REVIEW



# Carbon dots from eco-friendly precursors for optical sensing application: an up-to-date review

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### Abstract

Carbon dots (CDs) are zero-dimensional quasi-spherical nanoparticles endowed with excellent advantages including good luminescence features, photostability, low cytotoxicity, remarkable aqueous solubility, favourable biocompatibility, low risk to environment and great flexibility in surface modification. Fluorescent CDs that can selectively respond to specific inorganic/organic target molecules in environmental and biological samples are of prime significance amongst the new generation intelligent sensors due to the critical involvement of different ions/molecular species in not only human health, but also in environment processes. In this context, preparation of CDs from bioprecursors has immense significance due to the involvement of green principles, inexpensive, clean, nontoxic, easily accessible, renewable and large-scale production can be realized. This article aims at exploring different types of green raw materials including plant biomass, animal products, food items and waste materials as carbon sources for the synthesis of both undoped and doped CDs. The emphasis is given on different synthetic approaches adopted for improving the quantum yield without any chemical modification, the characterization techniques, mechanistic origin of photoluminescence and fluorescence response mechanisms involved in the sensing action towards various analytes. The significant benefits and limitations of CDs obtained from eco-friendly precursors through green approaches are summarized. Various challenges and the future prospects of these carbonaceous nanomaterials as sensors are also discussed.

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# **Graphical abstract**



Keywords Eco-friendly precursors  $\cdot$  Carbon dots  $\cdot$  Green strategies  $\cdot$  Fluorescence  $\cdot$  Optical sensing

Abbrevia	tions	Hg	Mercury
AIE	Aggregation-induced emission	IFE	Inner-filter effect
Ag	Silver	LA	Laser ablation
Al	Aluminium	LOD	Limit of detection
С	Carbon	LUMO	Lowest unoccupied molecular orbital
CDs	Carbon dots	Mn	Manganese
CEA	Carcinoembryonic antigen	6-MP	6-Mercaptopurine
CEF	Chelation-enhanced fluorescence	MTX	Methotrexate
Cr	Chromium	MW	Microwave
Co	Cobalt	NADP <sup>+</sup>	Nicotinamide adenine dinucleotide phosphate
Cu	Copper	Ν	Nitrogen
CQF	Chelation-quenched fluorescence	N-CDs	Nitrogen-doped carbon dots
Cys	Cysteine	0	Oxygen
EPA	Environmental protection agency	PBS	Phosphate-buffered Saline
FAM	Carboxyfluorescein	Р	Phosphorous
Fe	Iron	PL	Photoluminescence
FL	Fluorescence	Pb	Lead
FRET	Forster resonance energy transfer	PCT	Photo-induced charge transfer
FT-IR	Fourier transform infrared spectroscopy	PET	Photo-induced energy transfer
GSH	Glutathione	QY	Quantum yield
Н	Hydrogen	S	Sulphur
HOMO	Highest occupied molecular orbital	SASP	Salazosulphapyridine
HRTEM	High-resolution transmission electron	Sn	Tin
	microscope	UV	Ultraviolet
$H_2O_2$	Hydrogen peroxide	VOC	Volatile organic compounds

XPS	X-ray photoelectron spectroscopy
Zn	Zinc

#### Introduction

Though rapid industrial advancements represent the cornerstone for worldwide economy and enormously assist in improving the quality of human life, global policies continued to remain insensitive to its consequent impact on the environment. However, several mounting concerns emphasized the need for cleaner environment for a sustainable world lately. One among the solutions was the introduction of 'green principles' to the scientific world (Omran et al. 2021). The fundamental idea was either to exclude or to minimize the use of toxic solvents/chemicals in chemical processes in order to limit the hazards to the planet (Marco et al. 2019). Consequently, the need to limit the use of harmful materials became more significant than treating the previously created waste. In addition, several guidelines were framed regarding the utilization of harmful reagents and solvents in synthesis processes, with a factor to evaluate the eco friendliness of a method (Kharissova et al. 2019). In this context, nanotechnology has significantly impacted in evolving 'green' and 'clean' strategies with considerable environmental benefits. The green chemistry principles can be applied to develop more sustainable and safer nanomaterials through more viable as well as efficient manufacturing strategies. In the recent few decades, substantial advancements have been made in the evolving arena of nanotechnology with the aim of directing it to achieve its green potential. Green nanotechnology involves a clean production approach, such as preparing nanoparticles using green precursor materials, recycling of industrial/agricultural wastes into nanomaterials, etc.

The serendipitous discovery of carbon dots (CDs) during the purification of single-walled carbon nanotubes by Xu et al. in 2004 opened a new avenue to the development of a very useful quasi-0D functional material with a size lesser than 10 nm (Xu et al. 2004). These carbonaceous nanomaterials generally consist of a crystalline or an amorphous carbon core and an oxidized carbon shell with carboxyl groups. They have garnered extreme research interest owing to their attractive attributes including facile synthesis, easy functionalization, good aqueous solubility, excellent biocompatibility, low toxicity and photobleaching, excitation wavelength-dependent multi-colour emission, non-blinking, good thermal stability, chemical inertness and environmentfriendly nature, which render them appealing for wide-ranging applications (Humaera et al. 2021).

There are several reviews available collating the synthesis, outstanding physical and chemical properties, and applications of CDs including detection of toxic chemicals in food, explosives, chemicals, drug delivery, bioimaging, and biosensing, photocatalysis, solar energy conversion, electrochemical and biosensors, etc. (Gayen et al. 2019; Ashrafizadeh et al. 2020; Hassanvand et al. 2021). Ji et al. have summarized the recent advancements of CDs-based electrochemical and optical biosensors to analyse organic/inorganic molecules in living organisms (Ji et al. 2020). Due to their strong photo-luminescent, antibacterial, photo-induced electron transfer (PET) and good semiconductor capabilities, there are several reviews on CDs employed to detect heavy metal ions, remove organic and inorganic contaminants and photocatalytically degrade wastewater pollutants (Yoo et al. 2019; Laghari et al. 2021). Nazri and co-workers have reviewed the CDs for optical sensing applications based on colorimetry, optical fibres and surface plasmon resonance (Nazri et al. 2021). Later Peng et al. highlighted the emerging strategies in the fabrication of CDs with stimuli responsive afterglow luminescence and the key challenges to be overcome for their full real-life utility (Peng et al. 2021). Li et al. emphasized the unique electronic, fluorescent, photoluminescent, chemiluminescent and electrochemiluminescent properties of CDs for potential use in sensing applications (Li et al. 2019). Suresh and Janardan have reported the perspectives of magnetic CDs in analytical chemistry (Sursh and Janardhan Koduru 2022).

Though there are a few articles focussing on the sensing application of CDs, the objective of this comprehensive review is unique in highlighting the synthesis strategies and sensing applications of CDs obtained from eco-friendly precursor materials for the detection of metal ions and organic compounds. This article reviews different types of green raw materials including plant biomass, animal products, food items and waste materials as carbon sources for the synthesis of CDs. Besides, the review focuses on the photophysical properties, quantum yields and limit of detection (LOD) of the CDs prepared from natural raw materials. Moreover, the pictorial illustrations and the tables listing the main features of the synthesized CDs from various environment-friendly precursor materials enable easy understanding for the readers. Finally, the advantages, limitations and the future prospects while preparing CDs from natural sources are also summarized.

# Synthetic strategies for CDs

The well-known processes for the preparation of CDs with controllable characteristics are the top-down and the bottom-up approaches. The top-down approaches break down the larger bulk/nanomaterials to particles of size less than 10 nm. Among various top-down approaches, the electrochemical oxidation is the most widely recognized and hierarchically engineered synthesis of CDs because



Fig. 1 Representative images for characterization of CDs: A & B HRTEM, C particle size distribution histogram and D Raman spectra (Vandarkuzhali et al. 2018); (source: https://pubs.acs.org/doi/ 10.1021/acsomega.8b01146. Further permissions related to the material excerpted should be directed to the ACS.), E X-ray photo-

of the attractive benefits such as high purity, good yield and minimal effort (Zhou et al. 2007). Arc discharge is yet another method wherein the carbaneceous bulk undergoes decomposition at anodic region under high temperature (about 4000 K) to form high-energy plasma and eventually undergoes re-organization in cathodic region to form carbon nanoparticles. Though there are only few attempts to synthesize high quality CDs by this method, it is one of the best methods to produce nanoparticles (Arora and Sharma 2014). Laser ablation (LA) is another widespread technique used by scientists for the preparation of CDs (Thongpool et al. 2012). This technique involves removing molecules from a substrate surface with a pulsed laser to form nanostructures. LA has several advantages, including the capacity to create ligand-free noble NPs in a variety of solvents with minimal energy loss. Acid oxidation method in the presence of concentrated HNO<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub> with mild heating introduces numerous hydroxyl and carboxyl groups on carbon nanoparticle surface and thereby enhances their fluorescence (Desai et al. 2019; Zhang et al. 2017). However, the top-down synthesis strategies involve harsh experimental conditions, tedious operation steps, extended reaction durations, utilization of costly ingredients and expensive equipments, which greatly limit their practical application. Moreover, the CDs prepared using the top-down strategies without further purification and surface modification generally present weak

electron spectra; reprinted (adapted) with permission from Chatzimitakos et al. (2018) Copyright {2018} American Chemical Society, **F** absorption and emission spectra and **G** excitation wavelengthdependent emission spectra; reprinted (adapted) with permission from Hu et al. (2017) Copyright {2017} American Chemical Society

luminescence and low quantum yield (QY) (Wang and Hu 2014; Mikhraliieva et al. 2020).

The bottom-up approaches include the utilization of materials in nuclear scale, which is further developed to the required nanosize. There are additional approaches available for this method, allowing for more options in terms of using precisely designed precursors and preparatory processes. Consequently, CDs can be adapted to samples with well-defined molecular weights, sizes, shapes and characteristics. Bottom-up approaches are usually low-cost and efficient for creating luminous CDs on a large scale, which is necessary for practical applications. Ultrasonic treatment is a helpful bottom-up strategy as the carbonaceous materials might be separated using high-energy ultrasonic sonic wave (Kumar et al. 2016; Li et al. 2011). Sonochemical method involves generation of hot spots, which has higher temperature and pressure with faster heating and cooling rates. The collapse of this bubble results in generation of shock waves which can induce collision of reactant molecules and mass transport. The hydrothermal method is utilized majorly as an inexpensive and eco-friendly technique in achieving high QY to isolate CDs from saccharides, amines, natural acids and their subsidiaries (Baker and Baker 2010). This technique involves the reaction of solid material with an aqueous solution in a reaction vessel at high temperature and pressure, which results in the formation of CDs. It is also adopted mainly for its several advantages including mild temperature treatment compared to pyrolysis, enabling hydroxyl, carboxyl and amino groups on the surface of CDs and contribution to higher photoluminescence (Tan et al. 2017). Microwave (MW)-assisted synthesis is yet another bottom-up approach that usually requires lesser reaction time and involves homogenous heating. As MW treatment provides intense and efficient energy, the products can be obtained in a short duration with good crystalline structure and homogeneity (Yu et al. 2018). Pyrolysis method is accompanied by physical and chemical changes in a compound when subjected to high temperature and is generally employed to synthesize smaller sized particles, which mostly emit shorter wavelength (Li et al. 2010). The bottom-up approach is not only eco-friendly and costeffective, but also the CDs formed exhibit relatively higher fluorescence QY (Kang et al. 2020).

#### Structural and photophysical characterization of CDs

The characterization of CDs is required to better understand the structural features and unique properties exhibited by these carbonaceous nanoparticles and is generally performed using various analytical techniques. The structural properties of the CDs can be analysed using high-resolution transmission electron microscopy (HRTEM), X-ray diffraction (XRD), Raman spectroscopy, X-ray photoelectron spectroscopy (XPS) and Fourier-transform Infrared Spectroscopy (FTIR) (Jelinek 2017). HRTEM provides significant information on the morphology, particle size distribution and crystalline organization of the CDs, whereas XRD examines the crystalline nature of CDs and offers evidence on the unit cell dimensions and crystal spacing within the crystalline carbon cores. Raman spectra are yet another tool to gather information on the structural features of the carbon atoms within the CDs. A typical Raman spectrum of CDs features two peaks corresponding to the D band around  $1350 \text{ cm}^{-1}$ , attributed to the disordered sp<sup>2</sup> carbons and G band around 1600 cm<sup>-1</sup> due to the in-plane stretching vibration mode  $E_{2\sigma}$  of crystalline graphite carbons. The ratio of the intensities of these two characteristic Raman bands can provide insights on the carbon framework, primarily the degree of crystallinity and relative abundance of core carbon atoms as against the surface atoms. The surface functional units on the CDs can be elucidated using XPS and FTIR spectral analysis. XPS spectra provide information on specific atomic units present on the surface of CDs, and FTIR spectra usually complement XPS to reveal distinct information on functional groups. UV-spectroscopy can be used to measure absorbance of CDs, and generally the respective spectrum displays a strong band in the range of 230–282 nm

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corresponding to  $\pi$ - $\pi$ \* transition belonging to C=C group and another band around 300–355 nm corresponding to n- $\pi$ \* transition of C=O group. Excitation-dependent fluorescence is yet another striking feature of CDs, which can be studied using photoluminescence spectroscopy. The tuning of emission colour of CDs according to the different excitation wavelength is exploited in varied applications. Representative images on different characterization techniques of CDs adapted from various literatures are presented in Fig. 1.

