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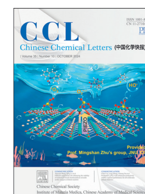

Fluorescence analysis of antibiotics and antibiotic-resistance genes in the environment: A mini review

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ABSTRACT

Antibiotics, as widely used antibacterial drug, exist in various environmental media. Antibiotic residues can affect biological metabolism and lead to bacterial resistance and the formation of antibiotic-resistance genes, posing a threat to human health and ecological safety. Establishing efficient detection methods for antibiotics and antibiotic-resistance genes has great environmental significance. Fluorescence detection methods, due to their fast response, high sensitivity and specificity, and low-cost, are widely used in chemical and biological sensing. This review first summarizes the pre-treatment methods for different types of environmental samples, and then focuses on the recent advances of fluorescence methods for the detection of antibiotics and antibiotic-resistance genes. Finally, main challenges and future research directions of fluorescence methods for antibiotic and antibiotic-resistance genes detection are discussed. This review highlights the promising prospect of fluorescence methods *in-situ* detection and monitoring of antibiotics and antibiotic-resistance genes, and provides guidance for the construction of overall risk assessment system of environmental media.

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1. Introduction

Antibiotics have been widely used to prevent or treat bacterial infections in humans and animals as well as in animal husbandry since the discovery of penicillin in 1929 [1,2]. Among the various categories, β-lactams (BLAs), fluoroquinolones (FQs), tetracyclines (TCs), macrolides (MLs), and sulfonamides (SAs) are the most common prescription antibiotics [3]. From 2000 to 2015, antibiotics consumption increased by 65% worldwide, where cephalosporins and broad-spectrum penicillins were the most consumed antibiotics [4].

It is well known that some antibiotics are easily degraded, such as penicillin, while others, such as sulfonamides and macrolides, are much more persistent in the environment, spread much farther, and can accumulate to higher concentrations [5]. When released into the environment, antibiotics can directly threaten human safety through disrupting normal metabolism and altering microbial communities [6]. On the other hand, the continuous release of antibiotics into the environment causes chemical contamination and promotes the production and transmission of antibiotic-resistance genes (ARGs) and antibiotic-resistant bacteria (ARB) [3–7]. Antibiotic resistance (AR) reduces the therapeutic potential of antibiotics against human and animal pathogens, posing a serious threat to global public health [8].

Wastewater from households, hospitals, the farm production industry [9–11], and the pharmaceutical industry [12] contribute to the ubiquity of antibiotics in the environment [13–15]. Due to the incomplete metabolism of antibiotics in humans or animals, as well as the partial degradation of antibiotics contamination by sewage treatment plants [16], large amounts of antibiotics are discharged into environmental media and widely exist in environmental media such as rivers [17], lakes [18], seawater [19], groundwater [20], and soil [21]. In 2021, researchers conducted a study on nine large-scale drinking water sources (including urban drinking water sources and township drinking water sources) in the Chongqing area of the Yangtze River and found that eight antibiotics residues were detected, with higher concentrations detected in the township drinking water sources [22]. Therefore, performing rapid and accurate *in-situ* detection is a prerequisite for effective monitoring and control of antibiotics and antibiotic-resistance genes contamination.

A variety of antibiotics detection methods have been developed in recent years including microbiological detection [23], chemical detection (chromatography, mass spectrometry, *etc.*) [24], and immunological analysis [25], which are widely used in various

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environmental media. By reviewing some of the standard methods for the detection of antibiotics (*e.g.*, China: GB/T 32951-2016, GB/T 22983-2008; USA: CLG-AMG4.02; Europe: EN 17299-2019), it is found that chromatography and mass spectrometry based on large-scale analytical instruments are the most widely used methods at present. The UHPLC-mass spectrometry (UHPLC/MS-MS), as a selective and sensitive analytical method, was developed for the simultaneous determination of fluoroquinolone antibiotics in sludge and water with detection limits at the ng/L level [26]. In another work, trace amounts of sulfadiazine can be detected in water samples collected from highly urbanized areas using UHPLC-tandem mass spectrometry with the limit of quantification being 0.40 ng/L [27]. As for ARGs, the main analytical methods are microbiological methods, PCR/quantitative PCR (PCR/qPCR) assays [28,29], and macro genomics methods [30,31], but there is no national standard currently. Ding *et al.* used high-throughput sequencing and high-throughput quantitative PCR to explore bacterial communities and antibiotic-resistance genes in soil [32]. Zhang *et al.* used macro genome assembly and crate analysis to explore potential antibiotic-degrading bacteria and their ARGs during pig manure composting [33].

In addition to the commonly used detection methods mentioned above, rapid detection methods have also been developed mainly including electrochemical methods [34], surface-enhanced Raman spectroscopy (SERS) [35], and surface plasmon resonance (SPR) [36]. Although the existing analytical techniques are capable of achieving high sensitivity and selectivity, most of them still have defects such as dependence on large instruments, and poor accuracy and stability for *in-situ* detection, which needs further improvement to fulfil their applications in practical environmental monitoring.

With the advantages of rapid, sensitive, and low-cost, fluorescence analysis is one of the most promising analytical methods currently [37–40]. The advantages of fluorescence analysis are conducive to combining it with portable detection devices. Rapid, *in-situ*, and highly sensitive analysis of the target substances can be achieved by selecting suitable fluorescence probes [41–43], which greatly expands the potential of fluorescence detection in practical applications.

Recently, some reviews discussed the applications of fluorescence analysis methods for antibiotics detection [44,45], but there is no review covers the antibiotic-resistance genes detection [46]. Moreover, most of these reviews focus on the fluorescence materials/probes [47,48] or on a specific class of antibiotics [49]. As shown in Fig. 1, in this review, we briefly summarize the pre-treatment methods for typical environmental samples, and highlight the progress of fluorescence sensors for antibiotics and antibiotic-resistance genes detection in environmental samples. Finally, the main challenges and future research direction for antibiotics and antibiotic-resistance genes detection using fluorescence methods are discussed. This review aims to enhance the understanding on the sensing capability of fluorescence sensors for antibiotics and antibiotic-resistance genes, and to highlight their feasibility as *in-situ* and real-time monitoring system for various environmental samples, which has high scientific value for the construction of a holistic assessment system for environmental media.

