



Organic pollutant sensing for human health based on carbon dots

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ABSTRACT

Organic pollutants are harmful and toxic chemical substances that adversely threaten human health and the living environment all over the world. More and more studies have been investigating the relationship between low level of human exposure of organic compounds and various internal diseases. For the sake of assessing disease risk due to organic compounds contact in a particular location, it is imperative for relevant government departments to make a human health risk assessment in view of the organic pollutants' bioavailability and their dosage-response correlations. It is inevitable to make use of an efficient method to detect organic pollutants, which is significant for public health and safety. Fluorescent assays based on carbon dots thus would provide a very plausible candidate method. After consulting a large number of literatures, we offer a comprehensive review of the sensing applications of carbon dots for organic pollutants.

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1. Introduction

With the fast development in economy and civilization, an increasing highlight has been proposed on sanitation survey and human health which are unfavourably influenced by critical environmental pollution. Organic pollutants are a kind of typical organic compounds which have strong resistance to natural degradation by biological, photolytic, and chemical processes to some extent. The chemical substances are toxic and harmful to human life and the ecological environment all over the world. Owing to their easy transportation by water and wind, most organic pollutants engendered in one country can have an adverse effect on people far away in another country once they are released. Organic pollutants mainly include pesticides, pharmaceuticals, solvents, and industrial chemicals. Although a portion of organic pollutants come from the nature (such as from volcanoes), the more are artificially synthesized or extracted [1].

Exposure to organic pollutants over a period of time would result in developmental defects, cancer, obesity, chronic diseases, diabetes, and death. Many organic pollutants can cause endocrine

disorders within the central nervous system, the reproductive system, and the defensive immune system, *etc.* [2]. Human beings have contact to organic pollutants mainly *via* occupationally, daily diet, or when growing and developing in the uterus. As for people without direct exposure to organic pollutants *via* occupational or accidental ways, more than 90% of exposed cases are from foods made with animal product because of bioaccumulation *via* the food chain. Generally, organic pollutant serum content would be augmented as people get older and incline to become richer in females group than that in the males [3].

Many studies have been conducted on the correlation between exposure level of organic pollutants and the development of various diseases. For the sake of evaluating disease risk from organic pollutants at a specific location, the relevant government departments could put forward a public health risk evaluation which accommodates the organic pollutants' bioavailability and corresponding dosage relationships [4]. In response to environmental degradation, it is imperative to make use of an efficient method to detect organic pollutants, which is significant for public health and safety. Carbon dots (CDs) have extensive and good application prospects in many kinds of fields, particularly in fluorescence imaging and the determination of organic pollutants, remains an interesting research object [5-7]. Fluorescent assays based on CDs thus would provide a very plausible candidate method [8-11]. Many traditional methods, such as

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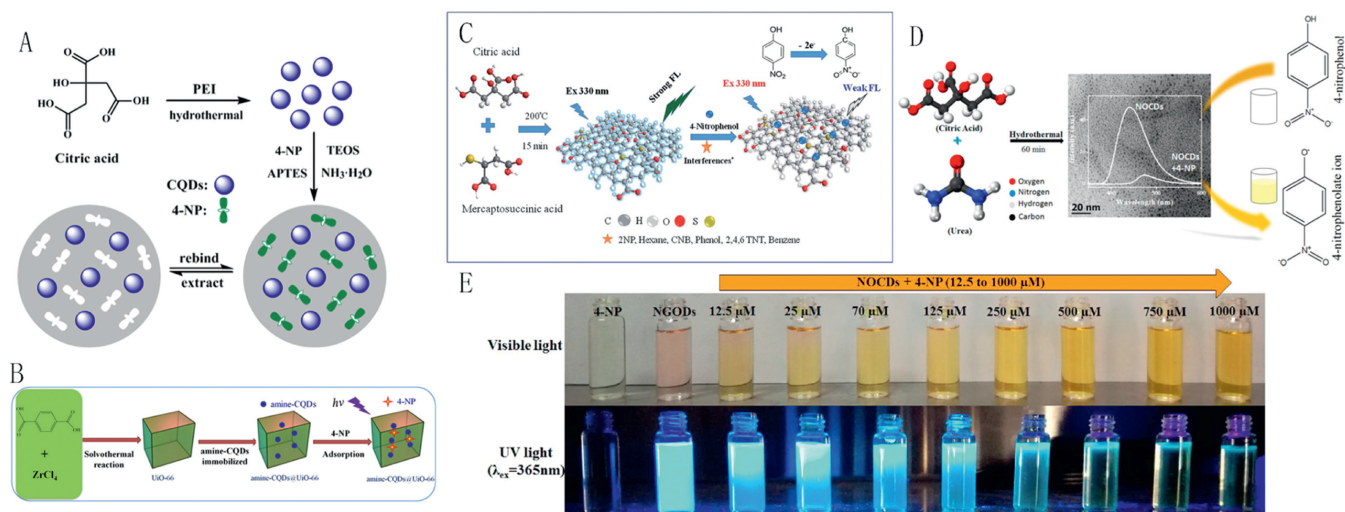


Fig. 1. (A) Scheme for the production of MIP-CDs. Reproduced with permission [14], Copyright 2016, Springer. (B) Scheme of the fabrication of amine-CQDs@UiO-66 composite along with its application to sense 4-NP. Reproduced with permission [15], Copyright 2018, Elsevier B.V. (C) Sketch of the sensing of 4-nitrophenol by fluorescent S-doped GQDs with different matrices. Reproduced with permission [16], Copyright 2019, The Royal Society of Chemistry. (D) 4-Nitrophenol fluorescence assay with N doped CDs. (E) Optical photos of NOCDs incubated with various concentrations of 4-NP solution under visible and UV light. Reproduced with permission [17], Copyright 2019, Elsevier B.V.

capillary electrophoresis, ion chromatography, mass spectroscopy, electrochemical methods, acoustic wave microsensors arrays, gas chromatography, enzyme-linked immunosorbent assays, wireless sensors, electro-chemiluminescence (ECL) method and high performance liquid chromatography (HPLC) have been adopted to sense organic pollutants [12–25]. Nevertheless, these methods are limited by pricey reagents, long time consuming, cumbersome specimen pretreatment, complex operations of professionals and potential generation of secondary pollution [26–30]. Compared to the traditional methods, CDs-based fluorescent detection methods exhibit superior features, including but not limited to high sensitivity, low toxicity, benign biocompatibility, robust photostability, facile preparation, low cost, easy surface functionalization and good water dispersity [31–40]. Herein, we offer a comprehensive review of the sensing applications of CDs for organic pollutants as following.

2. Common industrial chemicals

2.1. Nitrophenol

4-Nitrophenol (4-NP), has ever been regarded as the prior environmental pollutant by the U.S. Environmental Protection Agency, which could induce massive destruction to ecological environment and human health, due to its wide use in the preparation of pesticides, leather, explosives, dyes and drug as a common intermediate. It would accumulate in various organisms *via* food biological chains, causing long-lasting impairment [12,13]. Thus, it is very important to develop highly effective test methods for the measurement of the 4-NP.

Yan *et al.* developed a CDs based material, using environmentally friendly molecularly imprinted technology *via* a facile sol-gel polymerization process for specific fluorescence assay of 4-NP (Fig. 1A). Firstly, the amino-functionalized CDs were prepared by a hydrothermal method with poly(ethyleneimine) as the surface emollient, and subsequently going through a molecular sol-gel imprinting process, and finally the molecularly imprinted polymers (MIP)-CDs with fluorescence property were acquired. The MIP-CDs displayed bright fluorescence arising from CDs and has highly selective response to 4-NP. After optimizing the detection conditions, the relative fluorescence intensity F_0/F of MIP-CDs displayed a nice linear working curve vs. 4-NP concentrations in the concentration range of 0.2–50 $\mu\text{mol/L}$, whose detection limit was 0.06 $\mu\text{mol/L}$.

This approach was applied to determine a trace of 4-NP in Yangtze River water samples, which had decent recoveries (92.6%–107.3%). This study shows a common strategy to prepare materials taking advantage of CDs whose fluorescence property performs well for detecting organic pollutants in a selective fluorescence manner [14].