# Origin and further modulation of fluorescence in CDs

Diverse variety of fluorescent CDs including carbon nanodots, carbon nanotubes, graphene oxide/graphene quantum dots, nanodiamonds, polymer dots, etc. have been reported. The CDs generally exhibit intrinsic optical properties, especially tuneable luminescence based on their inner structure and surface chemical groups. The chemical structure varies with different CDs and generally consists of sp<sup>2</sup>/sp<sup>3</sup> carbon and oxygen/nitrogen-based groups or polymer aggregates (Yan et al. 2018). Therefore, the photoluminescence centre is also dissimilar for different types of CDs.

Although a variety of fluorescent CDs with different chemical structures from diverse precursor materials and different synthetic approaches have been reported, the mechanistic origin of fluorescence is still unclear till date and debatable (Zhu et al. 2015). Four common mechanisms have been proposed for the origin of fluorescence in CDs such as: (i) quantum confinement effect or conjugated  $\pi$ -domains that are determined by the carbon core, (ii) surface state depending on the hybridization of the carbon backbone and connected chemical groups, iii) molecular state determined exclusively by the fluorescent molecules connected to the surface or interior of the CDs and iv) crosslink enhanced emission effect. As per the first mechanism, generally attributed to graphene carbon dots and carbon nanodots, excitons in CDs have an infinite Bohr diameter, enabling them to exhibit quantum confinement effects and fluorescence emission by possessing a nonzero bandgap. It was inferred from theoretical calculations and the respective highest occupied molecular orbital-lowest unoccupied molecular orbital (HOMO-LUMO) gaps for CDs of different sizes that the quantum-sized graphite structure is responsible for the strong luminescence. Moreover, Jiang and coworkers demonstrated the red shift in the fluorescence exhibited by CDs based on the quantum size effect (Jiang et al. 2015). However, few investigations suggested that the surface defect states are responsible for the intrinsic fluorescence emission of these nanodots (Hu et al. 2009). Surface state

mechanism for the origin of luminescence is based on the functional groups that adorn the surface of CDs that have a series of emissive traps due to various energy levels. A redshifted emission has been perceived with a higher degree of surface oxidation or more surface defects realized through other surface modifications (Wang et al. 2017). The surface functional groups can be modified either covalently by various reactions including amide coupling, esterification, sulphonylation and copolymerization or noncovalently through electrostatic interaction and complexation to modify the optical performance (Yan et al. 2018). Moreover, the carbonization approach generally generates fluorescence in CDs even without surface passivation, probably due to the surface oxidation using strong acids, which introduces lattice defects onto the surface with different oxidation states (Esteves da Silva and Gonçalves 2011). These defects are significant enough to generate band gaps attributed to the small size and high surface area of the CDs, which allow electronic transitions leading to fluorescence. Further, in order to explain the molecular state mechanism, mainly ascribed to carbon quantum dots, the photoluminescence centre is generated by an organic fluorophore through carbonization process, and the fluorophore is linked either on the surface or interior of the carbon framework to exhibit direct luminescence (Krysmann et al. 2012). Finally, yet another new mechanism to enhance luminescence is crosslink enhanced emission generally observed in polymer dots, as chemical crosslinking generates new luminescent structures, enhanced immobilization and through-space interactions (Tao et al. 2020).

The size and surface functional groups of CDs can be prudently modulated to achieve desired fluorescence features. The emission of CDs could be precisely adjusted by controlling certain critical parameters including the synthetic approach, solvents, duration of reaction, temperature, pH, chemical manipulations or most commonly by doping with other elements (Yang et al. 2013). Various approaches that permit researchers to change the ground states of CDs to improve their properties such as dissolvability and good QYs based on their applications have been reported. On certain occasions, passivation or the introduction of surface functional groups is vital to improve the optical properties of CDs. Doping of various elements is an effective method to regulate the optical and electronic features, as well as the surface and local chemical reactivity of CDs (Wang et al. 2014a). Introducing atomic impurities, such as nitrogen (N), phosphorus (P) or boron (B), can change their intrinsic electronic structure and QY (Park et al. 2016). Therefore, surface engineering of CDs is highly crucial with respect to their use in various analytical applications such as sensing, bioimaging, drug delivery, photocatalysis, anti-counterfeiting, etc.

# Fluorescence-based sensing capabilities of CDs

CDs own great potential in serving as nanoprobes to detect various analytes based on any variations in fluorescence response phenomena including intensity, wavelength, QY, anisotropy, lifetime, quenching etc. Various mechanisms have been demonstrated for the selective sensing feature exhibited by these carbonaceous nanoparticles for realtime practical applications. The sensing mechanisms can be broadly classified as due to PET, Photo-induced charge transfer (PCT), Forster resonance energy transfer (FRET) and Inner filter effect (IFE) (Sun and Lei 2017). In PET, an internal redox reaction occurs between the excited state of the electron donating fluorescent CDs and electron accepting analyte species. The generated excited complex either returns to the ground state orbital without the emission of a photon, or in few instances exciplex emission is detected, and finally, the extra electron on the acceptor is returned to the electron donor. However, PCT involves electron transfer between the donor and acceptor to promote modifications in the electronic energy levels and leads to variation in fluorescence signals through complex formation. Longrange dipolar interactions are observed in FRET, wherein the excited donor CDs return to the ground level non-radiatively, simultaneously transferring the energy to an electron on the analyte acceptor via intermolecular long-range dipole-dipole coupling. The relative distance between the CDs and analyte species, their orientation as well as the extent of overlap of emission and absorption spectra of CDs and analyte, respectively, decide the rate of energy transfer. In the case of IFE, the absorption spectrum of fluorescence quenching of analyte overlaps with the excitation or emission spectra of CDs. The fluorescence intensity observed is linked to the excitation light intensity, whereas fluorescence lifetime is relatively independent of total intensity, and the QY is lower than that perceived for an infinitely dilute solution. Nevertheless, in few research investigations, IFEbased quenching using fluorescent CDs has been exploited for sensing applications.

In most cases, the sensing of analytes using CDs is based on quenching of fluorescence. The fluorescence quenching or reduction in QY can occur through different types of molecular interactions between the fluorophoric CDs and the quenching analyte molecule including energy or electron transfer, collision, excited-state reaction and groundstate complex formation. The quenching mechanisms can be generally categorized into dynamic type that results from collision and static type that is induced by the ground-state complex formation between the CDs and the analyte species. If the life time of the free CDs and its complex is different, then energy transfer occurs through collision and/or close interaction of  $\pi$ - $\pi$  overlap in excited state via resonance energy transfer, which results in dynamic quenching. In static quenching, the fluorescence life time of the free CDs and its complex is same (Kumari et al. 2018a). Explicitly, the interaction between CDs and quencher molecule forms a non-fluorescent ground-state complex, leading to static quenching.

The applications of CDs as fluorescent probes for selective and sensitive optical detection of metal ions, dyes, pharmaceutics, biomolecules, volatile gases, etc. have been studied by several researchers, and the outcomes of their studies were promising. Though highly selective and sensitive conventional analytical techniques such as ultraviolet-visible spectrometry, atomic absorption/emission spectroscopy, X-ray absorption spectroscopy, inductively coupled plasma mass spectrometry, Auger electron spectroscopy, stripping voltametry and polarography have been extensively used (Mura 2014), these methods require tedious sample preparation steps and sophisticated instrumentation, high operating cost and use of hazardous chemicals, which restrict their practical applications. Hence, long-term feasibility and sustainability in using these techniques are a challenge and there is a need to develop alternative analytical methods that are more economical, selective, sensitive, remain user-friendly and green to the environment. Optical sensing systems based on fluorescence responses are an ideal alternative attributed to its high sensitivity, rapid analysis and being non-sample destructing. CDs that exhibit unique and inherent fluorescence characteristics are a good choice for analytical applications due to their high water solubility, low toxicity profiles, excellent biocompatibility, resistance to photobleaching and low production cost. The presence of hydrophilic functional groups as binding sites for analytes and large surface area is responsible for their wide use in bio- and chemical sensing. The adsorption of inorganic/ organic ions/molecules on the surface of CDs induces variations in the optical signals, allowing their use as sensors.

# CDs from eco-friendly precursors for sensing application

The major advantage of CDs is the facile and one-step synthesis process on a large scale from plenty of carboncontaining precursor materials (Zhou et al. 2007; Arora and Sharma 2014; Thongpool et al. 2012). The raw materials for the preparation of CDs are plentiful and can be classified broadly into organic and inorganic carbon sources. As the fluorescence QYs of CDs synthesized from inorganic carbon sources are relatively low, further surface passivation is necessary. Hence, CDs are generally synthesized from organic synthetic precursors such as glucose, malic acid, citric acid, sodium citrate, urea, ascorbic acid (Gao et al. 2018; Zhi et al. 2018; Qu et al. 2012; Ding et al. 2016), etc. CDs with high QYs were obtained from these synthetic chemicals, and utilizing diverse precursors can introduce various functional groups in them, which facilitates enhancement in their luminescence. However, the key focus for a green synthetic strategy of CDs relies in using natural procedures, eco-friendly solvents and green precursors including organic natural products, biomass and waste materials. In contrast with various other fluorescent materials, CDs prepared through green chemistry approach from cheap carbon sources are abundant and are biofriendly.

Natural materials that are widely distributed in nature and are easily as well as continuously available, less costly and renewable can replace the commonly used chemicals to prepare CDs (Lin et al. 2019). Moreover, most natural materials possess complex components, which endow the CDs made from these green resources with abundant and various surface functional groups for specific sensing applications. Various materials used as eco-friendly precursors belong to the category of plant biomass, animal products, food items, biowastes and waste materials, which mainly contain proteins, starch and lipids and are ideal green and biocompatible carbon assets. There are multiple inherent advantages in utilizing these low-value precursor materials including natural bioresources or reusing the agricultural and industrial wastes. These raw materials are not only rich in carbon sources, but also permit varieties of heteroatom doping (N, S, P), thereby addressing the environmental issues caused by usage of costly/toxic chemical precursors, complex posttreatment processes, waste disposal, etc. (Kumari et al. 2018b).

Among various possible analytical and bioanalytical applications of CDs obtained from bioprecursors, sensing of metal ions and other organic molecules occupies foremost importance. CDs are both electron donors and receptors, and literature evidences exemplify quenching of their fluorescence in the vicinity of several target analyte molecules facilitated through the electron transfer between them. This section attempts to review different eco-friendly raw materials such as plant biomass, animal products, food items and waste materials used in the preparation of CDs through different approaches, mainly hydrothermal which uses milder conditions for sensing applications.

#### **CDs from plant biomass**

Plant biomass can be defined as the weight of an entire live plant both above and below the ground at a given time, which can serve as a suitable carbonaceous precursor material for the fabrication of CDs. Plant biomass has plenty of lignin, cellulose, and hemicellulose, carbohydrates such as sugar (xylose, mannose, galactose, and arabinose), starch and polysaccharides as primary heteropolymers. These constituents are rich in carbon (C), N, oxygen (O), sulphur (S) and phosphorous (P) and facilitate the synthesis of CDs with diverse functional groups (OH, COOH,  $NH_2$ , SH, etc.) on their surface (Meng et al. 2019; Shahraki et al. 2022; Wang et al. 2020; Zulfajri et al. 2020; Han et al. 2019). The as-prepared CDs derived from biomass have numerous advantages including the use of cheap raw materials, easy control of reactions, mass production and high yield. Different parts of the plant, edible or non-edible including root, leaf, stem, bud, flower and fruit, were efficiently used as cheap reservoirs of C-rich raw materials to prepare CDs with varying physicochemical features for sensing applications as illustrated below.

#### Metal ion sensing

Some heavy metals such as chromium (Cr), copper (Cu), cobalt (Co), iron (Fe), manganese (Mn), zinc (Zn) and tin (Sn) in trace amounts are essential to living systems, but at higher concentrations they are harmful and toxic to humans. Hence, detection of these metal ions is of prime significance and several researchers have exploited the fluorescence quenching phenomena of CDs for metal ion sensing.

The leaf extract of a popular and commonly used herb Ocimum sanctum (Tulsi) was used as a carbon source by Kumar et al. (Kumar et al. 2017) and Doshi et al. (Doshi and Mungray 2020) for the preparation of CDs using hydrothermal method. The raw material incorporated carvacrol, wide variety of acids including rosmarinic, oleanolic and ursolic and many more constituents with a variety of functional groups such as alcohols, aldehydes and ketones, which are rich in C, N and O that was tailored onto the surface of CDs without the aid of any surface passivating agents. The as-obtained uniform, spherical and water-soluble CDs displayed green fluorescence under 365 nm Ultraviolet (UV) illumination. These CDs were used for lead (Pb) ion detection in water samples and live cells, as they displayed Pb<sup>2+</sup>-induced fluorescence quenching. Bandi and co-workers developed highly fluorescent N-doped CDs from Lantana camara berries, which are abundant in C-rich components including polyphenols, glycosides and carbohydrates (Bandi et al. 2018). Ethylenediamine was used as the N-rich precursor. These N-doped CDs (N-CDs) demonstrated highly sensitive and selective fluorescence turn-off-based Pb<sup>2+</sup> detection. The nanoprobe with various surface polar groups was successfully applied for precise detection of  $Pb^{2+}$  in the real water and human serum as well as urine samples.