2. Pretreatment of environmental samples containing antibiotics and antibiotic-resistance genes

Since analytes accumulation in environmental samples is often at the micro or trace level, a series of pretreatments are often required to enrich, separate, and purify analytes in the sample before instrumental analysis. For aqueous samples, the commonly used pre-treatment techniques are liquid-liquid extraction (LLE) [50], liquid-phase microextraction (LPME) [51], solid-phase extraction (SPE) [52], magnetic solid-phase extraction (MSPE) [53,54], and solid-phase microextraction (SPME) [55,56]. While for solid samples liquid-solid extraction methods were used, mainly including pressurized fluid extraction (PFE) [57], microwave-assisted extraction (MAE) [58], and ultrasonic extraction (UE) [59]. The advantages and disadvantages of most pretreatment methods for environmental samples are listed in Table 1. While samples containing antibiotic-resistance genes require DNA extraction using DNA isolation kits before further analysis [60], but there are few studies that refer to the pre-treatment of samples containing antibiotic-resistance genes.

Solid phase microextraction (SPME) is a new sample pretreatment technique developed in 1990, which is best characterized by the integration of extraction, adsorption, enrichment, elution, and injection, and has the advantages of rapid, efficient, simple, and solvent-free [61] treatment of various types of samples. In the last decade, SPME method has been widely used in environmental sample pretreatment for both aqueous and solid samples [62]. The core of SPME is the coating material, and many novel sorbent materials have been investigated as coating material for SPME to enrich antibiotics. Using a flexible fabric substrate with a sol-gel derived hybrid organic-inorganic sorbent chemical coating as extraction medium, four penicillin antibiotic residues were extracted from milk without protein precipitation [63]. Dong *et al.* prepared carbon aerogel composites and catalyzed the carboxylation to obtain carboxylated carbon aerogels, which exhibited high extraction efficiency for six tetracyclines from egg and poultry farm wastewater samples [64].

In pretreatment, besides the utilization of one method, a combination of different methods can be adopted as required. Bajkacz *et al.* optimized the solid-liquid extraction (SLE) method and improved the clean-up procedure after solid phase extraction (SPE) by combining the two to separate antibiotics from digested manure and activated sludge samples with recoveries ranging from 45% to 85% [65].

3. Fluorescence methods for antibiotics and antibiotic-resistance genes detection

3.1. Background of fluorescence detection methods

Fluorescence was first recorded in 1565 by the Spanish physician and botanist N. Monardes [66]. In 1852, Stockes introduced the concept of fluorescence emission and derived the term "fluorescence" from the fluorescence mineral "fluorite" [67]. In 1867, Goppelsröder conducted the first fluorescence analysis study, using the fluorescence of aluminum-mulberry pigment complexes for the determination of aluminum [68]. Since the 20th century, fluorescence detection methods have been widely used and studied in environmental monitoring [69–71] and disease diagnosis [72–74].

The basic principle of the fluorescence detection is to convert information such as the concentration of the analyte into fluorescence signals using specific recognition elements for the analyte and to achieve the qualitative and quantitative analysis through analyzing the changes in fluorescence intensity, fluorescence lifetime or fluorescence emission wavelength [75–79]. The change of fluorescence signal in the sensing process is based on a variety of sensing mechanisms, including photoinduced electron transfer (PET) [80,81], Förster resonance energy transfer (FRET) [82–84], inner filter effect (IFE) [85,86], intramolecular charge transfer (ICT) [87,88], chelation induced enhanced fluorescence (CHEF) [89,90] and aggregation-induced fluorescence enhancement (AIE) [91,92]. Depending on different mechanisms, fluorescence sensing modes can be broadly classified into fluorescence quenching type (turn-off), fluorescence enhancement type (turn-on), and ratio fluorescence type (ratiometric) [93]. Ratiometric fluores-

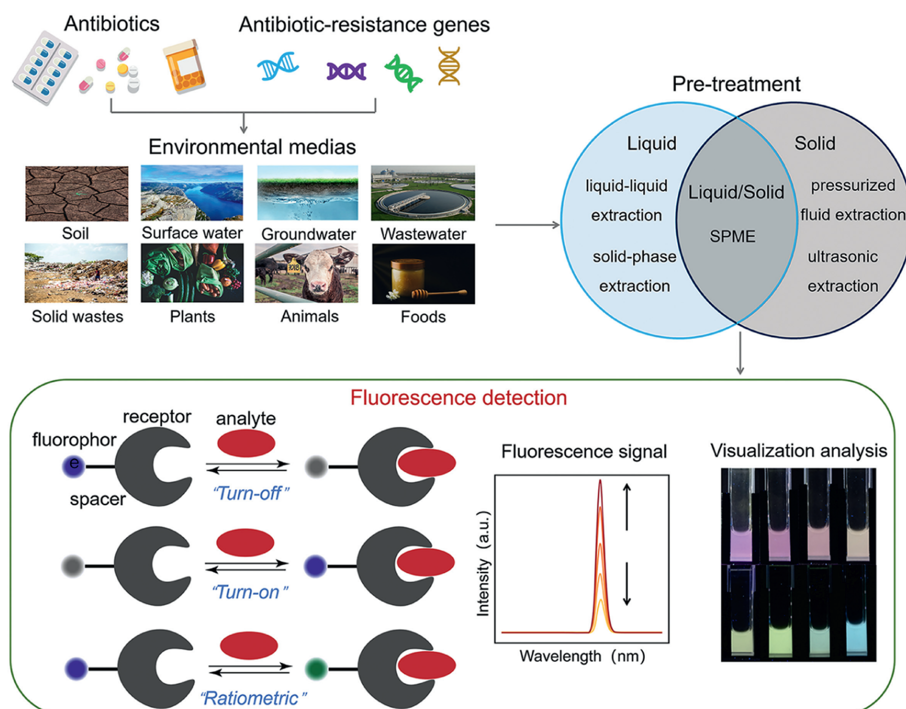

Fig. 1. Antibiotics and antibiotic-resistance genes analysis in environmental samples.

Table 1
Pre-treatment methods for antibiotics and antibiotic-resistance genes analysis in environmental samples.

