramphenicol affects the stability of AuNPs themselves and the adsorption of ssDNA onto AuNPs, but that chloramphenicol only weakly inhibits the adsorption of ssDNA onto AuNPs [64].

Visualized spectroscopic probes of aptamer-bound metal nanoparticles (especially AuNPs) will remain a hot topic of research in aptasensors in the future, because they have various advantages over others in terms of specificity, simplicity, and intuitiveness in sensing detection. As colorimetric sensing platforms incorporating metal nanoparticles have been intensively investigated in recent years, previously overlooked potential problems have been highlighted. Many problems are necessary to be considered in the design process of visualized spectroscopic probes, such as the way to remove the adsorption of the target itself on the surface of the metal nanoparticles, the effect that this adsorption has on the binding of the metal nanoparticles to the aptamer. In addition, in the actual environmental media, there are certain substances other than the target that may also be adsorbed on the metal nanoparticles, which may have an effect on the color change during the colorimetric detection, etc. Furthermore, the color of metal nanomaterials usually does not continue to change once they are fully aggregated; therefore, colorimetric aptamer sensing detection has good prospects for the qualitative detection of environmental pollutants, but has some limitations in quantitative detection. These are some of the key issues for future research on...
Table 2  Summary of partial detection methods of nucleic acid aptamer and metal nanoparticles binding spectroscopy

| Nanoparticles | Spectroscopy | Pollutants' target | Aptamer | Linear range/LOD | References |
|---------------|--------------|-------------------|---------|-----------------|------------|
| AuNPs         | UV           | Saxitoxin         | 5'-GGTATTGAGGTCGACCTCCCG TGAAACATGTCTCATTGGGCG CACGCGTTTCCTGATAGCTC TAACTCTCTCTC-3' | 10 fM–0.1 μM/10 fM | [36] |
| AuNPs         | UV           | Microcystin-LR    | 5'-GGCCAAAACAGGACCACCAT GACAAATTACCATACACCCTCAT AGCCCCCATCTCCCG-3' | 0.5 nM–7.5 μM/0.37 nM | [37] |
| AuNPs         | UV           | Bisphenol A       | 5'- GG TAG CGG GTT CC-3' | 97 nM | [38] |
| AuNPs         | UV           | Ag⁺              | 5'-SH-TCAAGGGCCGGC-3'/5'-SH-GCC CCCCTGA-3' | 1 nM–1 μM/0.236 nM | [39] |
| Au–MoS₂       | UV           | Cd^{2+}          | 5'-biotin-ACC GAC CGT GCT GGA CTC TGG ACT GTG GTA TTA TTT GTT GTG TAG CAG TAG TGA GCG TGG TTG CGG-3' | 1–500 ng·mL⁻¹/0.7 ng·mL⁻¹ | [40] |
| AuNPs         | FL           | 17β-estradiol     | 5'-GCTCTCCACGGCGTATT GAG TAATTAC ACCATTTACCAAGTCGTAATCACCACTGGGCGTGGA CGCGCAGAC-3' | 0.48–200 nM/0.48 nM | [41] |
| AuNPs         | FL           | Pb²⁺             | 5'-AAAGGGTGTTGGTTGGG T-FAM-3' | 0.5 nM–1 μM/0.28 nM | [42] |
| AuNPs         | FL           | Chloramphenicol   | 5'-SH-AGC AGC AGC GA TGC AGA TGA CCT CAG CCA GTT GTG GTA TTA TTT GTT GTG TAG CCT ATG GCT GCT ACC GTG AA-Cy3-3' | 26.0–277 μg·L⁻¹/8.1 μg·L⁻¹ | [43] |
| AuNPs         | FL           | Sulfadimethoxine  | 5'-GCC AAC GAG TGT TTA-3' | 2–300 ng·mL⁻¹/3.41 ng·mL⁻¹(water) | [45] |
| AuNPs         | FL           | Acetamiprid      | 5'-TGTAATGGGTCTGCAGCGGGTTTCT GATCCGCTGAACCATATTTAGA AG-3' | 5–100 μg·L⁻¹/1.08 μg·L⁻¹ | [46] |
| AgNCs         | FL           | Oxytetracycline  | 5'-CGTACGAGAATTCCGCTAGCAGA GGTAGGGGCGGCGGGTTAGCG GTACTGTGAATGTGGTGGGATCGCAGA GCTCCACGTGCCCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGCCGC
visualized spectroscopic probes combining aptamers with metal nanoparticles.