Liu et al. reported an effective fluorescent sensing platform using N-CDs obtained from grass through simple hydrothermal treatment for label-free identification of  $Cu^{2+}$ ions in water samples (Liu et al. 2012a). The chelation of paramagnetic  $Cu^{2+}$  with CDs quenched the blue fluorescence through electron or energy transfer, which was completely restored in the presence of ethylenediaminetetraacetic acid that had high  $Cu^{2+}$  affinity. Later Sabet et al. utilized these N-CDs for adsorption of Cd<sup>2+</sup> and Pb<sup>2+</sup> from water (Sabet and Mahdavi 2019). Pipe tobacco was chosen as yet another green starting material by Sha et al. as it contained many N-containing compounds that can serve as organo-nitrogen source (Sha et al. 2013). These N-CDs prepared via hydrothermal process allowed chemical passivation and could selectively detect  $Cu^{2+}$  ions with a LOD of 0.01  $\mu$ M, which interestingly is much lower than that of quantum dots based on CdS and ZnS (Gattás-Asfura and Leblanc 2003). Blue fluorescent CDs were prepared by Shi et al. through pyrolysis from leeks, which are rich in carbohydrates, protein and dietary fibre (Shi et al. 2016). These nanosensors were also applied as biosensing probes for both  $Cu^{2+}$  and pH in live cells. The pH detection range was from 3.5 to 10 and the LOD for  $Cu^{2+}$  was 0.05  $\mu$ M.

Bamboo leaves are rich in complex carbohydrates that carry hydroxyl, carbonyl/carboxylic moieties and hence were used as C source in the synthesis of CDs. Carbohydrates undergo dehydration, decomposition and aromatization during the hydrothermal treatment to form CDs via nucleation. Liu et al. in 2014 prepared CDs from bamboo leaves and further coated them with branched polyethyeneimine (PEI), a water-soluble cationic polymer, which contains primary, secondary and tertiary amino groups to achieve surface passivation and metal ion chelation (Liu et al. 2014). These CDs were used for the detection of  $Cu^{2+}$  at pH 4 with a LOD of 115 nM. The fluorescence quenching of the surface-modified and stable nanoparticles in the presence of  $Cu^{2+}$  was achieved through the formation of cupric amine complexes on the surface of CDs. Later, in yet another report in 2019 utilizing bamboo leaves, Liu et al. synthesized two types of dual- and three-emission hybrid CDs for ratiometric sensing of Pb<sup>2+</sup>and mercury (Hg<sup>2+</sup>)<sup>-</sup>ions (Liu et al. 2019). Flavonoids and chlorophyll were extracted from bamboo leaves via ethanol treatment, which further served as the starting materials for subsequent solvothermal synthesis of multiemissive nanohybrids under neutral and alkaline conditions, respectively. The fabricated blue emissive nanohybrids demonstrated specific binding of Pb<sup>2+</sup> and Hg<sup>2+</sup> to the flavonoid and porphyrin moieties with LODs of 0.14 nM and 0.22 nM, respectively.

The dry matter of *Prosopis juliflora* is characterized by high sugar content and protein (Santos et al. 2013). Pourreza et al. created an off–on CDs-based probe from *P. juliflora* leaves via carbonization technique for detecting  $Hg^{2+}$  and chemet drug (Pourreza and Ghomi 2019). The CD solution exhibited bright blue fluorescence under UV source, which was effectively quenched (LOD of 1.26 ng mL<sup>-1</sup>) due to the coordination of  $Hg^{2+}$  to the N-containing surface functional groups, C-O and COOH moieties. The fluorescence was restored rapidly in the presence of chemet, which has high affinity towards Hg<sup>2+</sup> and chelates through sulphur and carboxyl groups, freeing the Hg<sup>2+</sup> from the surface groups of the CDs. Easily available lotus root, which is rich in amino acids, alkaloids, glucoproteins and polysaccharides, was utilized as an economical C and N source for the preparation of Hg<sup>2+</sup> detecting nanodots by Gu et al. (Gu et al. 2016). These nanosensors obtained through MW-assisted synthesis had relatively higher QY due to the presence of N-containing groups from lotus root rather than at the expense of surface passivation reagents. The CDs demonstrated selectivity and sensitivity towards Hg<sup>2+</sup> with a LOD of 18.7 nM. The selective and static turn-off fluorescence behaviour compared to other interfering cations was due to the faster chelating kinetics of  $Hg^{2+}$  towards the hydroxyl, carboxyl and amino groups on the CD surface coupled with PET from conduction band to the complex states of Hg<sup>2+</sup> (Achmad and Budiawan 2017). Tamarindus indica leaves that contain vitamin-C, proteins and carbohydrates as constituents are abundant in C, O, N and S which were used to synthesize CDs through simple hydrothermal approach without the use of any surface-modifying agents (Bano et al. 2018). Fluorescent blue CDs with good stability and very high QY displayed highly selective binding capacity towards Hg<sup>2+</sup> through surface S atoms to enable turn-off fluorescence sensing via electron transfer process and turn-on sensing of glutathione (GSH) due to formation of Hg-S by freeing the CDs to restore the fluorescence.

Yu et al. used the hydrothermal approach to prepare CDs having surface hydroxyl and carboxylic groups from *Jinhua bergamot* as the carbon source for selective and rapid sensing of Hg<sup>2+</sup> and Fe<sup>3+</sup> with a LOD of 5.5 nM and 0.075  $\mu$ M, respectively, based on dynamic quenching of blue fluorescence of CDs (Yu et al. 2015). Similarly, Raja et al. presented large-scale production of CDs using betel leaves as cost-effective precursors using hydrothermal approach (Raja and Sundaramurthy 2018). The surface modification of the synthesized nanoparticles was not required due to the presence of O-rich hydroxyl groups. The sensing of Fe<sup>3+</sup> (LOD of 50 nM) was achieved through quenching of blue fluorescence emission via charge transfer from the surface carboxylic groups of CDs to the ions.

The flower extract of the medicinally important deciduous shrub *Magnolia liliiflora* contains plenty of phytoconstituents such as essential oils (trans- $\alpha$ -farnesene,  $\delta$ -cadinene) (Fujita 1989) and volatile components (1,8-cineole, farnesol, sabinene,  $\beta$ -pinene,  $\alpha$ -pinene, camphor). Atchudana et al. illustrated the successful utilization of the flower extract, which served as both N and C sources for the synthesis of CDs through hydrothermal technique without any surface passivating agents (Atchudan et al. 2018a). Initially, the hydroxyl group of phytoconstituents underwent dehydration to form furfural derivatives, whereas the N reacted with the carbonyl groups to form stable complex. Further polymerization and condensation of furfural derivatives generated water-soluble polymers. Finally, these polymers of furfural derivatives and complexes underwent carbonization to form N-CDs with good optical features and were well utilized for selective and sensitive detection of  $Fe^{3+}$ with LOD of 1.2  $\mu$ M. In another attempt to detect Fe<sup>3+</sup> ions, Kaur et al. fabricated N-CDs using Vigna radiate sprouts as the sustainable C source and ethylene diamine as the dopant via hydrothermal approach to minimize the surface defects and obtained CDs with very high OY of 58% (Kaur et al. 2019). These highly stable, haemocompatible, cell membrane-permeable and photocytotoxic CDs served as turnoff intracellular Fe<sup>3+</sup> sensor with LOD of 140 nM. Sachdev and Gopinath detailed an effortless one-pot hydrothermal treatment using coriander leaves as precursor material for obtaining fluorescent green CDs without any additional surface passivating agent. Dehydration, carbonization and subsequent in situ surface passivation of coriander leaves rich in carbohydrates and proteins containing C, N and O elements abundantly under high temperature and pressure generated CDs as selective Fe<sup>3+</sup> detection probes (Sachdev and Gopinath 2015). Atchundan and coworkers synthesized CDs by utilizing aqueous ammonia and Phyllanthus acidus, which is rich in vitamin C as the N and C sources, respectively (Atchudan et al. 2018b). These N-CDs were used for labelfree sensing of  $Fe^{3+}$  with a LOD of 0.9  $\mu$ M through fluorescence turn-off response. N-CDs were prepared from aqueous ammonia and Chionanthus retusus (Chinese fringetree) fruit extract rich in phenolic compounds and polysaccharides as N and C source, respectively, via hydrothermal-carbonization in yet another attempt by Atchundan et al. (Atchudan et al. 2017). These C nanoparticles displayed highly sensitive and selective fluorescence turn-off feature towards  $Fe^{3+}$ , with a LOD of 70 µM. Further, Edison and coworkers reported the synthesis of N-CDs through hydrothermal carbonization of Prunus avium (wild cherry) fruit extract, which was used as a fluorescence turn-off sensor for Fe<sup>3+</sup> in water with a LOD of 0.96 µM (Edison et al. 2016). The fruit extract served as a good C source as it had acids (malic, citric, fumaric, and shikimic), sugars (glucose, fructose, sucrose, and sorbitol) and phenolic compounds such as flavanols, hydroxycinnamic acids and anthocyanins. Glucose and malic acid were the major phytoconstituents for the generation of CDs, whereas aqueous ammonia was used as the N-dopant. Murugan and Sundaramoorthy prepared green CDs with carboxyl and hydroxyl surface groups from Borassus flabellifer flower through pyrolysis approach without any surface modifications. The CDs demonstrated selective and sensitive detection of Fe<sup>3+</sup> through fluorescence quenching (Murugan and Sundramoorthy 2018).

A green and facile strategy for preparation of CDs using *Catharanthus roseus* (white) leaves with monomeric indole alkaloids: vindoline, catharanthine, vinblastine and

Precursor materials	Synthesis approach	$\lambda_{abs}; \lambda_{ex}; \lambda_{em}$ (nm)	FL colour (λ <sub>ex.</sub> 365 nm)	QY (%)	Detecting ion/ molecule	LOD	References
Tulsi	Hydrothermal	280,330; 450; 500	Green	9.3	Pb <sup>+2</sup>	0.59 nM	Kumar et al. 2017; Doshi and Mungray 2020)
Lantana camara ber- ries	Hydrothermal	285,356; 360; 450	Blue	33.2	Pb <sup>+2</sup>	9.64 nM	Bandi et al. 2018)
Grass	Hydrothermal	280; 360; 443	Blue	6.2	Cu <sup>2+</sup>	1 nM	Liu et al. 2012a)
Pipe tobacco	Hydrothermal	-; 369; 450	Blue	3.2	Cu <sup>2+</sup>	0.01 µM	Sha et al. 2013)
Leek	Pyrolysis	-; 360; 450 (B CDs) -; 470; 520 (G-CDs)	Blue	5.9	Cu <sup>2+</sup>	0.05 μΜ	Shi et al. 2016)
Bamboo leaves	Hydrothermal solvothermal	491; 611, 665 (alkaline condition)	Blue	7.1	Cu <sup>2+</sup> Pb <sup>2+</sup> & Hg <sup>2+</sup>	115, 0.14 & 0.22 nM	Liu et al. 2014; Liu et al. 2019)
Prosopis juli- flora leaves	Carbonization	-; 350; 437	Blue	5	Hg <sup>2+</sup> & Chemet	1.26 & 1.4 ng mL <sup>-1</sup>	Pourreza and Ghomi 2019)
Lotus root	MW	280; 360; 435	Blue	19	Hg <sup>2+</sup>	18.7 nM	Gu et al. 2016)
Tamarindus indica leaves	Hydrothermal	280; 365; 433	Blue	46.6	GSH & Hg <sup>2+</sup>	1.7 µM & 6 nM	Bano et al. 2018)
Jinhua berga- mot	Hydrothermal	-; 330; 440	Blue	50.8	${\rm Fe}^{3+}$ & ${\rm Hg}^{2+}$	5.5 nM & 0.075 μM	Yu et al. 2015)
Betel leaves	Hydrothermal	215,275; 380; 456	Blue	-	Fe <sup>3+</sup>	50 nM	Raja and Sunda- ramurthy 2018)
<i>Magnolia lili- iflora</i> flower	Hydrothermal	275,315; 340; 405	Blue	11	Fe <sup>3+</sup>	1.2 μM	Atchudan et al. 2018a)
Vigna radiate sprouts	Hydrothermal	276,344; 360; 422	Blue	58	Fe <sup>3+</sup>	140 nM	Kaur et al. 2019)
Coriander leaves	Hydrothermal	273,320; 320–480; 400–510	Green	_	Fe <sup>3+</sup>	0.4 µM	Sachdev and Gopinath 2015)
Phyllanthus acidus	Hydrothermal	284; 350; 420	Blue	14	Fe <sup>3+</sup>	0.9 μΜ	Atchudan et al. 2018b)
Chionanthus retusus fruit	Hydrothermal	269,301; 340; 425	Blue	9	Fe <sup>3+</sup>	70 µM	Atchudan et al. 2017)
Prunus avium fruit	Hydrothermal	270,332; 310; 411	Blue	13	Fe <sup>3+</sup>	0.96 μΜ	Edison et al. 2016)
<i>Borassus flabel- lifer</i> flower	Thermal pyrolysis	282; 320; 403	Blue	11.7 (200 °C), 13.9 (300 °C & 10.8 (400 °C)	Fe <sup>3+</sup>	10 nM	Murugan and Sundramoorthy 2018)
Catharanthus roseus leaves	Hydrothermal	270,350; 330; 405	Greenish-blue	28.2	Al <sup>3+</sup> & Fe <sup>3+</sup>	0.5 & 0.3 µM	Arumugham et al. 2020)
Pseudo-stem of banana plants	Hydrothermal	284; 340; -	Green	48	$Fe^{3+} \& S_2O_3^2$	6.5 nM & 8.47×10 <sup>-7</sup> M	Vandarkuzhali et al. 2017)
Osmanthus Fra- grans flower	Hydrothermal	280; 340; 410	Blue	18.5	Fe <sup>3+</sup> & ascorbic acid	5 nM & 5 µM	Wang et al. 2019)
Cornstalk	Hydrothermal	234,280, 332; 410; 500	Blue	7.6	${\rm Fe}^{2+}$ & ${\rm H}_2{\rm O}_2$	0.18 & 0.21 µM	Shi et al. 2017)
Hibiscus sab- dariffa	Hydrothermal	267; 285; 429	Blue	-	Cr <sup>6+</sup>	-	Komalavalli et al. 2020)
Purple perilla	Hydrothermal	270; 360; 450	Blue	9.01	$Ag^+$	1.4 nM	Zhao et al. 2019)
Lychee seeds	Pyrolysis	-; 365; 440	Blue	10.6	Methylene blue	50 mM	Xue et al. 2015)
Chrysanthe- mum buds	Hydrothermal	240,342; 365; 450	Blue	28.5	Curcumin	21 ng mL <sup>-1</sup>	Bu et al. 2019)

