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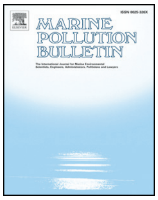

Focus

# Green oysters occurring in an industrial harbor in Central Taiwan

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## ABSTRACT

Green oysters were found within the Mailiao industrial harbor (MIH) located in Central Taiwan. The metal concentrations in seawater (dissolved and particulate phases), sediment and oyster samples collected within the MIH were analyzed to examine the green oysters. The dissolved and particulate metal concentrations ranged within 0.008–1.47 μg $l^{-1}$ and 0.97–799.2 mg $kg^{-1}$, respectively. The Cu concentration in seawater was dominated in the dissolved phase. In contrast, Cr, Pb and Zn in seawater were chiefly present in the particulate phase. The metal concentrations in sediment and oyster samples ranged within 0.02–148.4 mg $kg^{-1}$ and 0.18–1238 mg $kg^{-1}$ (dry W.), respectively. No anomalous values were found in the analyzed samples, except the Cu content in the oyster samples ranged within 274–1238 mg $kg^{-1}$. The relatively high bioconcentration factor of Cu in oyster and the longer exposure time induced the green oysters occurred within the MIH.

## 1. Introduction

The Mailliao Industrial Harbor (MIH) was constructed on artificial land, located in western central Taiwan at 120°8.9′ E/23°46.9′ N. This is the first privately run harbor in Taiwan. The local fisherman collected oysters within the MIH in October 2015, and found that some of the oysters were green, as shown in Fig. 1. The local fishermen were afraid that the MIH and its adjacent marine environment were seriously polluted by chemical compound discharge. However, the environmental monitoring program focused on the adjacent coastal water of the MIH did not show the anomalous values and the seawater quality were generally within the Taiwan EPA recommended quality criteria. A few exceptions of dissolved ammonia, phosphate and $BOD_5$ have been occasionally reported exceeding Taiwan's EPA guideline value. In spite of that, the As and Cr concentrations in sediments within the MIH also occasionally exceeded the effective range-low (ERL) biological effect guideline value (As, 8.2 mg $kg^{-1}$, Cr, 81 mg $kg^{-1}$; Long et al., 1995). (http://www.epa.gov.tw/np.asp?ctNode=32970&mp=epa). The MIH and its adjacent marine environment were not considered as the polluted environment. However, the green oyster was found within the MIH.

Green-colored oysters resulting from Cu contamination were reported more than a century ago (Wang et al., 2011). Green oysters have been reported in Taiwan since 1990 because the inshore area was seriously polluted by industrial discharges due to the substantial economic development during the beginning of 1970s (Han and Hung, 1990; Hung et al., 1993; Lin and Hsieh, 1999). The green oyster event was rarely reported in the last decade because the Taiwan government has paid a lot of attention to avoid the industrial wastewaters directly discharge into the marine environment and to reduce the pollution status along the Taiwan coast over the last two decades (Liu et al., 2015). The same problem as Taiwan is currently occurring in China due to the substantial economic growth over the past decades. Many studies have indicated that the green oysters are commonly found in the estuarine and coastal regions in the Southern China in this decade (Wang et al., 2011; Yu et al., 2013; Weng and Wang, 2014; Tan et al., 2015; Ji et al., 2015, 2016). Extremely high Cu and Zn concentrations, 19,000 mg $kg^{-1}$ and 23,000 mg $kg^{-1}$ (dry wt.), in the metal-contaminated oysters was found along the Jiulong River Estuary, Fujian Province. Surprisingly, no apparent signs of toxicity were observed in those green oysters (Weng and Wang, 2014). This is probably the highest Cu concentration in oysters in the existing literature. The green oyster events are seriously threatening the oyster farming which has been one of the largest coastal industries, with an annual production of around 1.0 million tons in recent years, in the Southern China (Yu et al., 2013).

Recently, the green oyster is rarely found in Taiwan. However, the green oyster was found within the MIH in recent. In order to understand the mechanisms causing the green oyster presence within the MIH and to eliminate the confusion of the local fisherman, seawater (including dissolved and particulate phase), sediment and oyster samples within the MIH were collected and analyzed for trace metals. The contents of trace metals in the particulate and sediment samples were chemically divided into the non-detrital and detrital fractions, with 1 N HCl acid, and aqua regia as well as HF acid, which are associated with the biological availability and non-availability, respectively. (Luoma, 1990). In addition, there are rarely reports focused on the survey of trace metals in

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**Fig. 1.** This green oyster (*Crassostrea gigas*) was collected from the Mailliao Industrial Harbor locating in the western central Taiwan.

the refinery harbor worldwide. The present work is also a pioneer study on trace metal contents in these components in the refinery harbor, such as the MIH in Taiwan.

