Supplementary Materials

Ultrasensitive detection of tetracycline using boron and nitrogen co-doped graphene quantum dots from natural carbon source as the paper-based nanosensing probe in difference matrices

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Table S1. Elemental analysis of passion fruit juice

| Produce              | Weight percentage of element (%) |
|----------------------|----------------------------------|
|                      | C      | O      | N    | H    | S    |
| Passion fruit juice  | 38.3   | 54.1   | 0.75 | 6.57 | 0.28 |
Table S2. Elemental weight percentage of elements in pure N-GQDs and B,N-GQDs estimated from survey scan of XPS.

| Materials    | Weight percentage of element (%) |
|--------------|----------------------------------|
|              | C  | O  | N  | B   |
| N-GQDs       | 60.1 | 33.4 | 6.5 | _   |
| B,N-GQDs     | 59.1 | 32.3 | 6.2 | 2.4 |

a: Not detected.
**Table S3.** Comparison of the quantum yield of 0-dimensional carbon-based nanomaterials and GQDs) synthesized from various natural products.

| Materials<sup>a</sup> | Precursors               | Reaction Conditions | Quantum yield (%) | Reference |
|------------------------|--------------------------|---------------------|-------------------|-----------|
| CDs                    | Manilkara zapota fruits  | 100 °C, 60 min      | 5.2 – 7.9         | [1]       |
| CDs                    | Watermelon peel          | 220 °C, 2 h         | 7.1               | [2]       |
| CDs                    | Ocimum sanctum           | 180 °C, 4 h         | 9.3               | [3]       |
| CDs                    | Thymus vulgaris L        | 180 °C, 5 h         | 5.2               | [4]       |
| CDs                    | Pomelo peel              | 200 °C, 3 h         | 6.9               | [5]       |
| CDs                    | Aloe                     | 180 °C, 11 h        | 10.4              | [6]       |
| P-CQDs                 | Pine wood                | 180 °C, 3 h         | 4.7               | [7]       |
| N, P-CQDs              | Eleocharis dulcis        | 120 °C, 5 h         | 11.2              | [8]       |
| CDs                    | Sewage sludge            | 700 W, 30 min       | 21.7              | [9]       |
| CQDs                   | Bamboo tar               | 170 °C, 15 min      | 19.3              | [10]      |
| PEG-CDs                | Gelatin                  | 600 W, 10 min       | 34                | [11]      |
| CDs                    | Milk protein             | 30 min              | 18.7              | [12]      |
| B,N-GQDs<sup>b</sup>   | *Passion fruit juice*    | 170 °C, 20 min      | 50                | This study |

a: CDs: carbon dots; CQDs: carbon quantum dots, GQDs: graphene quantum dots

b: B,N-GQDs
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**Table S4.** Zeta potential of B,N-CDs, tetracycline (TC), B,N-CDs/TC at various pHs.

| pH value | Zeta potential (mV) | B,N-CDs | Tetracycline (TC) | B,N-CDs/TC |
|----------|----------------------|---------|-------------------|------------|
| pH 3     | -11                  | -3.34   | -3.32             |            |
| pH 4     | -4.89                | -3.74   | -2.28             |            |
| pH 5     | -14.9                | -6.26   | -3.45             |            |
| pH 6     | -19.5                | -14.1   | -2.91             |            |
| pH 7     | -0.86                | -24.1   | -3.23             |            |
| pH 8     | -2.51                | -2.13   | -3.66             |            |
| pH 9     | -4.16                | -9.99   | -1.02             |            |
| pH 10    | -6.66                | -4.95   | -4.74             |            |
**Graph (a):**

- **G band:** 1584 cm\(^{-1}\)
- **D band:** 1357 cm\(^{-1}\)
- **ID/IG:** 0.89

**Graph (b):**

- **ID/IG:** 0.89
- **G band:** 1584 cm\(^{-1}\)
- **D band:** 1357 cm\(^{-1}\)
Fig. S1. (a) The full and (b) partial Raman spectrum of B,N-GQDs, and (c) the XRD pattern of B,N-GQDs on the Si substrate.
Fig. S2. (a) High resolution scanning electron microscopy (HRSEM) image, (b) energy dispersive spectroscopy (EDS) spectrum and elemental mapping of (c) C, (d) O (e), N and (f) B elements of B,N-GQDs.
Fig. S3. The (a) XPS deconvoluted C 1s and (b) O 1s peaks of N-GQD.
Fig. S4. The UV-visible spectra of B,N-GQDs, tetracycline (TC), and B,N-GQDs/TC.
Fig. S5. (a) (b) The change in fluorescence of B,N-GQDs in the absence and the presence of TC (30 µM) under visible light and (c) (d) under 365-nm UV light irradiation and in PBS solution.
Fig. S6. The linear relationship between fluorescence curve areas and absorbance for (a) pure N-GQDs, (b) B,N-GQDs, and (c) quinine sulfate standard.
Fig. S7. (a) The change in fluorescence emission spectra of B,N-GQDs at low TC concentration range of (0.06–14 nM) in urine and (b) human serum.
**Fig. S8.** The effect of pH on the fluorescence intensity of B,N-GQDs before and after the addition of 30 µM tetracycline. The pH is controlled at 3 – 10 in the presence of 0.1M PBS.