Table 1	Various plant sources as	s green precursors	for the synthesis of	f CDs utilized for se	ensing applications
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Tab	le 1	(continued)
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Precursor materials	Synthesis approach	$\lambda_{abs}; \lambda_{ex}; \lambda_{em}$ (nm)	FL colour (λ <sub>ex.</sub> 365 nm)	QY (%)	Detecting ion/ molecule	LOD	References
Lawsonia iner- mis powder	Hydrothermal	270,380; 360,440; 500	Green	28.7	Methotrexate	$7 \text{ nM } \text{L}^{-1}$	Shahshahanipour et al. 2019)
Lentinus edodes	Hydrothermal	240,290; 330; 410	Blue	5.5	Intracellular pH & haemin	$120 \text{ nM L}^{-1}$	Wang et al. 2016a)
Aloe	Hydrothermal	278; 441; 503	Yellow	10.4	Tartrazine	73 nM	Xu et al. 2015)
Rose flowers	Hydrothermal	-; 390; 435	Blue	13.4	Tetracycline	$3.3 \times 10^{-9} \text{ M L}^{-1}$	Feng et al. 2015)
Tagetes erecta flowers	Hydrothermal	320; 420; 495	Blue	63.7	Chlorpyrifos & quinalphos	2.1 ng mL <sup>-1</sup> &1.7 ng mL <sup>-1</sup>	Ghosh et al. 2021a)
Calotropis pro- cera leaves	Hydrothermal	320; 340; 416	Blue	72	Isoprothiolane	11.58 nM	Ghosh et al. 2021b)
Cauliflower	Hydrothermal	280; 325; -	Blue	43	Diazinon, ami- carbazone & glyphosate	0.25, 0.5, & 2 ng mL <sup>-1</sup>	Tafreshi et al. 2020)



Fig. 2 Graphical illustration of CDs obtained from various biomass sources through hydrothermal method for selective metal ion detection based on fluorescence responses under UV light source

vincristine as most abundant constituents was reported by Arumugham et al. (Arumugham et al. 2020). One-step hydrothermal carbonization of C source in the absence of passivating and oxidizing agents led to the formation of yellow suspension of CDs which exhibited pH-dependent fluorescence and aggregation-induced emission (AIE) upon UV-illumination. Prominent chelation-enhanced fluorescence (CEF) and decline in luminescence through chelationquenched fluorescence (CQF) mechanisms were observed in the presence of  $Al^{3+}$  and  $Fe^{3+}$  with an LOD of 0.5 and



**Fig.3** Graphical illustration of CDs obtained from various biomass sources: A through different preparation methods for  $Hg^{2+}$ ,  $Pb^{2+}$  and chemet detection and B through pyrolysis and hydrothermal approach

for selective sensing of different organic molecules based on fluorescence responses under UV light source

 $0.3 \,\mu$ M, respectively. The decrease in fluorescence lifetime for CD-Fe<sup>3+</sup> occurred due to the transfer of electron from excited CDs to the unfilled d-orbital of Fe<sup>3+</sup> resulting in strong nonradiative electron/hole recombination and subsequent AIE quenching. On the contrary, the electrostatic interaction and coordination of aluminium (Al<sup>3+</sup>) ions with CDs led to CD-Al<sup>3+</sup> conjugates with AIE and increased fluorescence lifetime.

Further endeavours were similarly accomplished using easily accessible pseudo-stem of banana plant having cellulose (43%), hemicellulose (16–20%) and lignin (12–16%) as primary constituents to obtain CDs with high QY of 48% by means of hydrothermal condensation (Vandarkuzhali et al. 2017). The CDs were highly selective to Fe<sup>3+</sup> with a LOD of  $6.5 \times 10^{-9}$  M, and their green fluorescence was quenched due to non-radiative electron transfer from the CDs to Fe<sup>3+</sup> ions. Furthermore, approximately 82% of the fluorescence was recovered upon addition of 1.25 M S<sub>2</sub>O<sub>3</sub><sup>2-</sup> ion with a LOD of  $8.47 \times 10^{-7}$  M. The high affinity of Fe<sup>3+</sup> towards the oxygen atoms of the carboxylate groups on the surface of CDs and the strong interaction between  $Fe^{3+}$  and  $S_2O_3^{2-}$  ions were responsible for the dynamic fluorescence off-on feature. Wang et al. used dried flowers of Osmanthus fragrans, rich in flavonoids, phenolic acids and henylethanoid glycosides (Wang et al. 2019) as natural raw material for the preparation of CDs using hydrothermal approach. The flower-derived CDs could sense Fe<sup>3+</sup> through fluorescence quenching via IFE mechanism in reallife water samples. In addition, the nanosensor could serve as a fluorescent "off-on" sensor to detect ascorbic acid based on the redox reaction between Fe<sup>3+</sup> and ascorbic acid. Shi et al. mentioned the usage of cornstalk as a bioprecursor for CDs via hydrothermal method without any additional surface modification (Shi et al. 2017). Hydrogen peroxide  $(H_2O_2)$  was added to oxidize Fe<sup>2+</sup> present in the sample to  $Fe^{3+}$ , which formed metal hydroxide complex with the hydroxyl and other oxygen functional groups present on the surface of the CDs to effectively quench the fluorescence.

The label-free probe had a LOD of 0.18  $\mu$ M and 0.21  $\mu$ M for Fe<sup>2+</sup> and H<sub>2</sub>O<sub>2</sub>, respectively, without the precipitation of iron oxyhydroxides.

In pursuit of further natural materials as C sources for obtaining fluorescent CDs via hydrothermal method, Komalavalli and team utilized Hibiscus sabdariffa leaves (Komalavalli et al. 2020). The CDs that owned excitation and size-dependent photo luminescent behaviour exhibited a phenomenal Cr<sup>6+</sup> detecting capacity through fluorescence turn-off. CDs synthesized from Purple perilla, a Chinese traditional medicinal herb as C and N source via the hydrothermal method, were used to detect silver ions (Ag<sup>+</sup>) through turn-off blue fluorescence (Zhao et al. 2019). The detection mechanism was through static fluorescence quenching attributed to the nonradiative electron transfer from the excited states of CDs to the d-orbital of Ag<sup>+</sup> and Ag<sup>+</sup>-induced conversion of -CONH - functional group from spirolactam structure to an opened-ring amide. The quenching was maximum at neutral pH. Table 1 summarizes the different plant biomass that has been utilized as precursor materials in the synthesis for CDs, which serve as selective fluorescent sensors for metal detection in water and biological samples. The schematic representation of the synthesis approaches of CDs obtained from different biomasses and their sensing mechanisms reviewed above is presented in Figs. 2 and 3A.

#### **Detection of organic molecules**

Besides the detection of metal ions, the fluorescence feature of CDs was well utilized for the sensing of other organic species. Xue et al. utilized a facile pyrolysis approach without any complex post-treatment procedures for the production of fluorescent CDs by utilizing lychee seeds as a green source (Xue et al. 2015). The bright blue fluorescence of CDs was selectively quenched by methylene blue, and this feature was utilized for precise quantification of toxic and pollutant methylene blue dye. The reduction in luminescence intensity was due to electrostatic adsorption of the dye on the surface of CDs with a LOD of 50 mM, and a maximum quenching response at pH 8. Bu et al. synthesized N-CDs by one-pot hydrothermal process using chrysanthemum buds as C source and ethylenediamine as dopant (Bu et al. 2019). The graphite-like crystalline CDs displayed self-quenching of fluorescence upon aggregation when their concentration exceeded beyond 40 µg/mL. Efficient detection of curcumin was possible because of the combination of IFE and static quenching, and these CDs can be used to develop a fluorometric assay for curcumin detection with a LOD of  $21 \text{ ng mL}^{-1}$ .

CDs were also obtained from traditional medicinal plant *Lawsonia inermis* (henna leaf) powder, which is a rich source of not only phenolic compounds including coumarins,

flavonoids, naphthalene and gallic acid, but also fatty acids, quinines, steroids and aliphatic hydrocarbons (Shahshahanipour et al. 2019). The carbonization of carbohydrates, which included hydrolysis, dehydration, decomposition, condensation, aromatization and passivation, finally generated the CDs with excitation-dependent fluorescence behaviour. The CDs with polar groups -COOH, -OH and NH<sub>2</sub> on the surface of henna CDs interacted via strong hydrogen bonding between with methotrexate (MTX) through FRET process and were used as a fluorescence quenching probe with a LOD of 7 nM  $L^{-1}$ . Wang et al. reported the use of dehydrated Lentinus edodes (shiitake mushroom) as a biomaterial for the preparation of CDs rich in O with hydroxyl, carboxyl and amine groups using hydrothermal approach (Wang et al. 2016a). The mushroom that contains abundant carbohydrates, lipid, proteins and amino acids served as the sole C and N source to achieve both surface passivation and N-doping simultaneously. The stable CDs exhibited intracellular pH sensing feature within a biological significant pH range of 4.0-8.0 and also acted as a sensitive sensor for haemin with a LOD of 120 nM  $L^{-1}$ .

Xu and coworkers developed CDs using aloe as a C source via hydrothermal method for the detection of tartrazine, a synthetic food colourant based on fluorescence quenching mechanism (Xu et al. 2015). The CDs displayed remarkable yellow fluorescence as opposed to conventional blue luminescence and was effectively applied towards routine analysis of tartrazine in food samples. The pH-dependent fluorescence quenching with a LOD of 73 nM was due to the ground-state complexes generated through interaction between tartrazine and CDs. Feng et al. reported the fabrication of CDs through hydrothermal approach from rose flowers for sensing tetracycline antibiotic based on fluorescence quenching mechanism with a LOD of  $3.3 \times 10^{-9}$  molL<sup>-1</sup> (Feng et al. 2015). The real-life application was extended to detection of the antibiotic in human urine samples.

Extensive use of pesticides to escalate the agricultural yield has posed increasing health risk to humans, even when used at lower concentrations. In this context, Ghosh et al. prepared CDs that mainly included C-, O- and N-containing surface functional groups from Tagetes erecta flower commonly called as Marigold flower via hydrothermal technique for selective detection of chlorpyrifos and quinalphos pesticides (Ghosh et al. 2021a). The as-synthesized CDs possessed negative surface charge (-14.91 mV) which changed to positive on addition of the pesticides due to aggregation effect. The sensing application was investigated by dispersing the CDs in different pesticide, but the CDs displayed marked specific response either as turn-off fluorescence in case of chlorpyrifos or fluorescence turn-on in the presence of quinalphos. The practical utility was confirmed through rapid detection of the two pesticides in rice and fruit samples with good accuracy. Further, the same team reported **Fig. 4** Graphical illustration of CDs obtained through microwave and hydrothermal methods from various animal products for selective sensing application based on fluorescence responses under 365 nm UV light source



the facile synthesis of CDs from the leaves of *Calotropis procera* by hydrothermal method for effective detection of isoprothiolane (Ghosh et al. 2021b). The aldehyde-, aminoand hydroxyl group-rich CDs could specifically detect the fungicide via a turn-off static fluorescence mechanism even in the presence of various other pesticides including thiamethoxam, clodinafop, carbendazim, andimidacloprid. The linkage of carbonyl group of isoprothiolane and amino group of CDs was confirmed for the rapid detection of isoprothiolane, and the nanoprobe was further successfully applied in rice and fruit samples effectively.

