

*Contamination of Marine Fish by Heavy Metals from Coastal Sewage Treated Effluent Runoff*

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Abstract

Arid regions with low rainfall are under continuous threat and pressure to maintain their stability and to meet public demands, specifically issues relating to the availability of fresh water. Due to the lack of rainfall and continuous population growth, many countries in the arid regions depend mainly on desalination of seawater and brackish water. Most countries in those regions have constructed sewage treatment plants (STP) due to the significant increase in sewage effluents. Many of the STPs constructed in the coastal area dump excess treated effluent into the sea. In this study, Oman was used as a model for the effect of the sewage effluent pollution. In 2006, it was reported that only 11% of sewage produce is recycled while the rest is unused. It is estimated that by 2035 the quantity of treated sewage effluent (TSE) will be more than 70 million m<sup>3</sup>. If the TSE contains pollutants, it will be disastrous to the environment. The main purpose of sewage treatment is to remove organic matter and microbial contaminants. However, heavy metals from industrial uses remain intact. In this investigation the marine fish around treated sewage dumping points were found to contain heavy metals. The highest heavy metals concentration in TSE was Ni followed by Cu, Mn, Fe, Co, Pb, and Zn. On the other hand, the dominant heavy metals in fish were Ni, Cu, Pb and Zn, although they were at permissible levels. If this trend continues the presence of heavy metals in fish will be a serious public health problem to human.

**Keywords:** Heavy metal, fish tissue, sewage treated effluent.

## INTRODUCTION

Heavy metal infiltration from sources such as treated sewage effluent (TSE) reaching the coastal region can contaminate marine wild life, mainly fish, since they are the major source of food in many coastal regions of the world (Bat *et al.*, 2012). In addition, sewage runoff and contaminated effluents were also the major sources of pollution in the terrestrial and aquatic environment affecting domestic animals and agricultural crops. Heavy metals and multiple antibiotic resistant bacteria (MARBs) were found to be released to the environment from TSE infecting agriculture crops, marine fish and sea turtles. In Oman, heavy metals and antibiotic resistant microbes are released to the environment, mainly through TSE (Al-Bahry *et al.*, 2007, 2009a 2009b, 2009c, 2011a, 2012a, 2012b; Al-Musharafi *et al.*, 2012, 2013a, 2013b; Mahmoud *et al.*, 2013).

Heavy metal contamination, including industrial waste products, mining, burning of fossil fuels, industry and the use of metals by consumers are the major sources of environmental pollution. In addition, heavy metal poor degradability results in the accumulation in water, sediments, soil and food chain (Svobodová *et al.* 1987). In terms of food safety, fish were used as indicators of pollutants, such as heavy metals and other toxic chemicals (Kenšová *et al.*, 2010).

Today, there is a steady increase in the usage of the heavy metals in industry inflicting a serious environmental problem and reaching toxic concentration levels in certain regions of the World (Güven *et al.*, 1999). Some of metals, such as Cu, Zn have essential physiological functions but they may accumulate reaching toxic levels (Al-Bahry *et al.*, 2011a, Hogstrand and Haux 1991; Rietzler *et al.*, 2001). Today, heavy metal accumulation in the industrial world can also affect aquatic environment which has become a serious problem, consequently a decline in fish population and quality (Holm *et al.*, 2002).

In addition, industrial waste and mining can also cause heavy metal pollution in the aquatic environment (Gumgum *et al.*, 1994). Heavy metals like Fe, Cu and Mn are essential in metabolic activity; however, Ar, Cd, Cr, Hg, Ni and Pb are usually toxic (Al-Bahry *et al.*, 2011a). Several studies reveal that there is a close association between MARBs and heavy metal accumulation which originate from several sources. The major source of such association is found in TSE, particularly in semiarid regions of the world where water shortage is one of the major problems. Researchers in Oman reported that the reuse of TSE specifically from industrial origin is becoming a major problem (Al-Bahry *et al.*, 2007, 2009a 2009b, 2009c, 2011b, 2012a, 2012b; Al-Musharafi *et al.*, 2012, 2013a, 2013b; Mahmoud *et al.*, 2013). The accumulation of heavy metals in various habitats is on the rise. Heavy metal discharge into the environment is very common in many regions in the world.

The aim of this study is to investigate the effect of heavy metal contamination in fish population in the coastal region of the Gulf of Oman. This investigation is essential since Oman is a major source of fishing industry. A gradual accumulation of heavy metals could affect both population and quality of fish. This condition will generate a major health and economic problems in the region.