Pre-treatment methods	Environmental media	Advantages	Disadvantages
SPE	Liquid	Simple operation with wide applicability	The recovery rate fluctuates greatly, and the cost of SPE columns is high
SPME	Liquid/solid	Easy to operate with minimal sample demand	The coating material has limited applicability, is prone to wear and tear, and has high cost
MSPE	Liquid	Short extraction time, multiple types of enriched substances	Not suitable for water samples with complex matrices
Disperse LLE	Liquid	Small sample volume and low amount of used organic solvent	High detection limit and time consuming
QuEChERS (quick, easy, cheap, effective, rugged, safe)	Liquid/solid	High recovery rate, low consumption of organic reagents	For samples with low water content or high-fat content, the extraction efficiency is low
PFE	Solid	High temperature for improved extraction efficiency	Require high temperature and pressure
MAE	Solid	High extraction efficiency, good reproducibility, and low generated waste	Limited extraction efficiency and ability for pollutants with weak polarity
UE	Solid	High extraction efficiency	Insufficient selectivity and sensitivity

cence can avoid the influence of environmental factors through a bimodal/multi-peak signal response, and provide richer and more obvious fluorescence color changes, which is conducive to visual detection [94]. The detection of antibiotic-resistance genes using fluorescence method mainly relies on the binding between DNA and fluorescent materials, which realizes the transduction and amplification of fluorescent signals.

Fluorescence analysis is becoming an emerging technique for the detection of antibiotics and antibiotic-resistance genes due to the advantages of rapid response time, high sensitivity, and low operational requirements [95–97]. In the following, we will classify antibiotics and antibiotic-resistance genes commonly detected in the environment, and review advances in fluorescence detection methods.

3.2. Antibiotics detection in environmental samples

3.2.1. Tetracyclines (TCs)

Tetracyclines (TCs) are natural compounds produced by Streptomyces species that were discovered by Benjamin Duggar in 1948 [98]. Due to their low cost and effective antibacterial properties, TC-based antibiotics have been successfully applied to combat bacterial infections in humans and animals [99]. TCs belong to polyketide antibiotics, which can be roughly divided into four different types, namely, tetracycline (TC), oxytetracycline (OTC), chlortetracycline (CTC) and doxycycline (DC).

In the past few years, many different types of materials such as metal-organic frameworks (MOF) [100], quantum dots (QDs) [101], and molecularly imprinted polymers (MIP) [102] have been developed and applied in the fluorescence sensors for the trace analysis of TCs. As shown in Fig. 2a, Wang *et al.* successfully synthesized an efficient bifunctional porous material (PCS-CZ-O-DCM) by combining a novel near-infrared emitting organic compound with octavinylsilsesquioxane (OVS). The hybrid material allows sensitive detection of trace tetracycline hydrochloride (LOD = 0.29 μmol/L) *via* the fluorescence quenching caused by IFE, and it can rapidly detect tetracycline hydrochloride in various solutions (artificial urine, bovine serum albumin and ovalbumin samples) [103]. Arkin *et al.* constructed a dual-channel ratiometric sensing platform (Stalk-TrpCDs@Eu) by utilizing the interaction of blue carbon

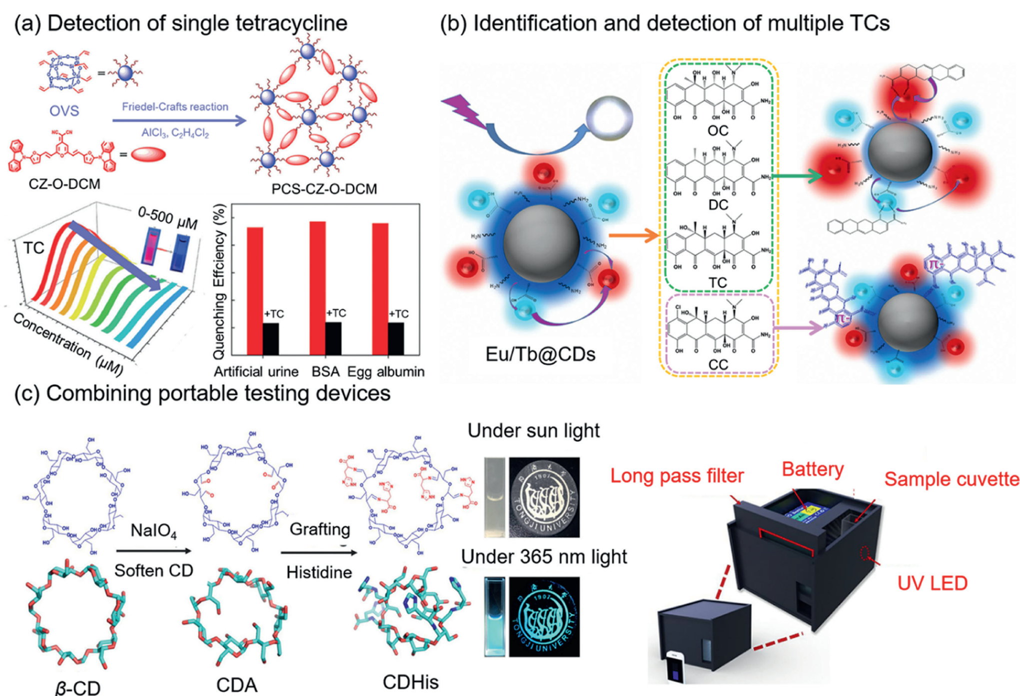

Fig. 2. Fluorescence detection of tetracyclines: (a) Fluorescence detection of single tetracycline. Reproduced with permission [103]. Copyright 2023, Wiley-VCH Verlag. (b) Fluorescence identification and detection of multiple tetracyclines. Reproduced with permission [105]. Copyright 2023, Elsevier. (c) Combining portable devices to detect tetracyclines. Reproduced with permission [109]. Copyright 2023, Springer Nature.

dots (Stalk-Trp-CDs), which have high fluorescence quantum yield, with Eu^{3+}. Under different pH conditions, OTC was detected sensitively, quickly, and selectively by using the color change caused by the IFE effect and Eu^{3+} coordination with water molecules, and the detection limit was reduced to 0.018 µmol/L. In addition, the results of OTC detection in honey and milk using the Stalk-Trp-CDs@Eu method were found to be in high agreement with the HPLC method recommended by national standards [104].