Tafreshi et al. reported cauliflower-derived CDs through hydrothermal approach for for application in food safety and environmental monitoring. The CDs could detect diazinon, glyphosate and amicarbazone pesticides (Tafreshi et al. 2020) through turn-off fluorescence without any interference from dialen super and bromacil. These carbonaceous nanoprobes demonstrated good fluorescence-based sensing behaviour for diazinon, glyphosate and amicarbazone in real fruit samples, though they failed to distinguish the three pesticides in their mixture. The summary of the CDs obtained from plant biomass for the detection of various organic molecules is provided in Table 1. The schematic representation of the synthesis approaches of CDs obtained from different biomasses for sensing of various organic molecules reviewed above is presented in Fig. 3B.

#### **CDs from animal products**

Animal products are rich sources of protein and saturated fat in addition to the presence of minerals such as zinc, calcium, vitamin  $B_{12}$  and iron. They have negligible amount of carbohydrate content with the exception of milk products and hence can serve as ideal raw materials for the synthesis of CDs (Scanes 2017). Hence, animal products can be an alternative over external doping agents (Murphy and Allen 2003). Yang et al. reported the synthesis of CDs using honey as C source via hydrothermal treatment (Yang et al. 2014). These CDs of 2 nm diameter exhibited blue fluorescence under UV light and were utilized as sensors for the measurement of Fe<sup>3+</sup> with an optimal efficiency at pH of 6.0 and a LOD of  $1.7 \times 10^{-9}$  M L<sup>-1</sup>. Fluorescence quenching was facilitated by aggregate formantion via coordination bonds with –COOH, –OH and –NH<sub>2</sub> surface functional groups.

 Table 2
 Various animal products and food items as green precursors for the synthesis of CDs utilized for sensing applications

Precursor mate- rials	Synthesis approach	$\lambda_{abs}; \lambda_{ex}; \lambda_{em}$ (nm)	FL colour (λ <sub>ex.</sub> 365 nm)	QY (%)	Detecting ion/ molecule	LOD	References
Animal products							
Honey	Hydrothermal	278; 338; 420	Blue	19.8	Fe <sup>3+</sup>	$1.7 \times 10^{-9} \text{ M L}^{-1}$	Yang et al. 2014)
Pork	Hydrothermal	235,281; 310; 412	Blue	17.3	uric acid	0.05 μΜ	Zhao et al. 2018)
Wool	MW	270; 320–420; 435- 480	Blue	16.3	glyphosate	$12 \text{ ng mL}^{-1}$	Wang et al. 2016b)
Food items					<b>D1</b> 2+		<i>a</i>
seeds	Carbonization	312; 250; 415	Blue	52	Pb <sup>2+</sup>	0.18 nM	Chauhan et al. 2020)
Chocolate	Hydrothermal	-; 280; 354	-	-	Pb <sup>2+</sup>	12.7 nM	Liu et al. 2016)
Mangosteen pulp	Calcination	283,350; 330; -	Blue	6	Fe <sup>3+</sup>	52 nM	Yang et al. 2017)
Rose heart radish	Hydrothermal	281; 330; 420	Blue	13.6	Fe <sup>3+</sup>	0.13 μΜ	Liu et al. 2017)
Papaya powder	Hydrothermal	250–290; 370; 450	Blue	18.9 & 18.3	Fe <sup>3+</sup>	$0.48 \ \& \ 0.29 \ \mu M \\ L^{-1}$	Wang et al. 2016c)
Konjac flour	Pyrolysis	230,320; 335; 393	Blue	22	Fe <sup>3+</sup>	-	Teng et al. 2014)
Garlic	Hydrothermal	282; 340; 428	Blue	13	Fe <sup>3+</sup>	0.22 nM	Chen et al. 2016)
Potato	Hydrothermal	272; 323; 405	Blue	15	Fe <sup>3+</sup>	$0.025 \text{ mM L}^{-1}$	Xu et al. 2014)
Sweet potato	Hydrothermal	266; 360; 442	Blue	8.64	Fe <sup>3+</sup>	0.32 µM	Shen et al. 2017)
Watermelon juice	Hydrothermal	282, 355; 355; 439	Blue	10.6	Fe <sup>3+</sup> & Cys	0.16 & 0.27 µM	Lu et al. 2018)
Strawberry juice	Hydrothermal	283, 344; 427	Blue	6.3	Hg <sup>2+</sup>	3 nM	Huang et al. 2013)
Cucumber juice	Hydrothermal	256; 370; 450 (100 °C) 269; 418; 505 (120 °C) 283; 514; 571 (150 °C)	Blue (100 °C) Greenish blue (120 °C) Green (150 °C)	3.25	Hg <sup>2+</sup>	$1.8 \times 10^{-7} \text{ M}$	Wang et al. 2014b)
Flour	MW	288; 365; 442	Blue	5.4	Hg <sup>2+</sup>	0.5 nM	Qin et al. 2013)
Citron	Hydrothermal	333; 265–355; 365–455	Blue	34.5	Hg <sup>2+</sup> & Cys	0.15 μM & 40 nM	Xavier et al. 2018)
Chinese yam	Hydrothermal	284; 320; 420	Blue	9.3	Hg <sup>2+</sup> & 6-MP	0.67 & 1.26 nM	Li et al. 2015)
Cornflour	Hydrothermal	282; 360; 441	Blue	7.7	Cu <sup>2+</sup>	1 nM	Wei et al. 2014)
Pakchoi juice	Hydrothermal carbonization	281; 380; 473	Blue	37.5	Cu <sup>2+</sup>	9.98 nM	Niu et al. 2015)
Eleusine cora- cana	Pyrolysis	230,305; 340; 425	Blue	-	Cu <sup>2+</sup>	10 nM	Murugan et al. 2019)
Groundnut	Hydrothermal	279; 360; 443	Blue	17.6	Cr <sup>6+</sup>	$0.1 \text{ mg L}^{-1}$	Roshni et al. 2019)
Enokitake mushroom	Hydrothermal	250–285; 360; 470	Blue	39	Cr <sup>6+</sup> & VOC	0.73 mM	Pacquiao et al. 2018)
Lemon juice	Hydrothermal	280; 420; 540	Green	21	V <sup>5+</sup>	3.2 ppm	Hoan et al. 2019)
Caffeine	Solid-state method	270 (o-CDs) 270, 360 (u-CDs): 340; 400 (o-CDs), 360; 420 (u-CDs)	Blue	69	Ag <sup>+</sup>	-	Dang et al. 2018)
Naked oats	Pyrolysis & MW	247,279; 310; 347,428	Blue	3	Al <sup>3+</sup> & pH	7.4 μΜ	Shi et al. 2015)
Tapioca sago	Hydrothermal	260; 350; 450	Blue	-	F <sup>-</sup>	_	Basu et al. 2015)

 Table 2 (continued)

Precursor mate- rials	Synthesis approach	$\lambda_{abs}; \lambda_{ex}; \lambda_{em}$ (nm)	FL colour (λ <sub>ex.</sub> 365 nm)	QY (%)	Detecting ion/ molecule	LOD	References
Water Chestnut, Onion	Hydrothermal	242,333; 370; 475	Green-blue	12	Coenzyme A	0.01 µM	Hu et al. 2017)
Lemon & onion juice	MW	280; 340; 425	Blue	23.6	Riboflavin	1.0 ng mL <sup>-1</sup>	Monte-Filho et al. 2019)
Yogurt	Pyrolysis & hydrothermal	282,333; 340; -	Blue	1.5	Formic acid	-	Moonrinta et al. 2018)
Sweet pepper	Hydrothermal	282; 360; 450	Blue	19.3	Hypochlorite	$0.05-0.06 \ \mu M$ L <sup>-1</sup>	Yin et al. 2013)
Tomato juice	Hydrothermal	-; 367; 440	Blue	13.9	CEA	0.3 ng mL <sup>-1</sup>	Miao et al. 2016)
Corn juice (Zea mays)	Hydrothermal	225,282; 353; 446	Blue	_	γ-aminobutyric acid	6.46 µM	Sangubotla and Kim 2019)



Fig. 5 Schematic representation of CDs obtained from various food items for the detection of A  $Pb^{2+}$  through carbonization and hydrothermal approach, **B** Fe<sup>3+</sup> and **C** Hg<sup>2+</sup> through hydrothermal approach based on fluorescence responses under 365 nm UV light source

The method was extended to practical use in real blood samples with little inference from other substrates. The study by Zhao et al. illustrated the facile use of pork that contains complex biomolecules including fat, proteins, vitamins (B, C and E) carbohydrates, cholesterol and minerals as source for fluorescent CDs via hydrothermal treatment (Zhao et al. 2018). Hydrophilic –COOH, –NH<sub>2</sub> and –OH groups, which decorated the surface of CDs facilitated uric acid detection via surface adsorption resulting in static fluorescence quenching at an optimum pH of 8.0. The adsorption process was assisted by the combination of electrostatic and hydrogen bonding interactions between CDs and uric acid and change in surface charge leading to the aggregation of CDs.

Wang et al. adopted a MW-assisted pyrolysis strategy for the fabrication of CDs using wool as an inert raw material (Wang et al. 2016b). The fluorescence of CDs was significantly quenched by Ag nanoparticles due to IFE. However, negatively charged glyphosate induced the aggregation of positively charged Ag nanoparticles via electrostatic interaction, which led to fluorescence recovery at an optimal pH of 4.0. These nanoprobes were effectively applied for precise and accurate detection of glyphosate in cereal samples through fluorescence recovery with a LOD of 12 ng mL<sup>-1</sup>. The reduction in the surface charge density and the crosslinking of the neighbouring Ag nanoparticles contributed to the high selectivity for glyphosate detection. Figure 4 schematically depicts the CDs obtained through different preparation methods from various animal products for sensing application based on fluorescence responses. Table 2 summarizes the synthesis of CDs for sensing application from animal products as precursor materials.

#### CDs from food items

Food items were chosen as C sources for the synthesis of CDs as they are easily available, and considerable OY is obtained upon thermal treatment or ultrasonication. These precursors contain small molecules like citric acid, ascorbic acid, polyethylene glycol, glucose, etc. (Chu et al. 2019). Pearl millet, which is one of the primary food crops grown worldwide, was chosen as a raw material for the preparation of value-added highly fluorescent CDs with water as the reaction medium (Chauhan et al. 2020). The synthetic approach was completely green without the use of any conventional solvents, and the CDs were successfully exploited as both colorimetric and fluorimetric sensor for  $Pb^{2+}$ . The yellow CD solution not only turned colourless visually, but also displayed a fluorescence enhancement in the presence of  $Pb^{2+}$  ions in an aqueous sample (Fig. 5A), exhibiting an LOD of 0.18 nM with 90% average recovery rate. The optical and luminescence changes were ascribed to the modulation of energy charge transfer process and complexation of Pb<sup>2+</sup> with the CDs. Later Liu et al. developed fluorescent CDs derived from chocolate, which is rich in C and O, without any chemical modification through one-step hydrothermal strategy for detection of  $Pb^{2+}$  ions in water sample (Liu et al. 2016). The chemosensor was highly selective and sensitive (Fig. 5A) at pH=7 with a low LOD of 12.7 nM. The fluorescence quenching effect was due to the efficient chelation between  $Pb^{2+}$  and surface hydroxyl groups of CDs.