**Table 2** (continued)

| Nanoparticles | Spectroscopy | Pollutants’ target | Aptamer | Linear range/LOD | References |
|---------------|--------------|-------------------|---------|-----------------|------------|
| Au@AgNP      | Raman        | Ag⁺               | 5'-Rox-CTCTCGGATCTTCTCGGTCCTTTCAACACAAGATCCCACTTTTCTTCTTCTTCTT-3’ | 0.1–100 nM/50 pM | [55]      |
| Au@AgNPs     | Raman        | Hg⁺               | 5'-Cy3-TTC TTT GTT CCC CTT CTTTGTGCCC CCC CC-SH-3’ | 0.001–1 nM/0.4 pM | [56]      |
| AuNR         | LSPR         | Saxitoxin         | 5'-TAGGGAAAGAAGGACATATGATGGCAACAGGCCCTACATGAAGGTATCATCAGGTTGACTAGTACATGACCACCTTG-3’ | 5–10,000 μg L⁻¹/2.46 μg L⁻¹ | [57]      |

**Fluorescence spectroscopy probes: fast analysis, high signal-to-noise ratio**

The principle of fluorescent aptasensors is that during the binding of an aptamer to a target, the properties of the

**Fig. 2** The principle of the AuNPs colorimetric process. **a** AuNPs are uniformly dispersed under electrostatic conditions. When NaCl or other electrostatic salt solutions are added, the AuNPs aggregate and the solution turns blue–violet or blue–gray. **b** Aptamer-coated AuNPs to protect AuNPs from aggregation under salt conditions. **c** When the target pollutant is added, a part of aptamers binds to it. Some AuNPs cannot be wrapped by the aptamer, causing them to aggregate in the presence of salt conditions and the color of the solution to change to purple.
fluorescent groups present in the probes change, such as extinction coefficient, resonance energy transfer, anisotropy, wavelength [65], etc. Then, the pollutants are detected qualitatively or quantitatively based on the corresponding fluorescent signals. In the construction of fluorescent spectroscopic probes, metal nanoparticles can act as fluorophores or bursts, with the latter being the more widely investigated. Metal particles such as AuNPs and AgNPs, in addition to the regularly utilized fluorescence burst materials like graphene oxide (GO) and quantum dots (QDs), have strong energy bursting capabilities [66].

Fluorescence spectroscopy probes can be separated into fluorescence-labeled probes and label-free probes based on whether the fluorescent groups are labeled on the aptamer. Labeled probes refer to the modification of fluorescent groups directly on the aptamer. The conformational change before and after the binding of the aptamer to the target leads to the fluorescent group near or away from the surface of the metal nanoparticle, which generates fluorescent signals of different intensities [67]. However, some researchers found that the modification of fluorescent groups on the aptamer may reduce the affinity of the aptamer to the target [68], and this behavior also greatly increases the cost of the probe. Therefore, the research on fluorescence spectroscopy probes and sensors has gradually tended to be unlabeled in recent years. The team of Eun-Song Lee et al. [69] constructed an aptasensor with higher sensitivity for BPA detection based on the AuNPs colorimetric method combined with a fluorescence method. They added an exogenous fluorescent group, SYBR GREEN I (SG-I), to the system. When the fluorophore SG-I bound to the double-chain part of the BPA aptamer, it emitted an intense green fluorescence at 520 nm. When the aptamer was bound to the target, SG-I had difficulty getting close to the aptamer and the fluorescence intensity was very weak. In a higher concentration of the salt solution, the addition of the BPA led to a weakening of fluorescence intensity and aggregation of AuNPs in the system, which was detected using fluorescence spectroscopy and UV spectroscopy (Fig. 5). The method exhibited high specificity and sensitivity and allowed for a wide range of quantitative analyses. Likewise, Su et al. [70] constructed a fluorescent aptasensor for carbendazim detection using rhodamine B as a fluorescent indicator combined with AuNPs. Homogeneously dispersed AuNPs were able to burst the fluorescence of rhodamine B. When the aptamers in the system were specifically bound to carbendazim, the AuNPs aggregated in the NaCl solution and had little effect on the fluorescence intensity of rhodamine B. The fluorescence intensity was then differentiated based on the fluorescence spectrum to quantify the concentration of carbendazim.