Zhang *et al.* reported a practicable tactic to fix amine-modified carbon dots (amine-CQDs) into the cavities of UiO-66 ($\text{Zr}_6\text{O}_4(\text{OH})_4(\text{bdc})_6$) to detect 4-NP through post-synthetic modification strategy (Fig. 1B). The response performance in terms of sensitivity and selectivity were ameliorated in two aspects; on the one side, the UiO-66 composite could capture target molecules massively, thus enriching the targeted substances specifically. On the other hand, fluorescent amine-CQDs, serving as functional monomers, were able to efficiently sense weak chemical force between UiO-66 and the target tested molecules. Therefore, a fast and specific detection means utilizing the amine-CQDs@UiO-66 fluorescent nano-probe was built for the sensitive assay of a trace amount of 4-NP in the linear concentration region of 0.01–20.0 $\mu\text{mol/L}$ with a rather lower detection limit 3.5 nmol/L. Its application development of MOF-based composite (amine-CQDs@UiO-66 composite) indicates its promising prospect in the toxic substance analysis [15].

Doong *et al.* developed a new and facile synthesis strategy to sensitively and selectively sense 4-NP in wastewater specimens under various matrices (Fig. 1C). The sulphur doped GQDs (S-GQDs) were with uniformly distributed diameter ranging from 1 nm to 5 nm, and could fluoresce with peak around 450 nm. The doping of S element improves the fluorescence emission quantum yield (QY). The response mechanism was built on the π - π interaction between 4-NP and the “huge” graphitic carbon planar backbone of GQDs, in which 4-NP could weaken the fluorescence. The limit of detection (LOD) of GQDs was 3.5 nmol/L in wastewater and 0.7 nmol/L in purified water respectively. Meanwhile, 4-NP can be detected in various matrices of water, avoiding the matrix interference. This method would pave an insightful way to prepare GQD-based sensor to detect nitroarenes in solution with different matrix effects [16].

Agarwal *et al.* reported a facile bottom-up approach which is affordable and readily available for 4-NP fluorescent and colorimetric sensing with highly fluorescent nitrogen doped oxidized carbon dots (NOCDs) (Figs. 1D and E). The addition of minute quantities

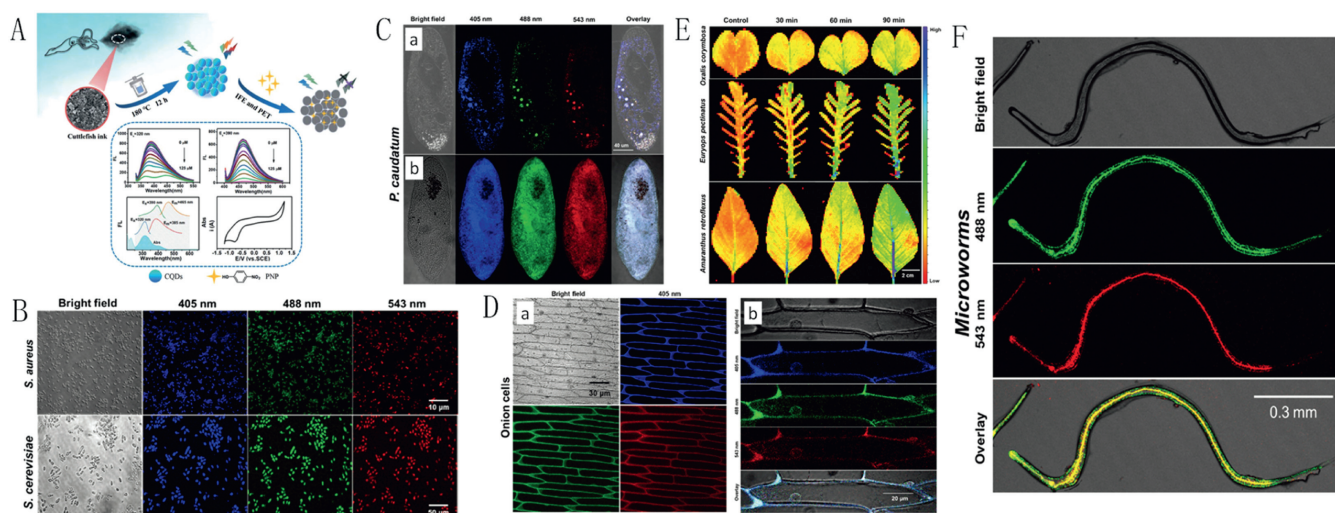


Fig. 2. (A) Schematic illustration for detecting PNP fluorescently with the N,S-CQDs made from cuttlefish ink. Reproduced with permission [18], Copyright 2021, Elsevier Inc. (B) Confocal laser scanning images of *S. aureus* and *S. cerevisiae* treated with CDs-KMR3 under excitation wavelengths 405, 488 and 543, respectively. (C) Fluorescence imaging of *P. caudatum* stained with CDs-KMR3 of 50 and 100 $\mu\text{g}/\text{mL}$ respectively under various excitation wavelengths. (D) Fluorescence imaging of onion cells after incubation with CDs-KMR3. (E) Fluorescence imaging of *Euryops pectinatus*, *Oxalis corymbosa*, and *Amaranthus retroflexus* injected with CDs-KMR3 under different time intervals. (F) Fluorescence imaging of CDs-KMR3 injected microworms. Reproduced with permission [19], Copyright 2021, Royal Society of Chemistry.

of 4-NP into the aqueous NOCDs solution would quench the fluorescence, which was ascribed to the reduction ability of 4-NP to NOCDs. The formation of reduced carbon dots (rNOCDs) was validated with X-ray photoelectron spectroscopy. The sensitivity performance test of NOCDs to 4-NP was carried out in spiked tap water, which had the minimum LOD of 2 $\mu\text{mol}/\text{L}$ in the linear concentration range of 2–100 $\mu\text{mol}/\text{L}$. The developed facile sensing platform could be applied to consume NOCDs and detect low concentration level of 4-NP simultaneously [17].

Zhou *et al.* developed a highly acute and specific fluorescent detection strategy for 4-NP with special cuttlefish ink as the raw materials for the synthesis of CQDs (Fig. 2A). The eumelanin rich cuttlefish ink was applied as the unique ingredient to prepare the CQDs by means of a one-pot hydrothermal approach. The obtained CQDs thus contained the considerable elements of nitrogen and sulphur, and showed admirable fluorescence properties. The fluorescent CQDs possessed two optimal emissions at the fluorescence emission wavelengths of 385 and 465 nm, separately. Upon the addition of 4-NP, these two emission peaks would be markedly quenched. 4-NP can be quantitatively detected in the range of 1.25–50 $\mu\text{mol}/\text{L}$ at emission 385 nm or 1.25–27.5 $\mu\text{mol}/\text{L}$ at emission 465 nm linearly with a LOD of 0.05 $\mu\text{mol}/\text{L}$. Important, when $\log(I_{385\text{ nm}} + I_{465\text{ nm}})$ was applied to quantitatively detect 4-NP, with linear response range 0.05 $\mu\text{mol}/\text{L}$ to 125 $\mu\text{mol}/\text{L}$, and the detection limit could be 0.039 $\mu\text{mol}/\text{L}$. The existence of inner filter effect and photoinduced electron transfer between 4-NP and N,S-CQDs was held responsible for the fluorescence quenching. The detection method is triumphantly applied to quantify 4-NP content in real water specimens, which provides another sustainable waste-prevention method for 4-NP [18].