Yang et al. devised a green approach to synthesize CDs using Mangosteen pulp via simple pyrolysis without addition of any surface modifying agents (Yang et al. 2017). Mangosteen pulp contains large number of organic entities like dietary fibre, amino acid, carbohydrates, vitamin etc. and has been proved as an abundant C reservoir. These nanoparticles not only exhibited good potential as a Fe<sup>3+</sup> sensor with a LOD of 52 nM in aqueous solutions, but also served as a sensitive temperature probe as their fluorescence intensity decreased linearly with increasing temperature from 10 °C to 90 °C with excellent recoverability. Liu et al. reported rose-heart radish rich in C, N and O containing anthocyanins, proteins, amino acids, carbohydrates and vitamins as a precursor to prepare N-CDs (Liu et al. 2017). The CDs prepared using hydrothermal treatment facilitated Fe<sup>3+</sup> sensing in environmental water samples with a LOD of 0.13 µM and acceptable recoverability. Yet another report on synthesis of two types of CDs with papaya powder in deionized water or 90% ethanol without any surface modification using green hydrothermal approach was reported by Wang et al. with good QYs of 18.98% and 18.39%, respectively (Wang et al. 2016c). The papaya-derived CDs obtained using both the solvents showed good detection capabilities for  $Fe^{3+}$ through fluorescence quenching, with LOD of 0.48  $\mu$ mol L<sup>-1</sup> and 0.29  $\mu$ mol L<sup>-1</sup>, respectively. These papaya-derived CDs demonstrated sensitive and selective detection of Fe<sup>3+</sup> in haem capsules. Konjac flour that constitutes heteropolysaccharide konjac glucomannan, protein, starch and inorganic salts such as Na<sub>2</sub>SO<sub>4</sub> and NaCl was used as a cost-effective precursor for preparing CDs using pyrolysis approach by Teng et al. (Teng et al. 2014). Further, Chen and coworkers prepared N and S co-doped CDs by hydrothermal treatment of garlic (Chen et al. 2016). Plentiful amount of C, N, S and O from crude protein, amino acid, allicin, niacin, lipid,



Fig. 6 Graphical illustration of CDs obtained from food items for detection of metal ions and organic molecules based on fluorescence responses under UV light source

carbohydrates, citral and vitamin enabled the fabrication of easily surface-functionalized CDs from garlic. The pyridinic-N, pyrrolic-N and thiophene-S formed on the surface of the CDs as a consequence of N and S co-doping facilitated Fe<sup>3+</sup> sensing through fluorescence turn-off response in environmental water samples with excellent sensitivity as well as repeatability with LOD of 0.22 nM. Xu et al. demonstrated the synthesis of CDs through hydrothermal method using potatoes, which are rich in starch as precursor material (Xu et al. 2014). Strong fluorescence quenching due to the complex formation between the CDs and Fe<sup>3+</sup> facilitated the selective ion sensing with a low detection limit of 0.025 mM L<sup>-1</sup> and was extended for real-life applications in environmental samples. Facile preparation of CDs from carbohydrate content-rich sweet potato through hydrothermal strategy for  $Fe^{3+}$  sensing with a LOD of 0.32  $\mu$ M was reported by Shen and coworkers (Shen et al. 2017). Lu and coworkers reported the green approach of preparation of N-CDs from watermelon juice using hydrothermal carbonization method (Lu et al. 2018), which exhibited turnoff fluorescence behaviour upon addition of Fe<sup>3+</sup> ions with a LOD of 0.16  $\mu$ M. Further the CD-Fe<sup>3+</sup> complex system could selectively sense Cys (LOD =  $0.27 \mu$ M) based on turnon fluorescence due to the binding preference of Cys towards  $Fe^{3+}$  ions. The schematic illustration in Fig. 5B depicts the food materials used for the fabrication of CDs that serve as selective probes for Fe<sup>3+</sup> ions.

Huang and coworkers reported N-CDs using hydrothermal method from strawberry juice (Huang et al. 2013). These CDs also enabled selective and facile sensing of Hg<sup>2+</sup> through dynamic fluorescence quenching with a LOD of 3 nM, which was further extended for practical use to measure environmental water samples with acceptable recovery. Cucumber has abundant C, N, O, P, S, and H (hydrogen) which constitutes carbohydrates, proteins, lipids and GSH. Therefore, Wang and coworkers described the synthesis of N, S and P co-doped CDs with plentiful O and N, but limited amount of P and S functional groups through hydrothermal approach from cucumber juice (Wang et al. 2014b). The CDs served as effective probe for Hg<sup>2+</sup> ion detection with a LOD of  $1.8 \times 10^{-7}$  M. Qin et al. prepared CDs from flour that was highly sensitive and selective towards Hg<sup>2+</sup> with a low LOD of 0.5 nM and was successfully extended for practical use in lake water samples (Qin et al. 2013). The selective pH-dependent fluorescence quenching due to energy or electron transfer in the presence of Hg<sup>2+</sup> was recovered on adding Cys, a strong Hg<sup>2+</sup> chelator that forms a Hg-S bond. Further, Xavier et al. reported selective detection of  $Hg^{2+}$  (LOD=0.15  $\mu$ M) and Cys via fluorescence turn-off-on mechanism using CDs derived from citron fruit extract, which is rich in citric acid and N-doping using human urine waste, which has abundant amino compounds (Xavier et al. 2018). The initial fluorescence quenching occurs when Hg<sup>2+</sup> coordinates with amine and carboxylic groups of CDs through an electron transfer process and the fluorescence switches on during the complex formation when Hg<sup>2+</sup> competitively binds with Cys through thiol- and N-containing functional groups. The considerable recovery in human urine samples validated the real sample analysis of the low-cost and biocompatible sensor system with rapid response based on dynamic fluorescence quenching and restoration.

Li and coworkers employed hydrothermal method to synthesize N-CDs using Chinese vam, which is a tuberous root that comprises of an assortment of chemical components including amino acids, alkaloid, glucoprotein and polysaccharides, which serve as abundant C and N sources (Li et al. 2015). This doped CD system was further modified by the carboxyfluorescein (FAM)-labelled ssDNA macromolecules, which were stabilized by strong  $\pi$ - $\pi$  stacking between the nucleobases and C = O/C = N groups for the sensitive analysis of the anticancer drug 6-mercaptopurine (6-MP) and  $Hg^{2+}$ . 6-MP chemically combines with CDs and FAM-DNA through hydrogen bonds with nitrogen base pairs of DNA and  $\pi$ - $\pi$  conjugations. The specific interactions between thymine (T) of DNA and Hg<sup>2+</sup> break the fluorescent yellow emissive complex between CDs, DNA and 6-MP resulting in the quenching of fluorescence. The hybrid nanosensor was used for the determination of 6-MP (LOD = 0.67 nM) in human serum and  $Hg^{2+}$  (1.26 nM) in water samples with reasonable results. The schematic illustration in Fig. 5C depicts the food materials used for the fabrication of CDs that serve as selective probes for  $Hg^{2+}$  ions.

Wei et al. reported amorphous photoluminescent CDs, which contain C, O and N that were sourced from amylum and protein of cornflour via hydrothermal method (Wei et al. 2014). These CDs with carbonyl, carboxyl, hydroxyl, epoxy and amino groups were used as fluorescent sensors for identifying Cu<sup>2+</sup> with a LOD of 1 nM. Similarly, Niu et al. demonstrated a hydrothermal approach to obtain N-CDs utilizing pakchoi juice as the sole C source without the use of any other solvent (Niu et al. 2015). These CDs also served as a selective and label-free sensing platform for Cu<sup>2+</sup> ions with a LOD of 9.98 nM at pH = 7-7.4. The substantial reduction in the fluorescence intensity of CDs in the vicinity of  $Cu^{2+}$ was due to its chelation with N and O (Fig. 6A). In addition, Murugan and colleagues reported the facile synthesis of graphitic and amorphous CDs, which had a higher affinity for Cu<sup>2+</sup> ions using Finger millet ragi (*Eleusine coracana*) as a carbon source (Murugan et al. 2019). The green CDs contained carbonyl and hydroxyl groups, which could facilitate the formation of coordination bonds with Cu<sup>2+</sup> ions. The practical applicability of the CDs was explored by sensing  $Cu^{2+}$  in real water samples with a detection limit of 10 nM.

Roshni et al. reported hydrothermal carbonization of groundnut powder, which is rich in protein and monounsaturated content for the synthesis of CDs that could detect  $Cr^{6+}$ 

(Roshni et al. 2019). The N-doping of CDs using ethylene diamine improved the QY from 7.87 to 17.6% and was highly selective to  $Cr^{6+}$  in neutral pH with a LOD of 0.1 mg/L. The deprotonation of the surface carboxylic acid to negatively charged carboxylate ion in neutral pH improved the radiative recombination of electrons and holes. An off-on type sensing mechanism was perceived as the fluorescence was recovered using humic acid and GSH as reducing agents as presented in Fig. 6A. Enokitake mushroom, an edible fungus containing bioactive compounds such as polysaccharides, lectins, sterols, protein-glucan complex, proteases, cellulases, laccases and peroxidases, was used as the precursor for fabrication of CDs through hydrothermal method by Pacquiao et al. The enokitake mushroom-derived CDs served as functional nanoprobes for  $Cr^{6+}$  (LOD of 0.73 mM) and volatile organic compounds (VOC) detection (Pacquiao et al. 2018). Surface passivation of CDs with tetraethylenepentamine improved the QY from 11 to 39%. The fabricated fluorescent colorimetric paper-based device exhibited Cr<sup>6+</sup> sensing with a LOD as low as 10 mM (Fig. 6A). The optical electronic nose with CDs integrated into it successfully enabled the detection of alcohol contents in aqueous solutions, and ethanol concentration in real spirit samples.

Hoan et al. developed CDs through hydrothermal treatment using lemon juice as C source (Hoan et al. 2019). These carbonaceous nanodots displayed luminescence quenching enabling sensitive detection of  $V^{5+}$  ions in water and serum samples even in the presence of high concentrations of other metal ions with a LOD of 3.2 ppm (Fig. 6B). The close coordination between the C = O, C–O–C, polar surface hydroxyl groups of CDs and  $V^{5+}$  ions permits the electron transfer from the excited level of CDs to the halffilled 3d orbital of  $V^{5+}$  to facilitate the non-radiative recombination of excitons. Higher thermodynamic affinity and faster binding process of  $V^{5+}$  with the surface functionalities of CDs compared to other metal ions were also responsible for the selective sensing.

A mixture of urea, ammonium persulphate and caffeine that consists of fused pyrimidine and imidazole rings was used as effective raw material for one pot solid-state synthesis of highly fluorescent N- and S-co-doped CDs (Fig. 6A) (Dang et al. 2018). The surface doping enhanced the QY of CDs drastically from 38% to as high as 69%, comparable to those prepared through hydrothermal approach using citric acid with N and S sources. These caffeine-derived CDs were efficient sensors for Ag<sup>+</sup> ions, and the quenching of fluorescence is attributed to the effective distribution of excitons or electron transfer process, which is facilitated by the strong binding affinity and rapid chelating kinetics between ions and the carboxylate, amino or hydroxyl groups of CDs. Dual emissive CDs were prepared by Shi et al. through pyrolysis and MW approach from naked oats for ratiometric sensing of  $Al^{3+}$  and pH (Fig. 6A) (Shi et al. 2015). The strong affinity between the hard acid Al<sup>3+</sup> and the hard base O of the surface carboxyl groups of CDs enabled the sensing response through electron or energy transfer mediated fluorescence quenching. The sensing was applied successfully for Al<sup>3+</sup> in real water samples and pH by monitoring the intensity ratios of the dual fluorescence bands at 428 and 347 nm when excited under 310 nm. Basu and coworkers reported starchrich tapioca sago as a C source for the preparation of CDs that could be used as an ultrafast chemosensor for F<sup>-</sup> ions based on simple fluorescence 'on-off-on' strategy (Fig. 6A) (Basu et al. 2015). The charge transfer process quenches the fluorescence of stable CD-Fe<sup>3+</sup> complex, which could be recovered rapidly on the removal of Fe<sup>3+</sup> from the surface of CDs in the presence of F<sup>-</sup> ions. The selectivity of F<sup>-</sup> is due to the formation of FeF<sub>3</sub>, which is stable in water solution due to high hydration energy.

Interestingly, Hu et al. demonstrated the synthesis of Sand N-codoped fluorescent CDs by utilizing water chestnut containing many nitrogenous compounds and onion containing a large number of thiol compounds as two natural biomass precursors (Hu et al. 2017). The S doping was responsible for the greenish-blue fluorescence of CDs. The OY of 12% was achieved using synergistic effects between N atom in water chestnut and S atom in onion as well as different functional groups, including -OH and -NH2, which can generate many surface defects on CDs and serve as excitation energy traps. These CDs were capable of acting as an off-on probe for Coenzyme A sensing with a LOD of  $0.01 \,\mu\text{M}$ . The surface carboxyl groups bind to Cu<sup>2+</sup> ions leading to fluorescence quenching, which is restored in the presence of coenzyme A as presented in Fig. 6C. Monte-Filho and coworkers utilized one-step MW assisted carbonization method for synthesis of CDs from two bioprecursors such as lemon, which is a rich source of citric and L ascorbic acids, and onion juice, which is an important source of S compounds with ammonium hydroxide as a N-dopant (Monte-Filho et al. 2019). Accurate and fast analysis of riboflavin was perceived due to efficient FRET process between the electron donating nanodots and electron accepting riboflavin (Fig. 6C). The strong hydrogen bonds of > C = O and –NH groups in vitamin B<sub>2</sub> to the surface OH and COOH groups of CDs increased the proximity of the electron donor and acceptor to act as an effective FRET pair. The CDs could selectively determine riboflavin in multivitamin/mineral supplements with satisfactory recovery values despite potential interferences with a LOD of 1.0 ng mL<sup>-1</sup>.

The application of CDs for the detection of vapours or gases is less developed. Moonrinta et al. prepared CDs from yogurt rich in vitamins, calcium, proteins, fats and carbohydrates using a two-step pyrolysis/hydrothermal approach (Moonrinta et al. 2018). These fermented food-derived CDs were integrated into an optical electronic nose for both qualitative and quantitative sensing of highly volatile formic acid



Fig. 7 Graphical illustration of CDs obtained from waste materials for selective sensing of various organic molecules and metal ions based on fluorescence responses under UV light source

vapour generated from aqueous and methanolic solutions at room temperature with a LOD of 7.3% v/v as depicted in Fig. 6C. Polar-polar interaction and hydrogen bonding between the CDs and formic acid facilitated the sensing action. Yin and coworkers prepared CDs in good yield from sweet red pepper for detection of hypochlorite through both up- and down-conversion fluorescence (Yin et al. 2013). The green raw material has abundant carbohydrate, ascorbic acid, carotenoids and other carbonaceous matter. The CDs were synthesized through low-temperature carbonization method and both up- and down-conversion emissions exhibited excellent photostability in varying pH solutions as opposed to CDs obtained from green pepper with low sugar content as C source. Measurement of wide concentrations of hypochlorite was achieved with a low LOD of 0.06 and  $0.05 \ \mu mol \cdot L^{-1}$  for up- and down-conversion fluorescence, respectively. This simple and sensitive detection method offered multi-dimensional signal and low-cost. Moreover, its real-world application was extended to a sensitive and rapid dual-readout assay (up- and down-conversion fluorescence) for hypochlorite in tap water in just one minute. The changed surface state of CDs due to the oxidation of reductive surface hydroxyl groups with hypochlorite resulted in fluorescence quenching, which could be recovered on addition of sodium borohydride as shown in Fig. 6C.