Antibiotics of the same type often have very similar structures and often coexist in real environmental samples, so it is difficult to conduct morphological recognition and quantitative detection of similar antibiotics. As shown in Fig. 2b, Che *et al.* developed white fluorescence Eu/Tb@CDs and successfully applied them to the identification and detection of tetracyclines (TC, OTC, CTC, DC) with detection limits below 1 nmol/L through IFE and AIE effects. The simultaneous morphological identification and accurate quantitative detection can be achieved in the actual water samples with four types of tetracyclines coexisting through the fluorescence sensing array structure combined with principal component analysis, which also confirms that other contaminants cannot interfere with the detection of the above four tetracyclines [105].

Due to the limitations of traditional detection methods in terms of time and space, in recent years, more and more researches have focused on the development of *in-situ* real-time monitoring methods [106,107]. Fluorescence detection methods is easy to integrate into portable detection systems due to their simple signal generation conditions and visualization capabilities, thus attracting much attention for their great application potential for *in-situ* monitoring. Li *et al.* developed an ultrasensitive luminescent lanthanide metal-organic framework (LnMOF) sensor for the immediate detection of OTC and TC in real environmental samples through the IFE effect with limits of detection (LOD) of 1.95 and 2.77 nmol/L, respectively. More importantly, sensitive test strip for the detection of OTC and TC has been manufactured, and allows easy differentiation of the presence of antibiotics using the naked eye, enabling real-time *in-situ* monitoring of OTC and TC [108]. As a low-cost fluorescence detection platform, test strips can achieve rapid visual semi-quantitative detection, but they may not be able to achieve high sensitivity and accuracy due to the inability of the naked eye to distinguish subtle color changes. As shown in Fig. 2c, Li *et al.* reported a novel aggregated nonconventional luminophores prepared by amino acid-modified cyclodextrins for rapid and specific identification of CTC in water with a detection limit of 0.012 µmol/L. In addition, they designed and constructed a smartphone device for real-time/*in-situ* visual detection of CTC, which uses a color scanning application to read the blue and green intensities (B and G values) of fluorescence images and performs quantitative detection of CTC based on the ratio of B/G values [109].

3.2.2. Fluoroquinolones (FQs)

Fluoroquinolones (FQs), the third largest class of antibiotics, account for 17% of the global market and are mainly used in the treatment of Gram-negative bacterial infectious diseases [110]. Fluoroquinolones are usually classified into first generation (cilnofloxacin and nalidixic acid (NAL)), second generation (ofloxacin, enrofloxacin), third generation (levofloxacin, moxifloxacin), and fourth generation (trovafloxacin) based on their synthesis [111].

Due to the similar heterocyclic conjugated structure, most FQs generate tight fluorescence emissions, making it impossible to be distinguished by their inherent fluorescence signals [112]. The interaction of FQs with fluorescence materials is usually used to enhance or quench the initial fluorescence for qualitative and quantitative detection of antibiotics (Fig. 3a) [113,114].

Fluorescence MOFs are considered to be one of the most promising sensing materials, with advantages such as high sensitivity and short response time for detection. Yang *et al.* synthesized new viologen functionalized Ln(III) complexes with three-dimensional structures. Norfloxacin (NOR) and ciprofloxacin (CIP) can be detected through dynamic quenching process, with detection limits of 7.90×10^{-7} and 9.48×10^{-7} mol/L, respectively. In addition, a dual-functional test paper for inkless printing media and rapid detection has been developed using the probe's excel-

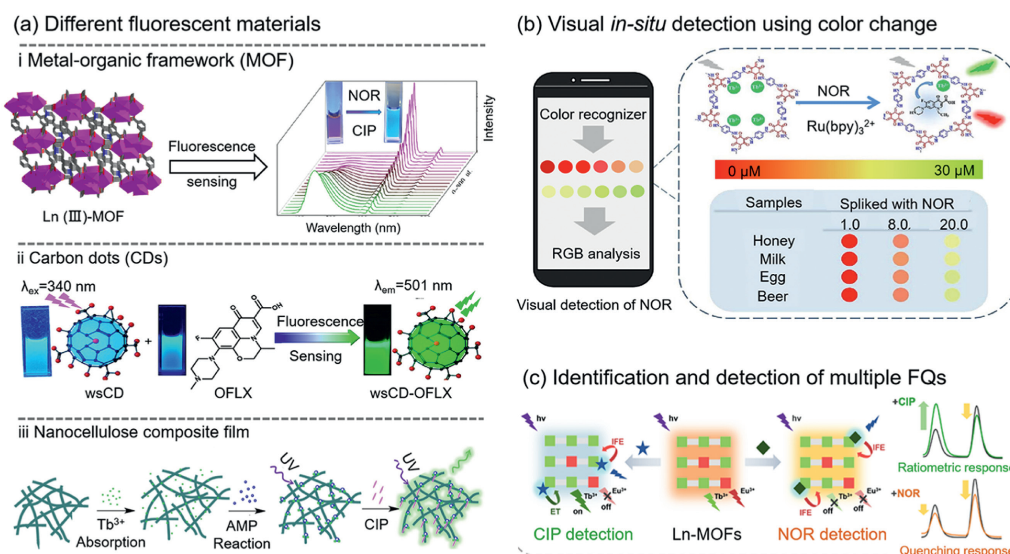

Fig. 3. Fluorescence detection of fluoroquinolones: (a) Detection of fluoroquinolones with different fluorescence materials: (i) Metal-organic framework. Reproduced with permission [115]. Copyright 2023, American Chemical Society. (ii) Carbon dots. Reproduced with permission [117]. Copyright 2023, American Chemical Society. (iii) Nanocellulose composite film. Reproduced with permission [118]. Copyright 2022, Elsevier. (b) Visual *in-situ* detection using color change. Reproduced with permission [120]. Copyright 2023, Elsevier. (c) Fluorescence identification and detection of multiple fluoroquinolones. Reproduced with permission [126]. Copyright 2022, Elsevier.

lent light response and fluorescence performance, providing a new strategy for the color recognition of antibiotics [115]. In addition to MOFs, carbon dots (CDs) have high quantum yields and are also commonly used for fluorescence sensing [116]. Aggarwal *et al.* prepared self-passivated fluorescence water-soluble CD (wsCD), which can be used for sensitive detection of ofloxacin (OFLX) through IFE effect, with the detection limit of 0.025 ng/L. The performance of wsCD in real soil and water samples was tested with spiking experiments, and the good recovery rate and relative standard deviation indicated that wsCD has the practical ability to detect OFLX as a sensor material [117]. Some novel materials have also been used as sensing materials for the detection of FQs. Wang *et al.* synthesized *in-situ* a novel fluorescence-responsive nanocellulose composite membrane doped with Tb at room temperature in aqueous media for detecting CIP. The CIP acted as an antenna to provide energy to Tb^{3+} to generate enhanced fluorescence signals with a low detection limit of 0.0392 μmol/L [118].

