

# Synthesis and Applications of Organic-Based Fluorescent Carbon Dots: Technical Review

*Musa Yahaya Pudza and Zurina Z. Abidin*

## Abstract

New ways of synthesizing organic-based fluorescent carbon dots (CDs) are required in environmental application. This is crucial for mitigation and control of pollutants without increasing the risk of releasing byproduct pollutants as the case with non-organic (metallic) quantum based substrate. Notably, this study provides current research on sustainable synthesis of CDs and their applications through analytical concept of recent and advance techniques for efficient and optimized processes. New scrutinized methods of synthesis and applications of CDs are beneficial and essential to optimize the state-of-art practices. The value distilled in this study adds to the field of sustainable production and application of CDs.

**Keywords:** carbon dots, fluorescent, organic-based, optimized, sustainable

## 1. Introduction

### 1.1 Origin of carbon dots

Carbon dots (CDs) are nanoparticles generated from organic/inorganic sources, was first discovered in 2004 when single-wall carbon nanotubes were electrophoretically purified [1]. CDs can be classified as carbon nanomaterials that are less than 10 nm in size, they are the latest class of fluorescent nanoparticles [2]. CDs have attracted the interest of researchers in diverse fields of science and technology such as; optoelectronics [3], environmental pollution and remediation [4], biosensor [5], bio-imaging and biomedical applications [6, 7].

CDs possess properties such as being dimensionless, durable, large surface area, enhanced porosity and stability, ease of being functionalized, fluorescence emission, biocompatibility and low toxicity [5, 8]. These properties of CDs can be applied to improve the environment and human health [4, 9, 10].

A toxic rival to the CDs is the popular semiconductor nanocrystals popularly known as quantum dots (QDs). The QDs are a type of semiconductor nanoparticles with diameter range from 1 to 10 nm [11]. More so, QDs normally are made from semiconducting materials, especially iron and cadmium, which are highly toxic and expensive to acquire [12]. Compared to QDs, CDs are considered best option with a high degree of biocompatibility, cost-effectiveness and non-toxic. It also serves as a suitable substitute to QDs in numerous areas of research such as bio-imaging, bio-sensing, pharmaceutical and fuel cells [13, 14].

Carbon dots (CDs) are suitable for the modification of electrode sensors. It combines fundamental aspects of biology, chemistry, and physical sciences, computer science and electrical engineering to meet various needs in a wide application field. Therefore, carbon as a sensor portrays several meanings, conditional upon what field the user subscribes [15, 16].

Over the years, various bulk materials and several processes and techniques have been developed and adopted by a wide range of researchers in the synthesis of CDs. These processes include the hydrothermal and microwave-assisted routes, heating, biogenic synthesis, thermal oxidation, ultra-sonification, subcritical water process (use of oil bath and salt bath), refluxing and chemical oxidation [17–26].

Three important factors must be considered in synthesizing CDs which are control of size, uniformity of CDs in solvents, and mitigated aggregation [27]. Wang and Hu [28] confirmed that CDs carbonaceous aggregation tends to form during carbonization but this can be prevented when synthesized by methods such as electrochemical synthesis, hydrothermal, or by pyrolysis method.

The application of biological and agro-waste to synthesize CDs have been advocated in numerous research such as; cooking oil waste [29], egg-white and egg-yolk [30], orange juice [6] as well as eggshells [31]. Though it is advantageous to use waste biomaterials in the synthesis of CDs to avoid competition with essential food production [32], however, the downside of the application of biomass in the synthesis of CDs is lacking of essential purity and structural homogeneity to obtain homogenous fluorescent CDs for purposes of sensing minute concentrations of analytes [21, 33]. These had caused the application of clean materials to be used in the synthesis of homogeneous fluorescent CDs [2].

A competent carbon source for soluble CDs synthesis is needed to comply with the goals of green chemistry and not be in direct competition with essential food production and should be cheap to synthesize [34–36]. Research in the synthesis of CDs must consider low price of additives and less purification steps in case of using biomass as a precursor material.

Thus, the emphasis is necessary on the cost of producing typical CDs, not to be a replica of the currently observed situation with semiconductor QDs, with the high cost and potential environmental negative impact and yet to achieve its full potential in commercial applications [37–39].

## **2. Green and sustainable carbon dots**

Carbon dots (CDs) have emerged to be attractive materials due to their excellent photoluminescence (PL) properties and wide surface areas, which are needed for sensitive and selective sensing of analytes [40]. These qualities are owed to the characteristics of the carbon element at nano-dimension and five valence electrons to bind carbon atoms [31, 41]. The green and sustainable carbons dots refer to CDs that are synthesized from agro and biomaterials that can be readily available without depleting their sources [42].

CDs can be obtained from various source [3]. These sources include plants and animal origins such as bamboo leaves, woods, green algae, sugar cane, mangosteen, carica papaya, saffron, gringko, neem gum, prawn shells, orange, cucumber and pineapple [32, 43–45]. Further interesting applications of CDs have been reported in diverse sectors of the environment and health fields of science and technology [3, 32].

For instance, Pattanayak and Nayak, in 2013 [43] presented an eco-friendly synthesis of iron nanoparticles from various plants and spices extract. The synthesis of nanoparticles from plant parts (leaf) is essential since this will not require expensive processes that are involved mostly in biomaterial processing. Iravani et al., [46]

demonstrated a green synthesis of metal nanoparticle using plants (*emblica officinalis* fruit extract) as a mean of mitigating the synthesis process of metal nanoparticles that are efficient and able to enhance green chemistry procedure for nanoparticles synthesis.

Liu [44], reported a research work on one-step green synthesized fluorescent carbon nano-dots from bamboo leaves for copper (II) ion detection and demonstrated the exploration of bamboo leaves as a carbon source. Carbon nano-dots were synthesized hydrothermally and a resultant high quantum yield quantum dots, with sensitive  $\text{Cu}^{2+}$  detection at a limit of detection as low as 115 nM on a dynamic range from 0.333 to 66.6  $\mu\text{M}$ . The zeta potential of the pristine carbon quantum dots was measured at  $-4.78$  mV which changes to  $+13.8$  mV after treatment with positively charged polyethyleneimine (a water-soluble cationic polymer).

