Environmental Pollution 230 (2017) 311-319

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Organochlorine pesticide, antibiotic and heavy metal residues in mussel, crayfish and fish species from a reservoir on the Euphrates River, Turkey^{*}

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ARTICLE INFO

Article history: Received 30 March 2017 Received in revised form 26 May 2017 Accepted 19 June 2017 Available online 28 June 2017

Keywords: Organochlorine pesticides Antibiotics Heavy metals Biota Keban dam reservoir Food safety

ABSTRACT

This study investigated the residues of 19 organochlorine pesticides, 37 antibiotics and 5 heavy metals in biota samples (one mussel species, one crayfish species, six wild fish species and one farmed fish species) from the Keban Dam Reservoir on the Euphrates River, Turkey. Among monitored OCPs, only p,p'-DDE was detected in biota samples. It was found only in mussel, fish muscle and fish gill samples. The highest concentration of p,p'-DDE (0.032 mg/kg ww) was determined in the gill of common carp, which was found to be below the maximum residue limit (MRL). Only sulfadimethoxine was detected among antibiotics in biota samples. Its maximum concentration (0.0044 mg/kg ww) did not exceed the MRL of 0.1 mg/kg. Sulfadimethoxine was found only in muscle and gill of common carp collected from site S6, where there are many rainbow trout cage farms. However, no detectable residue of sulfadimethoxine was found in farmed rainbow trout. The highest concentrations of As, Cd and Pb were detected in mussels, while the highest concentrations Cu and Zn were determined in crayfish. Metal concentrations in biota samples did not exceed the maximum permissible levels. According to these results, the consumption of biota samples from the Keban Dam Reservoir is safe for human health.

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1. Introduction

The use of organochlorine pesticides (OCPs) as insecticides and biocides has been widespread in agriculture and public health since the 1940s. However, these compounds have become a major environmental problem due to their long persistence, bio-accumulation and adverse effects on animals and humans (Yohannes et al., 2014; Wang et al., 2015; Kalyoncu et al., 2009; Dang et al., 2016). In aquatic environments, OCPs easily enter the food chain due to their hydrophobic and lipophilic properties, finally reaching humans by consumption of edible aquatic organisms. OCPs may act as endocrine disruptors and cause reproductive, developmental and growth abnormalities in aquatic organisms, wildlife and humans (Dang et al., 2016). Several of them are also potentially carcinogenic and mutagenic (Wang et al., 2015). Therefore, the use of the majority of OCPs was banned in most

western countries. Turkey also banned the use of OCPs in 1983 (Kalyoncu et al., 2009). However, OCPs can remain in aquatic environments over the course of many years due to their high environmental persistence. In addition, these pesticides are still used in some developing countries, for example, in Ghana and China (Li et al., 2016). Therefore, their residues and metabolic forms are still detected in aquatic biota (Wang et al., 2015; Yohannes et al., 2014).

Antibiotics are natural or synthetic origin drugs that have the ability to either kill or inhibit the growth of bacteria. They are commonly used in humans and animals to prevent and treat diseases, some of which are also used as growth promoters for farm animals (Zhao et al., 2015; Xu et al., 2014). The continuous use of antibiotics may promote antibiotic-resistance genes in bacterial populations in the environment, among humans, and farm animals (Rico et al., 2012). Furthermore, antibiotic residues in farm animals could pose a risk to those people who eat them (Chen et al., 2015).

In recent years, antibiotics have increasingly received attention due to their negative effects on humans, animals and ecosystems. Antibiotics are released into the aquatic environment from several sources. The majority of antibiotics is sewage derived, and they





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 $^{^{\}star}\,$ This paper has been recommended for acceptance by Maria Cristina Fossi.

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enter the aquatic environment through wastewater discharges (Xu et al., 2014). Antibiotics that are used for animals reach the aquatic environment via application of manure to fields as fertilizer. Surface runoff from agricultural fields is likely to contribute to the release of antibiotics to the aquatic environment. Aquaculture is another source of antibiotics entering the aquatic environment (Xu et al., 2014). In fish farms, antibiotics are used mainly for both therapeutic and prophylactic purposes. Thus, antibiotics used in fish farms may enter the aquatic environment and accumulate in the food chain (Kim and Carlson, 2006). In addition, antibiotic residues from the excessive use of antibiotics in fish farms can remain in the tissues of farmed fish, thus causing possible adverse health effects in humans (Chen et al., 2015).

Heavy metals are among the most important pollutants in the aquatic ecosystems because of their environmental persistence, adverse effects on organisms, bioaccumulation and biomagnification properties. Heavy metals accumulated in aquatic biota may cause negative health effects on humans when consumed at higher rates than maximum allowable consumption rates (Bosch et al., 2016; Ahmed et al., 2015; Tao et al., 2012). Therefore, in recent years, there has been increasing awareness within the scientific community to know the human health risks derived from the consumption of aquatic biota exposed to heavy metals (Griboff et al., 2017).

The accumulation of OCPs, antibiotics and heavy metals in different aquatic animals depends on their habitat and feeding habits. Mussels, crayfish and fish are reliable bioindicators for pollution monitoring because these aquatic animals concentrate these contaminants in their tissues directly from water and also through diet (Fisk et al., 2001; Lana et al., 2010). Therefore, data on the presence and distribution of OCPs, antibiotics and heavy metals in edible mussels, crayfish and fish species are important both from ecological and human health perspectives (Kalyoncu et al., 2009; Munaretto et al., 2013), because the consumption of contaminated mussels, fish and crayfish is a major pathway for transfer of OCPs, antibiotics and metals into humans (Dang et al., 2016).

