The Study of Toxic Metals Contamination (Pb, Cd, As, Al) on Medicinal Plants Cultivated Near Arak Industrial Manufactures

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Received: 25.05.2011 *Accepted: 02.07.2011*

ABSTRACT

Background: Medicinal plants use is growing in Iran and controlling their quality in terms of contamination with toxic metals is critical. City of Arak, due to its large industrial plants, is one of the most polluted cities of Iran; these pollutants can be transferred to the farms.

Methods: In this study, concentrations of metals such as aluminum, arsenic, lead and cadmium are measured by atomic absorption spectrometry in five medicinal plants (*Thymus vulgaris*, *Melissa officinalis*, *Achillea millefolium*, *Rosmarinus officinalis*, *Salvia officinalis*) cultivated around the city of Arak.

Results: According to the test results, the aluminum concentration levels were between 319.06 and 561.08 µg.g- and the amount of arsenic in the samples ranged from 0.246 to 0.293 µg.g- Minimum and maximum lead levels were 0.254 and 3.022 µg.g-, respectively. With regard to cadmium, the lowest and highest values obtained were 0.031 µg.g- and 0.144 µg.g-, respectively.

Conclusion: High level of aluminum concentrations can be associated with aluminum factory located 25km far from the cultivation area.

Keywords: Al, As, Cd, Contamination, Medicinal Herb, Pb.

INTRODUCTION

Medicinal herbs have been used since ancient times and their use has been increasing over time. World Health Organization (WHO) has revealed that 70 to 80% of world's population uses alternative remedies, especially medicinal herbs as their first step treatment and the tendency to use herbal products has recently grown (1). The first reason for this increase is that people believe these products are safe. Availability and lack of need for physicians' prescriptions are among other reasons (2). Development and widespread application of herbal medicine has increased the risks of such uses, and frequent risks have been reported for patients who use these herbal products. These risks include the prescription of wrong herbals, inappropriate consumption or instruction, and the concentration of heavy metals in them (3,4).

General pollution of the environment has been intensified with toxic metals. Water and air are the two most important factors in transferring lead to plants. These two were responsible for polluting vegetables studied in city of Sari (5). In another study, the concentrations of heavy metals were measured in vegetables cultivated around city of Shahrood and it was found that the concentrations of cadmium, lead, and chrome were above the standards set by WHO and FAO (6).

The adverse effects of cadmium manifest in kidneys and bones. Lead contamination occurs through air and food intake. In fact, air pollution causes contamination of soil, water, and food. Gastrointestinal absorption of the metal and its passing through BBB is associated with brain damage in children. Arsenic enters the body through food and water and causes digestive, nerve, and heart problems (7).

Aluminum, on the other hand, can cause inflammatory and oxidative reactions which may lead to damage to body tissues and, most

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importantly, the brain damage which give rise to Alzheimer's disease (8,9). Generally, low amounts (around ppm) of heavy metals (aluminum, lead, arsenic, cadmium) in herbal products may cause chronic diseases, such as liver damage, kidney, blood, nervous system, and muscle problems in the long run (10,11). There are different sources of environmental pollution, among which the presence of industrial plants such as the aluminum factory is highly associated with the presence of different concentrations of these metals.

It is noteworthy that the city of Arak started its way toward industrialization in the 1970s. The first major industry which entered the city was an aluminum factory that was located near the farms. All the factories and industrial centers which are located in the city somehow add to pollution (12).

Accordingly, this study aims to measure the concentrations of heavy metals in the medicinal herbs cultivated in a 25 km diameter from the industrial plants of the city of Arak in order see the presence or absence of these metals which may have adverse effects on medicinal plants users' health.

MATERIALS AND METHODS

Preparation of samples

Five samples, including *Thymus vulgaris*, *Melissa officinalis*, *Achillea millefolium*, *Rosmarinus officinalis*, and *Salvia officinalis* were collected from farms surrounding Arak from April to June 2010.

