

## Seasonal Monitoring of Cu and Zn in the Sewage Sludge of Malatya Advanced Biological Wastewater Treatment Plant

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### Abstract

In this study, Cu and Zn concentrations were monitored for 12 months in sewage sludge from the Malatya Advanced Biological Wastewater Treatment Plant (MABWTP). The obtained data were evaluated both monthly and seasonally and compared with the standard values given in the Regulation on the Use of Domestic and Urban Sewage Sludge in Soil. The highest Cu and Zn concentrations in sewage sludge from the MABWTP were  $181.74 \pm 9.0$  mg kg<sup>-1</sup> and  $653.8 \pm 32.6$  mg kg<sup>-1</sup> in September and the lowest Cu and Zn concentrations were  $103.69 \pm 5.18$  mg kg<sup>-1</sup> and  $436.2 \pm 21.8$  mg kg<sup>-1</sup> in December and March. The highest concentrations of Cu and Zn in the sewage sludge from MABWTP were found to be  $167.21 \pm 8.3$  mg kg<sup>-1</sup> and  $611.80 \pm 30.5$  mg kg<sup>-1</sup> in the summer and autumn, and the lowest Cu and Zn concentrations were  $109.39 \pm 5.4$  mg kg<sup>-1</sup> and  $440.13 \pm 22$  mg kg<sup>-1</sup> in the winter season. When the concentrations of Cu and Zn in the MABWTP treatment sludge were examined, it was found that they were lower than the limit values given in the regulation. As a result, it was determined that sewage sludge taken from MABWTP could be used for soil in terms of Cu and Zn concentrations.

**Keywords:** Heavy metal, legislation, monitoring, sewage sludge, treatment plant

## Malatya İleri Biyolojik Atıksu Arıtma Tesisi Arıtma Çamurunda Cu ve Zn'nun Mevsimsel İzlenmesi

### Öz

Bu çalışmada, Malatya İleri Biyolojik Atıksu Arıtma Tesisi'nden (MİBAAT) atılan arıtma çamurlarında Cu ve Zn konsantrasyonları 12 ay boyunca izlendi. Elde edilen veriler hem aylık hem de mevsimsel olarak değerlendirildi ve Evsel ve Kentsel Arıtma Çamurlarının Toprakta Kullanılmasına Dair Yönetmelikte verilen standart değerlerle karşılaştırıldı. Bu çerçevede, MİBAAT'nden atılan arıtma çamurlarında en yüksek Cu ve Zn konsantrasyonları Eylül ayında  $181.74 \pm 9.0$  mg kg<sup>-1</sup> ve  $653.8 \pm 32.6$  mg kg<sup>-1</sup> olarak, en düşük Cu ve Zn konsantrasyonları ise Aralık ve Mart ayında  $103.69 \pm 5.18$  mg kg<sup>-1</sup> ve  $436.2 \pm 21.8$  mg kg<sup>-1</sup> olarak tespit edildi. MİBAAT'nden atılan arıtma çamurunda en yüksek Cu ve Zn konsantrasyonları yaz ve sonbahar mevsiminde  $167.21 \pm 8.3$  mg kg<sup>-1</sup> ve  $611.80 \pm 30.5$  mg kg<sup>-1</sup> olarak, en düşük Cu ve Zn konsantrasyonları kış sezonunda  $109.39 \pm 5.4$  mg kg<sup>-1</sup> ve  $440.13 \pm 22$  mg kg<sup>-1</sup> olarak tespit edildi. MİBAAT arıtma çamurunda Cu ve Zn konsantrasyonları incelendiğinde yönetmelikte verilen sınır değerlerden düşük değerler aldığı tespit edildi. Sonuç olarak, Cu ve Zn konsantrasyonları açısından MİBAAT'nden atılan arıtma çamurlarının toprakta kullanılabileceği belirlendi.

**Anahtar Kelimeler:** Ağır metal, mevzuat, izleme, arıtma çamuru, arıtma tesisi

### INTRODUCTION

Sewage sludge is generated by metabolism of microorganisms in municipal wastewater treatment. It contains plenty of organic matters and is rich in nutrients. It could be used as organic amendment to improve soil fertility (Zhang et al., 2018). The addition of sewage sludge in the soil has beneficial effect both on soil (biological, physical, chemical properties) and on crop (growth and yielding) (Rehab et al., 2003; Vaca et al.,

2011; Ailincăi et al., 2012). However, the sewage sludge includes heavy metals. Using sewage sludge for fertilization can cause an increase in heavy metals content in the soil (Sobik-Szołtysek et al., 2017).

