



Development and evaluation of diffusive gradients in thin films based on nano-sized zinc oxide particles for the in situ sampling of tetracyclines in pig breeding wastewater

Nan You ^{a,1}, Hui Yao ^{b,1}, Yun Wang ^{c,1}, Hong-Tao Fan ^{a,*}, Chang-Song Wang ^{b,*}, Ting Sun ^{c,*}

^a College of Chemistry Chemical Engineering, and Environmental Engineering, Liaoning University of Petroleum & Chemical Technology, Fushun 113001, Liaoning, China

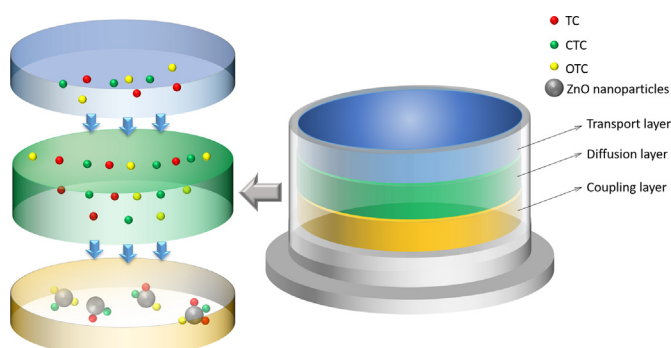
^b College of Applied Chemistry, Shenyang University of Chemical Technology, Shenyang, 100142, Liaoning, China

^c College of Sciences, Northeastern University, Shenyang 110004, China

HIGHLIGHTS

- A DGT sampler based on ZnO nanoparticles for in-situ sampling of tetracyclines in waters.
- An inert polyethersulfone membrane as the diffusion layer of DGT technique
- No notable influence on the uptake of nanoZnO–DGT for TCs in the range of pH 5–9 and pNaCl 1–3
- No interference of water matrices on the uptake of tetracyclines by DGT sampler
- Reliable and accurate results in field deployment with relative good precision

GRAPHICAL ABSTRACT



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ABSTRACT

The pollution of antibiotics, including tetracyclines (TCs), in aquatic environments has become an issue of concern in recent years. Herein, an in situ sampling of TCs in pig breeding wastewater that utilizes the technique of diffusive gradients in thin films (DGT), based on commercial nanosized ZnO (nanoZnO) particles as the potential effective binding agent and a polyethersulfone (PES) membrane as the diffusion layer, was developed. The diffusion coefficients of tetracycline (TC), oxytetracycline (OTC) and chlortetracycline (CTC) in a PES membrane at 25 °C were $(1.37 \pm 0.06) \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$, $(1.29 \pm 0.05) \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ and $(1.94 \pm 0.07) \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$, respectively. The results showed that the adsorption capacities of a gel disc containing 2.5 g L⁻¹ of nanoZnO particles were as high as $3.93 \pm 0.20 \text{ mg disc}^{-1}$ for TC, $3.21 \pm 0.20 \text{ mg disc}^{-1}$ for OTC and $4.62 \pm 0.22 \text{ mg disc}^{-1}$ for CTC. Both a solution pH in the range of 5–9 and an ionic strength (as pNaCl) in the range of 1–3 had an insignificant influence on the TCs uptake by nanoZnO–DGT samplers. There was no significant influence of fulvic acid or tannic acid on the TC uptake by nanoZnO–DGT samplers at the tested mass ratios. For all spiked freshwater samples, there was no notable interference of matrices on the performance of the nanoZnO–DGT samplers, suggesting that the nanoZnO–DGT samplers yielded satisfactory results for the uptake of TCs at concentrations existing in the spiked freshwater samples. Field deployment of the nanoZnO–DGT samplers in pig breeding wastewater

* Corresponding authors.

E-mail addresses: htyf_77@163.com (H.-T. Fan), Cswang_syict@163.com (C.-S. Wang), sun1th@163.com (T. Sun).

¹ The first three authors contributed equally to this paper.

also exhibited excellent precision and accuracy, indicating that the nanoZnO–DGT samplers could be used as a promising method for the in situ sampling of TC antibiotics in aquatic environments.

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1. Introduction

Tetracycline (TC) antibiotics, such as tetracycline (TC), oxytetracycline (OTC) and chlortetracycline (CTC), comprising fused aromatic rings in the molecule have been widely used as veterinary drugs and/or growth-promoting agents for swine feeding operations to control and prevent microbial infections in China (Huang et al., 2013; Zhang et al., 2015a). TC antibiotics were mostly excreted through swine manure with the prototype drug due to their poor absorption in the digestive tract (Barry and Braybrooks, 1975). In previous reports, high concentrations of TCs were found in swine wastes collected from concentrated swine feedlots (Zhang et al., 2015b). The excreted TC residues were delivered to the surrounding aquatic environment through the unregulated discharge or use of swine wastes, resulting in an increasing concern regarding the increase of pathogenic antibiotic-resistant bacteria (Li et al., 2016). To understand the potential environmental risks, it is thus of great importance to develop an efficient technology for the long-term monitoring of TC concentrations in aquatic environments.