Wembo et al., [47], researched on the economical and green synthesis of fluorescent carbon nanoparticles and their use as probes for sensitive and selective detection of mercury (II) ions. The adopted process by Wembo and colleagues was based upon the economy and green preparative strategy toward water-soluble fluorescent carbon nanoparticles with a quantum yield of 6.9% by a hydrothermal process using a low-cost waste from pomelo peel as a carbon source.

Piyushi et al., [45] cultivated *chlorella* (a genus of single-cell green algae belonging to the phylum *Chlorophyta*) on brewery wastewater for nanoparticle biosynthesis. The method of bio-nanoparticle synthesis using *chlorella* algal biomass grown in single water sample were harvested from the culture medium by centrifugation at 4000 rpm for 5 min followed by washing with ultrapure water to eliminate impurities. Iron nanoparticles were synthesized by mixing 0.5 g (dry weight) *Chlorella* sp. MM3 with 5 mL of 0.1 M  $\text{FeCl}_3$  solution followed by incubation at 37 C for 48 h which entails long and tedious process.

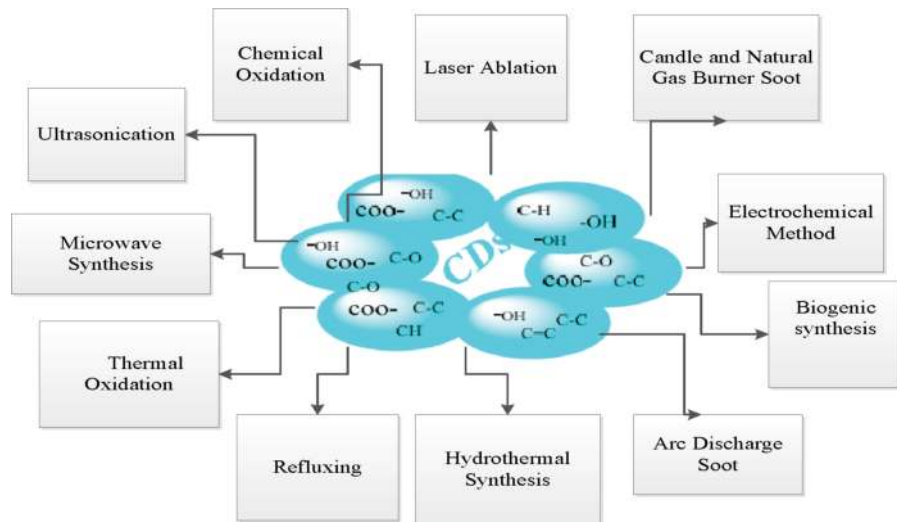
Till et al., [48] synthesized CDs by microwave-assisted hydrothermal treatment of starch and Tris-acetate-EDTA. The process confirmed that nitrogen-doped CDs have emerged to be complementary to starch-derived CDs. Addition of nitrogen to CDs improved the yield of photoluminescence from 19% to 28%, making them promising luminescent materials for improving fluorescence of CDs. However, there is no added value in incurring additional chemicals during synthesis process of CDs. Starch is a better alternative to the use of nitrogen for synthesizing CDs. Till and colleagues observed the effect of nitrogen (N) additives, through the use of ethylenediaminetetraacetic acid (EDTA); tris (hydroxymethyl) aminomethane (Tris) and a combination of both (TAE-buffer) on the photophysical properties of CDs. Temperature (45 min at 230°C) plays an important role in the improved nitrogen-doped carbon structures [48].

Some researchers have adopted nitrogen for fluorescence and photoluminescence enhancement, but this approach has shown indistinct composition which required extensive purification steps. This, however, is environmentally not suitable and contravenes the concept of green chemistry since it involves many chemicals in the synthesis process [49].

## 2.1 Methods of carbon dots (CDs) synthesis

Synthesis methods of CDs can be divided into two major parts; top-down and bottom-up as in **Figure 1**. Top-down starts from cutting the carbon materials into carbon particles or cleavage of larger carbonaceous materials such as carbon nanotube by laser ablation, arc discharge, electrochemical and candle/natural gas burner soot, and recently the hydrothermal route.

The bottom-up route involves the use of molecules as support for localizing the growth of CDs by blocking aggregation during high-temperature treatment.



**Figure 1.**  
Synthesis methods of carbon dots (CDs).

However, this study explores the top-down process of CDs synthesis. As earlier stated the top-down approach concentrates on precursor carbonization that include microwave-assisted method, chemical oxidation, heating, and hydrothermal process [50].

### 2.1.1 Laser ablation

It is a process removal of material from solid or liquid by irradiating it with a laser beam [51–54]. Material evaporates or sublimates when the laser flux is low and converted to plasma at high laser flux. Goncalves and colleagues [51] reported the synthesis of CDs from carbon targets immersed in deionized water by direct laser ablation (UV pulsed laser irradiation). CDs were optimized and synthesized after being functionalized with  $\text{NH}_2$ - polyethylene-glycol (PEG200) and N-acetyl-l-cysteine (NAC). To produce particles in tens of nanometer range by laser ablation, the energy is controlled within the incidence area of the precursor [54, 55].

Yu et al., [53] demonstrated the possibilities of relying on irradiating a toluene sample with a non-focused pulsed laser that is very different from the high powered laser irradiation employed in conventional ablation. This process by Yu and colleagues revealed an induced transformation of toluene into graphene sheaths, which subsequently produced fluorescent CDs. These nanoparticles can simply be functionalized using more than one molecule and stayed stable in an aqueous solution. It can also be applied to optical fiber devices through immobilization due to its stability in a specific optical nano-analytical sensor [56]. However, the equipment to conduct laser ablation is quite expensive and it needs technically skilled personnel to operate.

### 2.1.2 Arc discharge soot

CDs were first discovered through this method accidentally when the separation of single-walled carbon nanotubes (SWNTs) were made using gel electrophoresis from carbon soot by arc discharge method. Carbon is formed when direct current arc voltage is applied in an inert gas across two graphite electrodes. The biggest challenge of this method is that it generates impurities that are difficult to purify [1].

### 2.1.3 Electrochemical method

Electrochemistry is another top-down approach used in synthesizing CDs, this process is facile and the product yield is normally high [57]. CDs with a size of 6 to 8 nm and 2.8% to 52% can be obtained through exfoliation that utilizes graphite rods and Pt wire in ionic liquid or water solution.