Heavy metals are defined as elements with a density exceeding  $5 \text{ g cm}^{-3}$  (Arévalo-Gardini et al., 2017). Heavy metals are environmental toxins which harm the environment even at low concentration. They are persistent environmental

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contaminants (Gouda and Al Ghannam, 2016) and can accumulate in the food chain (Ahmad et al., 2015).

Heavy metal accumulation in the terrestrial environment has become a global problem (Xiao et al., 2017). Anthropogenic contributions to the heavy metal concentration in soil include past and current application of various soil conditioners (e.g. composts, manures and fertilisers, agrichemicals which contain metal, paints, emissions from vehicles, industries, and coal and fuel combustion (Paramashivam et al., 2016; Simmler et al., 2013; Szolnoki and Farsang, 2013; Ashrafzadeh et al., 2018). When excess amounts of metals enter the pedosphere, soil quality decrease which is followed by a reduction in soil productivity and food security (Wuana and Okieimen, 2011; Alloway, 2013; Xiao et al., 2017). Toxic metal ions can eventually reach the top of food chain and thus, become a risk factor for people's health (Katircioğlu et al., 2008). Heavy metals can be accumulated through ingestion of soil particles, dermal contact with soil particles or ground water, consumption of food and water, and inhalation of soil particles from the air (Liu et al., 2013; Li et al., 2014; Xiao et al., 2017). Children are particularly vulnerable because they have greater hand-to-mouth activities and gastrointestinal absorption (Calabrese et al., 1997; Ashrafzadeh et al., 2018). Zinc (Zn) is an essential trace element. It plays roles in all replications and acts as a cofactor in enzymes. The disturbances of Zn homeostasis can be associated with diseases (e.g. diabetes mellitus, cirrhosis of the liver, tumors). High concentrations of Zn can cause damages to human body (disturbances in energy metabolism or increase in oxidative stress and disruption in the homeostasis of other essential elements) (Samadi-Maybodi and Rezaei, 2014). The plants grown in soils which have high concentrations of copper (Cu) and Zn may accumulate these heavy metals in tissue resulting in toxicity symptoms (Kabata-Pendias, 2011; Tiecher et al., 2017). Excess Cu affects the function of membrane transporters and ion channels. Excess Cu and Zn may cause oxidative stress and negatively affect photosynthetic efficiency (Tiecher et al., 2017). Because of the above-mentioned problems, Zn and Cu have to be monitored in the sewage sludges which could be used in soil.

There is legislation in our country regarding the use of sewage sludge from domestic and urban wastewater treatment plants in the soil. The Regulation on the Use of Domestic and Urban Sewage Sludge in Soil (RUDUSS) came into force in 2010 by being published in the Official Gazette (date:03.08.2010 and no:27661). The maximum Cu and Zn concentrations allowed in the stabilized sewage sludge that can be used in the soil are given in Table 1.

**Table 1.** Maximum allowable concentrations in the sewage sludge

Heavy Metals	Limit Values (mg kg <sup>-1</sup> )
Copper (Cu)	1 000
Zinc (Zn)	2 500

In this study, it is aimed to determine and monitor seasonally the Cu and Zn concentrations in the sewage sludge of Malatya Advanced Municipal Wastewater Treatment Plant (MABWTP). In this frame, the obtained data were compared with the limit values given in the RUDUSS published in the Republic of Turkey. Also, the environmental importance in terms of Cu and Zn concentrations of the treatment sludge has been revealed.

## MATERIAL AND METHOD

Sewage sludge samples used as material in this study were obtained from MABWTP (Figure 1). MABWTP was established on an area of 183 923 m<sup>2</sup> with the aim of the treatment of the domestic wastewaters of Malatya central districts Battalgazi and Yeşilyurt. The project has been designed in three stages: 135 000 m<sup>3</sup> day<sup>-1</sup> for 2010, 180 000 m<sup>3</sup> day<sup>-1</sup> for 2020 and 220 000 m<sup>3</sup> day<sup>-1</sup> for 2030. The treatment plant was completed and put into operation in April 2004. MABWTP consists of pre-treatment, biological treatment and sludge removal units. The wastewater is taken to the treatment plant by two separate collectors. The west main collector has a diameter of 2 400 mm and the east main collector has a diameter of 1 000 mm.



