



## Influence of chemical and structural evolution of dissolved organic matter on electron transfer capacity during composting



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### HIGHLIGHTS

- Electron transfer capability (ETC) of compost-derived DOM was investigated.
- Composting treatment increased the ETC of DOM from municipal solid wastes.
- The ETC increase related to humic matter, and molecule weight, and N and S content.

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### ABSTRACT

Dissolved organic matter (DOM) can mediate electron transfer and change chemical speciation of heavy metals. In this study, the electron transfer capability (ETC) of compost-derived DOM was investigated through electrochemical approaches, and the factors influencing the ETC were studied using spectral and elemental analysis. The results showed that the electron accepting capacity (EAC) and electron donating capacity (EDC) of compost-derived DOM were 3.29–40.14  $\mu\text{mol}_e^- (\text{g C})^{-1}$  and 57.1–346.07  $\mu\text{mol}_e^- (\text{g C})^{-1}$ , respectively. Composting treatment increased the fulvic- and humic-like substance content, oxygenated aliphatic carbon content, lignin-derived aromatic carbon content, molecule weight, and N and S content of DOM, but decreased the aliphatic carbon content and the C and H content. This conversion increased the EDC and EAC of the DOM during composting.

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## 1. Introduction

Dissolved organic matter is a heterogeneous mixture of organic compounds that include organic acids, amino acids, humic substances, and enzymes. It can be used as food by microbes to improve organic matter degradation and enhance nutrient cycling [1,2], as well as interact with heavy metals or adsorb hydrophilic organic pollutants affecting the transport and fate of these pollutants [3,4]. In addition, DOM can function as electron shuttles and mediating electron transfer between reducing agents and electron acceptors [2], which facilitate the reduction of heavy metals such as Fe(III) and Cr(VI) [5] and the microbial degradation of persistent organic pollutants, such as chloro-organics and nitrophenols [6].

Composting is a biological transformation process in which organic matter is degraded to carbon dioxide or converted into humic substances [7]. DOM has a significant function in organic matter transformation during composting because it represents the most active organic fraction [1,7]. Labile organic substances such as aliphatics, polysaccharides, and proteins in DOM are used preferentially by microorganisms during composting, whereas compounds with aromatic structural units in DOM are not easily degraded and generally accumulate during the process [1,8]. The DOM from mature compost contains a low content of biodegradable organic matter and a higher concentration of organic macromolecules [9]. These aromatic-containing macromolecules consist mainly of humic substances, which are polyphenolic structures derived from the oxidation of lignin during composting [7,10]. The organic matter evolution during composting may change the complexation behaviors and electron transfer capability (ETC) of DOM in composts. However, to date, information on the influence of organic matter transformation on the ETC of DOM is limited.

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The ETC of DOM includes electron accepting capacity (EAC) and electron donating capacity (EDC), both of which can be determined through electrochemical approaches [2]. Quinine moieties, which can be produced by lignin oxidation, are responsible for the ETC of DOM [11]. Quinones can cycle between the oxidized state, semiquinone radical, and reduced or hydroquinone state [12] when they accept or donate electrons. Studies have demonstrated that some DOM fluorophores are a function of the redox state of the quinones in DOM, and use fluorescence spectra of DOM to detect the presence and redox state of quinines [12]. However, to our knowledge, the relationship between the fluorophore organic matter and the ETC characteristics of DOM is unclear. Apart from studying quinone-like moieties, Maurer et al. [13] showed that nitrogen- and sulfur-containing moieties may also be relevant to the ETC of DOM. Unfortunately, until now, no evidence has been provided to support the hypothesis.

Heavy metal contamination limits the use of compost products. The toxicity of heavy metals is related to their speciation. Compared with the heavy metals at the solid phase, dissolved heavy metals are more easily used by microbes and plants and exhibit higher toxicity. The ETC of compost-derived DOM may change during composting, which facilitates or delays the reduction of heavy metals and changes their speciation and toxicity. To our knowledge, the influence of the ETC evolution on the heavy metal speciation distribution in composts is seldom investigated.

The objectives of this study are to (1) investigate the ETC of compost-derived DOM, (2) analyze the influence of organic matter evolution on the ETC during composting, and (3) study the influence of the ETC evolution on the heavy metal speciation distribution in compost.

## 2. Materials and methods

### 2.1. Composting process and sample collection

Composting experiment was performed at a composting plant in Beijing, China. The composting raws were municipal solid wastes (MSWs), which were collected from Beijing. The MSWs were sorted manually and mechanically to remove metals, plastics, glass, and stones, and the residual MSWs were piled for composting. The total carbon, total nitrogen, organic matter and moisture content of the residual MSWs were 303.20 g kg<sup>-1</sup>, 12.55 g kg<sup>-1</sup>, 52.11% and 57.85%, respectively. The residual MSWs (more than 2 t each) were composted in trapezoidal piles (1.5 m high with a 2 × 8 m base). The composting process lasted for 51 d and divided into active and curing stages. The active stage was conducted for 21 d, during which the piles were turned every 2 d, and its humidity was maintained at around 50–65% with an addition of composting leachates. The curing stage lasted for 30 d, during which the piles were forked every 7 d. Five composts were sampled at 0, 7, 14, 21, and 51 days.

