Gaziosmanpaşa Üniversitesi Ziraat Fakültesi Dergisi
Journal of Agricultural Faculty of Gaziosmanpasa University
http://ziraatdergi.gop.edu.tr/

Araştırma Makalesi/*Research Article*

JAFAG
ISSN: 1300-2910
E-ISSN: 2147-8848
(2017) 34 (1), 129-136
doi:**10.13002/jafag1138**

The Heavy Metal Content of Some Herbal Plants on the Roadside of Adana-Gaziantep Highway

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Alındığı tarih (Received): 24.10.2016 Kabul tarihi (Accepted): 07.04.2017
Online Baskı tarihi (Printed Online): 17.04.2017 Yazılı baskı tarihi (Printed): 02.05.2017

Abstract: Oleander (*Nerium oleander)*, rosemary *(Rosmarinus officinalis)* and Pyracantha (*Pyracantha coccinea)* were planted for soil erosion protection purposes on the sides of Adana-Gaziantep highway. Humans and livestock consume the aforementioned plants for food although the consumption of this plants were forbidden by law. The highway emissions, mostly expected to be toxic on plants and soils, were determined in this study. Plant and soil samples were collected from the highway between Misis Industry Zone and Adana-Ceyhan highway, which have dense traffic throughout the year. Seven heavy metals (Cr, Cu, Fe, Mn, Ni, Pb and Zn) concentration were measured in plant and soil samples. The element components measured for plants for Cu, Fe, Zn, Mn, Cr, Ni, and Pb were 13.9, 172.9, 68.9, 71.3, 1.52, 62.06 and 7.3 µg g^{-1} respectively. Thus, the consumption of these plants should be prevented due to their heavy metal content over the allowable limits. However, the high density of those plants (*Pyracantha coccinea, Nerium oleander and Rosmarinus officinalis)* on the sides of highway may help to trap more heavy metals with reducing transportation to nearby agriculture and settlement areas.

Keywords: plant nutrient, soil, plants, medicinal plants, highway

Adana-Gaziantep Otoyolu Kenarlarında Yetişen Bazı Herbal Bitkilerin Ağır Metal İçeriği

Öz: Adana-Gaziantep otoyolu kenarında olabilecek erozyonun önlenmesi için Zakkum (*Nerium oleander)*,Biberiye *(Rosmarinus officinalis)* ve Ateş Dikeni(*Pyracantha coccinea)* bitkileri ekilmiştir. Otoyol kenarında ki bitkiler yasak olmasına karşın insanlar kendileri ve hayvanları için gıda olarak kullanmaktadırlar. Çoğu kez toksik etki göstermesi beklenen otoyol trafiğinin bitkilerin ve toprağın bitki besin elementleri ve ağır metal içeriklerine etkisi belirlenmiştir. Bitki ve toprak örnekleri Misis Organize Bölgesi ile Adana-Ceyhan otoyol gişeleri ile arasında kalan otoyol üzerinden toplanmıştır. Örneklerde yedi ağır metal konsantrasyonu (Cr, Cu, Fe, Mn, Ni, Pb ve Zn) incelenmiştir. Bitkilerde saptanan element içerikleri sırasıyla Cu, Fe, Zn, Mn, Cr, Ni, ve Pb için 13.9, 172.9, 68.9, 71.3, 1.52, 62.06 ve 7.3 µg g^{-1} olarak ölçülmüştür. Ağır metaller için saptanan düzeylerin izin verilen düzeylerden yüksek olması nedeniyle bu bitkileri gıda amaçlı kullanılmaları engellenmelidir. Bunun yanı sıra, bu bitkiler (*Pyracantha coccinea, Nerium oleander and Rosmarinus officinalis)* korunması hatta arttırılması ile eksoz gazlarından gelen ağır metallerin daha çok tutulmasında da kullanılabilecekleri belirlenmiştir.

Anahtar Kelimeler: Bitki besin elementi, toprak, tıbbi bitkiler, otoyol

1. Introduction

The 16 elements (C, H, 0, N, P, K, S, Ca, Mg, Fe, Zn, Mn, Cu, B, CI, and Mo) found in nature are essential nutrient elements for plants. The other 6 elements (Co, AI, Na, Si, Ni and V) are only found in some specific plants or used in biologic processes by plants (Yıldız, 2003). Pollution of air, water and soil by heavy metals have negative effect on human, animal and plant health. Moreover, the consumption of plants with high metal content has an additional damaging potency on majority of the living organisms.