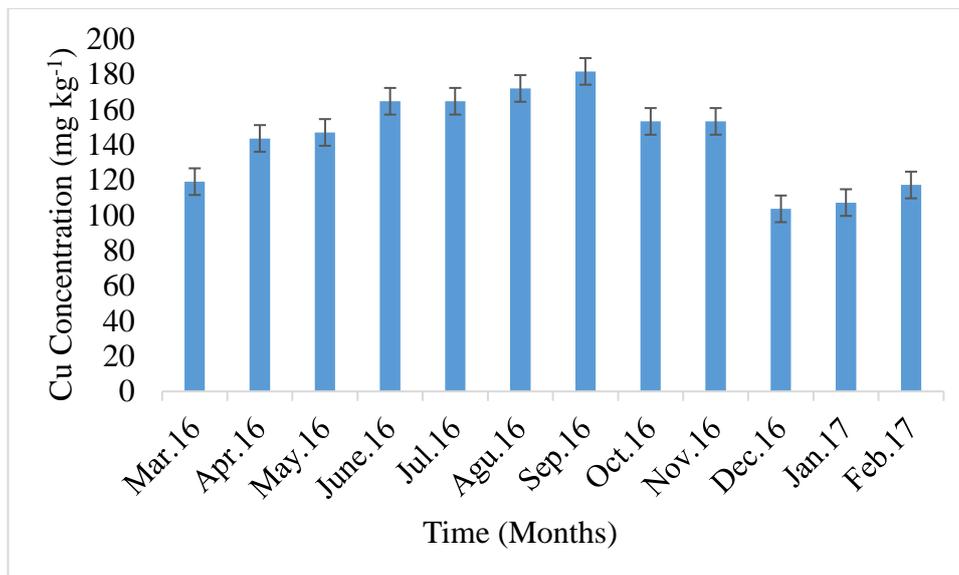
**Figure 1.** Overview of MABWTP (URL-1)

The wastewater is passed through the coarse grid at the entrance of the treatment plant to protect further equipment (entrance pumps). The wastewater is promoted with the aid of an inlet pump to allow attractive flow through the plant, and then the floating materials and coarse particles in the wastewater are held with fine grids and sent to the sand and oil retainer. Here inorganic substances and fine sand particles are precipitated and separated from the wastewater, and then the wastewater is sent to the anaerobic tank for biological phosphorus removal. The tank is equipped with submersible mixers to prevent sedimentation of the sludge in the anaerobic tank. After the anaerobic tank, the wastewater is sent to the aeration basins where biological treatment is carried out. The wastewater/sludge mixture in the aeration basins is taken to final settlement tanks

with gravity. In the final sedimentation basin, the activated sludge/water mixture enters the central section of each tank via a reversed siphon. The treated effluent is discharged to the Karakaya Dam Lake by Boran Stream with gravity by the help of collecting chambers and treated wastewater collecting channels. The sludge settled in the final sedimentation basin is swept into the mud tanks in the middle of the tanks by means of rotary sludge scrapers. From here, the sludge is sent to the sludge collection chamber and to the sludge pump station via gravity under hydrostatic pressure. The mechanical sludge thickening is applied to the pre-drying of the biological sludge to a dry solid concentration of 5-7%, after conditioning with cationic polymer (polyelectrolyte). Belt filter presses are applied to dewater the biological excess sludge to a dry solid concentration ratio of 22 to 25%. The dewatered sludge is mixed with lime to a dry solid concentration ratio of 36% and then sent to the final collection area, and disposed of in the municipal garbage area. It is thought that the sludge will be used in agriculture in the near future. In this study, the samples of the sludge examined were monthly taken from the final sludge collection area of MABWTP during 1 year (as 3 parallel). Cu and Zn metals were observed in the samples of treatment sludge. The samples taken for this purpose were analyzed by ICP/MS (ICP/MS-Perkin-Elmer ELAN 9000) in a laboratory with ISO 9001: 2000 accreditation.

## RESULTS AND DISCUSSION

The Cu concentrations detected in sewage sludge samples of MABWTP are given in Figure 2.

