



Risk assessment of marine environments from ballast water discharges with laboratory-scale hydroxyl radicals treatment in Tianjin Harbor, China



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ABSTRACT

For the majority of ballast water treatment system (BWTS) that employ active substances (e.g., oxidative compounds), relevant chemicals (RCs) formation is an issue owing to their potential adverse effects on aquatic organisms. Accordingly, BWTS must be approved by the International Maritime Organization (IMO), and the approval procedure requires environmental risk assessment. The most commonly employed harbor used to calculate predicted environmental concentrations (PECs) for RCs in treated ballast water is the GESAMP-BWWG (Group of Experts on Scientific Aspects of Marine Environmental Protection–Ballast Water Working Group) model harbor. However, there is very little assessment data available regarding the associated environmental impacts in ports and harbors of China. Therefore, in this study the concentration of fifteen RCs from the existing laboratory-scale BWTS using hydroxyl radicals was obtained and input into the MAMPEC (Marine Antifoulant Model to Predict Environmental Concentrations) model to compute PECs in Tianjin Harbor, China. The potential risks to the aquatic environment posed by treated ballast water in Tianjin Harbor were further assessed based on the calculated ratio of PECs and predicted no effect concentrations (PNECs). Only monochloroacetic acid and dichloroacetic acid were found to have potential risks, and the ratios of PECs and PNECs to the other measured RCs were less than 1, indicating that the environmental risk posed by treated ballast water discharged into Tianjin Harbor is of little concern. The concentration of total residual oxidant recommended by the IMO (<0.2 mg/L) in treated ballast water at discharge was found to be at levels that may pose a risk to the aquatic environment in Tianjin Harbor.

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1. Introduction

The concept of non-native organisms introduced through the discharge of ballast water was first proposed in the 1980s. Since then, the introduction of harmful aquatic organisms and pathogens to new environments via ships' ballast water has been identified as one of the four greatest threats to oceans worldwide, and as a source of adverse economic effects (Molnar et al., 2008; GloBallast, 2009). In recent years, aquatic nonindigenous invasive species have

become a significant and growing contributor to the spread of red tide blooms on the coast of China. To minimize the transfer of harmful aquatic organisms and pathogens via this route, the International Maritime Organization (IMO) adopted the International Convention for the Control and Management of Ships' Ballast Water and Sediments (Ballast Water Convention) in 2004 (IMO, 2004). Consequently, a variety of technologies have been developed by different vendors to remove living organisms from ballast water before it is discharged.

There are two generic technologies used to treat ballast water: solid–liquid separation and disinfection (Lloyd's Register, 2011). Solid–liquid separation is a mechanical process (e.g., hydrocyclone or filtration) that is usually applied prior to the actual treatment system to remove suspended solids, including larger suspended organisms. However, this method does not kill harmful organisms.

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Disinfection removes and/or inactivates organisms and bacteria using one or more methods, such as chemical inactivation (e.g., ozonation or electrolysis) or physicochemical inactivation (e.g., UV light, heat or cavitation) (Kazumi, 2007; American Bureau of Shipping, 2011). Such technologies are highly effective for the removal of harmful organisms; however, the majority of these chemical methods generate relevant chemicals (RCs) in varying amounts (Banerji et al., 2012; Werschkun et al., 2012). It is essential that systems releasing toxic substances undergo risk assessment because exposure of aquatic organisms with the probability of adverse effects cannot be excluded. Such systems employing oxidative substances must be approved by Procedure G9 of the IMO, which requires aquatic environment risk assessment to verify the environmental acceptability of the ballast water treatment system (BWTS) (IMO, 2008a). The OECD-EU (Organization for Economic Co-operation and Development–European Union) commercial harbor and/or GESAMP-BWWG (Group of Experts on Scientific Aspects of Marine Environmental Protection–Ballast Water Working Group) harbor were used in the application dossiers to perform aquatic environment risk assessment. However, previous environmental risk assessments are limited in China because of a lack of risk assessment approaches and/or models, limited toxicological information regarding many RCs, and insufficient information pertaining to ports and harbors.

China is one of the top ten ocean transportation countries in the world; however, the issue of adverse effects to the aquatic environment posed by treated ballast water associated with the Chinese shipping industry has been grossly neglected. In this study, Tianjin Port on the northern coast of China was selected for investigation of ballast water discharge. This port was chosen because it is the largest comprehensive sea-port with the highest level of artificial deep-water in north China in terms of cargo handled. Additionally, it is a semi-closed and poorly flushed bottleneck port because it is surrounded by land on three sides. Taken together, these factors make Tianjin Port a worst-case scenario for estimating the persistence of RCs released from treated ballast water at discharge.