## MATERIALS AND METHODS

### Study area

The study area is located in Muscat on the Gulf of Oman, near a treated sewage effluent which discharges its effluents directly into the sea.

## Sample collection

A total of 40 water samples (250 mL each) was collected from the TSE discharge point and immediately were stored in a cool box and transported to the lab immediately for further analysis.

The following fish species were sampled for heavy metal analysis (*Cephalopholis hemistiktos*, *Diodon liturosus*, *Lutjanus ehrenbergi*, and *Stephanolepis diaspros*) were collected from the dumping site because of their availability. A total of 40 fish was captured by line from the study area, stored in a cool box and transported to the lab. The weight ranged between 20–35 g ( $329.4 \pm 0.7$ ) and total length was 15–120 cm ( $13.1 \pm 1.35$ ).

## Sample preparation, analysis and quality control

Fish scales were removed and the fish were washed thoroughly with Milli-Q water. Tissue samples from gills, intestine, kidney, liver and muscles were dried in oven at 1000°C. The samples were grounded to powder for chemical analysis. Each sample (0.5 gm) was digested in a perchloric and nitric acid mixture in a Teflon beaker. Drops of 30% NaCl solution, 10 ml of 65% nitric acid and 70 % perchloric acid were mixed with fish powder. Each sample was placed at 70 °C water bath for 12 hrs to degrade the organic matter and to transfer the metals into the solution. The samples were centrifuged and the supernatant was analyzed (Kumar *et al.*, 2010).

Co, Cu, Fe, Mn, Ni, Pb and Zn concentrations were determined using Flame Atomic Absorption Spectrometry (Thermo, UK). Corrections of background were conducted by using standard addition method to compensate the matrix effects. For the precision of the instrument, standard references at six level dilutions were used to produce a calibration curve. All samples were analyzed in triplicates together with blank samples. Blank samples were used to check for cross contamination. The method accuracy was determined using reference material. Heavy metal concentrations were reported on dry weight mg/L. Heavy metal concentration in TSE was compared to the Minimum Permissible Levels (MPL) of the Omani Standard, which was adopted from FAO, for the waste water reuse and discharge (Table 1).

**Table 1.** Heavy metal standards of wastewater reuse in Oman (MEMWR, 1998).

Metal	Symbol	Minimum Permissible Levels (MPL)
Cobalt	Co	0.05
Copper	Cu	1
Iron	Fe	5
Manganese	Mn	0.5
Nickel	Ni	0.1
Lead	Pb	0.2
Zinc	Zn	5

## Statistical analysis

All data were statistically analyzed using SPSS statistical package. The *t* test at 5% levels of significance differences between the means was tested. The *p*-values of less than 0.05 indicated statistical significance.

## RESULTS

TSE heavy metal concentration varied in the seven metals. Co, Cu and Ni were at high concentrations but they do not vary significantly. While Mn concentrations were significantly higher than Fe and Zn ( $p < 0.05$ ). Pb was the lowest amongst the metals (Fig 1).

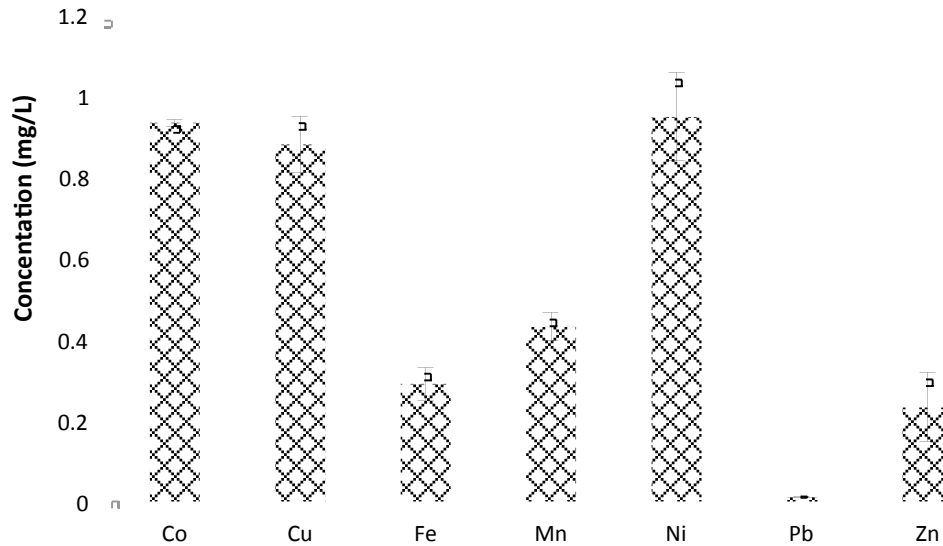


Fig 1. Concentration of heavy metal in TSE

Heavy metal concentration taken from sediment, Ni was significantly higher than the rest ( $p < 0.05$ ). However, Co concentration was next to Ni and was significantly higher than the rest of the heavy metals. Pb concentrations were the lowest, as in the case in TSE (Fig 2).