Herbal products were divided into two groups. The first group, after drying in the shade, was washed by de-ionized water and then dried in an 80^{oC} oven. The washing phase was absent for the second group; it was only dried in the shade after collection. Then each plant was grinded, mixed, and kept in a polyethylene container which was washed with acid (dipped for 24 hours in 10% nitric acid and then washed three times with de-ionized water) till the beginning of the next phase.

Mineralization of Samples

Pb, Cd

One gram of the powdered plant which had been washed by acid in a beaker was weighed and then 5 ml concentrated nitric acid was added. Then the beaker was put on an 80-90^{oC} heater for 2 hours. It is also possible to increase the heat up to 150^{oC}.

During this period, 3-5ml of concentrated nitric acid and 30% H_2O_2 were added. The digestion process of the samples continued until a clear solution was obtained.

After cooling, the samples were filtered with filter paper Whatman NO. 42 in 25 ml volumetric flasks and then they reached a certain volume through de-ionized water (13).

As

Five grams of the powdered herb was weighed in an acid-washed vessel and heated on the flame to burn up completely. Then it was put in a 350^{oC} furnace for 3 hours to be completely incinerated. After that, the vessel was taken out of the furnace to cool down. About 2ml of the concentrated nitric acid was poured on the obtained ash and they were mixed by an acid-washed beater. Then the vessel was put on the heater in order to vaporize the acid, and it was again put in a 350^{oC} furnace for one more hour.

After removing the vessel from the furnace and after it cooled down, the obtained ash was dissolved in 10% hydrochloric acid and filtered in a 50ml conical flask. Before the sample reached a certain volume, 0.5 grams of potassium iodide (equivalent to 1%) was added to the flask to convert the 5 valence arsenic to 3 valence arsenic, and then they reached a certain volume through hydrochloric acid of 10%.

Finally, the samples were put on a water bath for 10 minutes with less than 80^{oC} in order to complete the reactions (14).

Al

Five grams of the powdered herb was weighed in an acid-washed vessel and heated on the flame to burn up completely. Then it was put in a 450^{oC} furnace for 3 hours to be completely incinerated. Then after, the vessel was taken out of the furnace to cool down. About 2ml of the concentrated nitric acid was poured on the obtained ash and they were mixed by an acid-washed beater. Then the vessel was put on the heater in order for the acid to be vaporized, and it again was put in the furnace for an extra hour.

After removing the vessel from the furnace and when it cooled down, the obtained white ash was dissolved in 1M nitric acid and filtered in 25ml volumetric flasks. In order to prevent the aluminum from being ionized and to omit the interfering chemicals, 0.02g lanthanum chloride and 0.05 nitrate potassium were added to the flasks and reached a certain volume using de-ionized water (15).

Determination of toxic metals

Aluminum, arsenic, lead, and cadmium stock solution of 1000 ppm were purchased from Merck Co. After preparing the required concentration of standard solutions and reading the absorption standards on the device, a standard curve was plotted. After obtaining the samples absorption using a standard curve, the heavy metals concentrations in the plant samples were measured.

RESULTS

Table 1 shows the amount of heavy metals in the plants.

Table 1. The amount of metals ($\mu g.g^-$) in medicinal plants

Plant's name	Metal ($\mu g.g^-$)							
	As		Al		Pb		Cd	
	washed	unwashed	washed	unwashed	washed	unwashed	washed	unwashed
Thymus vulgaris	0.144± 0.003	0.274± 0.04	412.85± 2.55	503.86± 4.78	2.718± 0.007	3.022± 0.09	0.054± 0.003	0.057± 0.002
Melissa officinalis	0.244± 0.004	0.293± 0.004	154.51± 1.4	383.47± 3.09	1.27± 0.04	1.478± 0.02	0.029± 0.002	0.031± 0.002
Achillea millefolium	0.216± 0.004	0.281± 0.004	375.35± 4.14	561.08± 7.39	0.738± 0.02	0.753± 0.03	0.054± 0.004	0.081± 0.003
Rosmarinus officinalis	0.24± 0.004	0.246± 0.006	364.65± 0.41	365.55± 0.91	0.007± 0.01	0.254± 0.03	0.072± 0.003	0.129± 0.01
Salvia officinalis	0.28± 0.006	0.292± 0.006	165.13± 3.69	319.06± 2.57	0.003± 0.006	0.488± 0.04	0.053± 0.002	0. 144± 0.01