Accurate *in-situ* detection of FQs is of critical importance to ensure the safety of food as well as human health and to predict environmental contamination [43]. Yang *et al.* used triaminoguanidine hydrochloride (TAGH) to modulate the fluorescence of covalent organic frameworks (COFs) based on benzotrithiophene tricarbaldehyde (BTT) for real-time visualization and *in-situ* determination of enrofloxacin (ENR). With the increase of ENR, COBTT-TAGH can achieve a color shift from green to blue utilizing the changes in AIE effects. Therefore, test strips and gels were designed as real-time detection tools and combined with a smartphone application. The LODs obtained by RGB analysis were 106.2 nmol/L and 26.0 nmol/L for the test paper and gel, respectively, and the method has been successfully applied to the detection of ENR in fish and clam metabolites [119]. As shown in Fig. 3b, Zhou *et al.* proposed a lanthanide COF fluorescence sensing system (Tb@COF-Ru) using antenna effect and space confinement effect for RGB analysis *via* smartphone to achieve visual monitoring and quantitative analysis of FQs without any complex instrumentation. The detection limits of NOR fluorescence quantification and visual mode were 0.33 nmol/L and 7.3 μmol/L, respectively. The method is rapid (1 min) and intuitive, performing simple quantification analysis in various food matrices (honey, milk, eggs, and beef) and water samples [120].

The main challenge in the detection of real environmental samples is that different types or the same type of antibiotics usually coexist in the analytes, which makes it difficult to selectively analyze antibiotics with similar molecular structures [121,122]. Chen *et al.* prepared a new Tb^{3+}-ENR aptamer probe and established a sensitive, rapid, label-free fluorescence detection for three FQs (ENR, NOR, and CIP), with detection limits of 0.061, 0.020, and 0.053 ng/mL. Satisfactory recovery rates (80.10%–102.48%) were obtained by spiking honey and actual water samples, demonstrating the validity of the method [123]. Fluorescence signals are often accompanied by color changes, which means that species and concentrations of the analyte can be distinguished by color, where the Commission International Illumination (CIE) chromaticity space is commonly used to distinguish multiple targets [124,125]. As shown in Fig. 3c, Wang *et al.* constructed a dual-emitting LnMOF combining Tb^{3+} and Eu^{3+} as the luminescent centers. Due to the different sensitization effects to lanthanide metals and different inherent fluorescence emissions of FQs, the sensor exhibits characteristic color variations towards nine FQs (CIP; pefloxacin (PEF); difloxacin (DIF); ENR; enoxacin (ENO); NAL; NOR; OFLX; fleroxacin (FLE)) and enables the discriminative detection of multiple antibiotics with self-calibrated signals. For the first time, a polynomial surface fitting process is developed to correlate the coordinates of the color-coding map and target concentration for quantitative analysis [126].

3.2.3. Nitrofurans (NFs)

Nitrofurans (NFs) are broad-spectrum antibiotics that are widely used in humans and animals for the treatment of genitourinary, gastrointestinal, and surface infections, among others. For example, furazolidone (FZD) is used to treat bacterial diseases in fish farms, while nitrofurantoin (NFT) is mainly used as an oral treatment for human urinary tract infections [127–129]. The use of nitrofurans is prohibited in the EU, USA, and China due to their potential carcinogenic, teratogenic, and mutagenic properties [130].

Luminescent MOFs (LMOFs), as a novel fluorescence material, have been applied in environmental detection due to their ability to induce luminescence, rich structure, and mostly strong stability and anti-interference properties [131–133]. Wang *et al.* de-

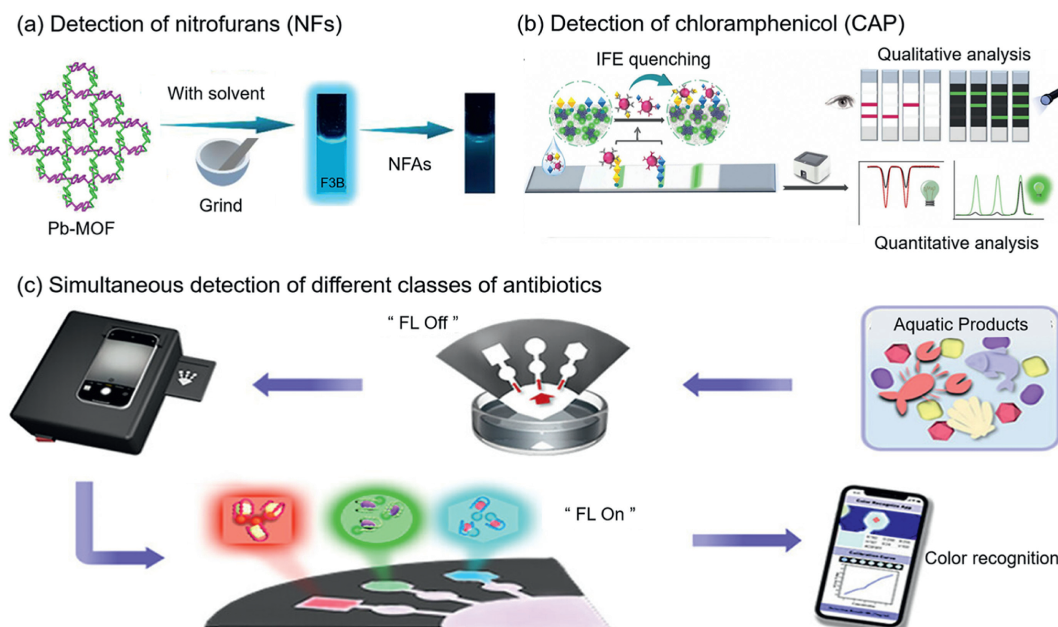

Fig. 4. Fluorescence detection of NFs, CAP, and coexisting antibiotics: (a) Detection of nitrofurans. Reproduced with permission [136]. Copyright 2022, American Chemical Society. (b) Detection of chloramphenicol. Reproduced with permission [143]. Copyright 2022, Elsevier. (c) Simultaneous detection of different classes of antibiotics. Reproduced with permission [145]. Copyright 2022, American Chemical Society.