The use of diffusive gradients in thin films (DGT), an in situ passive sampling technique, was first developed by Davison and Zhang (Zhang and Davison, 1994) and was previously used to monitor the concentration of the target pollutants in the environment (Öhlander et al., 2012). The first DGT device was composed of a diffusive layer with polyacrylamide hydrogel and a binding layer using polyacrylamide hydrogel impregnated with Chelex-100 resin as the binding agents (Zhang and Davison, 1995). For the application to effectively prosper, various functional materials were tested as the DGT binding agent, resulting in the extension of the target analytes from cationic heavy metals (Garmo et al., 2003; Wang et al., 2018) to nutrient elements (e.g., sulfide (Gao et al., 2015), dissolved reactive phosphorus (Ding et al., 2010), potassium (Tandy et al., 2012), ammonium (Huang et al., 2016) and nitrate (Huang et al., 2016)), metalloids (such as arsenic and selenium) (Bennett et al., 2010), oxyanion heavy metals (such as vanadium (Luko et al., 2017), uranium (Drozdak et al., 2016; Gregusova and Docekal, 2013), and chromium (Yao et al., 2016)), fluoride (Zhou et al., 2016) and organic pollutants (such as 4-chlorophenol (Dong et al., 2014) and bisphenols (Zheng et al., 2015)). Recently, a DGT device with a XAD18 resin as the binding agent has been developed and successfully tested for validation of monitoring 40 target antibiotics, including 16 sulfonamides, 12 fluoroquinolones, 6 macrolides, 2 ionophores, 2 diaminopyrimidines, 1 aminocoumarin, and 1 lincosamide, (Chen et al., 2013 and 2015; Xie et al., 2018), as well as pesticides (Challis et al., 2016), methcathinone and ephedrine (Zhang et al., 2018), in water. However, little is known about the sampling of TC antibiotics by the DGT method. Therefore, the development of new binding agents is beneficial for the optimization of the DGT method for the sampling of the available TC antibiotics in water.

Nanomaterials have specific physicochemical properties differing from those of bulk samples. In wastewater treatment, nanoZnO particles have attracted a great deal of attention for the removal of contaminants due to a large surface for adsorption (Ghaedi et al., 2014; Tran Thi and Lee, 2017) and a high reactivity for degradation (Guo et al., 2017; Panneri et al., 2016; Xue et al., 2015). To the best of our knowledge, this is the first study on in situ sampling of TCs using the DGT samplers based on nanoZnO particles (nanoZnO–DGT), which may have a potential significance for the monitoring of TCs in the aqueous environment. Under various pH conditions, ionic strengths and deployment times, the performance of nanoZnO–DGT samplers for TC analysis was assessed. The validation of the nanoZnO–DGT samplers for the in situ sampling of TCs in wastewater from animal husbandry was also examined.

2. Experimental

2.1. General experimental procedures

The nanoZnO particles (99.8% metals basis, 100 ± 10 nm) and tannic acid (TA) were obtained from Shanghai Aladdin Biochemical Technology Co., Ltd. Fulvic acid (FA) with purity of 90% was obtained from Shanghai Yuanye Biotechnology Co., Ltd. and used without further purification. All other chemicals were analytical reagent grade or better from Sinopharm Chemical Reagent Co., Ltd., or Sigma-Aldrich. White opaque polyethersulfone (PES, $0.1 \mu\text{m}$ pore size, $165 \pm 8 \mu\text{m}$ thickness, 25 mm diameter) membranes were obtained from Sartorius. The stock solution of Zn^{2+} (1000 mg L^{-1}) was obtained from the National Research Center for Standard Materials (Beijing, China). The stock solution of TCs at 1000.0 mg L^{-1} was obtained by deionized water. Prior to use, all devices and glassware were cleaned by soaking in 10% (v/v) HNO_3 solution overnight followed by rinsing with deionized water.

2.2. Interaction of the PES membrane with TCs

The PES membrane was immersed in 10 mL of TCs solution containing various concentrations (200 , 500 and $1000 \mu\text{g L}^{-1}$) with stirring for 24 h. The concentrations of TCs in the solutions before and after exposure were determined to obtain the TCs mass adsorbed by the PES membrane.

2.3. Preparation of binding gels

The binding gels were prepared following a published procedure (Zhang and Davison, 1995). The gel solution was comprised of 15% acrylamide and 0.3% N,N' -methylene bisacrylamide cross-linker. 25 mg of nanoZnO particles was added into 10 mL of gel solution to affect the dosage of 2.5 mg mL^{-1} . Subsequently, 70 μL of 10% ammonium persulfate and 25 μL of N,N,N',N' -tetramethylethylenediamine per 10 mL of mixture solution were added into the above. Binding gels were cast at 40°C for 1 h. The binding gel discs with a diameter of 20 mm and a thickness of 2 mm were cut and stored in 0.01 mol L^{-1} NaNO_3 solution at pH 6 prior to use at 4°C .

2.4. General procedure for assembly and deployment of nanoZnO–DGT samplers

The schematic diagram of a DGT sampler composed of a polytetrafluoroethylene enclosure and light-proof PES membrane is provided in the Supporting Information (Fig. S1). A nanoZnO binding gel disc was put into the polytetrafluoroethylene fillister, after which a PES membrane was covered on top of the nanoZnO binding gel disc and immobilized by a polytetrafluoroethylene cap with a window of 20 mm diameter. To avoid the adhesion of suspended particulates, nanoZnO–DGT samplers were deployed with the membrane side facing down during use. The laboratory and field deployments of nanoZnO–DGT samplers were carried out over a period of 24–120 h. Laboratory deployments were kept at $20 \pm 5^\circ\text{C}$. An aquarium pump with a current velocity of 100 cm s^{-1} for all trials was used to maintain constant and rapid motion of the solution in order to avoid the influence of diffusive boundary layer (DBL).

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