Miao and coworkers used tomato juice as the C source for the preparation of CDs for label-free sensitive detection of tumour-associated carcinoembryonic antigen (CEA) (Miao et al. 2016). The polymerization, dehydration and carbonization of the constituents of tomato juice including saccharides, citric and ascorbic acids during the hydrothermal process generated the fluorescent CDs. The sensing mechanism was based on the fluorescence quenching due to adsorption of ssDNA onto the carboxyl groups of CDs through  $\pi$ - $\pi$  stacking interaction and desorption of aptamers by a competitive mechanism through stronger binding affinity between CEA and CEA-aptamer to recover the fluorescence (Fig. 6C). CEA was quantitatively evaluated with LOD of 0.3 ng mL<sup>-1</sup> in practical samples in a continuous and recyclable way with high sensitivity and selectivity.

Sangubotla and Kim used the hydrothermal approach to prepare CDs from corn juice (Zea mays) and further functionalized them with 3-aminophenylboronic acid and NADP<sup>+</sup> (Sangubotla and Kim 2019). The surface-modified CDs could successfully detect  $\gamma$ -aminobutyric acid (GABA) by utilizing enzyme GABase. The detection was enabled via fluorescence quenching through electron transfer between GABase and substrate due to the formation of NADPH. Further, the sensor demonstrated better recovery results when applied to human cerebrospinal fluid and serum for the determination of GABA. Moreover, the modified CDs displayed good selectivity through fluorescence quenching even in the presence of interferents including acetylcholine, epinephrine, ascorbic acid, glutamate, serotonin and uric acid. The summary of the CDs obtained from animal products and food items as C sources for the detection of various metal ions and organic molecules is provided in Table 2. The schematic representation of the synthesis approaches of CDs obtained from different food products reviewed above is presented in Fig. 6.

#### CDs from biowaste/waste materials

Waste materials accumulate in the environment mainly due to exponential population growth and present life-style of the people. Moreover, effective waste management is energydriven and hence a costly process. Though non-biodegradable waste materials are recycled, the disposal of wet wastes poses environmental issues (Khanal et al. 2019). Biowaste materials including fruit and vegetable peels, non-edible plant parts, discarded food materials, as well as crustacean waste processing being costly, pile up in landfills. In addition, discarding or burning waste produces enormous amount of greenhouse gases such as carbon monoxide, carbon dioxide and methane, which influences global climate conditions. In synergy with the rising global awareness of crafting a sustainable community through minimization of waste materials, the option of advantageously reusing C-rich waste as inexpensive and renewable starting materials for synthesis of CDs has been reported by several researchers with fruitful results (Kaur and Dhillon 2015). Interestingly, these waste materials serve as copious C sources for their effective conversion to high-value functional CDs, besides aiding in waste management.

Peanut shells that are rich in fibres are an ideal reservoir of C and were explored for the preparation of CDs through pyrolysis method at 400 °C by Ma et al. (Ma et al. 2017). The chelation of  $Cu^{2+}$  to the N- and O-containing surface functional groups of CDs led to the decline in luminescence and hence could be used as a fluorescence sensor for detecting  $Cu^{2+}$ . Prawn shells which are an industrial byproduct rich in polysaccharide chitin and small amounts of protein, calcium, lipids and pigments were utilized as C source for fabrication of CDs by Gedda et al. (Gedda et al. 2016). The seafood waste-derived CDs with various polar functionalities including amino, secondary and primary hydroxyl groups served as a selective and sensitive label-free Cu<sup>2+</sup> sensing platform, especially at an optimum pH of 4 with a reaction time of 10–15 min. Moreover, they were applied in monitoring  $Cu^{2+}$ ions from seawater with a LOD as low as 5 nM. The CDs could distinguish the fresh seawater with Cu<sup>2+</sup> spiked samples in the presence of many minerals, organics and other components. IFE and cupric amine complex formation is the possible reasons for fluorescence quenching of CDs by Cu<sup>2+</sup> ions. Tan et al. reported the synthesis of CDs from bulk sago industrial waste using thermal pyrolysis (Tan et al. 2014). The fluorescence of CDs was quenched in the presence of  $Cu^{2+}$  and  $Pb^{2+}$  ions. The non-specific sensitivity of the CDs was due to the degradation of surface organic groups during high temperature pyrolysis treatment. The detection of Cu<sup>2+</sup> through fluorescence quenching using CDs obtained from the above reviewed literatures on waste materials as precursors is presented in Fig. 7A.

Onion waste is an excellent source of fibre and non-structural carbohydrates including glucose, fructose, sucrose and fructooligosaccharides, and alkenyl cysteine sulpoxides. Hence, Bandi and coworkers obtained fluorescent CDs through carbonization and subsequent passivation of aqueous extract of onion waste and ethylene diamine for detection of Fe<sup>3+</sup> ions in real-life applications through dynamic fluorescence quenching mechanism (Bandi et al. 2016). Coconut petioles that constitute cellulose, hemicellulose and lignin were successfully utilized as a suitable C-rich raw material for synthesis of CDs by Gao et al. (Gao et al. 2021). The simple hydrothermal treatment included dehydration, condensation, polymerization, and carbonization or aromatization to form numerous C skeleton and surface functional groups of coconut petioles-derived CDs. The synergetic effect of static and dynamic fluorescence quenching of CDs enabled selective detection of  $Fe^{3+}$  with a LOD of 2.3  $\mu$ M in real water samples. Biowaste material like dwarf banana peel was also used to prepare N-CDs via simple hydrothermal-carbonization treatment as proposed by Atchudan et al. (Atchudan et al. 2020). These nanoprobes could detect Fe<sup>3+</sup> ions efficiently with LOD of 0.66  $\mu$ M. Jiao et al. chose mango peel, which is rich in polyphenols, carotenoids, gallic acid, flavonoids, mangiferin and fibre as the C-rich source to fabricate CDs (Jiao et al. 2019). Mango peel-derived CDs were prepared via carbonization, oxidation, polymerization and nucleation processes during high temperature pyrolysis and concentrated acid oxygenolysis. They were successfully used as a quick fluorescent turn-off sensor for Fe<sup>2+</sup> in ferrous succinate tablets with LOD of 1.2 µM. Large amount of carbonyl, carboxyl, hydroxyl and amino groups adorned the surface of CDs due to self-passivation/N and S doping. The pictorial sketch depicting the sensing of Fe (II and III)

Precursor	Synthesis method	$\lambda_{abs}; \lambda_{ex}; \lambda_{em} \ (nm)$	FL colour (λ <sub>ex.</sub> 365 nm)	QY (%)	Detecting ion/ molecule	LOD	References
Peanut shells	Pyrolysis	-; 312; 413	Blue	10.6	Cu <sup>2+</sup>	4.8 mM	Ma et al. 2017)
Prawn shell	Pyrolysis	280,330; 330; 405	Blue	9	Cu <sup>2+</sup>	5 nM	Gedda et al. 2016)
Sago waste	Pyrolysis	-; 310; 429 (300 °C)	Blue	-	Cu <sup>2+</sup> & Pb <sup>2+</sup>	7.49 & 7.78 μM	Tan et al. 2014)
Onion waste	Pyrolysis	280,370; 380; 464	Blue	28	Fe <sup>+3</sup>	0.31 μM	Bandi et al. 2016)
Coconut petiole	Hydrothermal	278; 360; 437	Blue	1.4	Fe <sup>3+</sup>	2.3 μM	Gao et al. 2021)
Dwarf banana peel	Hydrothermal	272,320; 345; 413	Blue	23	Fe <sup>3+</sup>	0.66 μΜ	Atchudan et al. 2020)
Mango peel	Pyrolysis	-; 310; 425	Blue	8.5	Fe <sup>2+</sup>	1.2 μM	Jiao et al. 2019)
Coconut milk waste	Pyrolysis	276; 360; 440	Blue	-	Hg <sup>2+</sup>	16.5 nM	Roshni and Ottoor 2015)
Pomelo peel	Hydrothermal	280; 365; 444	Blue	6.9	Hg <sup>2+</sup>	0.23 nM	Lu et al. 2012)
Pineapple peel	Hydrothermal	280; 340; -	Blue	42	Hg <sup>2+</sup>	4.5 nM	Vandarkuzhali et al. 2018)
Human nails	Carbonization	275,330; 380,370; 450	Blue	81.4	Cr <sup>6+</sup>	0.3 nM	Chatzimitakos et al. 2018)
Jute caddies	Sonochemical	261, 322; 340; 458	Blue	_	Cr <sup>6+</sup>	0.03 µM	Das et al. 2020)
Lemon peel waste	Hydrothermal	270; 360; -	Blue	14	Cr <sup>6+</sup>	73 nM	Tyagi et al. 2016)
Rice husk	Carbonization, hydrothermal & calcination	-; 358; 439	Blue	3	Sn <sup>2+</sup> , alcohol & VOC	$18.7 \ \mu M \ L^{-1}$	Ngu et al. 2016; Thongsai et al. 2019)
Ginkgo leaves	Hydrothermal	230, 280; 350; 436	Blue	22.8	SASP	$40 \text{ nM } \text{L}^{-1}$	Jiang et al. 2019)
Egg shell mem- brane	MW	-; 275; 450	Blue	14	GSH	$0.48 \text{ mM L}^{-1}$	Wang et al. 2012)
Date kernel	Hydrothermal	275; 340; 430	Blue	12.5	Zoledronic acid	0.04 µM	Amin et al. 2018)

 Table 3
 CDs from biowaste/waste materials for sensing applications

ions using CDs prepared from four different waste materials discussed above is shown in Fig. 7B.

Waste by-product formed during the thermal pyrolysis of coconut milk that comprises primarily of lauric acid was used by Roshni et al. as a bio-precursor for the synthesis of CDs. These carbonaceous nanoparticles obtained through a simple thermal pyrolysis approach could detect Hg<sup>2+</sup> ions with LOD of 16.5 nM in water samples by its remarkably quenching effect (Roshni and Ottoor 2015). Lu et al. adopted hydrothermal process to synthesize hydrophilic, stable blue fluorescent CDs using pomelo peel as label-free, specific detectors of Hg<sup>2+</sup> ions with a LOD of 0.23 nM (Lu et al. 2012). The fluorescence quenching of CDs by  $Hg^{2+}$  was recovered due to the removal of  $Hg^{2+}$  from CDs when Cys, which is a stronger Hg<sup>2+</sup> chelating agent was added to form Hg-S bond. The practicality of the nanoprobe was demonstrated through determination of Hg<sup>2+</sup> in lake water. Vandarkuzhali et al. synthesized blue fluorescent CDs from pineapple peel waste that constitutes cellulose (20 - 25%) of the dry weight), hemicellulose, lignin and pectin by hydrothermal treatment (Vandarkuzhali et al. 2018). The CDs displayed dynamic fluorescence turn-off behaviour towards Hg<sup>2+</sup> ions and subsequent turn-on behaviour for L-Cys.

The CDs exhibited multiple logic gates such as NOT and IMP and were capable of mimicking a security keypad lock device at the molecular level with chemical inputs of  $Hg^{2+}$  ion and L-Cys as presented in Fig. 7C.

Chatzimitakos et al. successfully synthesized N and S codoped CDs from human nail by carbonization at 200 °C for 3 h with a very high QY of 81%, attributed to the intrinsic composition of the waste raw material (Chatzimitakos et al. 2018). Human nail mainly contains keratin that is rich in S-containing Cys amino acid and hence high content of C, N, O, S and H. The CDs were developed as an ultrasensitive nanoprobe for detecting Cr<sup>6+</sup> ions with a LOD of 0.3 nM via a combined IFE and static quenching mechanism. Jute caddies, an industrial waste, which comprises of cellulose, hemicellulose and lignin with amply oxygenated functional groups, were used as a precursor material for sonochemical generation of CDs by Das and coworkers (Das et al. 2020). Concentrated sulphuric acid was added to enable carbonization of cellulose, S-doping and oxidation. The surface modification was done with benzalkonium chloride to obtain surface-quaternized CDs, which served as a fluorescent nanoswitch to detect Cr<sup>6+</sup> in aqueous solutions with a detection limit as low as 0.03 µM. The luminescence that turns-off in the presence of the heavy metal can be recovered in the presence of antioxidant ascorbic acid (LOD of 0.04 uM) and hence can also be used for selective sensing of the vitamin with fast response time. The fluorescence quenching mechanism was solely based on IFE, rather than excited state electron transfer between surface-modified CDs and  $Cr^{6+}$ . Ascorbic acid reduces  $Cr^{6+}$  to  $Cr^{3+}$  to eliminate IFE, thereby turning on the fluorescence of the nanoprobe. Yet another nanoprobe with O-rich surface groups for detection of Cr<sup>6+</sup> was reported by Tyagi and coworkers from lemon peel waste using cost-effective hydrothermal method (Tyagi et al. 2016). The CDs with a LOD of ~73 nM were used to detect Cr<sup>6+</sup> in water purification processes based on fluorescence turn-off approach. The illustrative sketch depicting the sensing of Cr<sup>6+</sup> ions using CDs prepared from different waste materials is shown in Fig. 7C.