Several dual recognition probes have also been developed to improve the detection sensitivity and selectivity of sensing platforms. Li et al. [71] developed an aptamer-MIP (Molecularly Imprinted Polymer) fluorescence aptasensor for Cd$^{2+}$ detection using a molecularly imprinted polymer and an aptamer as a dual recognition system. The synthesized carbon QDs and gold nanoparticles were coupled to the surface of an indium tin oxide glass electrode (ITO) as fluorophores. And the MIP was synthesized on the surface...
of the SN-CQD/Au/aptamer-Cd$^{2+}$ modified ITO electrode by UV-induced polymerization, followed by the removal of Cd$^{2+}$ to obtain the imprinted blank sensor. SN-CQD/Au fluorescence bursts in the presence of Cd$^{2+}$ in the system. The sensor had excellent selectivity as the aptamer and MIP contained a dual recognition effect. The detection limit was 1.2 pM and was linear over a concentration range of 20–12 pM.

Fluorescence sensing methods possess the advantages of higher signal-to-noise ratio, higher sensitivity, and faster detection, which are very promising and powerful tools in the field of pollutants monitoring. Compared with labeled aptamer fluorescence spectroscopy probes, label-free probes have less impact on the recognition ability of aptamers and are less costly, but the sensing process is more complicated. Although AuNPs have good fluorescence bursting ability in the longer wavelength range, there are few metal nanomaterials with an ideal bursting effect in the near-infrared region [32]. Therefore, more new metal nanoparticles need to be investigated for improving the fluorescence quantum yield in the near-infrared region.

**Surface-enhanced Raman spectroscopy probes: sensitive, fast, and environmentally compatible**

Raman spectroscopy is an important spectroscopic technique for chemical identification purposes that can provide fingerprint information on molecular or lattice vibrations [72], but its detection sensitivity is so low that it has limited applications. Surface-Enhanced Raman Scattering (SERS) is a spectral technology based on Raman spectroscopy. The molecule is adsorbed or close to the rough metal surface, because the strong plasmon resonance effect and local photovoltaic field effect can be generated around the metal, which can enhance the Raman signal of the molecule, and then improve the detection sensitivity [73, 74].

The SERS technique has significant advantages over other analytical techniques. (1) The narrow bandwidth of the SERS characteristic peak makes it suitable for testing and analysis in complex environments. (2) SERS techniques incorporating nanoparticle structures with specific coupling effects are highly sensitive, even to the level of single-molecule detection [75]. (3) Fast signal acquisition speed and high detection efficiency. (4) SERS-enhanced substrates can be designed in a variety of forms, including liquid substrates, rigid solid substrates, flexible solid substrates, etc., to meet a variety of environments for analysis. SERS technology has led to significant research advances in chemistry, materials science, life science, and environmental analysis.

In surface-enhanced Raman methods, the creation of Raman-enhanced substrates is crucial. Since the 1970s, when Raman enhancement of rough silver surfaces was first discovered, various types of nanostructured Raman-enhanced substrates have been extensively investigated, mainly focusing on a small number of metal materials such as Au, Ag, Cu, etc. In the vicinity of nanoparticles, especially gold and silver nanoparticle dimers or heterodimers, significant Raman signal enhancement occurs, the phenomenon is known as the "hot spot" effect [76]. As a result, AuNPs and AgNPs and their dimeric nanoparticles are the most widely studied base metal nanomaterials due to their excellent SERS properties.