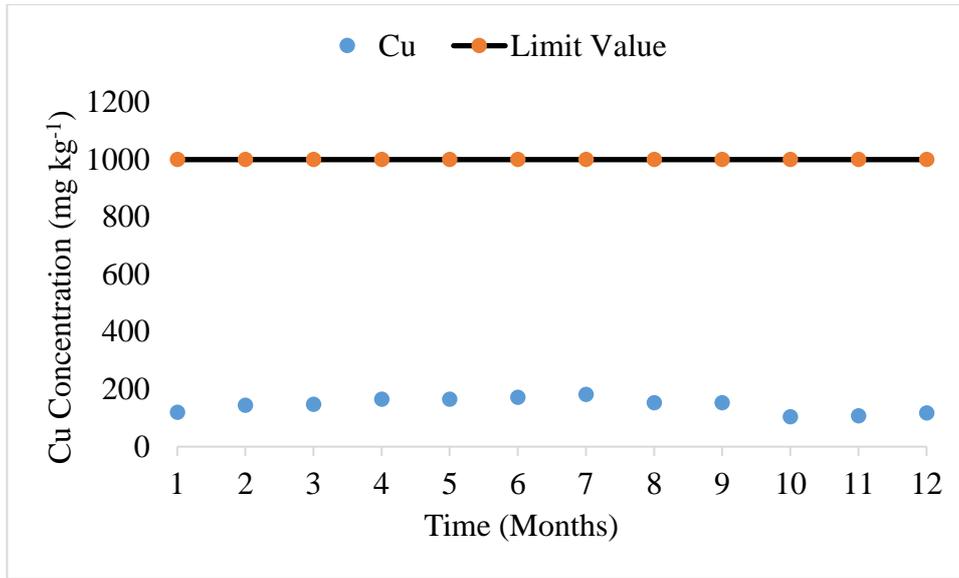


**Figure 2.** Cu concentrations in sewage sludge samples of MABWTP

According to Figure 2, the highest Cu concentration in the sewage sludge discharged from MABWTP was  $181.74 \pm 9.0 \text{ mg kg}^{-1}$  in September and the lowest Cu concentration was  $103.69 \pm 5.18 \text{ mg kg}^{-1}$  in December. The Cu concentrations detected in this study are in accordance with the values reported in sewage sludges by Gianico et al. (2013) ( $90\text{-}206 \text{ mg kg}^{-1}$ ) in Italy, Nikovski and Kalinichenko (2014) ( $200\text{-}300 \text{ mg kg}^{-1}$ ) in Russia, EECS (2012) ( $271 \text{ mg kg}^{-1}$ ) in Canada (Fijalkowski et al., 2017) and Rrong et al (2016) ( $235.6 \text{ mg kg}^{-1}$ ) in China, Alvarenga et al. (2015) ( $140.8\text{-}155.8 \text{ mg kg}^{-1}$ ) in Portugal. Higher results were also reported. Bergs (2015) reported Cu values between  $304\text{-}378 \text{ mg kg}^{-1}$  in Germany. Wiechmann et al. (2013) reported Cu concentration as  $292 \text{ mg kg}^{-1}$  in Germany.

Significantly higher values than ours reported by Healy et al. (2016) and Wang et al. (2016) were  $520 \text{ mg kg}^{-1}$  in Ireland and  $3\ 323 \text{ mg kg}^{-1}$  in China, respectively. Different values were given by Gawlik (2012) from EU countries. Gawlik (2012) reported Cu concentrations between  $27.3\text{-}578.1 \text{ mg kg}^{-1}$ . Gondek et al. (2014) reported values of  $127$  and  $324 \text{ mg kg}^{-1}$  in Poland. Souza et al. (2014) reported Cu concentrations between  $119\text{-}341 \text{ mg kg}^{-1}$  in Brazil. Öbek et al. (2004) reported Cu concentration as  $148.5 \pm 20.5 \text{ mg kg}^{-1}$  in Turkey (sewage sludge from Elazığ Municipal Wastewater Treatment Plant).

The comparison of the maximum Cu concentration given in RUDUSS and the Cu concentration determined in the sewage sludge taken from MABWTP are given in Figure 3.