The allowable levels for heavy metal varies for soil and plants. For soils, the toxic levels of Cu, Fe, Zn, Mn, Cr, Ni and Pb are >0.5>4.5>1>2>45>50 mg kg^{-1} respectively (Estefan et al., 2013; Stankovich, 2011; Swaine, 1955). The limits for plants are 10, 100, 20, 50, 1.3, 0.5-5, 2 mg kg^{-1} for Cu, Fe, Zn, Mn, Cr, Ni and Pb (Salazar and Pignata, 2014, Estefan et al., 2013)

The sources of heavy metals in urban soils, road dusts and agricultural soils are quite complex issue as there are several factors effecting the composition and amount of their presence, but their increment in any environment create public health risk (Johansson et al. 2009). In urban areas, heavy metals originated from polluted soils and road dusts can directly accumulated in human body via inhalation, ingestion and dermal contact absorption (Sezgin et al., 2003; Miguel et al., 2007). Among the most common heavy metals found in agricultural soils and road dusts are lead (Pb), chromium (Cr), arsenic (As), zinc (Zn), cadmium (Cd), copper (Cu), mercury (Hg), and nickel (Ni) (GWRTAC, 1997) originated from fuels and agro-chemicals (Christoforidis and Stamatis, 2009; de Vries et al., 2013). Several studies intended to outline the hazards of heavy metals in urban and rural areas as reported in United Nations World Urbanization, which outlined people preference to live in city center rather than live in rural areas (Satterthwaite, 2014). This figure as of 2014 is 54% of the world population and will most probably rise to in 2050. So, heavy metal borne human health problems in urban areas likely to increase in coming years.

There are also potential health risks associated with growing food in urban settings. Urban soils may harbor contaminants such as lead, petroleum products, and asbestos. People may meet these contaminants if they work or play in contaminated soil, or consume food that has grown in it. In some cases, exposure to soil contaminants can increase disease risks, especially for children (Hopkins, 2014). Thus, accumulation of heavy metals need detailed study for predicting serious health effects that may rise in short-term particularly with increasing number of cars. This is well documented by Adedeji et al (2014) who reported the accumulation of heavy metals in top soils is greatly influenced by traffic volume and all the heavy metals exhibited a significant reduction in the roadside soils with increasing distance from the road. Authors also ranked the metal concentrations in the roadside soils as follows Zn>Pb>Fe>Cu>Mn>Cd>Cr (Adedeji et al., 2014).

Roadside soils in urban areas all over the world are pointers of heavy metal contamination from a variety of sources mostly of anthropogenic origin. Major metal pollutants of the roadside environments are released from fuel burning, wear out of tires, leakage of oils, and corrosion of batteries and metallic parts such as radiators and muffles (Xia et al., 2010). However, all metals are not anthropogenic origin, they may be naturally occurring, and have shown potential health hazards especially in high concentrations in human and plant cells particularly in volcanic regions. Moreover, independent from origin, Cd, Pb, Cu and Zn are potentially toxic and pose great threat to food safety and human health even in minute concentrations (Abduljaleel et al., 2012). However, natural events that are hazardous to humans are very rare, after all researches have focused on heavy metals in urban soil, including those contributed by automobiles along transport routes (Lu et al., 2012; Świetlik et al., 2013). Poisonous metals such as Cd, Cr, Cu, Fe, Mn, Pb, and Zn are associated with ambient particles released by roadside dust and can cause the production and release of inflammatory mediators by the respiratory tract epithelium (Duan et al., 2012). Thus, research is needed to outline the heavy metal dynamics within the roadside impact area for heavy metal mitigation or prevention management studies. Tahar and Keltoum (2011) found that plant samples from the immediate environment of the dumpsite were highly contaminated with Zn, Cd and Mn. Three plants species: grape, artichoke and pepper, particularly, grape can be classified as accumulators for Zn, Cu, Cd and Fe, consequently, it revealed a health risk for human and livestock due to the spread of the metal pollution from waste dumpsites to agricultural areas. Thereby it can be said that urban areas particularly where road network is dense are prone to heavy metal contamination.