The mechanism of the exfoliation was due to complex interplay of anodic oxidative cleavage of water and anionic intercalation from the ionic liquid using titanium cathode and spectrum pure graphite in the center of electrolyzer to yield pure blue fluorescent CDs without the urgency of complex purification [28, 58–62].

### 2.1.4 Candle and natural gas burner soot

Application of carbon soot in the synthesis of CDs have been reported by Tian et al., [63], the carbon source was from a carbon-processing reaction. Due to the simplicity to obtain the starting material, this method has been used widely by researchers. It also provides a new use for a complicated by-product. At the same time, it possesses disadvantages such as uncontrolled chemical surface, production of many byproducts that can harm human health with a broad dispersion [64].

### 2.1.5 Microwave synthesis

Wang et al., [13] prepared CDs by microwave method. It proved reaction time can be shortened to 30–45 minutes with microwave-assisted technique. Similarly, Choi et al., [10] made effective use of lysine as a precursor to synthesize CDs within 5 minutes in a home type of microwave oven and the CDs were soluble in water with deep blue photoluminescence at a high mass yield of 23.3%.

Compared with other methods, the microwave route is more convenient since the heating of the carbon precursor is rapidly achieved within few minutes. It also exhibits high quantum yield and provides a long fluorescence lifetime. The procedure of microwave synthesis is much easier compared to others as it only utilizes heating via irradiation technique [65].

### 2.1.6 Chemical oxidation

This method is mostly applied to produce CDs on an industrial scale. CDs can be obtained through oxidation treatment of carbon precursors by a strong oxidant. CDs from natural products have been researched and developed, by the synthesis of large scale CDs from human hair, coffee, and biomass by adding it into concentrated sulfuric acid and then heating at different temperatures. The time range is from hours to days [66]. By varying the temperature of synthesizing CDs, the quality of CDs such as diameter and quantum yields can be controlled [67–69].

### 2.1.7 Hydrothermal synthesis

The hydrothermal route of synthesizing CDs is considered as environmental-friendly, low cost and involves few synthesis steps that are non-toxic [2, 70–74]. Musa et al explored the hydrothermal method at a temperature range between 75°C to 175°C where the researchers reacted the precursor in a sealed hydrothermal reactor that resulted into a high yield photoluminescent quantum yield at 34.9% [2].

As illustrated in **Figure 2**, tapioca was added to an aldehyde solvent (acetone + sodium hydroxide) to improve the mobility of glucose molecules in starch [2]. The mixture underwent stages of reactions such as hydrolysis, adsorption, and



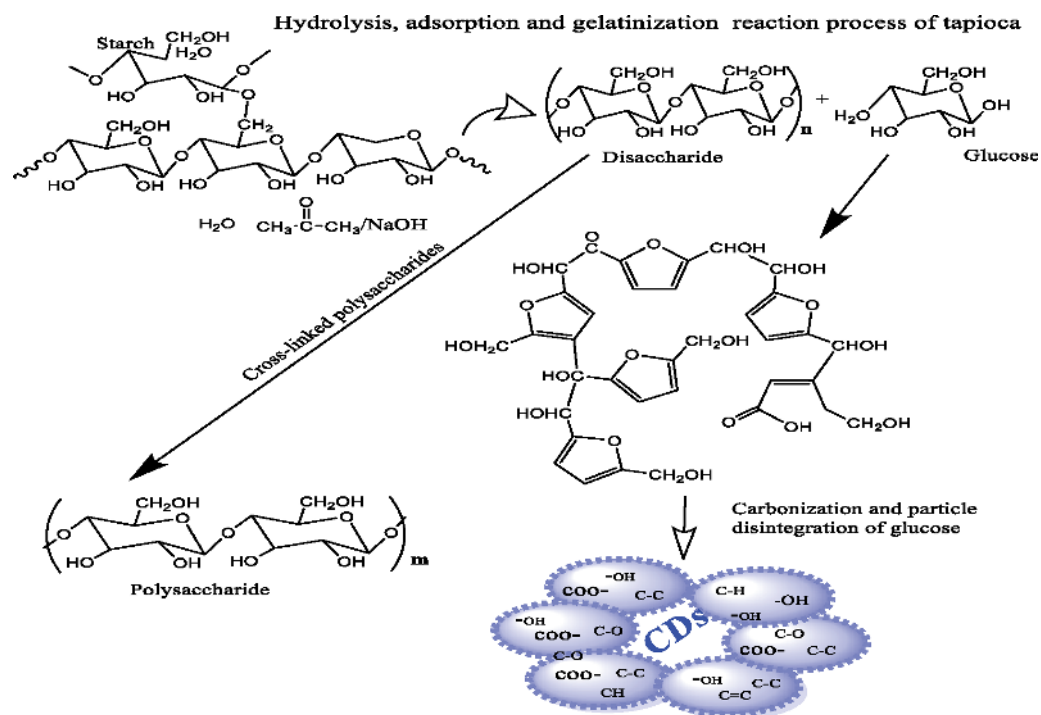
gelatinization to particle disintegration simultaneously [2]. The carbonization temperature breaks the bond between the starch, making it available for the reactive solvent which leads to hydrolysis to form disaccharide and gelatinized glucose. The disaccharides polymerized into polysaccharides and the gelatinized glucose yielded CDs for functional group characterizations [9].

Like all-natural products, starch undergoes seasonal changes and particularly the amylose/amylopectin ratio is influenced by plant species and area of plant cultivation, which could influence the CDs formation. Independently of seasonal changes and origin, a starch will provide CDs with highly reproducible photoluminescent properties [2].

Substances such as glucose, citric acid, banana juice, and protein are examples of many precursors used to prepare CDs by adopting the hydrothermal route of synthesis [75]. Success has been reported in the synthesis of CDs through one-step hydrothermal carbonization using chitosan applied directly as a bioimaging agent [76]. The hydrothermal method is promising in producing CDs and is suitable for industrial or large scale production [75]. However, it is notable in 2010 where Zhang et al. [77] first reported a one-pot hydrothermal method to synthesize CDs from ascorbic acid in the presence of ethanol as solvent. Quantum yield and average particle sizes of their synthesized CDs were 6.79% and ~2 nm, respectively [77].

Several methods of synthesizing CDs has been explored in this section, to prevent the use of expensive precursor and energetic systems like in laser ablation. The hydrothermal synthesis route is being recommended as foremost for the sake of ecological sustainability [25]. Chemical oxidation and exfoliations provide an inexpensive alternative although it employs large amounts of strong acid which is hazardous and undesirable [77].