Wei *et al.* successfully synthesized a kind of fluorescent CDs hydrothermally, taking *S. marcescens* KMR-3 as the unique raw materials. The prepared CDs-KMR3 had merits of nice solution dispersibility, favorable stability in different solution, excellent biocompatibility, and wide emission range. The CDs were effectively applied to image various kingdoms in biological taxonomy including fungi (*S. cerevisiae*), bacteria (*S. aureus*), protozoa (*P. caudatum*, Kingdom Protista), plant cells (onion cells), plant leaves (*Euryops pectinatus*, *Oxalis corymbosa*, and *Amaranthus retroflexus*), mammalian cells (HeLa cells) and lower animal (microworms) (Figs. 2B–F). In the meantime, the CDs were able to exclusively fluoresce-

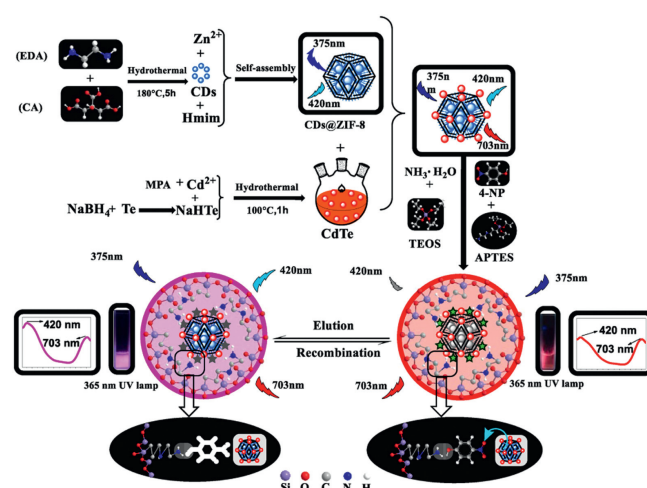


Fig. 3. Scheme of preparation process of CDs@ZIF-8/CdTe@MIP. Reproduced with permission [20], Copyright 2022, Elsevier B.V.

cently light the membranes of all above biological samples, with low biotoxicity cells. Additionally, the distinctive CDs-KMR3 was directly utilized to rapidly and selectively sense 4-NP. This work contributed greatly to the expanded foreground of bacteria-based CDs in the fluorescent bio-imaging and sensing of environmental pollutant 4-NP [19].

An imprinted nano sensor endowed with dual-response ratiometric fluorescence response established on visible/near-infrared emission was fabricated by Zhang *et al.* for ultrasensitive measurement of 4-NP selectively and visually (Fig. 3). The molecularly imprinted polymer, with the purpose of special recognition toward 4-NP was attached in the system comprising carbon dots@zeolitic imidazolate framework-8 (CDs@ZIF-8) with visible emission and cadmium telluride (CdTe) based quantum dots with near-infrared emission. The ZIF-8 was of huge specific surface area and rich porosity, which would enhance the emission of CDs by inhibiting the aggregation to avoid the self-quenching, and lower the detection limit by concentrating the analyte. The ratiometric sensor displayed dual fluorescence response to 4-NP at respective 420 nm

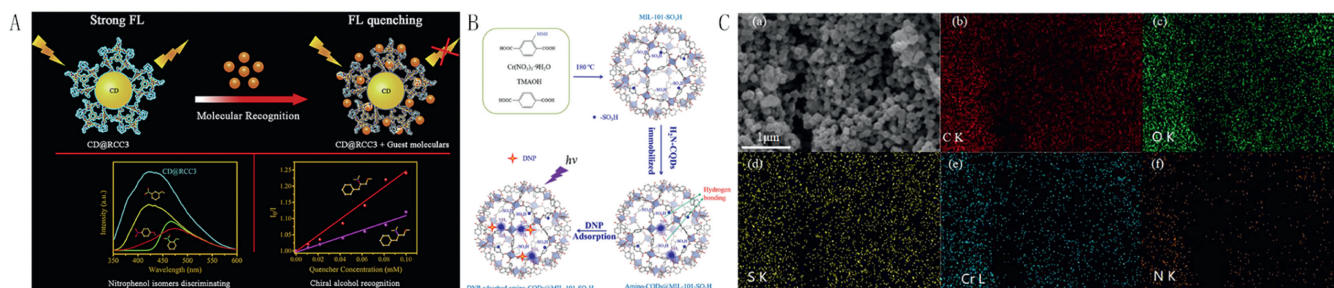


Fig. 4. (A) CD-embellished porous organic cage as nano-probe for quick discrimination of chiral alcohols and nitrophenol isomers. Reproduced with permission [21], Copyright 2019, Elsevier B.V. (B) Scheme of the fabrication of fluorescent amino-CQDs@MIL-101-SO₃H composite and its DNP adsorption. (C) Energy-dispersive X-ray mappings images of amino-CQDs@MIL-101-SO₃H: scanning electron micrograph (a) and elemental distributions of (b) C, (c) O, (d) S, (e) Cr, and (f) N. Reproduced with permission [22], Copyright 2021, Elsevier B.V.

and 703 nm. The near-infrared ratiometric response would eliminate the environmental interferences and improve the detection accuracy by self-calibration of the two fluorescence channels. What is more, there were two concentration ranges of good linear response for 4-NP, 0.1–3.0 pmol/L and 0.05 μmol/L–30 μmol/L with the LOD of 0.08 pmol/L and 0.05 μmol/L, respectively. Prominently, the fluorescence alterations could be spotted visibly from purple, pink to red using the naked eye observation. The fluorescence extinguishment mechanism was attributed to the electron transfer of the nanoprobe CDs@ZIF-8/CdTe@MIP and 4-NP, where the NH₂-from CDs@ZIF-8/CdTe@MIP interact with hydroxyl and nitro group from 4-NP. Its 4-NP detection with this method displayed the nice recoveries of 97.5%–106.3% in different real samples, thus providing an interesting approach for pollutant detection in real environment [20].

Isomers discrimination has been one of the most vital fields in modern analytical chemistry, pharmaceutical and biological science, and provision of effective and rapid strategy to accomplish this objective has aroused great attention. In Zhang's work, a new CD inlaid chiral hybrid nanocomposite called CD@RCC3 with porous organic cage structure was fabricated and applied to devise fluorescent sensor (Fig. 4A). CD@RCC3 displays excitation-dependent fluorescence phenomenon. The study of fluorescence and circular dichroism spectrum of the resultant CD@RCC3 showed that CD@RCC3 owns stable and brilliant fluorescent characteristics in frequently-used organic solvents, in particular it displays chiral property. The passivation by RCC3, resulting in decrease of nonradiative decay, was probably responsible for the higher QY of CD@RCC3 than that of CD. The latent application in fluorescence sensing of CD@RCC3 was illustrated by identification. The devised nano-sensor was then used for quick discrimination to nitrophenol isomers successfully. Besides, it was able to differentiate phenylalaninol and phenylethanol enantiomers. Fluorescence lifetime test indicates that the enantio-recognition processes of chiral alcohols of CD@RCC3 belong to static quenching. Their study enriches the variety of fabricating nano materials for organic pollutants sensing fluorescently, and supplies a facile approach for differentiating structural and chiral isomers [21].

2,4-Dinitrophenol (DNP) is another environmental pollutant, which is used in the manufacture and discharged in the industrial effluent. The development of ultrasensitive detection for DNP is imperative for human health. Based on the fact that composites decorated with metal-organic framework would contribute to surmounting the functional defects of single materials as a result of the synergism of the components, Yang *et al.* developed an amino-CQDs@MIL-101-SO₃H composite with fluorescence by decorating amino-modified carbon quantum dots on MIL-101 nanoparticles with sulfo-modification, which could serve as a sensor to sensitively detect DNP (Figs. 4B and C). MIL-101-SO₃H not only had the function to package the amino-CQDs into the pore cav-

ity *via* bonding interactions between the -NH₂ groups from the amino-CQDs and -SO₃H groups of MIL-101-SO₃H as a carrier, but also had great ability to enrich the DNP on the composite *via* the hydrogen bonding interactions between them. The composite amino-CQDs@MIL-101-SO₃H had the fluorescence emission peak at 440 nm, which originated from the original amino-CQDs. Considering the phenomenon that DNP could extinguish the composite fluorescence selectively and efficiently, the composite had the great potential to sense DNP, with a LOD of 0.041 μmol/L. The response time 10 min was short, which came from the strong driving force of DNP to adsorb on the composite. This is a pioneering work to suggest the decoration of amino-CQDs in mesoporous MIL-101-SO₃H fluorescently. The selectively fluorescent detection indicated in this work will open a new mind for testing organic hazardous pollutants [22].

2.2. Volatile organic compounds (VOCs)

VOCs with low boiling point are extensively used in agriculture, industries and appear in daily life, therefore it is unavoidable for human being to be exposed to VOCs atmosphere. Nevertheless, many VOCs are flammable and harmful to human body due to their toxicity and carcinogenicity even at quite low concentrations level. So, sensitive, real-time detection of VOCs quantitatively are very significant to human safety and environmental health [23].