Since another main component of this study is herbal plants, the source areas need utmost care as nearly 70–80 % of the world population still primarily relies on nonconventional medications, mostly derived from herbal plants (Sahoo, 2010). Therefore, the toxicity level of medicinal plants is crucial for human health that are grown nearby potential pollutant areas as they may most probably contain pesticides, microbes, heavy metals, chemical toxins, and adulterants (Saad et al., 2006). Collecting of herbal plants at roadside or nature is more profitable than their cultivation or growing other crops such as cereals and vegetables. So, roadsides decorated with medicinal plants are attracting low-income people who are collecting those plants for trading. This cause higher amount of heavy metal intake by humans as many consumers do not have any idea about the origin of these plants. On the other hand all metals are not harmful even required by plants at certain amounts (Barker and Pilbeam, 2015) such as zinc, copper, iron, manganese, and chromium are essential nutrients both for plants and even for humans (Prasad, 2013). They are important for the physiological and biological functions of the human body. However, as allowable and toxic levels of these metals are so close they can easily become toxic (Korfali et al., 2013).

Due to increasing level of pollution that is the result of rapidly increased number of automobiles in settlement areas, it is necessary to investigate the fate and effects of metals in components of ecosystems. Therefore, the present study has investigated whether the heavy metal accumulation in medicinal plants that grown on the edge of the highway and consumed by people.

2. Materials and Methods

Study Area

The study has been conducted on TEM (Trans European Motorway) highway, a major road connecting Europe to Middle East, namely O-52, located in South of Turkey at 36.989537^{o}N-35.611181^{o}E and 36.985438^{0}N-35.766565^{o}E. Adana has witnessed rapid population increase since 1990s, which triggered urban expansion. The increased vehicles and other socio-economic activities that caused attendant problems of traffic congestions and environmental pollution by heavy metals. Moreover, the O-52 highway that connects Middle East to Europe with dense traffic passes through the city. The number of light vehicles per day in 2014 was 12.956 and heavy vehicles was 8.739 on O-52 highway (TSD, 2015). Samples have been taken from two toll points on Adana-Gaziantep O-52 highway in during the dry season (August 2014)for avoiding rain-washing out the heavy metals from soil and plant leaves. The plant and soil samples have been taken from leaves of scarlet firethorn (*Pyracantha coccinea)* Oleander (*Nerium oleander),* rosemary *(Rosmarinus officinalis).*

Sampling

Prior to sampling, the areas were cleared from litter, and roadside soil samples have been collected from depths of 0-20 cm with stainless steel augers. A total of thirty (36) samples have been collected at varied distances of the highway in entrance of Adana Highway and Industrial Zone to the Ceyhan toll gates from side of the selected points.. Background or control samples have been collected in cultivation areas, which were about 1km away from the sampling locations. The geographical coordinates of these locations have been determined using a Garmin global positioning system (GPS) and recorded in geographical information system (GIS) for data processing. Soil samples for each sampling location have been thoroughly mixed after the removal of all debris and extraneous materials to give composite samples for each location.

Sample Treatment

Soil samples have been air-dried in a circulating air in the oven at 30°C to a constant weight and then passed through a 2 mm sieve and stored in dry labelled plastic and taken to the laboratory for pretreatment and analyses at controlled temperature (4°C) to prevent any microbial activity (Ahirwar et al., 2013).

Both plant and soil samples have been stored in sealed polyethylene bags. The soil and plant samples were measured by ICP Atomic Emission Spectroscopy (Zeiss, Plasmaquant 110).

The operating conditions of the ICP are as follows: plasma gas flow 12 L/min; carrier gas flow adjusted to 0.8 L/rnin and a gas input pressure of 0.5 MPa. Analytical grade reagents were used for digestion and standards.

3. Results and Discussion

Soil Analysis

Copper: The copper (Cu) concentrations varied in a wide range between 0.92 and 1.43 µg g^{-1} in soils (Table 1). The maximum concentration of copper is determined in Rosemary soils with 1.43 mg kg^{-1}. The critical limit of Cu for soils is ≥ 0.5 µg g^{-1} (Estefan, et. al., 2013). The concentration of Cu is higher than allowable limits in all soils.

Table 1. Soils of heavy metal concentration (µg g^{-1})in roadside different plants

Çizelge 1. *Yol kenarındaki farklı bitkilerde toprakların ağır metal konsantrasyonu (µg g-1)*

Plants	Cu	Fe	Zn	Mn	Cr	Ni	Pb
				µg g^{-1}			
P. coccinea	1.28 ab	69.6 b	3.760 a	1.700 b	45.27 b	43.08 c	33.70 b
N. oleander	0.920 b	75.7 a	2.400 b	0.770 c	55.28 c	66.05 a	25.56 c
R.osfficinali	1.430 a	63.7 c	1.520 c	3.183 a	56.88 a	61.19 b	35.76 a
Mean	**1.21**	**69.64**	**2.56**	**1.88**	**52.5**	**56.8**	**31.7**
LSD	**0.3859***	**1.920***	**0.7503***	**0.3776***	**0.0866****	**0.0866****	**0.1225***

*, **, ***, and NS indicate significance level at p=0.05,0.01,0.001, and not significant, respectively.

