



Four types of attenuation of phenol and cresols in microcosms under simulated marine conditions: A kinetic study



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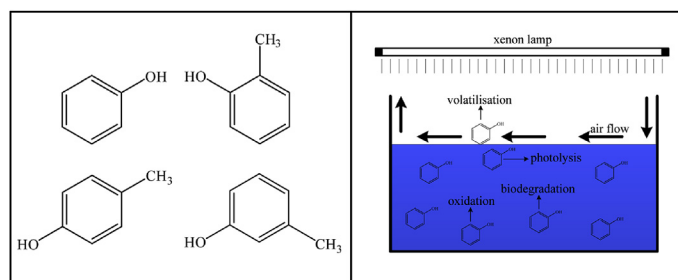
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HIGHLIGHTS

- Natural attenuations of phenols were simulated in microcosms.
- Half-lives of natural attenuation of phenols were obtained.
- Proposed kinetics explained the attenuation of phenols in detail.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 26 March 2017

Received in revised form

5 July 2017

Accepted 10 July 2017

Available online 11 July 2017

Handling Editor: Cianc-Cousins

Keywords:

Phenols
Attenuation
Kinetics
Marine
Microcosm

ABSTRACT

Phenol, *o*-cresol, *m*-cresol, and *p*-cresol were selected to conduct microcosm experiments to examine their attenuation under simulated marine conditions, aiming at estimating natural attenuation and the contribution of oxidation, photolysis, biodegradation, and volatilisation to total attenuation of phenol and three cresols in the marine environment. The development of attenuation in microcosms showed the relevance of the pseudo-first-order kinetic for all phenols. The half-lives of phenol, *o*-cresol, *m*-cresol, and *p*-cresol attenuation under optimal conditions were 7.9, 4.3, 5.3, and 4.4 d, respectively. Attenuation kinetics was proposed to analyse the natural attenuation of phenol and cresols. The leading attenuation type of phenol, *o*-cresol, and *p*-cresol was volatilisation, and the attenuation rate constants (K_v) were 0.0356, 0.0687, and 0.0710 d^{-1} . Photolysis (K_p : 0.0584 d^{-1}) was the major attenuation type for *m*-cresol. Biodegradation of phenol (K_b : 0.0021 d^{-1}) and *m*-cresol (K_b : 0.0049 d^{-1}) were extremely inhibited. The rank between the contributions of the four types of attenuation to total attenuation differed between phenol and the three cresols. The attenuation kinetics proposed in this study possibly demonstrated the attenuation of the phenol and cresols in microcosm. This new reaction kinetics can be used in the analysis of natural attenuation of chemical substances.

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1. Introduction

World transport of Hazardous and Noxious Substances (HNS) by

sea has increased significantly in recent years (Cunha et al., 2016), thus increasing the risk of accidental spillage of HNS in the marine environment. The frequency of occurrence of spillages of HNS has increased over time (Rocha et al., 2016).

The severity of an HNS spill impact depends on several parameters, such as: local marine environmental conditions,

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properties of the HNS, and the behaviour of HNS once released to the marine environment (Cunha et al., 2015). Hence, there is a need to understand, in more detail, the environmental fate of the HNS to support the development of strong, consistent contingency plans (Rocha et al., 2016). However, one of the major gaps in current knowledge is the limited information about the short-term behaviour of HNS once spilt at sea in real conditions. Models usually make use of the properties of HNS as obtained by laboratory testing: the time factor and meteorological conditions – two significant variables affecting post-spillage behaviour – have not been addressed (Cunha et al., 2015). The gap should be addressed through experiments in the laboratory involving priority HNS compounds. Phenolic compounds are toxic to fish and other aquatic organisms, and are listed as priority pollutants by the US Environmental Protection Agency (USEPA) (Du et al., 2009). Phenol is also one of the most common HNS chemicals involved in accidental marine spills (Cunha et al., 2015). Phenol and cresols are highly soluble in water (Wei et al., 2016), that increase the risk of environmental damage as the phenols can persist in an aquatic environment at high concentrations. It is vital to investigate the attenuation processes of phenol and cresols in the natural environment. In this study, phenol, *o*-cresol, *m*-cresol, and *p*-cresol were selected as priority HNS.

If an accidental spillage of phenol or cresols occurs in a semi-enclosed bay without effective water exchange capacity, high concentrations of phenol or cresols in seawater will accrue due to a lack of short-term dilution and diffusion. Phenol and cresols do not adsorb to suspended solids and sediment in the water (Badanthadka and Mehendale, 2014). Therefore, once spilt in a marine environment, the natural attenuation of dissolved phenol and cresols in the aquatic environment is governed by biodegradation, sunlight photolysis, volatilisation, and other abiotic transformations, such as oxidation (Callahan et al., 1979). Phenol and cresols are not expected to undergo hydrolysis in the natural environment due to the lack of hydrolysable functional groups (Lyman et al., 1990). The ability to simulate and model these processes is necessary to facilitate the management and understanding of any contaminated area.