Rice husk, an agricultural waste rich in cellulose, hemicellulose, lignin and silica, was used as the precursor for CDs prepared by Ngu et al. through carbonization in the presence of sulphuric acid (Ngu et al. 2016). The as-prepared CDs could detect  $Sn^{2+}$  ions with a LOD of 18.7  $\mu$ M L<sup>-1</sup>. The coordination complex formed between the CDs and Sn<sup>2+</sup> combined with the oxidation of  $Sn^{2+}$  to  $Sn^{4+}$  resulted in the fluorescence turn-off phenomenon. Later in 2019, Thongsai et al. also used the same precursor to prepare the CDs using hydrothermal and calcination methods (Thongsai et al. 2019). These rice husk-derived CDs were extended to real-life application in detecting the alcohol content of a commercial beverage. Moreover, the nanosystem was used as the sensing layer in an optical electronic nose system for not only to sense alcohol vapours (methanol and ethanol) at room temperature, but also to distinguish VOCs based on the modulation of their optical absorbance governed by polar-polar interfacial interactions (Fig. 7D). Jiang et al. used Ginkgo leaf as the C source for the preparation of N-CDs using hydrothermal method as an effective fluorescent sensing platform for label-free, sensitive and rapid detection of salazosulphapyridine (SASP), with a LOD of 40 nM L<sup>-1</sup> and maximum fluorescence turn-off response at pH=6 due to the IFE mechanism of SASP (Jiang et al. 2019). The system was successfully applied for SASP detection in mouse plasma, without any interference from other biomolecules and ions (Fig. 7D). Protein-enriched egg shell membrane ash was used for the green synthesis of CDs by Wang et al. via simple MW-assisted process (Wang et al. 2012). The synthesis of CDs involved fragmentation, polymerization, nucleation of CDs and final oxidization or surface passivation. These nanodots were implemented for rapid detection of GSH based on turn-off-on fluorescence system of the CDs-Cu<sup>2+</sup> system with a LOD of 0.48 mmol  $L^{-1}$ . When GSH is introduced,  $Cu^{2+}$  is removed from CD surface owing to the strong binding preference for biothiols from GSH forming a Cu<sup>2+</sup>–S bond to turn on the fluorescence as depicted in Fig. 7D. Amin and coworkers prepared N-CDs from date kernel through hydrothermal approach for assaying trace amounts of an anticancer drug, zoledronic acid in human biological samples (Amin et al. 2018). Date kernel that contains abundant amount of carbohydrates, fats and trace amounts of proteins and ash is a good C source for the preparation of CDs. The fluorescence of the nanoprobe with plenty of carboxyl, hydroxyl and amine groups was turned off due to interaction with Fe<sup>3+</sup> and was subsequently restored in the presence of zoledronic acid. The retrieval of luminescence occurred because of the generation of free CDs attributed to the strong affinity of the phosphate groups of the drug that resulted in competitive interaction with the Fe<sup>3+</sup> ions as portrayed in Fig. 7D. The label-free, rapid, selective and sensitive detection of the drug was possible with a LOD of 0.04 µM and the CDs could sense the drug with good recoveries. Table 3 displays biowaste materials used as C sources for the synthesis of CDs that could be successfully utilized as chemo- and biosensors and also as an alternative over disposing them.

#### Summary and critical analysis

Currently there is growing interest for one-pot synthesis of CDs, mainly to reduce wastage and by-products. Several sources including food items, plant biomass, biowaste and animal products have been explored as natural C sources as raw materials for the fabrication of CDs. Unlike synthetic precursors, eco-friendly raw materials are abundant in C and N sources in the form of proteins and carbohydrates and also serve as self-passivating agents for the fabrication of surface-functionalized CDs. In case of green sources, there is no need for external dopant as they are the reservoirs of organic molecules. The use of green sources has several advantages due to easy availability, minimum cost, comparatively clean reactions and non-toxicity. Generally, plant parts including flowers, fruits, seeds and stems containing numerous acidic, basic and neutral bioactive constituent molecules are fascinating as potent and sustainable biosources for the synthesis of carbonaceous nanodots in aqueous media. Moreover, different phytoconstituents present in a particular raw material not only play a vital role in the reaction kinetics, but also determine the surface functional groups on the CDs, and hence their reactivity. Besides, the QY of CDs depends on the type of phytoconstituents, particle size, solvent and dopants. Animal-derived homogenous CDs without much loss in fluorescence over subsequent storage have been prepared by subjecting precursors to hydrothermal/MW techniques. Besides, biowastes that have varying composition of cellulose, hemicellulose, lignin and other biomolecules were also well-utilized as precursors for the fabrication of CDs.

Hydrothermal treatment that involves dehydration, polymerization and carbonization of the carbohydrates is



Fig. 8 Schematic illustration of plausible mechanisms of fluorescence quenching in CDs in the vicinity of inorganic and organic analytes

attractive to develop CDs when the precursor material is a single natural renewable C source. In most studies reviewed above, mild and green hydrothermal method was the preferred synthesis route because it is simple, rapid and costeffective with low energy consumption and controllable reaction conditions that neither uses any strong acid nor needs post-synthesis surface passivation. Solvothermal, MW and solid-state synthesis of CDs from bioprecursors are seldom performed. Carbonization that employs higher temperatures was used in case of hard materials such as crustacean shells, while fruit and vegetable wastes were subjected to milder temperature conditions to facilitate the formation of CDs.

The sensing of heavy metal ions and certain organic molecules is of prime significance in environmental monitoring and clinical toxicology. Most of the bioprecursor-derived CDs displayed blue fluorescence, and very rarely even green emission under a UV source. These smart carbonaceous nanoparticles obtained from bioprecursors enabled label-free, simple, reliable, rapid, selective, sensitive and accurate detection of both inorganics and organic analytes, as their fluorescence is quenched by different mechanisms such as static and dynamic quenching, FRET, PET and IFE. The metal ion sensing principle is based on dynamic fluorescence quenching (determined by changes in the fluorescence lifetime before and after the action of the quencher) of CDs as presented in most of the reported literature. The CDs contain ample O and N atoms with lone pair electrons on their surface as a part of carbonyl, phenolic, hydroxyl, carboxylic and amine functional groups, which facilitates the formation of metal-CD coordination complex aggregates. However, the quencher (mainly inorganics) does not react with CDs. This results in transfer of excited state electron of CDs to the unfilled orbit of metal ions to form charge transfer complex for nonradiative electron/hole recombination and fluorescence quenching, thus making sensing feasible. The better the charge transfer complex, the greater will be the fluorescence quenching. Among all the transition metal ions, Fe<sup>3+</sup> and Cu<sup>2+</sup> are well known for stable metal-complex formation based on hard-soft-acid-base (HSAB) principle. Owing to the larger ionic radius of Hg<sup>2+</sup> and higher stability constant between the ion and carboxylic groups, the complex formation between Hg<sup>2+</sup> and CDs is energetically favourable (Huang et al. 2017). Besides,  $Hg^{2+}$  possess the considerable electron-withdrawing tendency with a higher reduction potential (Hg<sup>2+</sup> + 2e<sup>-</sup>  $\rightarrow$  Hg<sub>2</sub><sup>2+</sup>(+0.908 V)) that facilitates the non-radiative electron-hole recombination annihilation (Liu et al. 2012b).

In few investigations, static fluorescence quenching that occurs through the formation of a nonfluorescent ground-state complex via the reaction between CDs and the quencher is also reported. The design of an on-off type probe is convenient in this case as the substance with a stronger affinity can recover the fluorescence of CDs. Sensitive detection of Fe<sup>2+</sup>, Fe<sup>3+</sup>, Hg<sup>2+</sup> and Cu<sup>2+</sup> ions through maximum fluorescence quenching efficiency was perceived in the pH range 7–9, owing to the deprotonation of surface carboxylic groups of the CDs, which strengthens the CDmetal ion interaction. The quenching efficiency in acidic media was low due to the protonation of surface binding hydroxyl, carboxylic and amino groups. Moreover, at very high pH values, low quenching occurs due to complexation of metal ions to OH<sup>-</sup> instead of the CDs. Thus, neutral pH is optimal for the considerable exposure of functional groups for concomitant complex formation, subsequent fluorescence quenching and thereby efficient metal ion sensing. In addition to the metal ion detecting, the sensing of biomolecules and drugs using functional CDs derived from bioprecursors is also illustrated with real-life applications.

The analytes that were detected through PET quenching mechanisms of CDs include both inorganics, especially Pb<sup>2+</sup> and organics, particularly CEA and GSH. In the PET mechanism, the surface of CDs is usually rich in electron donor groups, and the quencher is an organic small molecule with an electron-withdrawing group on its surface. This mechanism may be extensively developed for organic small molecule detection. The analytes which were detected through FRET mechanisms of CDs also comprise of both inorganics and organics. Inorganics include S<sup>2-</sup> ions, Cu<sup>2+</sup>, etc., whereas organics involve GSH, tetracycline, glyphosate etc. FRET-based CD probe can recognize immense changes in Stokes shift and can provide a simple and effective method for visual detection of analytes. The process of confirming PET and FRET mechanism is complex as the values of molecular orbital energy levels, energy transfer efficiency and Förster distance, etc. need to be calculated. The IFE mechanism of fluorescence quenching does not entail any modifications of CDs, and the verification of the mechanism is less costly and complicated. Inorganics such as Cr<sup>6+</sup> and S<sup>2-</sup> and organics such as ascorbic acid can be detected through IFE quenching mechanism of CDs. Different mechanisms identified for the sensing mechanism of CDs based on fluorescence quenching by different analytes (Zu et al. 2017) are pictorially depicted in Fig. 8.

# **Challenges and future prospects**

A vast variety of CDs could be obtained by using a variety of bioprecursors and doping agents. But the key challenge remains to fabricate CDs with high QYs in ample amounts by using simple one-step methodologies. Most of the CDs emit blue fluorescence upon UV irradiation. However, blue fluorescence is generally less attractive for optical nanoprobes in biological applications due to autofluorescence. CDs that emit green luminescence are an attractive alternative, but there are only limited reports on them, and further research can be oriented towards green emitting CDs from bioprecursors for sensing application. Yet another area that requires attention is to develop CDs that emit in the deep-red or near-infrared region with good QY, where new synthetic strategies are still under intensive development.

Though it is well known that doping can improve the QY considerably, and natural sources are abundant with N, P and S, it is tedious task and requires extensive study to identify the amount of each constituents in a bioprecursor. Excellent auxochromic groups, especially N-containing groups, contribute to the high QY of the CDs, which might be caused by the doped nitrogen introducing a new energy level into the electronic structure of the N-CDs. Unfortunately, the N-CDs synthesized from some of these abundantly available and inexpensive precursors did not achieve high QY values, which demand better features for practical application. Therefore, there is still scope to search for low-cost precursors that can serve as both C and N sources and to develop a green, simple and rapid synthesis of highly photoluminescent N-CDs on a large scale. In comparison with CDs and N-CDs, N&S-CDs possess more abundant N- and S-containing groups, and these wide varieties of surface functional groups may bring the fluorescent materials novel performance and more extensive application prospect.

The mechanism of formation of CDs is still not clearly understood though several attempts are made to provide satisfactory explanation. Several factors like operating temperature, solvents employed, concentration of precursors, pH, etc. determine the yield, homogeneity and photoluminescent property of CDs. Every unique precursor with different composition of biomolecules has many parallel reactions occurring, which influences the formation of CDs. Hence, identifying the contributing parameters that decide the characteristic properties of these highly promising carbonaceous nanodots is a daunting task. Although CDs are considered as potential candidates to substitute quantum dots, few limitations such as their incapability to emit strong longwavelength fluorescence and dissolve in organic solvents besides water should be overcome. Furthermore, the reason for photoluminescence in CDs is still highly debatable, yet the presence of surface defects, particle size, different functional groups contribute highly to fluorescence properties.

Though nanotechnology has extended its reach in every avenue, it suffers from ease of reproducibility and largescale production. The use of sophisticated laboratory setup and expensive chemical compounds limits the economic use of nanoparticles; hence, the use of natural resources offers cost reduction and an alternative to bulk production. Future work could involve use of easily available natural resources to prepare CDs with excellent fluorescent properties and desired photostability. Therefore, more facile synthetic methods and green sources to obtain high photoluminescent CDs on a large scale are still urgently desired. Through this comprehensive review, we try to provide insights on various eco-friendly precursors for fabrication of CDs, their optical features and potential use as sensors. We anticipate that this appraisal would serve as a stepping stone for further research in this domain to develop intelligent and smart sensing technology.

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#### Declarations

**Conflict of interest** The authors declare no financial or non-financial competing interests.

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