The other methods of synthesizing CDs need multi-step experimental operations and some of them require post-treatments to improve their water solubility, stability, and luminescent. Besides, several other methods suffer from drawbacks



**Figure 2.**  
Mechanism for synthesis of carbon dots [2].

such as they require complex process and high temperature, time-consuming, harsh synthetic materials, and are expensive. This causes their applicability to be limited [78]. Several research successes proved that hydrothermal route to be a green method for the synthesis of CDs since the procedure produce soluble fluorescence CDs at reduce time and cost [2, 46, 79].

CDs derived from organic sources are excellent for the researcher and environment. Because the adoption of such material presents the choice to eliminate the need for metallic quantum dots, and any doping requirements, either through the use of sulfur (S) or nitrogen (N) agents. The use of metallic quantum dots and possible inclusion of S and N in enhancing their functionality contravenes the purpose of sustainable applications of nanomaterials in the modern field of nanotechnology [48].

**Table 1** is a list of different synthesis techniques that have been attractive to researchers in recent years. The table provides a list of interesting techniques such as hydrothermal, microwave assisted, biogenic synthesis, thermal oxidation, ultrasonication, refluxing and chemical oxidation with excellent particle sizes [80–91]. The hydrothermal synthesis of CDs proves to be efficient and effective since it provides relatively smaller sizes of the nanoparticles as synthesized by Du et al., [81] at 1.8 nm, when compared to other methods such as chemical oxidation by Thambiraj and Shankaran [85] at 4.1 nm, biogenic synthesis by Phadke et al., [20] at 5–8 nm, and refluxing by Himaja et al., [84] at ~50 nm.

## 2.2 Properties and characterization of fluorescent carbon dots (CDs)

One of the CDs properties is that it shows strong optical absorption in the UV region (200–800 nm) with a tail extending to the visible range, see **Figure 3**. CDs possess low toxicity with excellent photostability as compared to semi-conductor quantum dots [7, 23, 50, 71].

Absorption shoulders in the spectrum are due to the  $\pi$ - $\pi^*$  (pi to pi star transition) of C=C bonds or  $n$ - $\pi^*$  (n to pi star transition) of C=O and other fringe functional elements present [69, 92].

Method	Size (nm)	Reference
Microwave-assisted	2.7	[7]
Microwave	5–10	[10]
Biogenic synthesis	5.0–8.0	[20]
Thermal oxidation	5.0–10	[22]
Heating	3.0	[78]
Hydrothermal	2.3	[80]
Hydrothermal	1.8	[81]
Ultrasonication	5.0	[82]
“Oil bath”	2.59	[83]
Refluxing	~50	[84]
Chemical-oxidation	4.1	[85]
Chemical oxidation	2.5	[86]

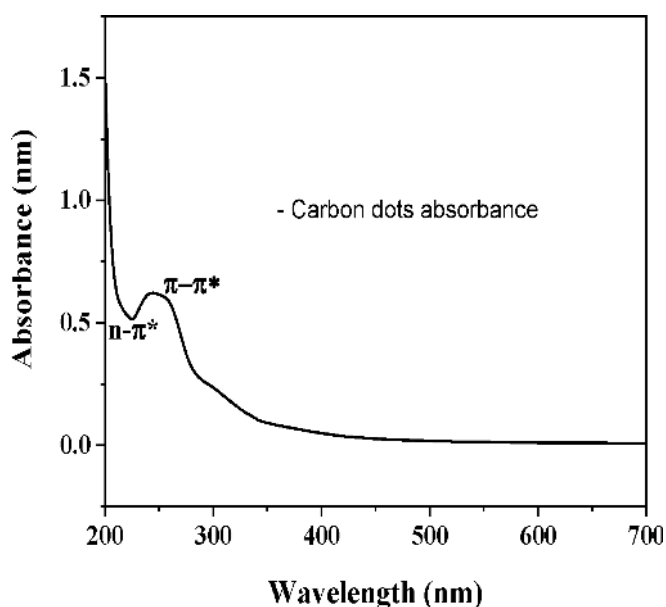
Note: n/a = not available.

**Table 1.**  
Carbon dots sizes and synthesis techniques.

The uniqueness of CDs is the availability of wide surface area for trace detection of analytes and provision of adsorptive sites through the availability of heteroatomic carbon in nano-dimension along with photoluminescence emission. Based on past study, CDs is dependent on intensity and wavelength emission towards its excitation wavelength [93]. This is due to the different sizes of particles and surface chemistry and/or different emissive traps on CDs' surface. The wavelength dependence behaviour makes CDs possible to be applied in multi-colour imaging and adsorptive purposes. Vinci et al., [93] suggest that CDs' core, surface states, and size are responsible for their emission and adsorptive properties [93].

**Table 2** shows the excitation wavelengths of CDs through the UV-lamp excitation process to obtain fluorescent characteristics [2].

The colour of CDs most of the time is related to the surface groups which corresponds to particle sizes [93]. Normally CDs show strong photoluminescence from blue to green wavelength. To enhance the quantum yield (QY) of CDs or change photoluminescence (PL) emission to meet desired applications, surface passivation and functionality play a vital role. Besides, CDs show great photostability as there are no reductions in PL intensity with continuous exposure to excitation. In terms



**Figure 3.**  
*Optical properties of carbon dots at UV-visible absorption and emission spectra.*

Colour	Interval of wavelength (nm)
Red	700–635
Orange	635–590
Yellow	590–560
Green	560–520
Cyan	520–490
Blue	490–450
Violet	450–400

**Table 2.**  
*The color range of visible light spectrum.*



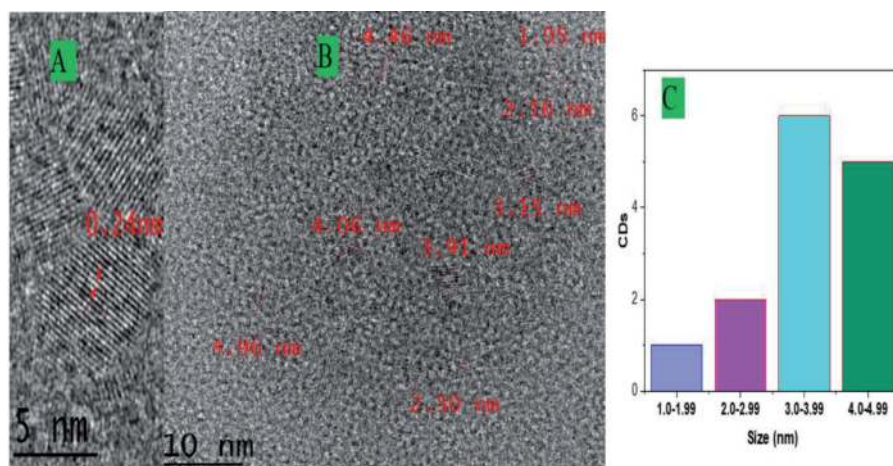
of chemical properties, different synthesis methods of CDs lead to different chemical structure and abundance of surface sites. They are usually connected or modified by polymer chains, oxygen-based, amino based groups, and others [93].