In Xie's study, amphiphilic CDs with intense solvatochromism property were developed for VOCs sensing. CDs were prepared using (Z)-4-(2-cyano-2-(40-(diphenylamino)-[1,10-biphenyl]-4-yl)-vinyl)benzotrile and polyethylene glycol with molecular weight of 2000 (PEG_{2k}) as precursors (Fig. 5A). As-resultant CDs display benign solubility in both polar media (for example acetonitrile, dimethylformamide, ethanol, methanol, water, and acetone) and apolar media (such as ethylacetate, dichloromethane and tetrahydrofuran). Meanwhile, the CDs exhibited highly fluorescent QY in apolar (30.7% in dichloromethane) and polar (19.4% in water) solvent. The tunable photoluminescence (color changes from blue, to green, to pink, and to orange) of the CDs in different media endows the CDs with ideal fluorescent sensing property for the VOCs detection (Fig. 5B). The CDs solution could be casted on the strip paper conveniently to prepare test paper. This type of solid device was able to serve as an optical sensor and smell out different VOCs rapidly through different fluorescence color changes. The CDs in this study shows the tremendous application potential for the on-site assay of many VOCs, which does not need to seek complicated processes or equipments [24].

Acetone is another most extensively used and controlled organic solvent in the pharmaceutical, chemical, and cosmetic research laboratories and industries. It is a rather volatile, water-soluble with irritating smell and could do harm to human health. It is very flammable, toxic and hence hazardous as well, once it

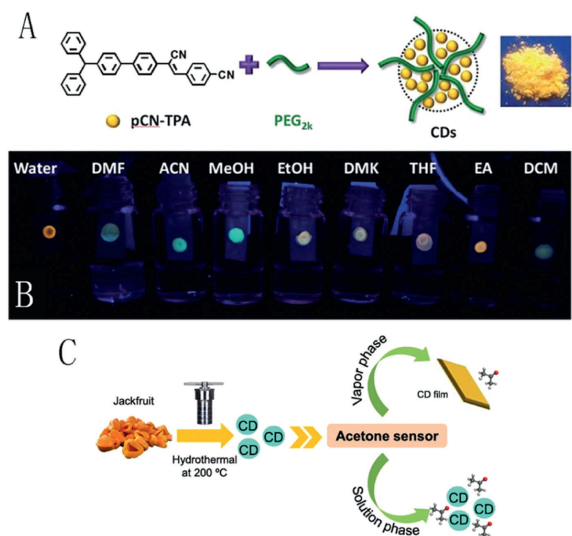


Fig. 5. (A) Fabrication route of CDs. The digital image as inset of CDs shows their solid state under 365 nm illumination. (B) Images of the strip as paper sensor printed with CDs upon the exposure to the vapor of different solvents under the 365 nm UV lamp. Reproduced with permission [24], Copyright 2016, Royal Society of Chemistry. (C) Schematic illustration of the preparation and application of CDs from jackfruit. Reproduced with permission [28], Copyright 2019, Elsevier B.V.

is leaked out. Time lasting exposure to or high concentrations of acetone would lead to dryness of the skin, irritation, cutitis, fatigue, cephalalgia, nausea, kidney and liver dysfunction, and even lethal case if the acetone level is over 1000 ppm [25,26]. Except for safety surveying, acetone vapour assay is helpful for health examination objective. Patients with obesity, diabetes, or hyperglycemia, have a disposition to express a high level of acetone concentration in their exhaled gas [27]. Hence, detection of acetone is beneficial to a series of biomedical and analytical chemical fields. The quantification of gas samples is especially challenging when the gas concentrations are ultralow. Paoprasert *et al.* used jackfruit to prepare CDs, which was then processed as a sensing layer of an electronic nose to detect acetone vapour at room temperature in real time (Fig. 5C). The CDs exhibited blue fluorescence with excitation wavelength-dependent emission, admirable photostability, and a QY of 5.2% in water. Based on component analysis strategy, the CDs-integrated optical electronic nose could differentiate acetone from ethanol, hexane, water, and methanol and quantify various content of acetone in aqueous and ethanol solutions. The optical electronic nose data agree well with the increase results in CDs' extinction coefficients in bulk solvents, which were supported by the time-dependent density functional theory calculations. Interestingly, the special ambipolar and optoelectronic properties of CDs were demonstrated to be able to serve as both electron donor and electron acceptor, according to the frontier molecular orbital calculations. In addition, the CDs could detect acetone fluorescently in aqueous solutions. Their jackfruit-resultant CDs were illustrated to be multifunctional substances for acetone sensing in solution and vapor, and their theory calculation means pointed out the significance of interfacial electronic coupling to endow CDs with special sensing properties [28].

Through a hydrothermal method, Xia *et al.* synthesized a fluorescent nano-sensor MCM/CQDs for acetic acid (HAc) successfully by means of synthesizing CQDs within MCM-41 materials. There were some differences of CQDs produced between in MCM-41. First, the fluorescent and ultraviolet-visible spectroscopy (UV-vis) spectra MCM/CQDs deviated from those of CQDs. Second, the CQDs and the MCM/CQDs have a contrary fluorescence response to HAc. Both aspects indicated special fluorescent properties of

the MCM/CQDs, owing to the mesoporous structure of MCM-41. The produced MCM/CQDs integrated the beneficence capability of MCM-41 and the fluorescence of CQDs, enabling the fast and sensitive assay of acidic gases with the MCM/CQDs fluorescently. The means used for the synthesis of MCM/CQDs in this study is for other CQDs to be embedded into mesoporous structure with new potential sensing applications [29].

2.3. Other industrial chemicals

Perfluorinated compounds (PFCs), a kind of fully fluorinated compounds, primarily consist of hydrophilic end and hydrophobic carbon chain groups [30]. PFCs have been extensively applied in civil and industrial fields, including adhesives, firefighting foams, shampoos, refrigerants, cleaners and since floor polishes 1950s [30-33]. In the PFCs family, perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) are the typical representatives, which have been most widely studied [34,35]. PFOS, as a widely persistent organic pollutant, could get into the atmosphere, water, sludge, soil and other forms of environmental media. PFOS is strongly resistant to hydrolysis, photolysis and biodegradation in the natural environment, which is very hard to be broken up once entering the environment [36]. Meanwhile, PFOS can enter the bladder, liver and serum of human beings, which would bring harm to the body such as hepatotoxicity [37,38], immunotoxicity [39] or neurotoxicity [40]. Some companies such as 3M have even asserted to give up the production of PFOS in view of its serious and unfavorable environmental pollution for human health [41]. Hence, it is very significant to develop a facile PFOS analysis technology. A sensing means for fluorescent turn-on measurement of PFOS visually was devised by Tan *et al.* Berberine chloride hydrate is able to turn off the fluorescence of CDs upon generating a ground state complex, through the electrostatic interaction and the inner filter effect. When PFOS was put into the CDs-BH solution, there would be an apparent fluorescence enhancement at 533 nm. As a result, the blue fluorescence changed to light yellow fluorescence remarkably. The new design platform is convenient, rapid and selective with low cost to distinguish PFOA from PFOS. Moreover, the proposed strategy was used to the detect PFOS successfully in environmental water specimens with favorable results, suggesting satisfactory potential for assessing the environmental governance process of harmful PFOS in the water specimens [42]. Some synthetic organic dyes are often toxic emerging organic pollutants released in to environmental water in agricultural and textile industries, posing a serious threat [43,44].

Agarwal *et al.* synthesized a sort of N doped CDs (NOCDs) which have excitation wavelength independent fluorescent behaviors one-step hydrothermally. Their fluorescent NOCDs showed a wonderful sensing property with sensitive detection towards toxic dyes like bromophenol blue, methyl orange, rhodamine 6G, and methylene blue with a LOD of 2.0 $\mu\text{mol/L}$, 38.3 nmol/L, 1.0 $\mu\text{mol/L}$ and 1.5 $\mu\text{mol/L}$, respectively. The fluorescence of NOCDs has strong dependence on temperature and pH. After incubation with NOCDs from 0.1 $\mu\text{g/mL}$ to 1000 $\mu\text{g/mL}$, the CDs indicated nice biocompatibility. The high sensitivity of the prepared NOCDs provides probable chances from laboratory research to practical sample detection applications to devise them as promising nanoprobe to fluorescently sense and image toxic organic dyes [45].