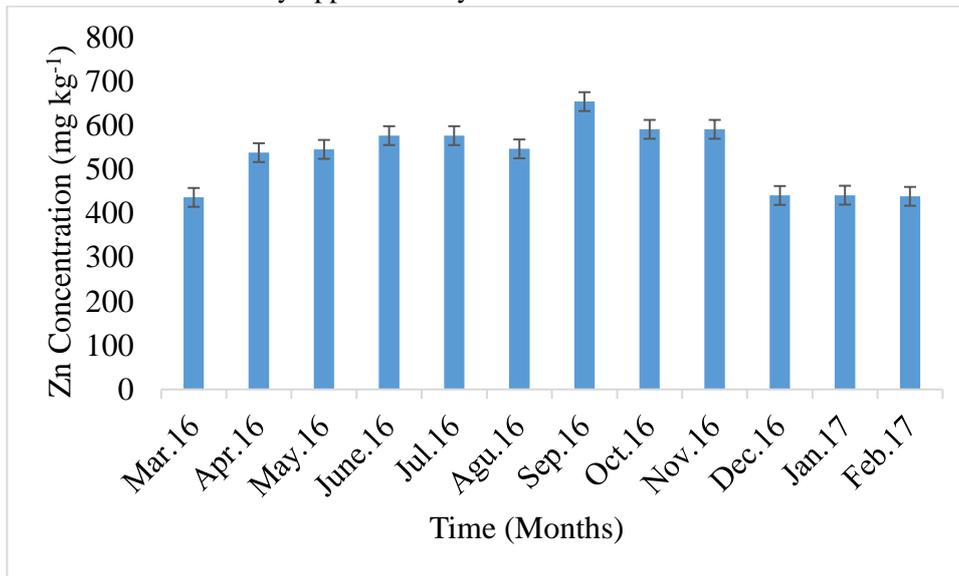


**Figure 3.** The comparison of the limit value and the detected value of Cu

The maximum Cu concentration that can be allowed in the stabilized sludge which can be used in the soil is 1 000 mg kg<sup>-1</sup> according to the RUDUSS (Figure 3). When Cu concentrations in the MABWTP treatment sludge were examined, it was found that they were lower than the limit value given in the regulation. In our study, Cu concentrations were found to vary approximately

between 100 and 200 mg kg<sup>-1</sup>. As a result, it has been determined that sludge from the MABWTP can be used in the soil in terms of Cu concentration.

The Zn concentrations detected in sewage sludge samples of MABWTP are given in Figure 4.



**Figure 4.** Zn concentrations in sewage sludge samples of MABWTP

As shown in Figure 4, the highest Zn concentration in sewage sludge discharged from MABWTP was 653.8±32.6 mg kg<sup>-1</sup> in September

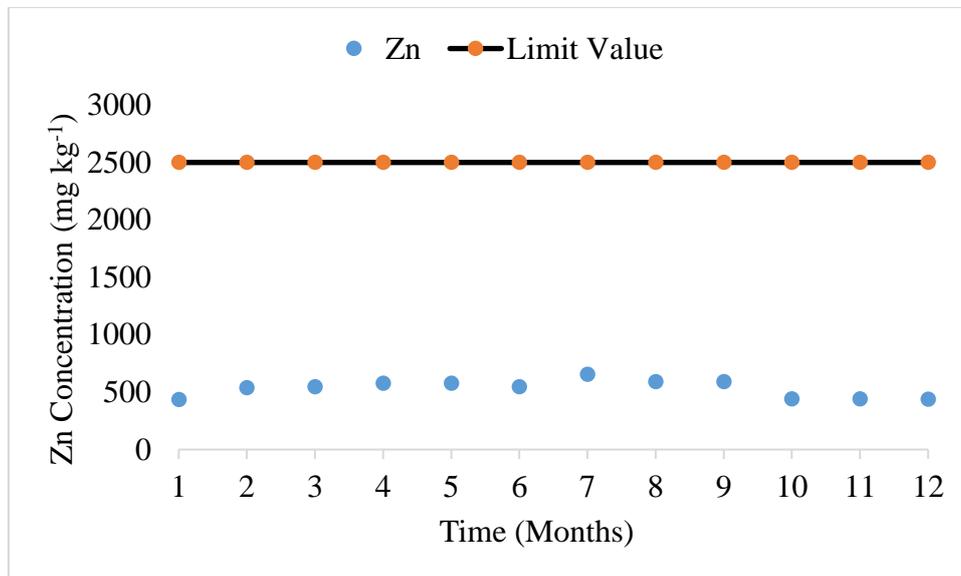
and the lowest Zn concentration was 436.2±21.8 mg kg<sup>-1</sup> in March. Zn concentrations did not change very much in December, January and