Characterization of CDs by high resolution transmission electron microscopy (HRTEM), X-ray photoelectron spectroscopy (XPS), Fourier transform infrared (FTIR), Atomic force microscopy (AFM) and Zeta Potential provide deep insights into the attributes of CDs such as hybridization and coefficient between functional groups and carbon core that take parts in the provision of the abundance of surface sites and the photoluminescence behaviour [94]. In comparison to graphene and metallic quantum dots, the CDs serves as the way out of toxicity concerns in environmental monitoring and medical applications [95].

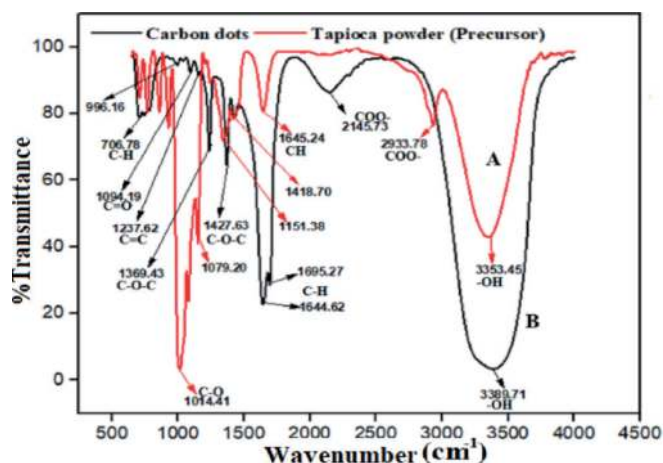
### 2.2.1 High-resolution transmission electron microscopy (HRTEM)

The sizes and texture of CDs are important for fundamental applications in the field of environmental science and nanotechnology. **Figure 4(A–C)**, shows the HRTEM images of CDs at different resolutions between 1 nm to 10 nm. Synthesized CDs revealed amorphous quasi-spherical morphology with a lattice spacing of ca 0.24 nm (**Figure 4A**), CDs characteristics are suitable absorbent of pollutants that are larger than 0.24 nm [2, 71, 89].

High-Resolution Transmission Electron Microscopic images of the CDs characterized in magnifications of 5 nm and 10 nm (**Figure 4A and B** respectively). **Figure 4A** is the lattice spacing for carbon dots at 5 nm magnification. While **Figure 4B** is the size distribution within 10 nm magnification. **Figure 4C** is the histogram chart, demonstrating the nanoparticle sizes of CDs. The synthesis of nanoparticle with low lattice space is needed for research applications of CDs in environmental chemistry, pollutant entrapment in aqueous media and water purification [74]. The interplanar distance (lattice spacing) of 0.24 nm (**Figure 4**) is lower than the lattice spacing planes of graphitic materials (0.34 nm), the larger interlayer spacing could be attributed to the abundant oxygen-containing groups. In other words, the oxygen-containing groups could expand the layer spacing. The synthesized CDs is in consonance with recent reports by Arumugam and colleagues, CDs was hydrothermally synthesized from broccoli [79], ginkgo fruits [87], and cabbage [8].



**Figure 4.** High resolution transmission electron microscopic (HRTEM) (A) Lattice space of carbon dots (CDs) characterized in magnifications of 5 nm. (B) Images of CDs at 10 nm. (C) Size distribution of CDs within 10 nm magnification [2].



**Figure 5.**  
FT-IR spectrum of the carbon dots and tapioca [2].

### 2.2.2 Fourier-transform infrared spectroscopy (FTIR)

Fourier-transform Infrared spectroscopy (FTIR) portrays the functional structure of CDs. It reveals the intrinsic functional groups and other useful compounds present in CDs. **Figure 5** provides functional groups that exist before and after the hydrothermal treatment of tapioca as a precursor for CDs.

As shown in **Figure 5(A)** representing tapioca. Peaks associated with the stretching vibrations of hydroxyl ( $\text{-OH}$ ) and carboxylic ( $\text{COO-}$ ) groups are at  $3353.45$  and  $2933.78 \text{ cm}^{-1}$  [75]. Further stretching vibration of  $\text{C-H}$  occurred from  $1645.24$  to  $1341.82$ . The peaks at  $1151.38$ ,  $1079.20$ ,  $1014.41 \text{ cm}^{-1}$  can be due to the  $\text{C-O}$  stretching vibrations and out-of-plane bending modes of  $\text{sp}^2$  and  $\text{sp}^3$   $\text{-CH}$  group [75].

There were substantial changes observed in the spectra of CDs (**Figure 5B**). The hydroxyl ( $\text{-OH}$ ) group of  $3389.71 \text{ cm}^{-1}$  increased on the carbon structure as a result of hydrolysis. While the carboxylic ( $\text{COO-}$ ) group  $2145.73 \text{ cm}^{-1}$  reduced by thermal destruction of saccharides structure [34]. The peaks at  $1695.27 \text{ cm}^{-1}$  and  $1644.62 \text{ cm}^{-1}$  showed the increase in the  $\text{C-H}$  stretching vibrations of the bending modes of the  $\text{sp}^2$  and  $\text{sp}^3$   $\text{-CH}$  group. The peaks around  $1427.63 \text{ cm}^{-1}$  until  $1369.43 \text{ cm}^{-1}$  are due to  $\text{C-O-C}$  [34]. The peak at  $1237.62 \text{ cm}^{-1}$  corresponds to the  $\text{C=C}$  stretching vibration while  $1094.19 \text{ cm}^{-1}$  and  $996.19 \text{ cm}^{-1}$  represents the  $\text{C=O}$  stretching vibration and the last group at  $706.78 \text{ cm}^{-1}$  denotes the  $\text{C=C}$  bond of the unsaturated glucose structure in the starch. These attributes were responsible for the water-soluble nature of CDs [34]. The FTIR graph shows the formation of unsaturated carbon. Along with oxygen-rich groups such as hydroxyl, carboxyl, and carbonyl situated on the CDs surface, which agree with the hydrothermal synthesized CDs from the organic origin [23, 25, 26, 81, 90].