signed and synthesized two Zr(IV)-based water-stable LMOFs (BUT-12 and BUT-13) for the detection of NFs. The fluorescence properties of MOFs are derived from the fluorescence emission of organic ligands, and a trace of nitrofurazone (NZF) can effectively quench the fluorescence of the sensor through PET and FRET, thus making the sensor highly sensitive. The detection limits of BUT-12 for NZF were estimated to be 58 µg/L, while those of BUT-13 were 90 µg/L, respectively [134]. Xian *et al.* synthesized two Zn-MOFs (LCU-113 and LCU-114) that can provide sensitive detection of NZF through IFE effect, with detection limits of 0.35 and 0.46 µmol/L, respectively. At the same time, spiking experiments were also conducted in real water samples, and the good recovery rate represents the reliability of the two Zn-MOFs in detecting real samples [135]. As shown in Fig. 4a, Liu *et al.* designed a novel lead-based anionic 2D MOF with an interlocking structure (F3B) and obtained high quantum yield (QY = 73.77%) and blue fluorescence through the solvent-protected grinding method. With the synergistic effect of PET and competitive energy absorption, F3B exhibited high selectivity and rapid response to NFs with a LOD of 0.26 µmol/L. The method has been successfully applied in the detection of NFs in actual fish tanks and pond water samples with good recovery rates and low relative standard deviations [136].

While rapid and sensitive detection of a certain nitrofurans residue is essential, it is also crucial to distinguish between coexisting similar antibiotics with similar structures. Yu *et al.* synthesized a host-guest complex (RhB@Tb-dcpcpt) that enables sensitive and selective detection of NFs (NZF and NFT) by a fluorescence quenching process caused by PET and IFE, with detection limits of 99 µg/L and 107 µg/L, respectively [137]. Yue *et al.* successfully synthesized fluorescence spindle-shaped Al(III) nanosheets containing a metal-organic framework (Al-MOF) by one-step hydrothermal method, which are used for the sensitive and selective detection of NFs through IFE effects, with detection limits of 0.53, 0.838 and 0.583 µmol/L for NZF, NFT, and FZD, respectively. The practical application of the system was verified by HPLC in spiked milk samples with a recovery of 88.14%–126.21%. In addition, fluorescence test strips were designed for the semi-quantitative detection of NFs by the naked eye [138].

3.2.4. Chloramphenicol (CAP)

CAP is a white crystalline antibiotic, which was originally isolated from streptomyces venezuelanus [139]. It can be used as an effective antibacterial agent for many Gram-positive and Gram-negative bacteria (including most anaerobic bacteria), but it may be carcinogenic and genotoxic to humans [140].

Liu *et al.* presented a ratiometric fluorescence sensing strategy based on an aptamer labeled with a fluorescence dye and a highly stable zirconium-porphyrin MOF (PCN-222) as a quencher for the high-efficiency detection of CAP. PCN-222 demonstrated a high quenching efficiency *via* FRET and PET processes. In the presence of CAP, dye-labeled aptamers were released from the PCN-222 surface, resulting in the recovery of fluorescence. The sensor can completely detect CAP within 26 min with a detection limit of 0.08 pg/mL [141]. Dong *et al.* proposed a fluorescence immunoassay based on $CaCO_3$ nanospheres and magnetic nanoparticles (MNPs) encapsulated by CDs for high sensitivity and rapid analysis of CAP in chicken, with a detection limit of 0.03 µg/kg [142].

The visualization of the fluorescence method can be used for rapid, sensitive, and economical analysis of CAP residues in the environment. As shown in Fig. 4b, Xiong *et al.* developed a novel fluorescence quenching immunochromatographic test strip (FQICTS) for the simultaneous detection of CAP and amantadine (AMD) using gold nanoparticles (AuNPs) and highly luminescent green gold nanoclusters (AuNCs) as quenching agent/donor pairs. Under optimal conditions, the LODs of the AuNCs-based dual-readout FQICTS in the “turn-on” mode were 0.043 ng/g and 0.45 ng/g for CAP and AMD in chicken samples, respectively [143]. Wu *et al.* synthesized a ternary-emission fluorescence imprinted polymer in a one-pot by the sol-gel method after mixing luminescence metal-organic framework, green CdTe, and near-infrared red CdTe for visual detection of CAP in food by PET effect. The response time of the ternary emission fluorescence imprinted sensor for CAP was 3 min and the detection limit was 3.8 pmol/L. The method was applied to the detection of trace CAP in food with recoveries of 98.2%–101.2%, which provided a new way for rapid visual detection of CAP [144].

Table 2
Fluorescence methods for antibiotics detection.

Types of antibiotics		Materials	Detection time	LOD	Portable testing	Environmental samples	Ref.
TCs	OTC	Carbon dots	3 min	0.018 μmol/L	Yes	Honey Milk	[104]
	TC OTC CC DC	Carbon dots	<1 min	TC (0.69 nmol/L) OC (0.81 nmol/L) CC (0.14 nmol/L) DC (0.54 nmol/L)	No	Real water samples	[105]
	CTC	Non-conventional luminophores	120 min	0.012 μmol/L	Yes	No reported	[109]
FQs	OFLX	Carbon dots	within seconds	0.025 ng/L	No	Real soil and water samples	[117]
	NOR	LnCOF	1 min	0.33 nmol/L (fluorescence quantification) 7.3 μmol/L (visual quantification)	Yes	Various food matrices Water samples	[120]
	CIP PEF DIF ENR ENO NAL NOR OFLX FLE	LnMOF	3 min	CIP (17 nmol/L) PEF (17 nmol/L) DIF (23 nmol/L) ENR (16 nmol/L) ENO (56 nmol/L) NAL (29 nmol/L) NOR (1.1 μmol/L) OFLX (1.0 μmol/L) FLE (0.95 μmol/L)	Yes	Tap water and river water	[126]
NFs	NZF	Zn-MOF	30 min	NZF (0.35 μmol/L)	No	Real water samples	[135]
	NZF NFT	Pb-MOF	20 min	NZF (0.35 μmol/L) NFT (0.33 μmol/L) NFZ + NFT (0.26 μmol/L)	No	Actual fish tank and pond water samples	[136]
CAP		AuNCs	20 min	0.043 ng/g	Yes	Chicken samples	[143]
		Fluorescence molecularly imprinted polymers (FMIPs)	3 min	3.8 pmol/L	No	Food samples	[144]
Multi-antibiotics	SMZ OTC CAP	Carbon dots	15 min	SMZ (0.47 μg/L) OTC (0.48 μg/L) CAP (0.34 μg/L)	Yes	Shrimp samples	[145]