Iron: The observed range of iron (Fe) in the current soil has varied from 63.7 to 75.7 µg g^{-1} (Table 1). The maximum concentration of iron was determined insoils of *Oleander*with. The critical limit of the Fe for soils is ≥ 4.5 µg g^{-1} (Estefan et al., 2013). Thus, the Fe concentration were fifteen times more than the threshold value reported in studied soils.

Zinc: The concentration of zinc (Zn) in the analyzed soil samples ranged between 1.52 and 3.76 µg g^{-1} (Table 1). The maximum concentrations of zinc was observed in *P. Coccinea*as. When Zn concentration is greater than or equal to 1 µg g^{-1} in soils it is considered (Estefan et al., 2013). Thus, the roadside soils have high Zn content for health limits.

Manganese: The Mn concentrations of soils have been found between 0.77 and 3.183 µg g^{-1} (Table 1). Estefan et al. (2013) reported that the ≥ 2 µg g^{-1} Mn asthe critical limit in soils Results revealed that Mn accumulation on roadsides reached dangerous proportions in the study area.

Chrome: According to Abii and Okorie (2011), the Cr critical level is 40 µg g^{-1}. Cr concentrations observed higher than critical levels in plants than soils. Cr levels varied from 45.27 and 56.88 µg g^{-1} in studied soil samples (Table 1).

Nickel: The Ni concentrations observed between 43.08 and 66.05 µg g^{-1} (Table 1). The critical level is 50 µg g^{-11} in soil (Stankovich et al., 2011).

Lead: The lead level is 22 µg g^{-1} according to some source critical level is 15 µg g^{-1} (Swaine, 1955). The lead values are between 25.26 and 35.76 µg g^{-1} in soil (Table 1). Overall the results of the analysis showed that heavy metals were present in varied concentrations in the three traditional plants commonly consumed in the Turkey. The concentration of heavy metals in many cases exceeded the internationally accepted permissible levels. Especially, elements of Cr, Pb, Ni and Fe are accumulation (Figure 1). Generally, the longer the highway usage history, the higher the concentration in the roadside soil (Bai et al, 2009), because it is positively related to traffic volume (Chen et al., 2012). Heavy metals being high in the wayside are because of exhaust gases (Cheraghi et al., 2012).

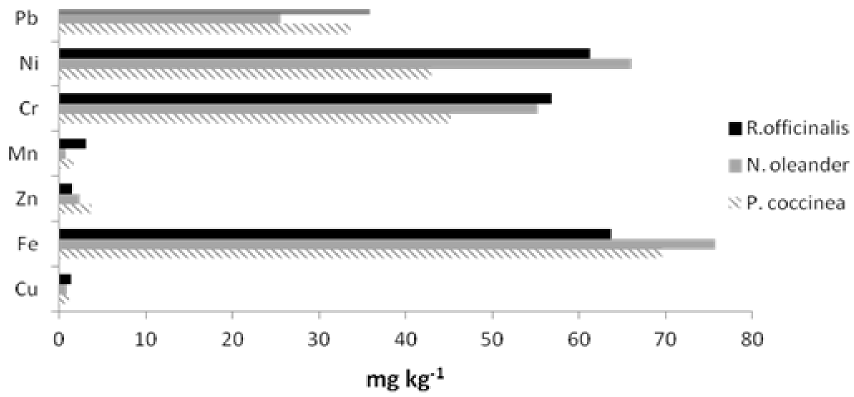

Figure 1. Heavy metal concentration in various plants growing in roadside
Şekil 1. *Yol kenarında büyüyen çeşitli bitkilerdeki ağır metal konsantrasyonu*

Plant analysis

Copper: The copper (Cu) concentrations of the plants varied in a wide range between 11.8 and 15.5 µg g^{-1} (Table 2). The maximum concentrations of copper in *Pyracantha coccinea* was 15.5 µg g^{-1}. The threshold value of the Cu for plants is ≥ µg g^{-1} (WHO, 2002). Thus, the concentration of Cu is quite high than allowable limits in all studied plants. Copper is an essential component of many enzymes, however its excessive intake can cause dermatitis, irritation of the upper respiratory tract, abdominal pain, nausea, diarrhea, vomiting, and liver damage (Ulla et al., 2012). The high amounts of Cu is highly related to traffic volume as reported by Chen et al. (2012).