Nitrogen-containing organic alkali compounds, for instance, aromatic amines, aliphatic amines, five-membered heterocyclics, six-membered heterocyclics, amino acids, fused-ring heterocyclics, and antibiotics have been extensively applied in chemical, agricultural, biomedicine, and other industries [46,47]. These nitrogen-containing organic bases are not just vital semi-finished fine chemical material, and also have various biological activities. For example, spermine, as a kind of natural polyamine, has a signif-

icant function in cell function of growth and proliferation [48]. Nitrogen-containing organic bases could also bring serious issues of environment pollution, food contamination, and adverse biological reactions if they were not appropriately used and monitored because they can be accumulated in body once released into the environment. Hence, it is necessary to find a reliable sensing way for the sensing of these nitrogen-containing organic bases compounds. Peng *et al.* fabricated sensors which have both colorimetric and spectral sensors in solution and test paper for the rapid and sensitive assay for common nitrogen-containing organic bases; the LOD could arrive at as low as 0.50 nmol/L. Nitrogen-containing organic bases included 11 samples, including aromatic and aliphatic amines, antibiotics, five- to six-membered heterocyclics, amino acids, and fused-ring heterocyclics. Moreover, the detection manifested high reliability in sensing nitrogen-containing organic bases and splendid ability to distinguish nitrogen-containing organic bases from other heteroatomic atoms such as oxygen and sulfur containing organics. The assays developed could find important applications for the detection of nitrogen-containing organic bases in the fields of chemistry, biomedicine, and agriculture [49].

3. Insecticide

In virtue of their efficient advantage for insect extermination, cheap cost, and easy synthesis, organophosphorus compounds (OPs) have already been widely employed as pesticides or insecticide in modern farming, forestry, animal husbandry, and fishery *etc.*, which are also possible to be used as chemical warfare agents negatively in military actions or terrorist attacks. Around a billion pounds of OPs flow into the atmosphere, soil, water supplies and food globally each year, causing tremendous hidden dangers to human health and the environmental ecosystem. As a result, the acute and high toxicity of OPs would irreversibly hold-up the biological activity of neurotransmitter acetylcholinesterase in the peripheral nervous and central system [50,51], leading to the excess accumulation of the acetylcholine specific part in the animal and human bodies that could cause organ failure and final lose life. A number of OPs have been banned to be used in farming industry owing to their hypertoxicity, for example, methamidophos, methyl parathion, azodrin, and parathion. Nevertheless, there remains the problem of the illegal use of these OPs in agricultural and animal husbandry products. Hence, there is an imperious need for the detection of OPs.

Tao *et al.* made an effort to take advantage of a novel fluorescence resonance energy transfer (FRET) method to determine chlortoluron using CDs part as energy donors, while CdTe part works as an energy acceptor part. The intuitive assay method is shown in Fig. S1A (Supporting information), where energy transfer between CDs and CdTe brings the fluorescence reduction of CDs while fluorescence enhancement of CdTe. When the chlortoluron was introduced, the intensity would be quenched, owing to the production of ground state complex. The quenching degree was proportional to the added chlortoluron content with high selectivity and sensitivity, and LOD was 7.8×10^{-11} mol/L. The FRET system also been used to sense chlortoluron in water samples satisfactorily [52].

A facile fluorescent nano sensor for methyl parathion is explored in view of L-tyrosine methyl ester modified CDs (Tyr-CDs) which is setup by tyrosinase (Fig. S1B in Supporting information). The CDs are first prepared *via* an easy hydrothermal reaction with carbon resource citric acid and then modified with L-tyrosine methyl ester. The obtained CDs are characterized by a series of testing methods for nano materials, including transmission electron microscope (TEM), high resolution transmission electron microscopy (HR-TEM), X-ray diffraction (XRD) spectrum, X-ray photoelectron spectroscopy (XPS), and Fourier transform in-

frared (FTIR) spectroscopy. The Tyr-CDs possess stable strong fluorescence with a QY of 3.8%, on which the tyrosine methyl ester could be oxidized to its quinone products and weaken the fluorescence under the catalysis of tyrosinase. OPs are able to inhibit the tyrosine activity, thus can be introduced into the system and reduce the fluorescence weakening rate. In their work, methyl parathion was taken as the OPs model, whose fluorescence test indicated that the methyl parathion concentration is proportional to the enzyme inhibition rate logarithmically in the concentration range from 1.0×10^{-10} to 1.0×10^{-4} mol/L with the LOD of 8.8×10^{-11} mol/L. This proposed system has been triumphantly used to determine the methyl parathion in milk, cabbage and fruit juice specimens, suggesting its excellent repeatability, accuracy and selectivity [53].

Ding *et al.* displayed a green route to fabricate fluorescent CDs for glyphosate assay (Fig. S1C in Supporting information). They chose wool, a nontoxic natural raw material, to fabricate CDs through a microwave-auxiliary pyrolysis method which was simple and facile without addition of other substances, avoiding the complex post-purification process. The CDs have bright fluorescence with high QY 16.3%, whose fluorescence intensity would be reduced by silver nanoparticles (AgNPs) through inner filter effect action. The glyphosate can make the AgNPs aggregated; therefore, the fluorescence would be recovered of the original quenched CDs. Using the response principle, a fluorescent sensing complex (CDs/AgNPs) was built for the assay of glyphosate. After the optimization of the test conditions, a linear concentration range data of 0.025–2.5 $\mu\text{g}/\text{mL}$ for quantifying glyphosate was obtained, whose LOD was 12 ng/mL. In addition, the proposed means was used to sense glyphosate in the cereal samples triumphly [54].

Lin *et al.* fabricated CQDs with tunable fluorescence from precursor chlorophyll to selectively sense paraoxon (Fig. S1D in Supporting information). The synthesized CQDs are relatively uniform spherical dots. Through FRET process, the fluorescence from CQDs could be weakened sharply by Au NPs. Under the efficient and dedicated hydrolysis of butyrylcholinesterase (BChE), acetylthiocholine (ATC) would be hydrolysed to produce thiocholine which could make the Au NPs aggregated and the initial FRET-quenched fluorescence would thus be recovered. The OPs would inhibit the catalytic activity of BChE irreversibly and reduce the recovery effect. Among the different OPs, paraoxon exhibited the highest inhibition percentage, thus was selected the representative of OPs whose LOD could reach 0.05 $\mu\text{g}/\text{L}$ in the concentration from 0.05 $\mu\text{g}/\text{L}$ to 50 $\mu\text{g}/\text{L}$ with this sensing platform. Actual sample test in river and tap water suggested that the fluorescent sensor was reliable in terms of repeatability and accuracy, which is a meaning OP sensor in food safety and environmental assay [55].

Preparation of red emissive CDs is very attractive for detection purpose, as key points remain in the aspects of raw material fabrication and final product purification. Lu *et al.* proposed a special approach for obtaining effective red fluorescent CDs with maximum fluorescence at 610 nm and QY of 28.0% by solvothermal treatment of the mixture of thiourea and citric acid in dimethylformamide (Fig. 6A). The CDs with a uniform diameter dispersion in the range of 4–7 nm have high chemical and optical stability. Deeper investigations manifest that the conjugating sp^2 -domain modification with elements nitrogen and surface doping are probably associated with the red fluorescence of the obtained CDs. Using fluorescent properties and rich surface functional sites of CDs, a ratiometric fluorescent platform could be constructed easily for detecting trace content of OPs. Based on the principle that the acetylcholinesterase could adjust the polymerization degree of dopamine and the pesticide could inhibit the acetylcholinesterase activity, the final outcome is that the polymerization degree of dopamine is directly related to the OPs content. By quantifying the emission intensity ratio at 503 and 610 nm, the proposed sensor displayed

