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Modelling environment contamination with heavy metals in flathead grey mullet *Mugil cephalus* and upper sediments from north African coasts of the Mediterranean Sea

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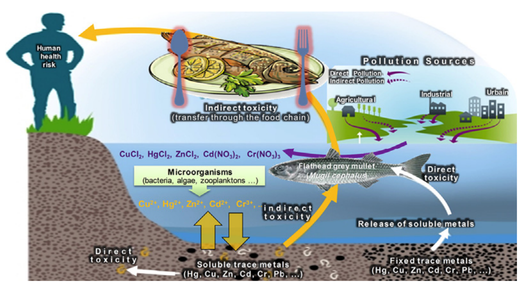
HIGHLIGHTS

- The contamination of Annaba Gulf by trace metals (TM) is linked to human activities.
- TM accumulation in muscles affects the weight growth of *M. cephalus*.
- Sediment-muscle relationships of TM follow logistic regressions, but a linear in Zn.
- The consumption of *Mugil cephalus* is dangerous due to the high levels of Pb, Zn and Cd.
- Sediments and *M. cephalus* are good indicators of TM pollution in coastal habitats.

GRAPHICAL ABSTRACT



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ABSTRACT

Heavy metals are a serious hazard for aquatic ecosystems and human health. They negatively affect aquatic life functioning through accumulation resulting physiological/growth disturbances in aquatic lifeforms. This survey focused on the assessment of heavy metal pollution in the Gulf of Annaba (northeastern Algeria), the largest and most diversified industrial hub in Africa, using a multi-compartment approach (water–sediment–biota). The study aims to characterize the spatiotemporal variation of trace metal (TM) contamination and its effects on the growth of the Flathead grey mullet (*Mugil cephalus*). It reviewed TM concentrations in upper sediments and organs of *M. cephalus* from various hydrosystems worldwide. Five sites distributed along the Gulf were sampled to determine water physicochemical parameters as well as the contamination of surficial sediments and muscles of *M. cephalus* by zinc, copper, lead, cadmium and mercury. The spatiotemporal variations of the measured parameters were tested and discussed following the synergetic effects of water, sediment and muscle variables on fish biometrics. The sediments at the Port, Joinoville and Sidi-Salem sites were classified as heavily polluted by lead, copper, zinc and cadmium, whereas only at the Port by mercury. Muscular lead concentrations exceeded international standard values in Joinoville and Port, and zinc in Port. The increase of water dissolved oxygen induced a significant decrease in sediment TM. The increase of sediment TM caused a significant increase in muscle TM levels. The S-shaped logistic models indicated that muscle contaminations reached a saturation plateau following the current sediment pollution. TM concentrations in fish muscles negatively affected fish weight,

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but only copper and cadmium significantly influenced fish length. The consumption of fish from the Port, Joinoville and Sidi-Salem can be dangerous because concentrations of lead, zinc and cadmium exceeded the international standards. This study validates the effectiveness of biomonitoring using *M. cephalus* as bioindicator in polluted coasts.

1. Introduction

For more than half century, pollution has been one of the most serious problems threatening human well-being and the integrity of ecosystems of our planet (Langston, 1990). Pollution is commonly defined as the presence in the environment of dangerous manmade chemical products with harmful effects that can last long periods (Gochfeld, 2003). The aquatic ecosystem is increasingly threatened by various sources of pollutants that may reduce its economic potential, disrupt its ecological functioning and consequently generate negative repercussions on human health (Papagiannis et al., 2004; Yakimov et al., 2007). During the last decade, the fight against water pollution represents the core of global discussions and debates (Tarras-Wahlberg et al., 2001; Cheng, 2003; Malik et al., 2010; Chasek et al., 2016). Heavy metals are among the chemical substances that constitute a serious environmental and health hazard to living beings specifically in aquatic environments (Wu et al., 2013a, 2013b; Khemis et al., 2017). These elements are very toxic and are increasingly used in industrial and agricultural sectors (Bisone, 2012; Chang et al., 2014; Pradhan and Kumar, 2014). Some non-toxic metals become hazardous because of their bio-accumulative capacity and their persistence in the environment and in particular in hydrosystem sediments, given their non-biodegradation (Larrose et al., 2010; Diop, 2014; Saher and Siddiqui, 2016). The accumulation of metal residues in aquatic producers and micro-organisms affects food webs, and may triggers toxicity at various trophic levels, which increases the vulnerability of flora and fauna and concerns ultimately human health (Türkmen et al., 2008; Yi et al., 2011; Medeiros et al., 2012).