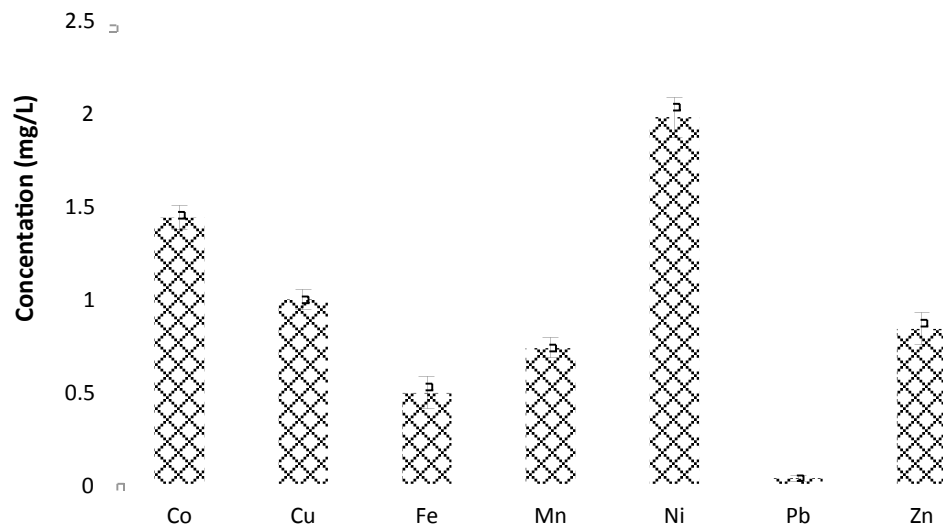
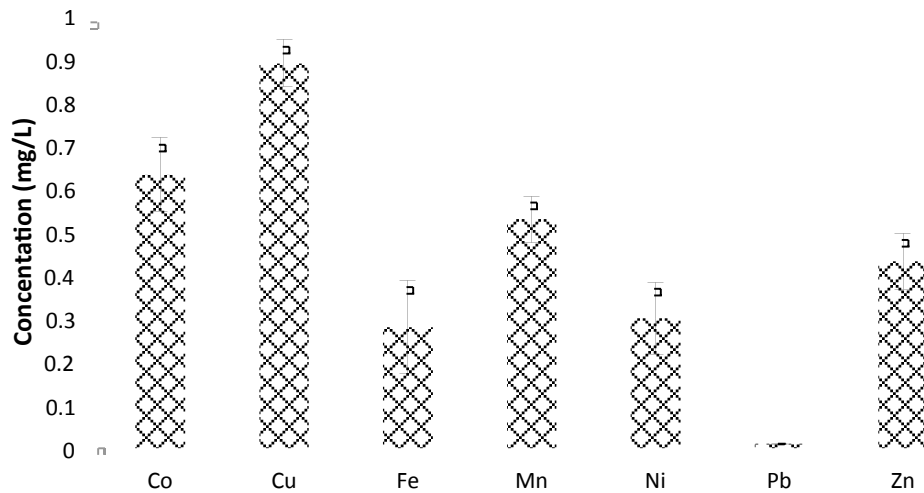