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February (in the range of about 438-440 mg kg<sup>-1</sup>). The Zn concentrations detected in the present study are in accordance with the values reported in sewage sludges by Bergs (2015) and Wiechmann et al. (2013). Bergs (2015) reported Zn value was detected as 756.7 mg kg<sup>-1</sup> in Germany in year 2005. Wiechmann et al. (2013) reported Zn concentration as 762 mg kg<sup>-1</sup> in Germany. Higher results were reported by Bergs (2015) between 794-2 140 mg kg<sup>-1</sup> in Germany in years from 1977 to 2001. Gondek et al. (2014)

reported values of 1 151 and 1 478 mg kg<sup>-1</sup> in Poland. Lower result (235.6 mg kg<sup>-1</sup>) was reported by Rrong et al (2016) in China. Also, Grobelak et al. (2017) reported value of 288.9 mg kg<sup>-1</sup> in Poland. Souza et al. (2014) reported Zn concentrations between 11.5-62.2 mg kg<sup>-1</sup> in Brazil.

The comparison of the Zn concentrations detected in sewage sludge from MABWTP with the Zn concentration given in the regulation is given in Figure 5.

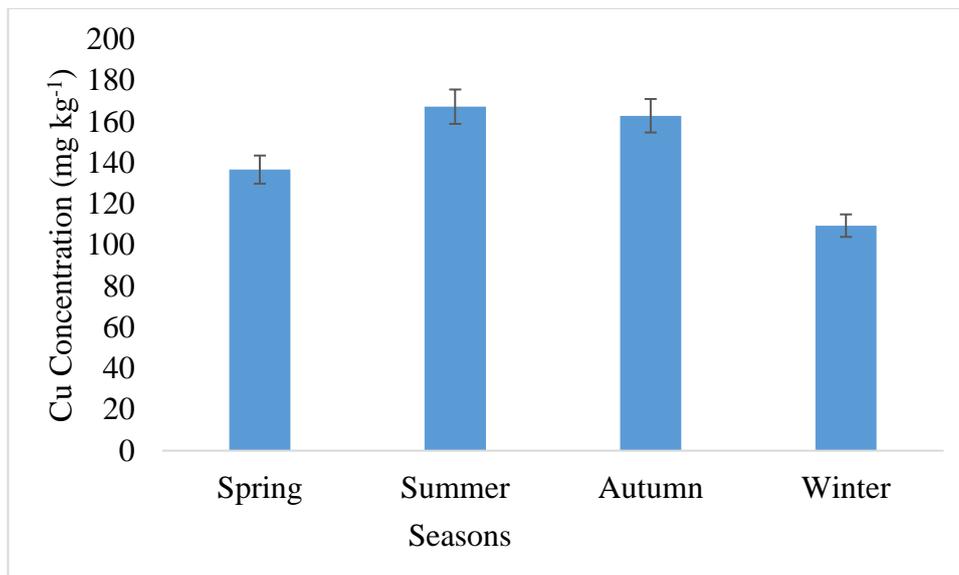


**Figure 5.** The comparison of the limit value and the detected value of Zn

The maximum permissible Zn concentration in the stabilized sludge that can be used in the soil is 2 500 mg kg<sup>-1</sup> as shown in Figure 5. In this context, it was determined that Zn concentrations in the MABWTP treatment sludge were lower than the limit value given in the regulation. Zn concentrations in our study ranged approximately from 430 to 655 mg kg<sup>-1</sup>. As a result, it was

determined that the sewage sludge from MABWTP can be used in soil in terms of Zn concentration.

The seasonal variation of Cu concentrations detected in MABWTP sewage sludge is given in Figure 6.