3.2.5. Simultaneous detection of different classes of antibiotics

The overuse of antibiotics leads to their residues in water, soil, plants, and animals, ultimately circulating to humans through ecological cycles. In real samples, antibiotics often exist as mixtures, so multiplex detection of antibiotics is important but challenging [137].

As shown in Fig. 4c, Tong *et al.* developed a laser-printed paper-based microfluidic chip loaded with multicolor fluorescence nanoprobes (mCD-μPAD) to rapidly and *in-situ* detect sulfamethazine (SMZ), OTC, and CAP. These “fluorescence off” probes composed of CDs conjugated with aptamers (donor) and MoS_2 nanosheets (acceptor) were based on FRET. Upon the addition of target antibiotics, the significantly recovered fluorescence signal on the μPAD can be sensitively perceived by employing a 3D-printed portable detection box through a smartphone. Under optimal conditions, this μPAD showed a rapid response (15 min) toward SMZ, OTC, and CAP with considerable sensitivities of 0.47, 0.48, and 0.34 ng/mL, respectively. In shrimp samples, the recoveries were 95.2%–101.2%, 96.4%–105%, and 96.7%–106.1% with RSD below 6% [145].

Tang *et al.* proposed a smartphone-integrated trichromatic fluorescence sensing platform based on acid-sensitive fluorescence imprinted polymers for dual-mode visual intelligent detection of ibuprofen (IP), CAP, and florfenicol (FF) through PET and IFE effects, with detection limits of 10 pmol/L, 8.5 pmol/L, and 5.5 nmol/L for IP, CAP, and FF, respectively. Additionally, a smartphone was used to capture fluorescence colors and read out the RGB values for IP, CAP, and FF, in which the detection limits were calculated as 15 pmol/L, 12 pmol/L, and 7 nmol/L, respectively [146]. Geng *et al.* synthesized two new Co(II)/Ni(II) coordination polymers, CP 1 and 2, under hydrothermal conditions. They can be used as ultra-versatile fluorescence sensors for erythromycin (ERY) and oxacillin (OXC) through fluorescence enhancement caused by PET and furaltadone (FTD) through fluorescence quenching caused by FRET, PET and static quenching, separately. The sensors have a rapid luminescence response, good recyclability, and excellent fluorescence stability [147].

Table 2 summarizes the above-mentioned fluorescence detection methods for different types of antibiotics. Fluorescence detection methods can provide rapid and sensitive detection of one or multiple antibiotics residues in the environment, and can also be combined with portable detection equipment to achieve *in-situ* detection. However, current research mainly focuses on the detection of a small portion of antibiotics, and there is still a need to detect other types of antibiotics in actual environmental samples to determine their contamination levels.

3.3. Antibiotic-resistance genes detection in environmental samples

The continuous release of antibiotics into the environment causes chemical pollution and promotes the production and transmission of ARGs and ARB [7]. ARGs refer to genes that confer antibiotic-resistance (AR) to the protein products in host cells (mainly microorganisms). Once pathogens acquire ARGs, AR may spread further between pathogens and symbionts, posing a serious threat to human health [148]. Therefore, sensitive and selective detection of the species and levels of ARGs in the environment can help to develop effective mitigation strategies to enhance environmental resistance.

Table 3
Fluorescence methods for antibiotic-resistance genes detection.

Antibiotic-resistance genes	Materials	Detection time	LOD	Portable testing	Environmental samples	Ref.
tet-M	ZFPs GO	10 min	1 nmol/L	No	No reported	[151]
tet-A, *sul-1*	AgNCs	55 min	*tet-A* (0.45 nmol/L) *sul-1* (0.32 nmol/L)	Yes	Farm effluent, sewage treatment plant, and surface water	[152]
mecA, *ermC*	$[Ru(phen)_2dppz]^{2+}$	5 min	*mecA* (100 copies) *ermC* (285 copies)	Yes	MRSA in diluted broth resuspended in human serum	[153]

Note: *tet-M, tet-A:* Tetracycline-resistance gene. *sul-1:* Sulfamethoxazole-resistance gene. *mecA*: Methicillin-resistance gene. *ermC*: Macrolide-lincosamide-streptogramin-resistance gene. $[Ru(phen)_2dppz]^{2+}$: phen = 1,10-phenanthroline, dppz = dipyridophenazine. MRSA: Resistant strains methicillin resistant *Staphylococcus aureus*.

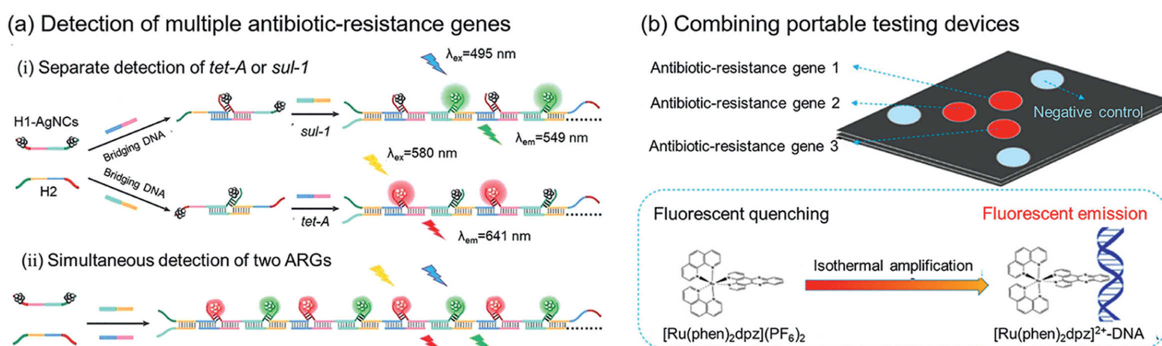

Fig. 5. Fluorescence detection of antibiotic-resistance genes: (a) Detection of multiple antibiotic-resistance genes. Reproduced with permission [152]. Copyright 2023, Elsevier. (b) Combining portable devices to detect antibiotic-resistance genes. Reproduced with permission [153]. Copyright 2018, American Chemical Society.