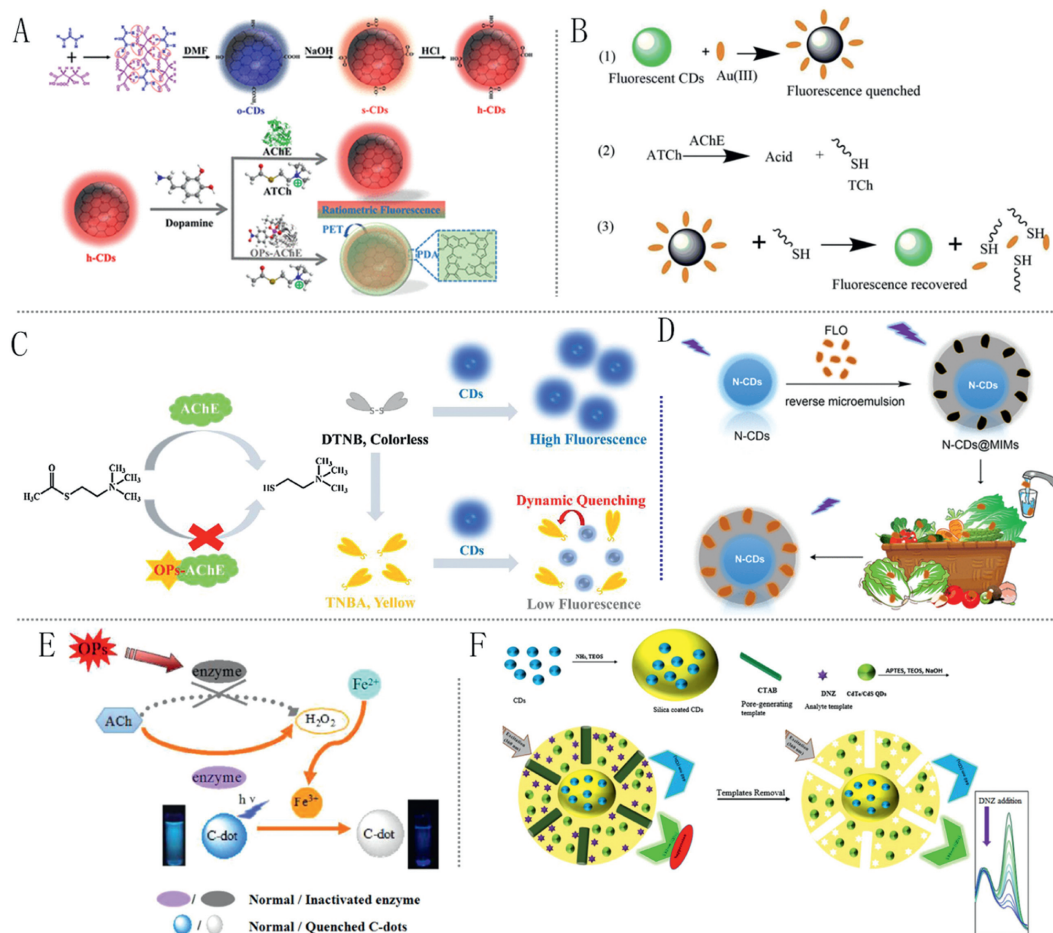


Fig. 6. (A) Fabrication route of CDs and sensing illustration of h-CDs/DA for OPs. Reproduced with permission [56], Copyright 2020, American Chemical Society. (B) Mechanism illustration of CD/Au(III) sensor for organophosphate pesticide sensing. Reproduced with permission [57], Copyright 2018, The Royal Society of Chemistry. (C) The scheme of CDs for OPs detection with the dual-signal response system. Reproduced with permission [58], Copyright 2017, Elsevier B.V. (D) N-doped CDs peeled from covalent organic framework to detect fonicamid. Reproduced with permission [59], Copyright 2020, American Chemical Society. (E) The detection mechanism of modification-free CDs for organophosphorus pesticides detection. Reproduced with permission [60], Copyright 2018, Elsevier Ltd. (F) Schematic illustration of the architecture of the mMIP/CDs/QDs and the working mechanism for the detecting DNZ. Reproduced with permission [63], Copyright 2017, Elsevier B.V.

high selective and sensitive response to OPs, with an ultrasensitive sensing concentration in the pg/L level. In addition, its practicalities were validated into the spiked sample of real tap water, soil, river, apple, rice and serum, which endows CDs as a common paradigm for a sensing application *via* modular programming [56]. CDs fabricated hydrothermally from egg white were proposed by Wang *et al.* to design a sensor for sensing organophosphate pesticides (OPs) (Fig. 6B). First, the fluorescence of CDs was weakened by a trace of Au(III) ions; then the fluorescence were eliminated by thiocholine produced by acetylcholinesterase-catalyzed hydrolysis of acetylthiocholine, recovering the fluorescence. Organophosphates were able to reduce the acetylcholinesterase activity, which changed fluorescence recovery. Thus the pesticide concentration could be detected based on the pre-established working calibration curve. In the pesticide-spiked sample determination, its test recovery was 99.85% in the response range of 0.45–48.80 $\mu\text{mol/L}$ [57].

A label-free bioplatform was built by Su *et al.* for sensitive determination of OPs by means of dual-readout including fluorescence spectroscopy and colorimetry taking advantage of acetylcholinesterase (AChE)-governed extinction of fluorescent CDs, which can transform molecular interaction events into fluorescence or color alteration (Fig. 6C). In their work, the N-CDs were obtained by stripping a covalent organic framework, and their fluorescence sensitivity and stability were ameliorated by the added ionic liq-

uid. The multiporous imprinted microspheres can be used to sense fonicamid based on the principle of the resonance charge-transfer principle caused fluorescence quenching between the fonicamid molecules and N-doped CDs. The microspheres displayed purple fluorescence, whose intensity can be decreased sharply in the presence of fonicamid. The fluorescence weakening degree has a nice linearity relationship with the fonicamid concentration ranging from 0.02 $\mu\text{g/g}$ to 0.2 $\mu\text{g/g}$, whose LOD was 0.0059 $\mu\text{g/g}$. The study flourishes the fundamental research of fonicamid residues, and also widens the practical applications of multiporous imprinted microspheres in monitoring environment pesticide in agriculture and food science [58].

CDs doping with controlled nitrogen can adjust the band gap and lead to the emission change from the excitation-dependent mode to excitation-independent emission mode. With a one-pot reverse micro-emulsion surface-imprinting means, Liu *et al.* prepared multi-porous imprinted microspheres from nitrogen-doped CDs (N-CDs) (Fig. 6D). In their work, the N-CDs were stripped from a covalent organic framework, and their sensitivity and the fluorescence stability were ameliorated by the added ionic liquid. The multiporous imprinted microspheres can be used to sense fonicamid based on the principle of the fluorescence-resonance charge-transfer strategy caused fluorescence quenching between the fonicamid molecules and N-doped CDs. The multiporous imprinted microspheres displayed purple fluorescence, whose inten-

sity can be decreased sharply in the presence of flonicamid. The fluorescence quenching degree has a good linearity relationship with the flonicamid concentration in the range of 0.02–0.2 $\mu\text{g/g}$, whose LOD was 0.0059 $\mu\text{g/g}$. This study not only flourishes the fundamental research of flonicamid residues but also widens the practical applications of multiporous imprinted microspheres in monitoring environment pesticide in food and agriculture [59].

In Yu's study, the burned waste paper ash was taken as raw material to prepare CDs (Fig. 6E). The fluorescence from resultant CDs would be declined by Fe^{3+} which was transformed from Fe^{2+} under the oxidation of H_2O_2 . In the biological system, H_2O_2 can be produced from acetylcholine by acetylcholinesterase. When the organophosphorus pesticides are present, the acetylcholinesterase activity would be destroyed efficiently, thus inhibiting the coming into being of H_2O_2 . Taking advantage of above mechanism that the fluorescence intensity alteration of CDs has direct ratio with the pesticides concentration, a universal approach for pesticides assay was created. Herein, chlorpyrifos was taken as an example, whose linear range of fluorescence was in the 0.01–1.0 $\mu\text{g/mL}$ range with LOD 3 ng/mL . When applied in to actual samples test, this method was sensitive and reliable. The fluorescent imaging of chlorpyrifos on cabbages leaves suggested this means could be applied to detect organophosphorus pesticides in vegetables visually [60].

Triazole fungicides are synthetic organic pesticides extensively applied to control fungus diseases development in field crops. However, they are a double-edged sword. These pesticides are mutagenic and carcinogenic, and could disturb human endocrine activity [61]. Diniconazole is one of the triazole fungicides with high activity, and its residue has posed some environmental problems owing to extensive usage [62]. On account of these influences, it is important to find rapid and facile ways to detect diniconazole in different environmental samples. Amjadi *et al.* reported a molecularly imprinted mesoporous probe to fluorescently sense diniconazole in ratiometric mode (Fig. 6F). The probe displays two fluorescent maximum peaks at 440 and 530 nm, excited with 350 nm wavelength. The green fluorescent QDs (CdTe/CdS) are embedded into the gaps of mesoporous silica to play the role of response signal, while the blue fluorescent CDs are encapsulated into the silica core to provide the reference signal. The mesoporous pores have the merits of nanosize and high pore volume, which endow the sensors with rapid response and high sensitivity. The analysis application was carried out in soil and water samples for diniconazole detection [63].