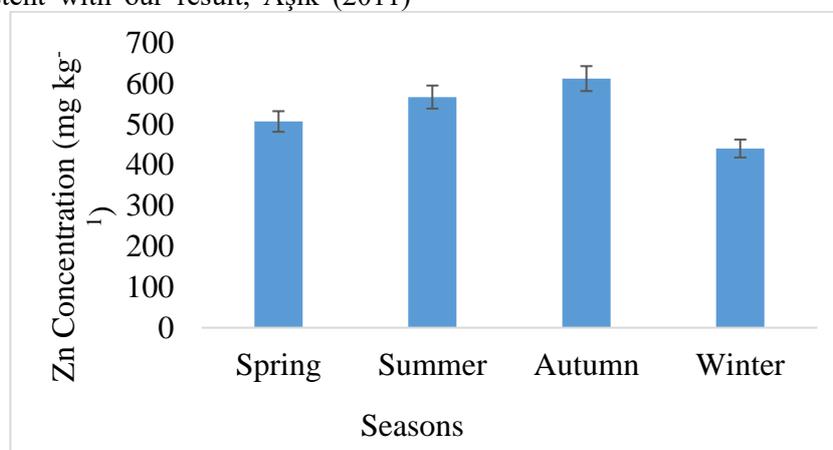


**Figure 6.** The seasonal variation of Cu concentrations in sewage sludge samples of MABWTP

The Cu concentrations in the sewage sludge from MABWTP were examined seasonally and the highest mean Cu concentration was found to be  $167.21 \pm 8.3$  mg/kg in the summer. Similarly, Aşık (2011) reported that the highest Cu concentration in the sewage sludge from the treatment plant of Bursa Metropolitan Municipality Water and Sewerage Administration (east region) was detected in the summer ( $203.8$  mg kg<sup>-1</sup>). Also, Aşık (2011) reported that the highest Cu concentration in the sewage sludge from the treatment plant of Yenice Municipality was detected in the summer ( $74.79$  mg kg<sup>-1</sup>). In our study, the lowest mean Cu concentration was found to be  $109.39 \pm 5.4$  mg kg<sup>-1</sup> in the winter season. Consistent with our result, Aşık (2011)

reported that the lowest Cu concentration in the sewage sludge from the treatment plant of Bursa Metropolitan Municipality Water and Sewerage Administration (east region) was detected in the winter ( $137.1$  mg kg<sup>-1</sup>). Also, Aşık (2011) reported that the lowest Cu concentration in the sewage sludge from the treatment plant of Yenice Municipality was detected in the winter ( $67.63$  mg kg<sup>-1</sup>). In our study, the mean Cu concentration was  $136.64 \pm 6.8$  mg kg<sup>-1</sup> in spring and  $162.81 \pm 8.14$  mg kg<sup>-1</sup> in autumn (Figure 6).

The seasonal variation of Zn concentrations detected in sewage sludge of MABWTP is given in Figure 7.



**Figure 7.** The seasonal variation of Zn concentrations in sewage sludge samples of MABWTP

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The highest mean Zn concentration in the sewage sludge from MABWTP was found as  $611.80 \pm 30 \text{ mg kg}^{-1}$  in the autumn season. Contrary to our results, Aşık (2011) reported that the highest Zn concentration in the sewage sludge from the treatment plant of Bursa Metropolitan Municipality Water and Sewerage Administration (east region) was detected in the summer ( $822.9 \text{ mg kg}^{-1}$ ). In our study, the lowest mean Zn concentration was found as  $440.13 \pm 22 \text{ mg kg}^{-1}$  in the winter season. Contrary, Aşık (2011) reported that the lowest Zn concentration in the sewage sludge from the treatment plant of Bursa Metropolitan Municipality Water and Sewerage Administration (east region) was detected in the spring ( $616.1 \text{ mg kg}^{-1}$ ). In our study, mean Zn concentrations were found to be  $506.47 \pm 25.3 \text{ mg kg}^{-1}$  in spring and  $566.47 \pm 28.3 \text{ mg kg}^{-1}$  in summer.

In our study, Zn concentrations were higher than the Cu concentrations in the sewage sludge samples taken from MABWTP. Similarly, Shrivastava and Banerjee (2004) reported that Zn and Cu in sewage sludge form the series of:  $\text{Zn} > \text{Cu}$  (Sobik-Szoltyssek et al., 2017).

## CONCLUSIONS

The production of sewage sludge which originates from the wastewater treatment plants increase day by day and become a global problem. However, the sewage sludge is cheap, suitable and valuable organic amendment for soils. Nevertheless, when using sewage sludge in soil, it should be noted that heavy metal content of the sewage sludge can cause harmful and toxic effects on all livings. In the present study, due to the content of Cu and Zn, the examined sewage sludge met standards in the RUDUSS. Therefore, the sewage sludge could be used in agriculture in terms of Cu and Zn.

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