## 2. Materials and methods

### 2.1. Study area

In 1995 the Mailiao Harbor Company was authorized to invest in, build and administer the MIH, which is the first privately run harbor in Taiwan. The total area, navigation depth and channel width in the harbor are approximate 1.594 ha, 24 m and 380 m, respectively. This harbor is designed with twenty berths dedicated to serve the Formosa Refinery Company, which has already been completed, with a further 10 berths planned to serve the shipping needs of nearby Yulin County. This harbor is run by the Formosa Refinery Company under the Mailiao Harbor Company, which holds administrative authority over the harbor. The Taiwan government supervises navigation and manages the port authority and other issues. The annual handled volumes topped 70 million metric tons in 2009.

The investigated results focused on the marine environment around the MIH area show that the dissolved concentrations of As, Cd, Cr(VI), Cu, Ni, and Pb, in seawater were generally lower than 1 μg $l^{-1}$. While, the higher dissolved concentrations of Fe and Zn, 5–10 μg $l^{-1}$, were occasionally determined during the winter season. The sediment grain sizes within the MIH were dominated by mud (<63 μm), accounting for >80% of the sediment weight. The very fine sand (63–125 μm) was the second important grain sizes. The concentration of total organic carbon (TOC) within the MIH sediment generally exceeded 0.5% due to the finer grain size. The total concentrations of sedimentary trace metals within the MIH were generally lower than the effective range-low (ERL) biological effect guideline value (Long et al., 1995), with the occasional exceptions of As and Cr concentrations exceeding the ERL value (http://www.epa.gov.tw/np.asp?ctNode=32970&mp=epa). It has been shown that total As concentrations in the estuarine and coastal sediments of the southwestern Taiwan commonly exceeded the ERL value due to the geologic sources (Hung et al., 2009). The survey results also indicate that the trace metal concentrations within MIH sediments generally correlated well with the TOC concentrations and with the finer grain size (http://www.epa.gov.tw/np.asp?ctNode=32970&mp=epa).

### 2.2. Sampling and analysis

Oyster (*Crassostrea gigas*) samples were collected at ten stations within the MIH by a MIH investigation vessel on 16 December 2015 (Fig. 2). The sampling stations were selected to encompass the MIH. Stations 1 and 2 are located in the investigation vessel berth where is the inside corner of the harbor and the seawater exchange here is probably the poorest within the MIH. Stations 4–8 are located in the oil tanker berth of the east bank and station 10 is located near the entrance of the harbor. The shell length of the collected oysters ranged within 4.5–10.5 cm, indicating that the growth age of oyster was estimated approximately 1.5–4 years (Status Review of the Easter Oyster, Crassostrea virginica, 2007). The color, shell length and the estimated growth age of oyster collected in each station are shown in Table 1. For logistical reasons water and sediment samples were collected from seven stations. The water samples in the surface layer (ca. 1 m depth) were collected at the studied stations using a Go-Flo bottle and stored in acid cleaned 1-liter Nalgene low density polyethylene bottles (LDPE). The LDPE bottles were rigorously cleaned by sequentially soaking them in a detergent solution and a mixture consisting of 50% (v/v) hydrochloric acid and 50% (v/v) nitric acid, prior to use. The sediment samples on the surface sediment were collected using grab catching at generally <5 cm depth. The sediment and oyster samples were stored in clean sealed PVC bags. All collected samples were stored in a cold-box and immediately sent back to the laboratory.

The water samples were filtered using an acid-cleaned Nuclepore membrane (0.45 μm) under a laminar flow hood prior to analysis. The filtered water sample was acidified with ultrapure nitric acid (J.T. Baker Ultrex Brand) 2 ml $l^{-1}$. The suspended particulate matter (SPM) loaded filters were rinsed and dried to constant weight in a flow hood at room temperature. The SPM was obtained from the difference in filter weight before and after filtration. The sediment samples for trace metal analysis were freeze dried using a low-temperature vacuum instrument. The dried sediment samples were ground with a mortar and pestle and stored in clean sealed PVC bags. The oyster samples were rinsed with the MQ water and dissected in the laboratory. The tissues were dried at 80 °C in an oven for about six days until they were dried.

The determination of dissolved trace metals in seawater were preconcentrated using the APDC-DDDC/Freon (ammonium pyrrolidine dithiocarbamate/diethyl diammonium dithiocarbamate) technique (Statham, 1985; Muller, et al., 1994; Fang and Lin, 2002). A two-step sequential extraction method was employed to analyze the trace metal concentrations in the particulate and sediment samples. The first step used 1 N ultrapure HCl acid to extract the weakly bound or non-detrital trace metals for 24 h at room temperature. Afterwards, the residual samples were digested with aqua regia and HF acid on a hot plate. The first step chemically attacks the trace metal speciation associated with the weakly bound or non-detrital trace metals which correlate well

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