Although the commonly used detection methods (such as high-throughput quantitative PCR and macro genomics approaches) are very popular, they are also time-consuming and require various preprocessing steps and tedious operations by skilled personnel [149]. In contrast, the fluorescence detection method for the ARGs as an emerging technology has attracted widespread attention due to its simplicity, rapidity, and selectivity [150]. Ha *et al.* designed a simple and rapid sensing platform to detect ARGs in bacteria using engineered zinc finger proteins (ZFPs) and 2D nanosheets of graphene oxide (GO). The quantum dot (QD)-labeled ZFPs were adsorbed on the GO sheets, and their fluorescence signals were quenched *via* FRET. In the presence of the target DNA, the ZFPs bound to the target DNA induce GO dissociation, thereby restoring the fluorescence signal. This sensing system can detect the specific dsDNA of the tetracycline-resistance gene *tet-M* with high specificity after just 10 min of incubation, and provide detection limits to be 1 nmol/L [151].

The ability to simultaneously detect multiple ARGs is crucial for improving detection efficiency as well as practical *in-situ* applications. As shown in Fig. 5a, Chen *et al.* developed a novel fluorescence sensing strategy based on chameleon DNA-templated silver nanoclusters (AgNCs) to achieve simultaneous detection of two ARGs (*tet-A* and *sul-1*). The bridging of target ARGs and probes allows the formation of an infinitely extended linear DNA structure containing multi-branched AgNCs beacons with chameleon AgNCs close to the fluorescence-enhancing sequences, thus enabling the transduction and amplification of green and red fluorescence signals. This strategy successfully achieved highly specific detection of two ARGs with LODs of 0.45 nmol/L for *tet-A* and 0.32 nmol/L for *sul-1*. In addition, the strategy remains good applicability for the detection of real samples containing complex components [152]. Li *et al.* developed a paper-based chip (Fig. 5b) that integrates loop-mediated isothermal amplification (LAMP) and the "light switch" molecule $[Ru(phen)_2dppz]^{2+}$, to conduct turn-on fluorescence detection of antibiotic-resistance genes. The LOD of this paper-based assay is as few as 100 copies, and it can detect antibiotic-resistance genes in various bacteria [153].

The proliferation of antibiotic-resistance genes can lead to the emergence of antibiotic resistant bacteria, thus posing a great threat to public health. Currently, very few studies have been conducted on the detection of antibiotic-resistant bacteria using fluorescence methods. Gupta *et al.* reported a special class of water-soluble metal-based aggregation-induced emission luminogens (AIEgens), which had high selective and rapid sensing properties for endotoxins (such as lipopolysaccharides and lipoteichoic acid) that were released by the bacteria. The detected bacteria included carbapenem-resistant *A. baumannii* (CRAB) and methicillin-resistant *S. aureus* (MRSA) with a concentration as low as 1.2 CFU/mL [154].

We have summarized the above fluorescence detection methods for antibiotic-resistance genes in Table 3. The research on the use of fluorescence detection for antibiotic-resistance genes is still relatively limited, and most of them are focused on laboratory research scale Therefore, it is necessary to further explore the applications of fluorescence detection methods in detecting more types of antibiotic-resistance genes from actual environmental samples.

The development of *in-situ*, accurate, and efficient techniques for identifying antibiotics and antibiotic-resistance genes in the environment is of great practical significance to evaluate their environmental risks. Considering the demand for *in-situ* and real-time analysis, fluorescence sensing methods have great potential in practical applications due to their easy combination with portable devices, and the development of reliable, anti-interference, and portable detection solutions based on sensitive probes has great applicable prospects in the field of rapid qualitative and precise quantitative detection of antibiotics and antibiotic-resistance genes.

4. Summary and outlook

In this review, we provide a detailed description of the development of fluorescence sensors for antibiotics and antibiotic-resistance genes in environmental samples. Firstly, a brief introduction is given to the current situation and hazards of environmental pollution caused by antibiotics and antibiotic-resistance genes. Sec-

ondly, the pre-treatment methods commonly used for environmental samples are described. Finally, the advances in the fluorescence detection methods for antibiotics and antibiotic-resistance genes are summarized. Overall, fluorescence detection methods have the advantages of rapid, sensitive, and low-cost, and it can be easily combined with portable detection devices, which have the potential to provide a solution for *in-situ* monitoring of antibiotics and antibiotic-resistance genes.

Although fluorescence methods have been widely used in the analysis of antibiotics and antibiotic-resistance genes, there are still some unresolved challenges, which deserve more research attention in the future.

(1) Need more investigations about fluorescence detection of antibiotics and antibiotic-resistance genes in various actual environmental samples (water, soil, atmosphere, *etc.*).
(2) The differentiation and anti-interference ability of fluorescence detection methods should be further improved. There may be more than one type of antibiotics in the actual environment, and most of them have similar structures, which can cause significant interference.
(3) More concern should be given to new antibiotics. There are many antibiotics newly approved for use in current therapy that need to be detected urgently to determine their pollution levels.
(4) The compromising of pre-treatment and detection technology should be developed especially for solid samples (especially hazardous waste). Before harmless treatment and resource utilization of hazardous waste, such as mycelial fluids/dregs, it is necessary to detect its pollution.
(5) More advanced portable devices should be developed. The commonly used methods are test strips and smartphone-based camera devices, but the accuracy of these color-based and visual recognition methods still needs to be improved.

Declaration of competing interest

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

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Supplementary materials

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