The photolysis, biodegradation, oxidation, and volatilisation of phenol and cresols have been investigated by various researchers (Southworth, 1982; Devlin and Harris, 1984; Hwang et al., 1986; Rayne et al., 2009; Stefanakis et al., 2013; Hantoosh, 2016; Wei et al., 2016); Thornton et al. (2001) and Mayer et al. (2001) evaluated the distribution and natural attenuation of phenol and cresols in a deep sandstone aquifer; however, no information regarding the analysis of the natural attenuation of phenol and cresols under environmentally relevant conditions could be found. On the other hand, most models focussing on the degradation of a substance cannot be used to infer the effect of factors such as degradation by microbial action, oxidation, photolysis, hydrolysis, volatilisation, etc., on the rate of attenuation of the chemical substance. The aim of the present study is to investigate the attenuation of phenol and cresols in a simulated marine environment with a view to proposing a kinetic model that takes into account biodegradation, oxidation, photolysis, and volatilisation. The natural attenuation of the phenol, and three cresols, was explored in microcosm under simulated marine conditions.

2. Materials and methods

2.1. Chemicals

The *o*-cresol, *m*-cresol, and *p*-cresol were purchased from Tokyo Chemical Industry Co., Ltd (Tokyo, Japan). Phenol was obtained from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). All of

the other solvents and reagents were of HPLC or analytical grade.

2.2. Seawater

The natural seawater using in this study was collected from the coastal region near Qingdao, and the samples were filtered through a 200 μm nylon mesh to remove large detritus and metazoans. The physicochemical properties of the seawater were as follow: pH (7.92), Salinity (32.0), Dissolved Oxygen (7.6 mg L^{-1}), Conductivity ($4.52 \times 10^4 \text{ us cm}^{-1}$), Turbidity (2.85 NTU), Cl^{-1} (492.5 mM), SO_4^{2-} (28 mM), Dissolved Inorganic Nitrogen (0.095 mg L^{-1}), $\text{PO}_4\text{-P}$ ($5.8 \times 10^{-3} \text{ mg L}^{-1}$).

2.3. Experimental devices and conditions

The marine microcosm used throughout this study referred to the apparatus described in Macnaughton et al. (2003) (Fig. 1). Considering that the acute toxicity (LC_{50}) of phenol or cresols to the majority of fish species is not higher than 50 mg L^{-1} (Kegley et al., 2008), a spill of 50 mg L^{-1} of phenol or cresols will have a significant impact on marine ecology. 50 mg L^{-1} was selected as the concentration of phenol and cresols in microcosm. To simulate this phenol and cresol slick, $500 \pm 50 \text{ mg}$ phenol, *o*-cresol, *m*-cresol, or *p*-cresol were added to each microcosm, which containing 10 L seawater. The air flow was then switched on (mean average velocity

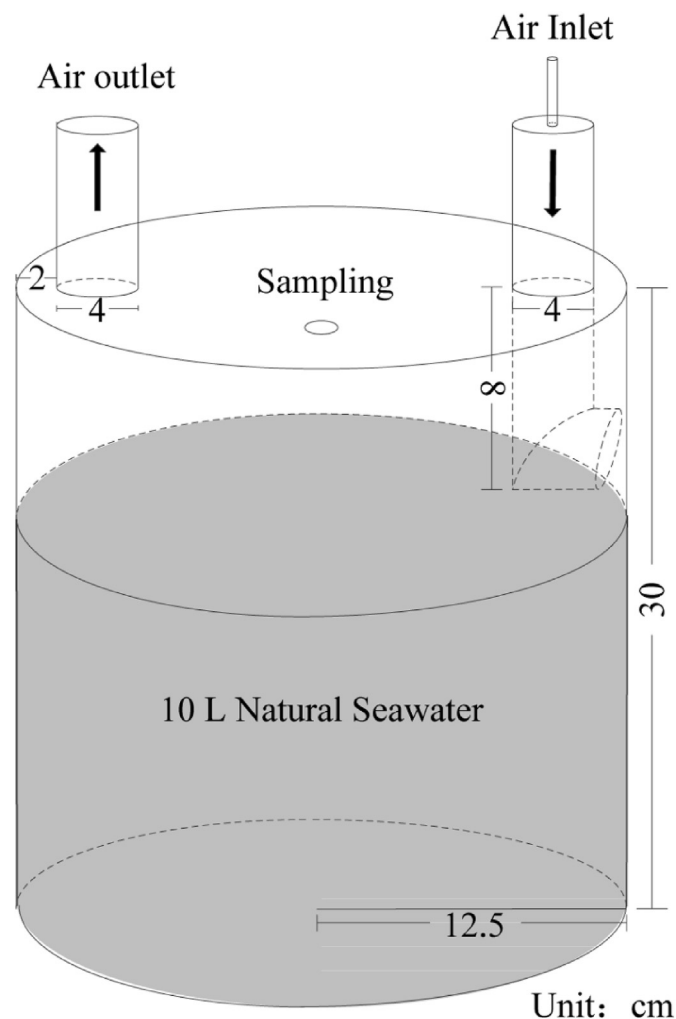


Fig. 1. Schematic of the laboratory microcosm.

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