We previously developed a BWTS based on a hydroxyl radicals ($\cdot\text{OH}$) technique with biological treatment efficacy sufficient to meet the D-2 performance standard of the Ballast Water Convention (Bai et al., 2012). In addition, we conducted aquatic toxicity tests for aquatic organisms of three trophic levels to confirm that the effects of treated ballast water were acceptable when discharged (Zhang et al., 2012). However, no environmental risk assessment of $\cdot\text{OH}$ -based BWTS has been performed in any ports or harbors of China. Therefore, the present study was conducted to focus on RCs that are of high concern and required to be measured by the GESAMP-BWWG, including tribromomethane (TBM), trichloromethane (TCM), dibromochloromethane (DBCM), bromodichloromethane (BDCM), 1,2,3-trichloropropane (1,2,3-TCP), monobromoacetic acid (MBAA), dibromoacetic acid (DBAA), tribromoacetic acid (TBAA), monochloroacetic acid (MCAA), dichloroacetic acid (DCAA), trichloroacetic acid (TCAA), bromochloroacetic acid (BCAA), dibromoacetone (DBAN), bromate and sodium thiosulphate ($\text{Na}_2\text{S}_2\text{O}_3$) as a neutralizer in discharged ballast water. Predicted no effect concentrations (PNECs) were derived from short-term and/or long-term aquatic toxicity results for aquatic species by dividing the lowest available effect concentration by appropriate assessment factors (AF). Predicted environmental concentrations (PECs) of the measured RCs in Tianjin Harbor were then calculated using the MAMPEC (Marine Anti-foulant Model to Predict Environmental Concentrations) model with the conception of worst case emission scenario (e.g., the maximum concentration of each chemical detected among the samples). Finally, environmental risk assessment was performed by comparing PECs with PNECs.

2. Materials and methods

2.1. Study area

Tianjin Port is located on the northern coast of China, at $38^\circ 59' \text{N}$ and $117^\circ 42' \text{E}$. A map of the port is shown in Fig. 1. This port is the largest artificial sea-port in north China with cargo throughput of 477 million tons in 2012, making it the fourth largest in the world. Additionally, there are nearly 500 flights per month originating from the port and shipping to more than 500 ports worldwide. The port consists of four harbors and is equipped to handle multiple types of cargo including dry bulk, liquid bulk, containers and break bulk. This port is a semi-closed bottleneck port; therefore, its natural purification ability is very limited.

2.2. Experimental procedures

The schematic treatment process diagram for laboratory-scale BWTS with a flow rate of $10 \text{ m}^3/\text{h}$ is shown in Fig. 2. Natural seawater was collected from Tianjin Harbor, after which the microorganisms of interest (*Thalassiosira rotula*, *Skelrtonema costatum*, *Prorocentrum micans*, *Karenia mikimotoi* and *Heterosigma akashiwo*) representing five species from three different phyla/divisions with an initial algal cell density of approximately 1.0×10^4 cells/mL were added to simulate the influent ballast water. The initial salinity, temperature and pH of the influent ballast water were 29.7 PSU, 13.8°C and 8.37, respectively, and these parameters were measured each day during 5-days of storage.

The experimental procedures were described in our previous study (Zhang et al., 2012). Briefly, the influent ballast water was filtered to remove particles and organisms larger than $50 \mu\text{m}$, after which the water was subjected to an inactivation step employing a plasma reactor. A mixture of oxygen (99.5% purity) and water in the gas phase (3.5 vol.%) was allowed to run through the plasma reactor at a flow rate of 2.0 L/min to form chemically active species such as $\cdot\text{OH}$, $\cdot\text{H}$, H_2O_2 , O_2^+ and O_3 . These species were then mixed with incoming water via a venture ejector, which resulted in strongly active substances (mainly $\cdot\text{OH}$, $\text{HO}_2\cdot$, $\text{O}_2^{\cdot-}$, O_3 and H_2O_2) being produced immediately due to cavitation and shock waves. However, it is not convenient to detect these highly active species in-situ; therefore, active oxygen compounds combined with hypobromous acid (HOBr) and/or hypobromite (OBr^-) due to the reaction between highly active species and bromide ions in seawater were expressed as total residual oxidant (TRO), and measured online using a TRO analyzer during treatment. During de-ballasting, the filter and the plasma reactor were by-passed. Treated ballast water was neutralized using $\text{Na}_2\text{S}_2\text{O}_3$ upon discharge if the TRO level was greater than 0.2 mg/L. According to Guideline G8 of the IMO (IMO, 2008b), treated ballast water was stored in the tank for 5 days. Three sampling points were arranged as shown in Fig. 2. Control water samples (i.e., untreated) were collected from S.P.1, while treated water samples were collected from S.P.2 immediately after treatment. Samples of discharged water were collected from S.P.3.

2.3. Analytical methods

TRO was determined by a colorimetric DPD (N,N-diethyl-p-phenylene diamine) method based on US EPA method 330.5 (US EPA, 1978). An iodometric method was used to determine $\text{Na}_2\text{S}_2\text{O}_3$ in seawater. Ion chromatographic method performed on a ion chromatograph system (DIONEX ICS-1500, Thermo Fisher Scientific Inc., United States) was applied to determine bromate based on US EPA method 317.0 (US EPA, 2001). According to US EPA methods 524.2 and 552.3 (US EPA, 1995, 2003), TBM, TCM, DBCM, BDCM, DBAN, 1,2,3-TCP, and MBAA, DBAA, TBAA, MCAA, DCAA,

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