## 3. Applications of carbon nanoparticles

There are numerous applications of carbon nanoparticles due to the abundant properties they possess [3]. These applications are being discussed in the subsequent sections of the report.

### 3.1 Application in bioimaging and biomedical

Carbon dots have shown great potential to act as a sensor and can be used for environmental monitoring and control of pollutants, more so in the medical field

for biosensor applications. It can donate or accept electrons that make it suitable for detection of ions, vitamins, nucleic acid, protein, enzyme and biological pH value [7, 11, 96–98]. Even though different materials are used to detect specific ions, the detection mechanisms are identical [99].

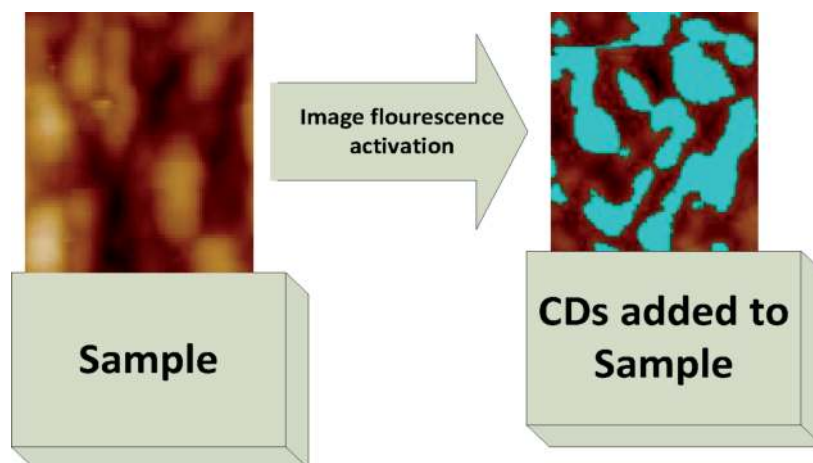
The functional groups on the surface of CDs specify distinctive affinities to different target ions, through an electron or energy transfer process and high selectivity to other ions [100]. CDs have been involved in the detection of 2,4,6-trinitrotoluene (TNT) and also applied as a dual-sensing platform for fluorescent and electrochemical detection of TNT [101]. Other reports utilized CDs as pH sensors for in-vitro and in-vivo investigations [102].

Research showed CDs able to detect intracellular pH inside a living pathogenic fungal cell and has been developed to sense nucleic acid in the DNA [103]. In other cases, CDs have been used in bioimaging because of their low toxicity and excellent photostability compared to semi-conductor quantum dots that posed health problems and environmental concerns [8]. Its visible excitation, emission wavelengths, and high brightness confirm CDs as a suitable candidate in this area. Several studies have been conducted using CDs in cell imaging, including pig kidney cell line [104], *Escherichia coli* [105], HeLa Cells [106], liver diseases [95], see **Figure 6**.

Chengkun et al., [98] discovered photoluminescence in CDs synthesized from Nescafe original instant coffee and applied it in the field of bioimaging. From their investigation, CDs from Nescafe are found to be amorphous and the cytotoxicity study revealed that the CDs did not cause any toxicity to human hepatocellular carcinoma cells at a concentration as high as 20 mg/ml. Yang et al., [107] also worked on novel green synthesis of high-fluorescent CDs from honey for sensing and imaging. It was an innovative and green approach towards a CDs of high fluorescent quantum yield and excellent photostability, employed for HeLa cells imaging and coding. Rui-jun et al., [108] produced photoluminescent CDs from polyethylene glycol (PEG) for cellular imaging. The PEG is a biocompatible non-conjugated polymer, used as both carbon source and passivating agent [108].

### 3.2 Application in heavy metal ions detection

The application of CDs in the selective detection of heavy metals have been reported in several scientific and experimental research [34, 40, 79]. However, there are gaps and lapses needing redress, such applications are predominantly in



**Figure 6.**  
Graphical description of fluorescence images of carbon dots.

photoluminescent quenching of heavy metals. whereas, current section looks into reliable and robust CDs for applications in electrochemical sensing of multiple ranges of heavy metal ions.

The development of a convenient and sustainable technique for detecting and identifying human and environmentally toxic metal ions is of great interest. The following are reports concerning CDs application in heavy metal detection.

Zhang and Chen [109] worked on nitrogen-doped carbon quantum dots application as a turn-off fluorescent probe for the detection of  $\text{Hg}^{2+}$  ions at a detection limit of  $0.23 \mu\text{M}$ . The fluorescent quenching mechanism is attributed to the surface-state triggered by the mercury-induced conversion of special functional group ( $-\text{CONH}-$ ) from spirolactam structure to an opened-ring amide [109].

Sandhya et al., [110] applied nanostructures for heavy metal ion sensing in water using surface plasmon resonance of metallic nanostructures. They reviewed on techniques to improve selectivity and sensitivity of surface plasmon response sensors with attention to homogeneity. Effects of particle size, shape, material type, and surrounding environment were found to be effectual in the surface plasmon surface frequency.

Similarly, Qu et al., [111] developed CDs to detect  $\text{Fe}^{3+}$  ions by using dopamine as a starting material with a detection limit of  $0.32 \mu\text{M}$ . Quenching of photoluminescence intensity occurred when there was an interaction between CDs and ions. Meanwhile, Liu [44] reported a research work on one-step green synthesized fluorescent carbon nanodots from bamboo leaves for copper (II) ion detection and demonstrated the exploration of bamboo leaves as a carbon source with high carbon constituent. Carbon quantum dots were synthesized hydrothermally with sensitive  $\text{Cu}^{2+}$  detection at limit of detection as low as  $115 \text{ nM}$  and a dynamic range from  $0.333$  to  $66.6 \mu\text{M}$ . The zeta potential of the pristine carbon quantum dots was measured at  $-4.78 \text{ mV}$  which improved to  $+13.8 \text{ mV}$  after treatment with positively charged polyethyleneimine (a water-soluble cationic polymer). More so, Rao et al., [112] reported on the ability of CDs generated from citrus acid anhydrous to detect heavy metal such as  $\text{Fe}^{3+}$ , with a detection limit of  $0.239 \mu\text{M}$ .