### 2.2. DOM extraction

As described by Said-Pullicino et al. [7] and He et al. [1], compost DOM was extracted with distilled water by preparing the compost sample with a solid-to-water ratio of 1:10 (w/v, dry weight basis) and then shaking the mixture at 200 rpm in a horizontal shaker for 24 h at 20 °C. The suspension was subsequently centrifuged at 10,000 rpm for 10 min, and then filtered through a 0.45 μm membrane filter. Filtrates obtained were divided into two parts: one part was freeze-dried for solid-state <sup>13</sup>C NMR and elemental analysis, and the rest was stored at 4 °C before fluorescence, UV–vis spectra, and heavy metal analysis. The dissolved organic carbon content of DOM was measured using an Analytik Jena Multi N/C 2100 TOC analyzer.

### 2.3. ETC measurements

The EDC and EAC of compost-derived DOM were measured through electrochemical experiments. The experiments were conducted on an electrochemistry workstation CHI660D (Chenhua, Shanghai, China) with a conventional three-electrode cell as described by Yuan et al. [5] and Bi et al. [2]. The cell consisted of a counter electrode, a reference electrode, and a working electrode. The Pt net and Hg/Hg<sub>2</sub>Cl<sub>2</sub> electrodes were used as the counter and reference electrodes, respectively, and a graphite plate electrode with a projected surface area of 17.5 cm<sup>2</sup> was chosen as the working electrode. Amperometric *i*–*t* curve analysis was conducted to evaluate the EDC and EAC of compost-derived DOM in a nitrogen-saturated phosphate buffer solution (pH 7.0) [2,5]. The process involved the following steps: 1 mL of the DOM with a dissolved organic carbon concentration of 150 mg L<sup>-1</sup> was carefully transferred into the gastight electrochemical cell and the EDC was determined at a positive potential of +0.5 V (versus Hg/Hg<sub>2</sub>Cl<sub>2</sub>); then, the EAC was measured at a negative potential of –0.6 V (versus Hg/Hg<sub>2</sub>Cl<sub>2</sub>). The process was repeated at least three times per sample at an ambient temperature. The ETC was calculated using the following formula:

$$\text{ETC (EDC or EAC)} = \frac{A_p}{(e \times N_A \times M_C)}$$

where ETC was the amount of electrons donated by the DOM for the EDC measurement or the amount of electrons transferred to the DOM for the EAC measurement. The unit of the ETC was μmol e<sup>-</sup> (g C)<sup>-1</sup>. *A<sub>p</sub>* was the coulomb integral of the oxidized wave of DOM (area below the recorded curve) for the EDC measurement or the coulomb integral of the reductive wave of DOM (area above the recorded curve) for the EAC measurement. The unit of the *A<sub>p</sub>* was coulomb. *e* was the charge on an electron (1.6 × 10<sup>-19</sup> C) and *N<sub>A</sub>* was the Avogadro constant (6.02 × 10<sup>23</sup> mol<sup>-1</sup>). *M<sub>C</sub>* was the amount of carbon transferred to the electrode cell (2 × 10<sup>-4</sup> g).

### 2.4. Fluorescence spectra

Prior to spectral analysis, the DOC of all samples was adjusted to 7.89 mg L<sup>-1</sup>. Excitation–emission matrix (EEM) spectra were recorded with a Hitachi model F-7000 luminescence spectrophotometer equipped with a 150 W xenon arc lamp as the excitation source. To obtain the EEM spectra, we set the slit widths of the excitation and emission monochromators as 5 nm, adjusted the voltage of the photomultiplier tube to 400 V, and scanned the emission (Em) wavelength from 280 to 520 nm by increasing the excitation (Ex) wavelength in 5 nm increments from 200 to 440 nm with a scan speed of 2400 nm min<sup>-1</sup>. The EEM spectra of distilled water were obtained and subtracted from the EEM spectra of the compost-derived DOM.

The EEM spectra data of the compost-derived DOM and another 45 EEM spectra data of the DOM from the other compost and landfill leachates comprised a three-dimensional data array (50 samples × 41 Ex × 49 Em). Parallel factor (PARAFAC) analysis was applied to the three-dimensional data array using MATLAB 7.0 (Mathworks, Natick, MA) with the DOMFluor toolbox ([www.models.life.ku.dk](http://www.models.life.ku.dk)), and the array was decomposed into three matrices: score, Ex loading, and Em loading matrices [14,15]. The scores in the score matrix, which were expressed as *F<sub>max</sub>* values, were used to evaluate the concentration of the fluorescence components determined through EEM–PARAFAC analysis in accordance with previous reports [14–16].

In addition, emission spectra were obtained over a range of 300–500 nm at an excitation wavelength of 254 nm, and humification index (HIX) of compost-derived DOM was calculated by dividing the area at 300–345 nm by the sum of the areas at

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