Heavy metals from natural or anthropogenic sources released to an aquatic environment are largely trapped in sediments (Sin et al., 2001). Sediments are often used to assess the degree of pollution in aquatic environments (Pekey, 2006; Buggy and Tobin, 2008; Belabed et al., 2017). Seabed sediments are known for their ability to accumulate trace metal elements (TM) introduced into seas and oceans via direct, industrial and/or urban tributaries, fluvial and atmospheric inputs, and soil leaching (Belabed et al., 2013a; Diop et al., 2014a). Therefore, they play the role of a real carrier of these metals to aquatic organisms (Türkmen et al., 2008; Yildirim et al., 2009; Diop et al., 2015). The negative and toxic effects of TM on aquatic organisms influence various physiological functions that result in decreased rate of individual growth and reproduction, and increased mortality rate (Canli and Atli, 2003; Farkas et al., 2003; Bird et al., 2008). TM can enter fish bodies via three possible ways: through body surface, gills, or digestive tract by three sources: water, food and sediment (Ben Ameur et al., 2012).

Nowadays, seas and oceans receive large volumes of pollutants such as heavy metals and pesticides from various sources such as industrial and urban wastewater discharges (Gueddah, 2003; Tessier, 2012; Diop, 2014; El-Zrelli et al., 2015; Belabed et al., 2017). Semi-enclosed seas, like the Mediterranean Sea, are more sensitive to the impacts of pollution when the ratio of coastline length to sea surface is high and the renewal of water masses is slow (Pekey et al., 2004). Moreover, because coastal areas are generally highly urbanized and industrialized, they are therefore subject to anthropogenic TM discharges, sometimes at large scale (Pekey, 2006; Waltham et al., 2011).

The Gulf of Annaba is one of the most important touristic and economic centers on North African and Algerian coasts. The pollution related to a booming economic activity threatens fisheries resources of the region, which become the receptacle for all toxic residues produced by different industrial units located along the coast (Belabed et al., 2013b, 2017). The choice of this region as a study area is motivated by the socioeconomic importance of its ecosystems at national and regional scale and the intense human pressure exerted on its ecosystems in terms of pollution and degradation.

Fish are widely used as sentinel species of metallic contamination in aquatic environments (Chen, 2002; Yilmaz, 2003; Uluturhan and Kucuksezgin, 2007; Diop et al., 2016a, 2016b). Thus, the present study aims to evaluate the degree of metallic contamination in sediments of the Gulf of Annaba using the contamination index and through a biological indicator i.e. Flathead grey mullet *Mugil cephalus* L. (Pisces: Mugilidae); given its high capability to concentrate TM and its wide use as a good indicator of metallic pollution (Omar, 2013; Fazio et al., 2014; Khemis et al., 2017). Fish attract the scientific attention in assessing the health of aquatic habitats because they are at the top of trophic chains and can therefore directly affect human health (Bervoets et al., 2005; Yilmaz, 2009). Muscle is usually analyzed during biomonitoring surveys as it is the main part of fish consumed by humans and is involved in health risks (Stancheva et al., 2013). The study fish species, the Flathead grey mullet, is a pelagic omnivorous species, in close relationship with sediment, characterized by low mobility and its diet rich of algae, polychaetes, crustaceans, gastropods and small fish (Yilmaz, 2009; Stancheva et al., 2013).

This study complements and adds new insights to previous works carried out in the region to deepen our understanding of the effects of pollution on North African and Mediterranean aquatic ecosystems (Abdennour et al., 2000; Gharsallah, 2005; Belabed et al., 2008; Ouali et al., 2008; Belabed et al., 2013b, 2017; Boucetta et al., 2016). The pollution problem of the Gulf of Annaba is approached and discussed by answering the following questions: (i) do water characteristics influence TM concentration in the upper sediment? (ii) are water characteristics and sediment TM the determinants of TM concentration in fish muscle? (iii) are fish biometrics (length and body weight) influenced by water parameters, sediment TM concentrations, and fish muscle contamination? We also test the hypothesis whether TM concentrations are length/weight-dependent and investigate through a statistical modelling approach the relationship between sediment TM concentrations vs. muscle TM concentrations. In addition, this study compares metal contamination recorded in the Gulf of Annaba with the reviewed and documented metal concentrations in upper sediments from various hydrosystems worldwide. It also reports and reviews all studies at global scale that measured heavy metal concentrations in different organs of the Flathead grey mullet (brain, gills, gonads, liver, muscle, and skin), which facilitate the determination of muscle contamination status in the study area compared to food guidelines and anterior surveys and then decide whether fish consumption poses risks to human health or not.

2. Materials and methods

2.1. Study area

Located at the extreme eastern part of the Algerian coast, at 600 km from Algiers and 100 km from the Tunisian border, the Annaba Gulf is a