Methionine has been used as a material for the synthesis of CDs [113]. These CDs were co-doped with nitrogen and sulfur to enhance surface functionalization for the detection and environmental monitoring of heavy metal pollutants [113]. Similarly, Shen et al., [4] applied fresh pomelo in the synthesis of CDs co-doped with nitrogen and sulfur for the detection of chromium (Cr (VI)).

A fluorescent probe for selective detection of metal ions such as mercury ( $\text{Hg}^{2+}$ ,  $1.00 \times 10^{-8} - 1.50 \times 10^{-3} \text{ M}$ ,  $1.00 \times 10^{-7} \text{ M}$ ) with wide linear range and satisfactory detection limits was discovered when citric acid monohydrate was used for the synthesis of fluorescent CDs [114]. More essentially and effective is the burning of ash from waste paper and further utilized as a source of CDs by Lin et al., [115]. They succeeded in synthesizing CDs without any surface modification and subsequently, the fluorescent CDs were quenched by  $\text{Fe}^{3+}$ .

Simpson et al., [21] synthesized carbon nanoparticle from glycerol and phosphoric acid mixed in a Berghof high-pressure reactor at  $250^\circ\text{C}$  for 4 hours. Afterward, glassy carbon electrodes were fabricated by drop-casting the carbon nanoparticles, and further applied for heavy metal ( $\text{Cu}^{2+}$  and  $\text{Pb}^{2+}$ ) detection by square wave anodic stripping voltammetry [21]. Heavy metals such as  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Cr}^{3+}$ ,  $\text{Co}^{2+}$ ,  $\text{Ag}^+$ ,  $\text{Hg}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{Fe}^{2+}$ , and  $\text{Fe}^{3+}$  have been tested on CDs synthesized from carbon source of mangosteen pulp and a ground discovery was made. Among the listed heavy metals,  $\text{Fe}^{3+}$  was the favourite in detection with a detection limit of  $52 \text{ nM}$ . Further application was found for cell imaging, which reveals their diverse potential applications [89].



Abhishek et al., [14] made a paper strip based live cell ultrasensitive lead sensor using CDs synthesized from biological media. They reported a formulation of a sensor through microwave heating of potato-dextrose agar (PDA) for the detection of lead ( $Pb^{2+}$ ) in solution but again involved a long and laborious process.

Pajewska et al. [116] explored the fluorescence of synthesized CDs from citric acid with glutathione for the sensing of mercury ( $Hg^{2+}$ ) ion. A high recovery of  $Hg^{2+}$  was achieved at 115.1%. The method of synthesizing CDs with low toxicity is embedded in the green chemistry principles. Thus, it fulfills the criteria of being eco-friendly. **Table 2** provides a list of applications of CDs in the detection of heavy metals ions.

As seen in **Table 3**, the mechanism of action for the application of CDs largely depends on the analyte of concern. In the case of CDs from citric acid monohydrate for application in fluorescence quenching of  $Hg^{2+}$ , it relies on Förster resonance energy transfer (FRET) [114]. This is similar to CDs synthesized from biomass [117], polyacrylamide [118], lotus root [119], degreased cotton [120], gold nanoclusters [111], and Petroleum coke [127].

Fluorescent carbon nanoparticle sensing is largely dependent on changes or disturbances that are caused by an analyte that interacts with a fluorescent probe. This shift mostly will lead to a measurable change in the emission characteristics of the probe (emission wavelength, intensity, lifetime, or anisotropy), which can be directly linked to analytes (e.g heavy metal) concentration. More so, fluorescence probe strategies are based on quenching (turn-off) or enhancing (turn-on) emission, and surface-enhanced Raman scattering (SERS) techniques [33, 125].

Source of carbon nanoparticles	Sensing mechanism	Type of metal ions and linear range	Sensing (LOD)	Reference
Biomass	Fluorescence	$Hg^{2+}/Fe^{3+}$ $0.002 \text{ mol L}^{-1}$	10.3 and 60.9 nM	[117]
Polyacrylamide	Fluorescence	$Hg^{2+}$ $0.25\text{--}50 \text{ }\mu\text{M}$	13.48 nM	[118]
Lotus plant	Fluorescence	$Hg^{2+}$ $0.1 \text{ to } 60.0 \text{ }\mu\text{M}$	18.7 nM	[119]
Degrease cotton	Fluorescence	Cr(VI) $1.00\text{--}6.00 \text{ mmol/L}$	0.12 $\mu\text{g/mL}$	[120]
Gold nanoparticles	Luminescence	$Pb^{2+}$ $1 \times 10^{-5} \text{ M}$	n/a	[121]
Biomass from peanut shells	Fluorescence	$Cu^{+2}$ $0\text{--}5 \text{ mM}$	4.8 mM	[122]
Metal oxides	Electrochemical oxidation.	$Cu^{2+}$ $0.1 \text{ to } 1.3 \text{ }\mu\text{M}$	0.04 $\mu\text{M}$	[123]
Metal nitrates	Isotherm	$Cd^{2+}$ $10 \text{ mg/L}$	12.60 mg/g	[124]
Mushroom	Fluorescent	$Hg^{2+}$ $0 \text{ to } 100 \text{ nM}$	4.13 nM	[125]
Penaeus merguensis enzyme	Isotherm	$Cu^{+2}$ $1\text{--}5 \text{ mM}$	2 mM	[126]
Coke	Fluorescent	$Cu^{+2}$ $0.25\text{--}10 \text{ }\mu\text{M}$	0.0295 $\mu\text{M}$	[127]

**Table 3.**  
Carbon nanoparticles for heavy metal sensing.



Other notable techniques for the detection and quantification of heavy metal ions include, inductively coupled plasma mass spectrometry (ICP-MS). This instrumentation is efficient among several other methods, but it is expensive. It was developed since the 1980s [128–130], used mostly by multivariate analysis along with the ICP-MS technique to unravel heavy metal elements present samples. However, inductively coupled plasma atomic emission spectroscopy (ICP-AES) have also been used to identify heavy metal pollutants. But, the method is expensive and requires sophisticated instrumentations and a highly trained technician [131].