Table 2. Heavy metal concentrations (µg g^{-1}) in different plants
Çizelge 2. *Yol kenarında büyüyen çeşitli bitkilerdeki ağır metal konsantrasyonu*

Plants	Cu	Fe	Zn	Mn	Cr	Ni	Pb
				µg g^{-1}			
P. coccinea	15.5 a	170 ab	88.5 a	71.4 b	2.82b	2.63 b	8.94 a
N. oleander	11.8 c	181.4 a	42.9 b	96.6 a	0.62 c	1.81 c	4.86 c
R.officinalis	14.4 ab	167.4ab	75.4 a	28.7 c	10.7 a	30.2 a	8.16 d
Mean	**13.9**	**172.9**	**68.9**	**71.3**	**1.52**	**2.06**	**7.3**
LSD	**3.010****	**17.98****	**8.18****	**1.88****	**0.00274****	**0.00274****	**0.00274****

*, **, ***, and NS indicate significance level at p=0.05,0.01,0.001, and not significant, espectively.

Iron: The observed range of iron (Fe) in the study was between 167.4 and 181.4 µg g^{-1} in plant samples (Table 2). The maximum concentration of iron in nerium is similar to soils, which was ≥ 100 µg g^{-1} (Estefan et al., 2013). The plants' Fe concentration was about two times more than the allowable levels expressed in the literature (Ray et al. 2016). Iron has emerged very high in soil too. This result also shows that the Fe is readily transported from the soil to the plants. Including oxygen supply, energy production, and immunity. Excess iron is associated with symptoms of dizziness, nausea and vomiting, diarrhea, joints pain, shock, and liver damage. Iron toxicity has also adverse effects on various metabolic functions and cardiovascular system (Martin and Griswold, 2009**).**

Zinc: The concentration of zinc (Zn) in the analyzed plant samples ranged between 42.9 and 88.5 µg g^{-1} (Table 2). The maximum concentrations of zinc was determined in *Pyracantha coccine*. The critical limit of the Zn for plants is ≥ 20 µg g^{-1} (Estefan et al., 2013).

Manganese: The Mn concentrations range for the studied samples were 28.7 to 96.6 µg g^{-1} (Table 2). The critical limit of the Mn for a plant tissue is ≥ 50 µg g^{-1} (Estefan et al., 2013). Manganese accumulation in the human body leads to neurological disorders (Neal and Guilarte, 2013).

Chrome: According to WHO (2002), the Cr critical level for plants is 1.3 µg g^{-1}. Cr concentrations observed between 0.62 and 10.7 mg kg^{-1} in plant samples (Table 2). The highest value was measured as 10.7 mg kg^{-1}in Rosemary. This excess Cr values are threating human health in the study area as the highway passes from densely populated city center, and people can obtain these plants easily from roadside collectors. People use these collected plants for treatment of some illnesses and making herbal tea. This situation caused the transition of excess Cr to the human body. Moreover, people can exposed to chromium through breathing, eating or drinking and through skin contact with chromium or chromium compounds which was plants in this study.

Nickel: The Ni concentrations observed between 1.81 and 30.2 µg g^{-1}in plants (Table 2). The critical level of Ni for plants is 0.5-5 µg g^{-1}(Hussain and Khan, 2010;Salazar and Pignata, 2014). In case of Ni, the concentration in different plants is in the order of; A. vulgaris 4.35 µg g^{-1}, G. aparine 4.16 µg g^{-1}, M. pruriens 3.18 µg g^{-1}, C. tetragonoloba 2.79 µg g^{-1}, W. somnifera 2.64 µg g^{-1}, S. rebaudiana 2.36 µg g^{-1} and A. Adscendens 0.94 µg g^{-1}. According to results the highest value observed by 30.2 from Rosemary. All values measured were above critical level. Khan et al., (2008) reported that high concentration of Ni in plants are mainly related to anthropogenic activities.