DISCUSSION

According to the data presented in Table 1, washing the medicinal plants resulted in lowering the metal contamination in the samples, which indicates the source of contamination had been the air-born particles. As it can be seen, the amount of reduction in all plants was not the same, and since the same washing method was applied to all the plants, it can be concluded that the differences may be a result of morphologic nature of the plants surface, plant tissues being even or uneven, and the amount of fluff and absorbents on the outer surface of the plants.

Joint Expert Committee on food additives from the World Organization of FAO and WHO has presented the index of PTWI (provisional tolerable weekly intake) for each metal. In this article, this index was compared to the concentration obtained from the plants, the comparison was carried out according to the estimation of the amount of daily use of medicinal plants.

Cd

Cadmium concentrations obtained from the samples in this study vary from 0.031 to 0.144 $\mu g.g^-$ plants' dry weight. As seen in Table 1, there was not a significant difference in contamination between washed and unwashed samples. This may indicate that air pollution does not have a significant effect on the amount of cadmium in plants grown in the area (16). Accordingly, the observed cadmium in the samples may have been absorbed through water and soil and that is why washing did not affect the cadmium amount significantly.

Some research has been carried out on this issue throughout the world. For instance, the amount of cadmium in 10 different medicinal plants were measured in Egypt and the amount was reported to be between 0.3 to 0.05 µg.g^{-} (17). In Pakistan, 24 samples of medicinal plants had at least 0.05 µg.g^{-} and at most 12.06 µg.g^{-} of cadmium (18). Maleki's study shows that vegetables in outskirts of Sanandaj have <0.2 to 0.65 µg.g^{-} cadmium (19). The amounts of cadmium in medicinal plants in Italy were shown to be 0.01 to 0.75 µg.g^{-}(20). A study in Turkey indicated that there were 0.24 to 0.97 µg.g^{-} of cadmium in the studied vegetables (21). In these studies, the amounts of cadmium were more than the amount found in this study, and it seems that water and soil in the area of this study were less polluted than the areas of the above mentioned studies.

There are also some studies in which cadmium concentration is similar to the

ACKNOWLEDGEMENTS

concentration reported in this study. A good example for this is the vegetables studied in Pakistan that their cadmium levels were between 0.033 and 0.073 µg.g^{-} (22). Fytianos *et al.* (2001) reported that lettuce and spinach grown in an industrial polluted area had small amounts of cadmium (23).

The obtained cadmium levels in samples in this study were below the PTWI presented by WHO (60 µg/day/60 Kg) (24). Of course, cadmium concentrations in plants should be considered since in some parts of the world, people receive 60% of the PTWI from water and food intake. Also, patients with kidney problems are exposed to 50% of the presented PTWI (26).

Pb

As shown in Table 1, the levels of lead in studied medicinal plants vary from 0.254 to 3.022 µg.g^{-}. This table illustrates that lead contamination is more in unwashed than washed samples. It is said that the lead concentration in plants depends on air pollution and the absorption of this metal varies in different parts of the plants (27). This is why the difference between the washed and unwashed samples like *Salvia officinalis* is significant; it seems that washing removes lead contamination from the plant.

