Adsorbate preparation

Ultrapure water was obtained from a Milli-Q water filtration station (18.2 MΩ·cm at 20 °C). Aqueous solutions of diquat dibromide [DqDb] (Sigma-Aldrich, 97%) were prepared at concentrations of 15.0 and 25.0 µg/mL with a pH of 6.8 monitored with a Mettler Toledo, FG2 pH-meter.

Adsorbents preparation and characterization

Granular AC and hydroxyl(OH)-functionalized SWCNTs containing 3.9% OH groups were purchased from Calgon Carbon Corp. (Filtrasorb 100) and Cheap Tubes Inc., respectively. Their purity level is higher than 90 wt% and they were used as-received without further treatment to preserve the functional groups on their surface. Pristine single-wall and multi-wall CNTs were obtained from Cheap Tubes Inc. and further purified as follows. SWCNTs and MWCNTs were soaked in separate hydrochloric acid bath (Sigma-Aldrich, 37.5%) overnight at room temperature to remove metal particles. The suspensions were then filtered through aqueous membranes (90 mm, 113 Whatman) and thoroughly washed with deionized water, before being placed in a muffle furnace (Thermo-scientific, Thermolyne 1300) at 560 ºC for 10 minutes to remove amorphous carbon. Prior to adsorption analysis, all samples (i.e. granular AC, SWCNT, MWCNT and SWCNT-OH) were maintained at 110 ºC for at least 48 hours to limit the competitive Coulombic stability of the deprotonated carboxylic acid.

All specimens were characterized using a field emission scanning electron microscope (FE-SEM, Hitachi S-900) operated at 2 kV. Similar morphological features without any distinctive differences were observed between the pristine SWCNTs and the OH-functionalized SWCNTs. Raman spectra were acquired using a JY-Horiba Labram spectrophotometer equipped with a 632.6 nm He/Ne laser as the excitation radiation.
**Fixed bed adsorption studies**

A gravity-fed fixed-bed column was operated with DqDb feed concentrations of 15 and 25 µg/mL. The solution feed was continuously agitated with a magnetic stirrer, and a peristaltic pump (Langer instruments BT100-2J) was used to maintain a set hydrostatic head above the bed to achieve a constant flow rate of 2 mL/min through the column. Slight adjustments of the pump speed were necessary to compensate for the pressure drop in the column over time. The adsorbent bed was created by packing the various carbonaceous adsorbents (200-900 mg) to a 2 cm depth in a portion of a transparent glass tube having a total length of 65 cm and an internal diameter of 1.4 cm (Fig. 1). The bed was supported and surrounded by 5 cm layers of glass wool (Sigma Aldrich), while the top of the column was filled with glass beads (Sigma Aldrich, 0.5 cm diameter) to promote liquid distribution across the column cross-section. Prior to adsorption, the column was flushed with de-ionized water for 1 hour and no adsorbent was detected in the effluent at any time using absorption spectroscopy. The performance of the column was evaluated at 20 ºC by measuring the effluent concentration at the outlet of the column at different time intervals by UV/Vis absorption spectroscopy (Perkin Elmer Lambda 950 UV/Vis/NIR) using a measured extinction coefficient from a Beer's law analysis. Control experiments indicated that the adsorption of DqDb on glass wool was negligible. For statistical soundness, all data corresponded to the average among triplicate trials with typical errors being less than 10%.

**Batch adsorption studies**

In batch adsorption studies, 0.5 mg ±0.05 mg of dried adsorbent was submerged into a constant volume of the solution of interest. Isotherm adsorptions were studied at 20 ºC in a batch adsorption vessel (10 mL) placed on an orbital shaker platform operated at 120 rpm (Bel-Art Spindrive). Equilibrium was declared when there was no appreciable change in solution concentration with additional contact time. Blank adsorption experiments were performed without any adsorbent to ensure that no molecule was adsorbed on the wall of the container. The amount of DqDb adsorbed per mass of adsorbent, $q$, was deduced by subtracting the mass of adsorbate in solution at a given time from the initial mass of adsorbate in solution.

From the equilibrium isotherms presented in Fig. S1, the maximum adsorption capacity of DqDb can be extracted, leading to maximum uptake values of 0.085 and 0.120 mg/mg for AC and carbon nanomaterials, respectively. These values are higher than those for fixed bed adsorption. This is due to fact that the feed concentration is in the flat part of the equilibrium isotherm. In this regime, the advantage of fixed bed is lost and mass transfer limitations along with axial dispersion limit the degree of adsorbent utilization in the column, hence decreasing its overall efficacy. However, it is worth noting that for a limited time, all fluid exiting the fixed bed column has zero contaminant. In contrary, it is not possible to achieve complete removal of diquat in a well-mixed batch process since there is always a
residual concentration.

**Figure S1.** Adsorption isotherms of DqDb onto granular AC (●) and carbon nanomaterials (♦) at 20 ºC. The straight and dashed lines are curvefits corresponding to Langmuir and Freundlich isotherms, respectively.