The synthetic dye malachite green (MG), as a very effective parasiticide and fungicide, has been widely used in fish farming fields [64]. Nevertheless, the overdose usage of MG is harmful to fish which have chronic MG exposure and through the subsequent food chain cause side effects on the immune and reproductive systems of human [65–67], which have chronic MG exposure and through the subsequent food chain cause side effects on the immune and reproductive systems of human [68,69]. Hence, it is vital to develop a sensitive and fast strategy for the analysis of MG. A new molecular imprinted fluorescence sensing platform (CDs&ZIF-8@MIPs) possessing core-shell construction was fabricated by Yan *et al.* via the sol-gel means that possessed high accuracy and selectivity to determine MG. The excellent specificity is attributed to molecular imprinting template molecule using MG, the functional monomer using 3-aminopropyltriethoxysilane, the cross-linker using tetraethoxysilane, and the initiator using ammonia respectively. The excellent sensitivity is attributed to CDs with bright blue fluorescence as the ZIF-8 worked as the host to improve the CDs dispersibility. What is more, the CDs&ZIF-8@MIPs was studied for the MG detection quantitatively by means of fluorescence dynamic quenching principle in a range 20–180 nmol/L , low LOD (2.93 nmol/L) and quick response (within 2.0 min) ability [70].

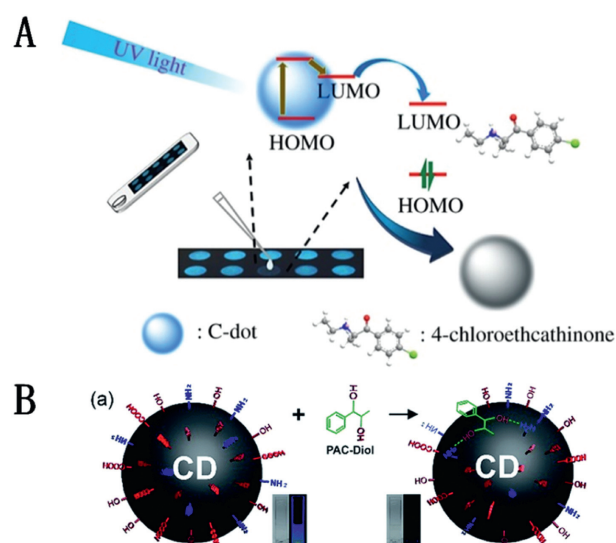


Fig. 7. (A) Scheme of CDFP for 4-chloroethcathinone detection. (B) Proposed interaction between CD's surface and phenylpropan-1,2-diol. Reproduced with permission [72], Copyright 2015, Royal Society of Chemistry.

4. Abused drugs

Narcotic drugs, also known as abused drugs, such as methamphetamine and morphine are illegally used in large quantities and their addiction is a worldwide problem. Thus, their qualitative and quantitative detection are very necessary to control their abuse. Chang *et al.* fabricated CDs from as raw material L-arginine via a hydrothermal treatment and the resultant CDs was used for the assay of 4-chloroethcathinone both in water solution and on C-dot-functionalized papers (CDFPs) (Fig. 7A). Following the Stern-Volmer equation, 4-chloroethcathinone could quench the fluorescence. The lifetime test of CDs in the presence of 4-chloroethcathinone, and the determination of the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels of the 4-chloroethcathinone and CDs together indicated that the fluorescence quenching of CDs by 4-chloroethcathinone was attributed to the electron transfer between CDs and π -conjugated 4-chloroethcathinone. Chromatography papers was cut in to the pattern of 8×12 circles and then coated with CDs in the wells via a solid-ink printing approach. Due to weak solubility of cocaine and heroin at pH 11.0, the C-dot exhibited priority selective fluorescent response to cathinones. The CDs in liquid aqueous solution detection system and CDFPs at pH 11.0 showed LOD 1.73 mmol/L and 0.14 mmol/L for 4-chloroethcathinone. Their detection system composed of a smartphone, a hand-held UV-lamp and a low-cost CDFP was applied to sense cathinones, heroin and cocaine at pH 7.0 successfully, displaying great potential to detect these drugs from crime scene [71].

Li *et al.* prepared the amine-modified carbon dots (A-CDs) hydrothermally with urea and citric acid (Fig. 7B). The CD sample after purification, whose surfaces are covered with rich $-\text{NH}_2$ groups, displays a surface states dominated excitation-independent fluorescence emission with a QY in water of 30%. The obtained A-CDs display high sensitive response to precursor phenylpropan-1,2-diol and precursor phenylpropan-2-one, and an extraordinary selectivity to common drug cutting agents including aspirin and paracetamol. It is first time to report the unique selectivity of A-CDs to this kind of drug precursor molecules. The sensing mechanism of charge transfer is investigated by a minute analysis such as the changes of lifetime and zeta potential on the dynamic and static fluorescence test. They further illustrated an immobi-

lization method for preparing solid-state platform for detecting phenylpropan-2-one and phenylpropan-1,2-diol, and the solid A-CDs displayed more sensitive response to phenylpropan-1,2-diol than that to phenylpropan-2-one upon immobilization. The work displays the potential of the CDs as a portable optical sensor and test kits for illicit drugs, to satisfy the urgent test requirements [72].

Masteri-Farahani *et al.* fabricated the graphene quantum dots (GQDs) possessed brilliant blue fluorescence *via* pyrolysis method to differentiate and determine two very common narcotic drugs namely methamphetamine and morphine in very low concentrations. The blue emission fluorescence around 460 nm of the resultant GQDs with the maximum excitation wavelength 362 nm would be weakened upon the addition of methamphetamine, while would be enhanced upon the addition of morphine whose found LOD was respective 1.48 and 0.5 $\mu\text{g}/\text{mL}$. The low-cost sensing system possesses some merits including fast response time (less than 1 min), low LOD and as well as nontoxicity [73].

5. Food additives

It is of high importance to analyse the food and cosmetics unauthorized organic additive, because they are prone to cause various health issues [74]. Melamine ($\text{C}_3\text{H}_6\text{N}_6$) is generally applied to the fabrication of formaldehyde resins for producing plastics, coatings, laminates, adhesives, dishware, *etc.* However, due to the high nitrogen contents (66 mass%), melamine is notorious to be illegally added into animal feed and baby dairy products to artificially raise the measured protein content by some illegal businesses mistakenly using the defects of the nitrogen content test standard. Very unfortunately, it has been verified that melamine will cause the production of crystals in kidney and result in the formation of kidney stones [75]. The ingestion of melamine with concentration beyond the safety limit would lead to renal failure and even death in infants. Therefore, it is rather urgent to propose a sensitive and reliable approach for the qualitative and quantitative detection of melamine.

Li *et al.* developed a FRET platform between CDs and AuNPs for melamine detection (Fig. S2A in Supporting information). The obtained CDs have fairly broad size, which is probably responsible for the broad absorption peak. AuNPs have the typical characteristics of high extinction coefficient and a wide absorption wavelength which overlay a considerable part of the fluorescence emission spectrum range of the CDs. In this system, CDs played the role of energy donors, and AuNPs worked as energy acceptors, thus the introduced AuNPs would weaken the fluorescence of CDs through their FRET process. Once melamine was added, it could combine with AuNPs through covalent binding and effectively prevent the absorbance of CDs onto the surface of AuNPs, thus reducing the FRET process and increasing the fluorescence intensity. The impact factors such as AuNPs concentration, incubation time and solution pH were optimized before the melamine determination. Using the linear relationship from 50 nmol/L and 500 nmol/L of melamine, a LOD of 36 nmol/L was obtained, which was satisfactorily used to determine melamine in milk samples in terms of accuracy and reliability. Compared with previous studies, the proposed strategy represented great advantages including high sensitivity, rapid analysis speed, low budget and easy operation [76].

Zou *et al.* designed a kind of Au nanoparticles@Carbon quantum dots composites (Au@CQDs) for sensing melamine in milk visually (Fig. S2B in Supporting information). The fluorescent emission intensity of Au@CQDs would rise with the added dose increase of melamine concentration. A fluorescence standard array and working calibration curve ($R^2 = 0.9856$) were founded for the assay of melamine under the concentration range from 1 $\mu\text{mol}/\text{L}$ to 10 $\mu\text{mol}/\text{L}$. The quantification limit and detection limit for

melamine were 12 and 3.6 nmol/L, respectively. The close detected concentration of melamine mingled in milk specimens was determined in visual *via* the fluorescence standard array and commercial smartphone. This detection performance coincides with that of the gold standard high-performance liquid chromatography assay (recoveries of 105.56%–107.95%). The Au@CQDs-based detection means for fluorescently and precise assay of melamine visually in milk was displayed and possessed great application potential [77].