Lead: The allowable lead level for plants is 2 µg g^{-1} (Salazar and Pignata, 2014). However, lead values in studied plants varied from 4.86 to 8.94 µg g^{-1} (Table 2). Medicinal herbs are easily contaminated by lead during growth, development and while processing them for consumption. After collection of the polluted plants from roadsides, the high heavy metals easily enter the human body and disturb the normal functions of central nervous system, liver, lungs, heart, kidney and brain, leading to hypertension, abdominal pain, skin eruptions, intestinal ulcer and different types of cancers (Khan et al., 2008). Pb concentration that must be existent in herbal organisms in normal conditions has been reported as 0,05-3 µg g^{-1} (Demirezen, 2002).

The heavy metals (Cu, Fe, Zn, Mn, Cr, Ni and Pb) concentrations in the studied soils and plants are above threshold levels (Figure 2).

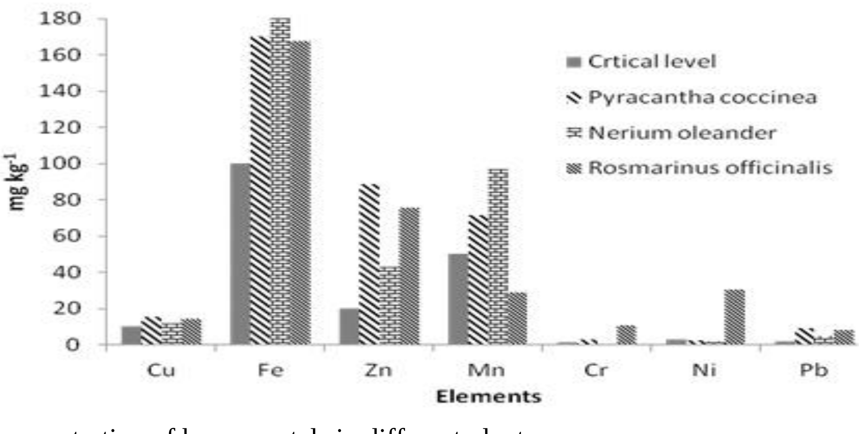

Figure 2. The concentration of heavy metals in different plants
Şekil 2. *Farklı bitkilerdeki ağır metal konsantrasyonu*

Along with muffle emissions, bitumen and mineral filler materials in asphalt road surfaces also contain various heavy metals, including Cu, Zn, Cd, and Pb (Winther et al., 2010). Heavy metals can be transported to the roadside soils by winds, water runoff and atmospheric precipitation

(Nabuloa et al., 2006) which amplifies the heavy metal problem with roadside environment and needs a intentive mitigating or prevention management for human and animal health.

The Potential Risk Assessment of heavy Metals soil and plants

Metal contaminants in soils can have serious implications for the soil ecosystem, soil organisms and finally for human health. Soil and plant contamination is occurring on a global scale due to combustion of fossil fuels. Exposure to emitted heavy metals are causing fatal effects on humans and the environment. It is therefore desirable to obtain and monitor a quantitative analysis of the potential risk of heavy metals contamination particularly on roadsides where fossil fuels are excessively used. According to our analysis, the heavy metals in plants of the study are Pb, Zn, Ni, Fe, Mn, Cu and Cr respectively. All these are well-known for their toxicity to environment.

People used as medicinal and food herbal of plants. Low-income people pick up these plants unconsciously from the highway roadsides, and sell at low prices which increases their consumption by public. When we looked at the heavy metal values in our study, we identified that heavy metals are over threshold levels for both plants and soils. Thus, we highly recommended not to use plants grown on road sides of O-52 highway as these plants can adsorb and accumulate heavy metals that are toxic to human health. Unfortunately**,** the roadsides of O-52 highway is also used for pasture area by herders as no rangeland is allocated in adjacent agricultural lands.

4. Conclusion

The results suggest that medicinal plants used for human consumption or for preparation of herbal products and standardized extracts should be collected from a natural habitat that are far away to roads which is also mentioned in health regulations. This study illustrated metal accumulation in various herbal plants planted on roadside that are used in traditional healing culture in Turkey, can cause lethal health issues if they are collected from roadsides.

However, these plants are capable of reducing heavy metal concentration that may pollute soils, water and air. But, the total amount of emitted heavy metal concentration should be studied for O-52 highway. Then sufficient amount of plants will be planted to trap heavy metals. Thus, the studied plants namely are Oleander (*Nerium oleander),* rosemary *(Rosmarinus officinalis)* and Pyracantha (*Pyracantha coccinea)* are suggested to employed for trapping heavy metals in recreation areas, city centers that are located close to highways since this study demonstrated their suability for heavy metal mitigation management.

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