The reported lead levels in other articles are varied. In some studies in Egypt (17), Pakistan (18), Saudi Arabia (28), Iran (19), and Taiwan (29), the amount of lead in the samples of plants in these countries were 0.046-0.308, 0.07-103, 0.22, 11.23-16.99 (maximum), and 0.06 µg.g^{-}.(maximum), respectively. In another study in Pakistan, the amount of lead concentration in leaves of vegetables under the study was between 1.331 and 2.652 µg.g^{-} (21). The lead concentration in 6 out of 10 samples in a study carried out in Brazil were reported to be higher than the allowed level presented by WHO (10,30).

The lead concentrations in samples of this study were less than the accepted PTWI level by WHO (214 µg/day for a 60 kg person) (24). It is noteworthy that lead is considered as a toxic metal that its intake from food in most countries is higher than the allowed PTWI level (25). As the cities are getting more and more industrialized, the amount of lead contamination increases in the environment. So community exposure to lead has to be controlled, and one of the ways for such control is through medicinal herbs intake.

As

According to Table 1, the highest arsenic level is 0.293 and the minimum is 0.246 µg.g-. As it can be seen from Table 1, the arsenic level of the washed samples is less than the unwashed ones. Also, the effect of washing in all plant samples is similar.

Similar studies have been carried out on arsenic contamination in plants in other parts of the world, but the number is not too large. In a study in Taiwan, the maximum arsenic in 13 samples of medicinal plants was reported to be 8.78 ppb which is much less than the obtained levels in our study (29).

The amounts of arsenic in medicinal plants in most of the studies are more than the amounts found in this study. For example, a study conducted by Dzung on some medicinal plants reported a contamination between 0.173 and 15.62 µg.g^{-}(31). Another study on vegetables conducted in Bangladesh, reported their arsenic levels between 0.47 to 3.24 µg.g^{-} (32). Perhaps this difference is because of the difference in the source of arsenic contamination.

The arsenic levels of the present samples of this study are within the limits stated by the WHO (128µg/day for 60 kg person) (24).

Al

Aluminum concentrations in samples in this study were between 319.08 to 561.08µg.g^{-}. Aluminum levels in all washed samples were less than unwashed ones. The differences between washed and unwashed samples in aluminum levels were more than those in other samples. An explanation for these differences is the fact that the samples were cultivated around the aluminum factory and the high level of aluminum in the air. Therefore, its levels on the plants were high which explains why washing was more effective.

Unfortunately, few studies have been carried out on aluminum contamination in medicinal plants and food. An example of such studies is that of Lopez in Spain. This study was carried out on 17 plants and spices in which the aluminum levels ranged from 3.74 to 56.50 µg.g^{-} (33). In another study on foods carried out by the Nabrzyski, the highest levels of aluminum contamination were found in vegetables and tea plants (34). Hayacibara in Brazil reported that the level of aluminum in tea is within the allowed range provided by WHO (35). In another study which was conducted on nature-based raw materials, the levels of aluminum in the studied medicinal plants were 32 to 222 µg.g^{-} (36).

PTWI presented for aluminum is 60 mg/day for a 60kg person (24). Hence, the aluminum levels in the samples in this study were within the allowed (safe) range. Of course, the aluminum which is present in beverages, canned foods, medicines such as anti-acid syrups, food additives, and so on should also be taken into account (37). Noticing the irreparable risks of this element on nervous system, it is necessary to monitor the aluminum concentration in food and plants.

The toxic metals (aluminum, arsenic, lead, and cadmium) concentrations in the samples of this study were within the safe range. However, these medicinal plants are used by patients, who may have a number of problems in active body organs, infants, children, pregnant women, or those who continuously use such herbal products which may result in chronic poisoning. Therefore, medicinal plants should be cultivated in areas with the maximum distance from industrial centers.

CONCLUSION

All plants in this study were contaminated with lead, cadmium, arsenic, and aluminum, but they were within the safe range. If these plants are to be used for a short period of time or in small doses, washing can reduce the surface contamination to a great extant. Hence, it is suggested that consumers wash the medicinal plants prior to use.

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