Nowadays, marine pollution is becoming a global phenomenon and seafood safety has played a crucial role in human health [129]. Fatema et al., [132] applied atomic absorption spectroscopy (AAS) to measure the absorbed quantity of  $Pb^{+2}$ ,  $Cd^{+2}$ , and  $Hg^{+3}$  in shrimps. Heavy metals have been detected by other means such as energy dispersive x-ray fluorescence (EDXRF), electrothermal atomic absorption method (ETAAS), and flame atomic absorption spectroscopy (FAAS) [133, 134]. But, all of the aforementioned techniques have disadvantages in the detection of heavy metals, such that they are expensive and require strenuous experimental steps [135]. Therefore, environmental researchers have continued to strive to develop a cheap, simple, sensitive, specific, accurate, user-friendly, and eco-friendly means of detection for heavy metal pollutants.

### 3.3 Application in non-metal detection

CDs are very useful in detecting non-metallic elements. Several types of research have been reported, CDs synthesized from potato are well able to detect phosphate [106]. Zhaoxia et al., [136] utilized CDs with tunable emission and controlled size for sensing hypochlorous acid. As a class of carbohydrate that is widely distributed in a living organism, sucrose was chosen as a carbon source with assistance of microwave irradiation. A strongly fluorescent CDs without post-passivation was produced. By increasing the concentration of phosphoric acid as fluorescence enhancer under UV lamp, various fluorescent emissions of CDs of variable sizes were obtained. It was found that green CDs have excellent sensitivity for the detection of hypochlorous acid.

Kuo et al., [85], experimented with percutaneous fiber-optic nanosensors for instant evaluation of chemotherapy efficacy for in-vivo strategy of assay design aimed at monitoring non-homogeneously distributed biomarkers. They identified optimal exogenous fluorophores for the cell distribution indicators that are independent of the treatment of the apoptotic initiator and without interfering with the optical characteristics of fluorophores.

Huilin et al., [137], investigated on CDs as a fluorescent probe for off-on detection of sodium dodecyl-benzenesulfonate (SDBS) in aqueous solution. The pristine CDs were synthesized from sodium citrate through a simple, convenient, and one-step hydrothermal method. Fluorescent recovery was achieved with the application of SDBS. Detection of SDBS in real water samples was proportional to the concentration in the range of 0.10 to 7.50  $\mu\text{g/mL}$ . Furthermore, fluorescence sensing probe has been used to detect kaempferol (flavonoid that is present in a variety of plants and plant-derived foods) using fluorescent CDs synthesized from chiefly acetic acid with a detection limit of 38.4 nM in the concentration range of 3.5–49  $\mu\text{M}$ . Finally, organophosphorus as pesticides have been detected through the use of CDs as a detector for pollutants without surface modification [115].

### 3.4 Application in adsorption studies

CDs and carbon structured nanoparticles have attracted researchers to explore their effectiveness and optimization ability in the fields of pollution research [3].

Adsorbent material	Adsorbate/analyte	Reference
Gold nanoparticles (AuNPs)	4-nitrophenol	[145]
Carbon dots (sodium citrate)	Mercury (II) ions.	[146]
Fluorescent carbon dots from o- phenylenediamine	Cell imaging and sensitive detection of Fe <sup>3+</sup> and H <sub>2</sub> O <sub>2</sub>	[18]
Silica gel	Aromatic volatile organic compounds (VOCs)	[147]
Graphene oxide	Nitrobenzene in sulfide	[148]
TiO <sub>2</sub> , SiO <sub>2</sub> , and ZnO nanoparticles	Neptunium (V)	[149]
Polystyrene latex nanoparticles	Alumina	[150]
Graphene oxide	Radionuclide removal	[151]
Polyaniline modified graphene oxide	Uranium(VI)	[152]
Carbon nanotubes	Mingle-ringed N- and S-heterocyclic aromatics	[153]
Graphene oxide	Minerals such as montmorillonite, kaolinite, and goethite, in aqueous phase	[154]

**Table 4.**  
*Nanostructured materials in adsorption processes.*

Because of their excellent properties; carbon material performs concurrently as adsorbent and a transducing-agent [138–141].

Due to abundant surface sites provided by CDs, it is a suitable candidate for studies in the detection and adsorption of heavy metals [142, 143]. For instance, Ghiloufi et al., [144] used gallium doped zinc oxide (ZnO) nanoparticle in the adsorption of heavy metals (Cd<sup>2+</sup> and Cr<sup>6+</sup>) in aqueous solution. The adsorption of heavy metals was analyzed through the effect of pH and it revealed favourable adsorption at a low pH level, less than pH-3 and temperature of 298 K [144].

**Table 4** provides harmonized presentation of nanomaterials applied for the purpose of absorbing environmental pollutants and contaminants in aqueous systems [145–154].

So far the concept of applying nanoparticles for environmental objectives have been successful. Meanwhile it is recommended that comparisons be made with bulk counterparts of the same substance to measure efficiency. On this note a study on the application of bulk agro material from *jatropha curcas* demonstrated efficiency in adsorption of pollutants and is recommended for comparison with its nano-dimension counterparts [155]. Similarly, a report on the application of sesame straw biochar in adsorption of heavy metal analyte concluded that further adsorption studies for nano-range agro-based materials are necessary for accurate estimation of adsorption in natural environments [156].

#### 4. Conclusion

A suitable carbon source for CDs synthesis should be soluble in water (green chemistry), accessible worldwide (i.e. geographical abundance) with defined and well-known properties (i.e. functional attributes), should not be in direct competition with essential food production (i.e. sustainable), and it should be cost-effective (i.e. cheaply accessible). While the price of additives or carbon source plays a minor role in fundamental research, it may play a major role when large quantities are considered.

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## **Author contributions**

M.Y.P., as the first author; made the study conception and design acquisition of reports and drafting of manuscript. Z.Z.A., contributed in the study conception and design, critical revision of major scientific ideas through clinical experience.

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## **Conflicts of interest**

The authors hereby declare that there is no conflict of interest.


## **Author details**

Musa Yahaya Pudza\* and Zurina Z. Abidin\*  
Department of Chemical and Environmental Engineering, Faculty of Engineering,  
Universiti Putra Malaysia, Serdang, Selangor, Malaysia

\*Address all correspondence to: pudzamusa@gmail.com and zurina@upm.edu.my

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