Nie et al. [51] proposed a label-free surface-enhanced Raman scattering (SERS) sensor of aptamers for trace malathion residue detection (Fig. 6). AgNPs were used as Raman-enhanced substrates and modified with positively charged spermine molecules to capture the negatively charged malathion aptamer. This approach compensates for the poor biocompatibility of silver nanoparticles and their ability to adsorb aptamers. The detection of trace malathion residues was achieved by recognizing the binding of malathion molecules to the aptamer. However, it has been proved after numerous studies that gold–core–silver–shell nanoparticles (Au@AgNPs) have more significant Raman enhancement than monometallic gold nanoparticles, silver nanoparticles, and even gold–silver nanoparticles formed by simple self-assembly of gold and silver nanoparticles, as well as silver–core–gold–shell nanoparticles [77, 78]. Liu et al. [79] constructed an aptasensor for the rapid and sensitive detection of 17β-estradiol in the environment. They bonded 4-mercaptobenzoic acid (4-MBA) (Raman signal molecule) and single-chain aptamers to the surface of Au@AgNPs as Raman substrates. The 4-MBA was attached to the Au@AgNPs through the Ag–S bond, causing a "hot spot effect". When the aptamer was attached to the surface of Au@AgNPs, the "hot spot effect" would be reduced. When 17β-estradiol is introduced to the system, it binds to aptamers selectively. As a result, residual aptamers on the substrate surface are decreased, the 4-MBA "hot spot effect" is improved, and the Raman signal is boosted as well. The method had a linear detection range of 0.1 pM–10 nM with a detection limit of 0.05 pM. The team of Chung et al. [80] developed a labeled aptamer sensing platform based on surface-enhanced Raman scattering. They embedded double-stranded DNA labeled with Raman signal molecules into Au@AgNPs to construct Raman spectroscopy probes (Fig. 7). They achieved ultra-sensitive detection of BPA in tap water in the range of 100 nM–10 fM. The total detection time of the method was about 40 min, and in combination with a portable Raman spectrometer was expected to enable rapid on-site detection of environmental pollutants. Compared with the previous method, this method used a double-stranded aptamer to label fluorescein-binding nanoparticles, which makes the cost higher, but has the advantages of lower detection limit and better specificity.
Meanwhile, with the continuous development of spectroscopic probe-based sensing means, some researchers are beginning to fuse different optical sensing methods together, which can improve measurement accuracy and circumvent the limitations of a single technology. Li et al. [81] proposed an aptasensor for bimodal detection of microcystins using a combination of real-time fluorescence and surface-enhanced Raman scattering techniques. The sensing platform was based on the fact that single-stranded DNA and double-stranded DNA have different affinities to gold nanostars. The Cy3-modified aptamer was modified to the surface of gold nanostars with a complementary sequence-bound bilayer. When microcystin is present in the system, it binds to the aptamer specifically, the bilayers unspin, the Cy3 dye approaches the gold nanostar, the Cy3 fluorescence fades, and SERS is activated. The fluorescence intensity and surface-enhanced Raman intensity are proportional to the concentration of microcystin. This dual-mode detection technology is more precise and repeatable than single-mode detection.

Great progress has been made in the application of aptamer and surface-enhanced Raman spectroscopy for pollutant detection, and the combination of this method with hand-held Raman spectroscopy has great potential for in situ pollutant detection and analysis. However, there are still several issues and limitations of surface-enhanced Raman aptasensors that need to be addressed. In these sensors’ designs, the enhancement of the Raman signal is mainly achieved by the "hot spot" effect of the metal nanoparticles. However, in the preparation of metal nanoparticles, uniformity and reproducibility are not easily controlled. This results in a less uniform distribution of the hot spot effect and therefore a less stable SERS intensity, which affects the accuracy and stability of the sensor for the determination of pollutants. Second, as real samples often contain multiple contaminants, future researchers can work on developing new SERS probes that can simultaneously identify multiple targets. This will enable simultaneous analysis of multiple contaminants, thus saving time and sensor fabrication costs. As Raman signals are generated by molecular vibrations, it is also possible that signals from other molecules in the system may interfere with the detection during actual sample detection. However, the introduction of external Raman signal molecules, which are commonly used in existing studies, can effectively mitigate this interference.

**Reusable plasmon resonance spectroscopy probes: unlabeled**

The collective resonance of many free electrons present on a metal’s surface in the presence of an incident electromagnetic wave is called surface plasmon resonance (SPR). When the particle size of precious metal particles is smaller than the incoming light’s wavelength, the optical phenomena caused by the collective oscillation of metal particle-free electrons are known as local surface plasmon resonance (LSPR) [52]. Surface plasmon resonance spectroscopy probe-based aptasensors consist of a surface plasmon optical system and a molecular recognition element (aptamer). The substance is detected by the interaction of the aptamer fixed to a metal surface with the analyte in the sample, based on the refractive index change of the incident light in the surface plasmon field [82]. Metal nanoparticles,
such as gold, silver, and aluminum, have a strong localized surface plasmon effect. With the ability to quantitatively detect low-molecular-weight substances, surface plasmon resonance aptasensors have good prospects for applications in environmental detection, food safety, and clinical therapy.