You *et al.* proposed a time-saving, cost-effective and facile fluorescent turn-on detection method for melamine using CDs as the fluorescence reporter (Fig. S2C in Supporting information). Melamine is easy to take coordination reaction with Hg^{2+} owing to the existence of a multi-nitrogen containing heterocyclic ring of melamine. This principle is responsible for anti-weakening capability of Hg^{2+} towards CDs *via* reducing the adsorption interaction between CDs and Hg^{2+} . In this way, a LOD of 0.3 $\mu\text{mol}/\text{L}$ for melamine is gained. Moreover, the sensing system was used to sense melamine successfully in some real specimens such as milk powder and raw milk without pricey instruments and complicated modification [78].

He *et al.* prepared a cost-saving, without background interference and smartphone-based micro apparatus with UV lamp, a sample slot, and a smartphone for quantitative assay of target melamine (Fig. S2D in Supporting information). Carbon nitride quantum dots (abbreviated as CNQDs) were chosen as the fluorophore, and the sodium carboxymethyl cellulose (abbreviated as CMC) was set as the matrix to form CNQDs-CMC composite film to produce the room temperature phosphorescence of CNQDs. The polymer matrices can suppress the k_{nr} and k_{q} , thus activating the temperature phosphorescence efficiently. Temperature phosphorescence detection is advantageous in terms of longer lifetime to facilitate building of low cost visual sensing systems, negligible background and larger Stokes' shifts. A confinement effect induced by the internal hydrogen bonding interactivity between CMC and CNQDs could restrain the non-radiative relaxation process and improve the QY effectively. The apparatus equipped with temperature phosphorescence CNQDs-CMC film was used to visually and rapidly detect melamine. The temperature phosphorescence color of CNQDs-CMC film would be weakened by the melamine and taken pictures by the smartphone. The self-contained APP was able to recognize the red (R) value to detect melamine quantitatively. Thus, digital image colorimetry (DIC) assay method for melamine was realized owing to the visible temperature phosphorescence color alteration of CNQDs-CMC film. The real-sample applicability of the device was illustrated by the quantitative assay of melamine in milk [79]. Lemon yellow azo dye tartrazine, full name trisodium 1-(4-sulfonatophenyl)-4-(4-sulfonatophenylazo)-5-pyrazolone-3-carboxylate, once used as a synthetic food color, is still used in some countries but confined in more countries in the food industry [80]. Though azo dyes are able to be eliminated normally from kidneys and liver in human body, tartrazine may give rise to some diseases such as cancers, liver disorders, kidney problems, generative toxicity, allergy, asthma, hyperactivities in children and neurobehavioral poisonousness [74,81]. In consideration of the above concerns, developing efficient detection sensors for tartrazine is in great need.

Using aloe as the starting material, Liao *et al.* developed a green and economical means for the synthesis of high-fluorescent bluish green CDs *via* hydrothermal process (Fig. 8A). Various characterizations showed that the CDs were in 5 nm sized spherical shape with a fluorescence QY of nearly 10.37%. The rich hydroxyl units on the surface of CDs contributed to the satisfactory solubility, excellent photostability and high QY. The work found that tartrazine could weaken the fluorescence of CDs *via* a static quenching principle, which made it possible to detect trace tartrazine in food specimens purchased from markets using the linear range of 0.25–

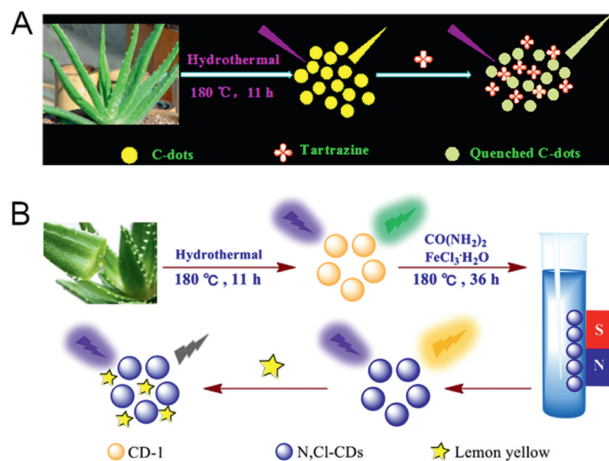


Fig. 8. (A) Scheme of the synthesis for CDs and the mechanism for tartrazine sensing. Reproduced with permission [82], Copyright 2015, American Chemical Society. (B) Scheme illustration of the fabrication of N,Cl-FCDs and the mechanism of tartrazine sensing. Reproduced with permission [83], Copyright 2019, Elsevier Ltd.

32.50 $\mu\text{mol/L}$. The great potential for food additive routine analysis sheds light on the positive action of fluorescent CDs with nice biocompatibility [82].

Zhao *et al.* prepared heteroatoms N,Cl co-doped carbon dots (N,Cl-FCDs) to specifically detect tartrazine fluorescently (Fig. 8B). Urea was used as the N source, while FeCl₃·6H₂O was used as the Cl source to hydrothermally synthesized N,Cl-FCDs with precursor CDs which was first prepared with aloe. In contrast to pure CDs, the fluorescence peak of N,Cl-FCDs would blue shift by 60 nm, whose fluorescence QY 60.52% was about 5 times of that from pure CDs, owing to the synergistic effect from heteroatoms doping. Based on the FRET mechanism, N,Cl-FCDs display a LOD 48 nmol/L in the working range of 0.1–30 $\mu\text{mol/L}$. Then the proposed platform was applied to sense tartrazine in beverages successfully, demonstrating the potential in food analysis [83].

Carmine (CRM) is an *artificially* synthetic azo red colorant which has been widely used in the food field [84]. CRM is reported to have a considerable genotoxic potential to serve as a DNA targeted intercalator to break the DNA chain, thus its long-time exposure would inevitably detriment the organisms [85]. The maximum usage has been strictly restricted in foodstuffs use by many countries. The maximum usage of CRM in China for daily food such as fruit juices, candies, hawthorn products and pickles is restricted to be less than 50 mg/kg or 50 mg/L [86]. So it is importantly crucial to develop convenient and reliable means to ensure food safety for human health. Hu *et al.* developed an efficient spectrophotometric sensor utilizing nitrogen-doped carbon dots (N-FCDs) for the highly fluorescent sensitive detection of CRM in foodstuffs. Using *m*-phenylenediamine as the raw material, the N-FCDs were prepared through a hydrothermal approach. The obtained N-FCDs possess strong fluorescence, which could be quenched by CRM owing to the inner filter effect caused static quenching with the production of ground state complex without fluorescence. There is a low LOD of 11.2 nmol/L based on the good linear relationship from 0.1 $\mu\text{mol/L}$ to 10.0 $\mu\text{mol/L}$. The method founded in this work was facile, sensitive and reliable for determining CRM in a food matrix such as strawberry jam, watermelon frost lozenges, haw flakes and dried waxberry with high precision and accuracy, which had great potential as a powerful sensor for the analysis of additive food colouring agents [87].

In summary, organic pollutants, as the toxic chemicals, would affect human health and the environment adversely around the world. More and more researches have studied the correlation be-

tween long-term low level exposure of organic compounds and a variety of diseases. For the sake of assessing disease risk coming from organic compounds in a particular location, related government agencies should propose a human health risk evaluation which considers the pollutants' bioavailability and their content-response relationships a lot. It is imperative to make use of an efficient method to detect organic pollutants, which is significant for public health and safety. Fluorescent assays based on CDs thus would provide a very plausible candidate method. After consulting a large number of literatures, we offer a comprehensive review of the sensing applications of CDs for organic pollutants.

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.

CRediT authorship contribution statement

Quan Zhang: Writing – original draft. **Shunjie Xing:** Formal analysis. **Jingqian Han:** Formal analysis. **Li Feng:** Investigation. **Jianchun Li:** Funding acquisition. **Zhaosheng Qian:** Writing – review & editing. **Jin Zhou:** Writing – review & editing.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ccllet.2024.110117.

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