There are many studies on the organochlorine pesticide contamination in aquatic organisms in many countries (Barnhoorn et al., 2015; Yohannes et al., 2014; Covaci et al., 2005; Wang et al., 2015; Rose et al., 2015; Eqani et al., 2013; Veljanoska-Sarafiloska et al., 2013; Peterson and Boughton, 2000; Gitahi et al., 2002). However, only some reports are available on the OCP contamination in aquatic organisms collected from Turkish waters (Barlas, 1999; Erdogrul et al., 2005; Kalyoncu, 2009; Aksoy et al., 2011). A large part of these studies performed in Turkey and other countries indicated that aquatic organisms were contaminated by the OCPs. Investigations on antibiotic residues in aquatic organisms collected from nature are very few in number (Li et al., 2012; Zhao et al., 2015; Gao et al., 2012). However, some investigations have been performed on the presence of antibiotics in farmed aquatic organisms because of the common use of antibiotics in aquaculture production (He et al., 2016; Kim et al., 2017; Done and Halden, 2015; Choi et al., 2016). These investigations revealed that some antibiotics were detected in aquatic organisms, especially in farmed aquatic products. Studies on heavy metal contamination in aquatic biota in Turkey and many other countries are available in plenty (Türkmen and Ciminli, 2007; Kuklina et al., 2014; Cheng et al., 2013; Djedjibegovic et al., 2012; Mendil et al., 2010; Alamdar et al., 2017; Ahmed et al., 2016; Rahman et al., 2012; Alipour et al., 2015). These studies reported that heavy metal contamination in aquatic biota is a major problem in many countries due to rapid industrial development, and the increased population and urbanization.

Keban Dam Reservoir (KDR), one of the most important dam reservoirs on the Euphrates River in Turkey, has a significant potential in terms of fisheries and aquaculture production. Although some reports have been published on the heavy metal levels in mussels, crayfish and fish from the KDR, there has not been any study on organochlorine pesticide and antibiotic residues in mussels, crayfish and fish from the KDR. The main objectives of the present study were to determine residue levels of OCPs, antibiotics and heavy metals in mussel, crayfish and fish species in the KDR in order to compare concentrations of OCPs and antibiotics in the different tissues (gill, gonad and muscle) of fish, and to assess the human health risks associated with biota consumption. For these purposes, in this study, residue levels of 19 OCPs, 37 antibiotics and 5 heavy metals were investigated in one mussel species, one crayfish species, six wild fish species and one farmed fish species from the KDR.

2. Materials and methods

2.1. Study area

The Keban Dam Reservoir (KDR) was formed on the Euphrates River in eastern Anatolia. It is the second largest reservoir in Turkey. Located between latitudes 35°20′ and 38°37′ N, and longitudes 38°15′ and 39°2′ E, the KDR has a surface area of 675 km² and a volume of 30.6 km³ at 845 m above sea level (Fig. 1). The KDR has a significant potential in terms of fisheries and aquaculture production. Rainbow trout have been cultured in net cages on the dam reservoir since 2000 (Güner, 2015).

2.2. Mussel, crayfish and fish collection

In this study, one mussel species, one crayfish species, six wild fish species and one farmed fish species were collected from sampling sites in May 2015. The collected mussel species in the study included *Unio elongatulus eucirrus*, crayfish species included *Astacus leptodactylus*, wild fish species included common carp (*Cyprinus carpio*), mangar (*Luciobarbus esocinus*), trout barb (*Capoeta trutta*), Tigris scraper (*Capoeta umbla*), Euphrates barbell (*Luciobarbus mystaceus*) and escaped rainbow trout (*Oncorhynchus mykiss*), and farmed fish species included farmed rainbow trout (*Oncorhynchus mykiss*) (Table S1).

In the present study, six sampling sites were selected (S1-S6) on the Keban Dam Reservoir (Fig. 1). Detailed information describing the sampling sites is given in Table S2. It was planned to collect 4–8 individuals per species per site. However, the planned species and individual numbers were not met at all sites: crayfish, common carp, mangar, trout barb and Euphrates barbell were caught at all sites, mussels at sites S1-S5, Tigris scraper at sites S1-S5, escaped rainbow trout at sites S5 and S6. Farmed rainbow trout were taken from three different cage farming facilities (sites S4-S6) on the KDR (Table S3).

At sampling sites, crayfish were caught by fyke nets, while wild fish and escaped rainbow trout were caught by using gill nets. Mussels were collected by hand. Soon after collection, all samples were transferred to the laboratory on ice boxes within 5 h. Total lengths and body weights of mussels, crayfish and fish were measured (Table S4). The soft tissues of mussels were dissected from shells. Crayfish were dissected to obtain abdominal muscle tissues. Fish were dissected to obtain muscle, gill and gonad. A composite sample (consisting of 2–4 individuals) for each target tissue type of each species from each site was prepared. In the study, OCP, antibiotic and heavy metal analyses were performed only in soft tissues of mussel and muscle tissues of crayfish samples. In fish samples, OCP and antibiotic analyses were performed in gill, gonad and muscle, while metal analyses were performed only in muscle tissues of them (Table S3). For OCP analyses, 3 mussel samples, 4 crayfish samples, 15 fish muscle samples, 7 fish gill

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