Luo et al. [83] established a surface plasmon resonance (SPR) method for the detection of BPA based on the principle that salt-induced aggregation of AuNPs causes a change in the refractive index of the solution. The detection was achieved by competitive binding of small molecules causing aggregation of AuNPs, and the change in the refractive index of the solution was measured using a silver-plated u-type fiber optic surface plasmon resonance (FOSPR) probe. This method had a higher sensitivity than the colorimetric method constructed by simple visualization probes. The team of Zhang [84] combined specific aptamer reactions and nanocatalytic amplification of signals to establish a novel SPR detection method for the detection of trace arsenic ions in water, which greatly improved the detection sensitivity (Fig. 8). The study started with the preparation of gold-doped carbon dot nanosol. It could catalyze the reduction of AgNO3 to silver nanosol, which has a strong surface plasmon resonance absorption peak. The catalytic activity was inhibited when the aptamer was adsorbed on the surface of the gold carbon dots. When arsenic ions were added, the arsenic ions were bound specifically to the aptamer, the gold carbon dots were released, the catalytic activity was restored, the nanosilver in the system increased, and the SPR absorption peak was enhanced. The detection limit for arsenic ions in this method was 0.02 μg L⁻¹. Upconversion nanomaterials (UCNPs) have many advantages, such as the ability to improve the signal-to-noise ratio of the system. Xu et al. [36] prepared upconversion nanomaterials NaYF4-coated AuNPs along with aptamers as probes for the sensitive detection of mercury ions (Hg²⁺) in combination with SPR spectroscopy.

In recent years, plasmon resonance spectroscopy probes constructed based on the combination of aptamers and gold/silver nanoparticles have shown greater potential in the field of pollutant detection and analysis with their advantages of no labeling and high detection sensitivity. However, plasmon resonance scattering spectroscopy is not so widely used among all the spectroscopic means described, which may slightly limit its development and application on a large scale.

**Conclusion**

Environmental testing has always occupied an important position in the field of environmental pollution control. The detection methods of environmental pollutants are also constantly developing and updating, among which
A review of spectroscopic probes constructed from aptamer-binding gold/silver nanoparticles...

The development of biosensors provides many new ideas for rapid detection of environmental pollutant residues. This paper reviews the application of spectroscopic probes of nucleic acid aptamers with gold/silver nanoparticles or their dimers in the detection of environmental pollutants. These probes combine the unique advantages of aptamers and metal nanoparticles with spectroscopic techniques. With the popularity of portable spectrometers, they will lay the foundation for rapid and accurate, on-site real-time monitoring of environmental pollutants and have a broad application prospect.

Although some progress has been made in this type of research, there are still many shortcomings and challenges in a relatively new field. (1) The technology still has a long way to go before it can be matured and translated into commercial products. In terms of aptamers, due to the wide variety and number of environmental pollutants,
there are still many important pollutants for which specific aptamers have not been screened. Second, the ability of aptamers to recognize targets is susceptible to environmental factors such as pH, temperature, and time. (2) In the case of metal nanoparticles, the signal changes in spectroscopy-based aptasensors are largely dependent on the optical properties of the metal nanoparticles and, therefore, the synthesis of metal nanoparticles needs to be more consistent, stable, and with good reproducibility. (3) In the construction of spectral-based probes, the combination of different optical sensing modes may be able to circumvent the limitations of a single mode for better application in the detection of environmental pollutants. (4) In the sensing application of spectral probes, although a large number of studies have demonstrated their feasibility. However, most of them focus on single-factor analysis, with few studies on multi-target analysis. The complexity of the actual samples and the influence of environmental factors also impose more stringent analytical requirements on the stability and sensitivity of spectroscopic probes. In future research, these issues need to be further addressed and refined to provide better quality, reliable, and simple aptasensors for future pollutants detection areas.

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Declarations

Conflict of interest The authors declare that they have no competing interests.

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