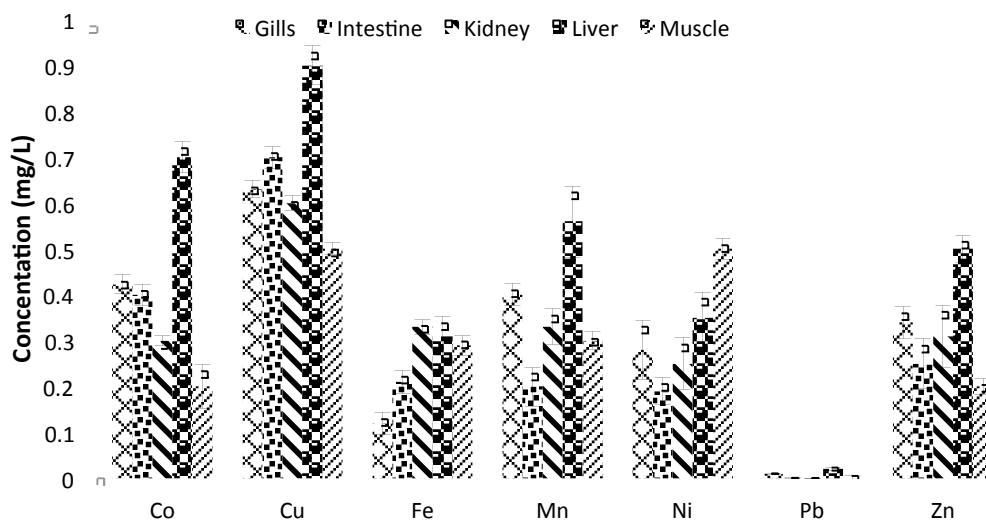
Fig 2. Concentration of heavy metal in sediment

Heavy metal concentrations of combined fish tissues taken from gills, intestine, kidney, liver and muscle varied. Cu concentrations were the highest ( $p < 0.05$ ) followed by Co, Mn, Zn and Ni. Pb concentrations, as in the case of TSE and sediment were the lowest among the group (Fig 3).



**Fig 3.** Total heavy metal concentration in fish tissues (gills, intestine, kidney, liver and muscle).

Heavy metal concentrations from gills, intestine, kidney, liver and muscle were analyzed (Fig 4). There is no specific concentration pattern for all the metals taken from five organs. However, liver has the dominant metal concentrations over the rest of the organs except for Ni. Pb was the lowest in all organs. In general, Co, Cu, Ni concentrations were higher than the rest in both TSE and sediment. In the combine fish tissues, taken from gills, intestine, kidney, liver and muscle, Cu was the dominant heavy metal. In the individual tissues Cu was the dominant. Looking at the individual organs, liver was the dominant in containing most of the heavy metals followed closely by gills, kidney, intestine and muscle.



**Fig 4.** Heavy metals concentration in different fish organs.

## DISCUSSION:

The heavy metal accumulation in sediment and aquatic, both fresh water and salt water habitats, is becoming a serious problem in Oman. One of the reasons for such accumulation is the reuse of TSE from industry which is not entirely removed from toxic heavy metals (Al-Musharafi *et al.*, 2012).

Heavy metals are naturally found in marine environment at certain concentrations. However, domestic, industrial, agricultural and mining activities have resulted in increasing concentration of toxic metals affecting marine habitats (Bat *et al.*, 2012).

The main purpose of this investigation is to analyze the heavy metal concentrations in three different ecological niches in order to compare the degree of heavy metal infiltration and concentration in the sampling areas. The sampling areas were chosen because TSE dumping site is located on the shoreline contaminating aquatic environment including sediment and fish.

The most significant aspect in this investigation is that the fish samples were contaminated with the same heavy metals which were found in the TSE and the sediment. In addition, the toxic heavy metals found in the TSE, sediment and fish had similar concentration patterns throughout the study period. Based on this data it is crucial to assure that sewage effluent does contain heavy metals even after treatment. This indicates that TSE is one of the sources of heavy metal contamination to the environment.

During the last few years in Oman, there is a steady increase in toxic heavy metals such as Hg, Zn, Pb, Sn, and Co (Al-Musharafi *et al.*, 2012, 2013a, 2013b). Although Co is essential for vitamin B12, it can be toxic at high concentrations. On the other hand, Hg, Pb are nonessential metals (Al-Rawahy *et al.*, 2007). Due to slow process of elimination and by the ageing process these metals have the ability to bind and bioaccumulate in tissue (Harrison, 2001; Nigro and Leonzio 1996). Some of the metals have a tendency to bind to proteins (Shahidul and Tanaka, 2004).

In this study, the level of heavy metals in fish was slightly lower than the sediments. However, others reported that the level of heavy metals in marine fish were many times higher than in sediments (Bat *et al.*, 2012; Boran and Altinok, 2010). Probably the site of the present study is less contaminated compare to the others.

In conclusions, based on the data from this research there is heavy metal contamination in the marine environment originated from TSE. Efficient methodologies for removing toxic heavy metals from sewage effluents are urgently needed before releasing the effluent to the environment to avoid environmental pollution. Periodic examinations of wild life such as fish must be implemented before fish consumption which may have impact on public health.

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