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# Mercury uptake and phytotoxicity in terrestrial plants grown naturally in the Gumuskoy (Kutahya) mining area, Turkey

**Merve SASMAZ[1], Bunyamin Akgül[2], Derya Yıldırım[2], Ahmet SASMAZ[2]***

***Abstract*--This study investigated Hg uptake and transport from the soil to different plant parts by documenting the distribution and accumulation of Hg in the roots and shoots of 8 terrestrial plant species, all of which grew naturally in surface soils of the Gumuskoy Pb-Ag mining area, with continental climate.**

***Key Words:*** *Mercury uptake, wild plants, enrichment coefficient, translocation factor, phytoremediation, mining area*

## I. Introduction

Mercury is one of the most toxic heavy metals and is considered as a global contaminant. Its toxic level varies as a function of the exposure pathway and the chemical species in which mercury occurs [1],[2],[3]. It is also very expensive to clean up, because of its accumulative and persistent character in the biota [4]. As a highly bio-accumulated toxic metal in the food chain, mercury (Hg) possesses increasingly environmental concerns worldwide. There are many large Hg mines abandoned recently because of lower Hg prices and low demands. The presence of those abandoned Hg mines continues to impact the local environments through mine-wastes, drainage and elemental mercury vapor. Because the ecological and toxicological effects of Hg are strongly dependent on the chemical species present, the primary concern about those Hg mines is the biological accumulation [5]. Direct Hg contamination is usually the result of releases from abandoned Hg mines, gold-silver-thallium and other mining activities or the chlorine-alkali industry, while indirect (non-point source) contamination is largely attributed to atmospheric deposition originating from coal-fired power plants [5],[6],[7],[8]. It has been estimated that it would cost 40,000 to 70,000 US$ to remove each pound of Hg from the environment with currently available technologies; thus, there is an urgent need for the development of alternative Hg remediation strategies. The potential application of phytoremediation to Hg contamination has been explored in several environmental settings. There is evidence that certain plant species have the ability to extract and accumulate Hg both from atmospheric and soil sources, although no species with Hg hyperaccumulating properties has been identified [9]. The accumulation of mercury in terrestrial plants has been reported to be related to soil characteristics, including concentration of the element [10], but also the uptake of Hg has been found to be plant-specific [11], [12]. Soil characteristics such as high pH value, abundant lime and accumulated salt reduce its uptake by plants. A highly significant correlation exists between mercury and organic matter content in the top layer of forest soils [13]. In Turkey, many researchers have studied the distribution and speciation of Hg and heavy metals in foods, drinking and river waters, agricultural soils, and environmental samples [14], [15], [16].

The aim of the present study was to investigate Hg uptake and transport from soil to plant parts by studying distribution and accumulation of Hg in the roots and shoots of 8 wild plant species growing naturally in Hg-contaminated surface soils of the Gumuskoy Ag-Tl-As mining area in order to assess its Hg pollution degree and to contribute to the knowledge about the Hg soil/plant relationship.

## II. Material and Method

### A. The study area

In the present study, the plants and the associated soil samples were collected from an area by polymetallic ore deposits in the Gumuskoy mining district, Kutahya, Western Turkey. In this region, outcrops comprise of metamorphic, volcanic, and sedimentary rocks ranging from Permian to present-day. A number of polymetallic ore deposits represented by Ag, As, Tl, Pb, Zn and Sb occur between the Gümüşköy and Şahin villages. Soil and plants in the study area are naturally polluted by these heavy metals. This region has at least 3534±24 years of mining history, according to $^{14}C$ absolute age determinations on charcoal discovered in mining waste by [17]. Consequently, the area has been heavily charged with different metals arising from both ancient and modern mining activities [18],[19],[20]. Intensive mining operations continue in this region in the present day.

### B. Plant and soil samples

The plant samples, consisting of their roots-shoots together with their associated soils, were taken from forty-one sites in the study area. The plant species in the Gumuskoy region can grow under severe climate conditions due to their massive and deep-reaching root systems. These systems also give them the ability to live in areas deficient in organic matter. The Hg content was measured in 8 plant species that grow in the area: *Alyssum saxatile* L. (AL), *Anchusa arvensis* L. (AN), *Centaurea cyanus* L. (CE), *Glaucium flavum* (GL), *Onosma* sp. (ON), *Phlomis* sp. (PH), *Silene compacta* (SL) and *Verbascum thapsus* L. (VR). These plants were chosen because they are native and dominant species in the study area.

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[1]Department of Environmental Engineering, Firat University, Elazig, Turkey
*[2]Department of Geological Engineering, Firat University, Elazig, Turkey

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## *C. Preparation of Samples*

Soil Samples: Soil thicknesses in the study area vary between 30-40 cm. and 5-6 m. The soils are generally light-dark brown and black color, with a loamy and peaty clay texture (23.6% sand, 51.4% silt and 19.3% clay), with pH between 6.4 and 7.2, and with an organic matter content of 2.32-6.48 %. An X-ray diffraction study on the clay minerals was not performed. Soil samples were collected from around the roots of the plants at a depth of 30-40 cm. After drying in an oven at 100 $^oC$ for 4 h and removing rocks, the soil samples were ground using hand mortars. Soil samples were digested in a mixture of $HCl:HNO_3:H_2O$ (1:1:1, v/v; 6 ml per 1.0 g of soil) for one hour at 95 $^oC$. This treatment dissolved all soil samples except for silicates, and the digests were analyzed using ICP/AES & MS techniques for Hg at the ACME Analytical Labs, Vancouver, Canada (www.acmelab.com).

Plant samples: Plant samples were randomly collected from sites that were chosen based on representative characteristics of the Gumuskoy mining area. Three samples of shoots and roots were taken from each sampling site. The root samples were taken at a depth of 30-40 cm below the surface. The shoot and root samples of the studied plants were thoroughly washed with tap water, rinsed with distilled water, and dried at 100 $^oC$ in an oven for thirty minutes and then at 60 $^oC$ for 24 hours. A chelating EDTA wash was applied, and no differences were observed between EDTA washing and without EDTA washing. The dried plant samples (approximately 2.0-3.0 g) were ashed by heating at 300 $^oC$ for 24 hours. The ashed samples were digested in $HNO_3$ (Merck, Darmstadt, Germany) for one hour, followed by digestion in a mixture of $HCl:HNO_3$: $H_2O$ (1:1:1, v/v; 6 ml per 1.0 g of the ashed sample) for one hour at 95 $^oC$. The digests were analyzed using ICP/AES & MS techniques for Hg and the plant concentrations were calculated on a dry matter basis.

## *D. Enrichment coefficients of roots (ECR)*

Enrichment coefficients were found by calculating the ratios of specific activities in plant roots and soils (concentration in ppb of plant root divided by concentration in ppb of soil). This value is used as an index to determine accumulation of trace elements in plant parts or to establish the transfer of elements from soil to plant root [21].

## *E. Enrichment coefficient for shoots (ECS)*

Enrichment coefficients were also calculated for shoots (ECS) (concentration in ppb of plant shoot divided by concentration in ppb of soil). The ECS is a very important factor, as it indicates the capacity of a given species for phytoremediation [22] and this value is also used as an index to characterize the transfer of elements from the soil to the plant shoot. The ECS therefore characterizes the capability of a plant to absorb and transport metals from sediment and then to store them in the above-ground parts [23], [24], [25].

## *F. Translocation factors (TLF)*

Translocation factors (TLF) are obtained by calculating the ratio of metal in plant shoot to that in the plant roots (concentration in ppb of plant shoot divided by concentration in ppb of root). In a metal accumulator species, a translocation factor greater than 1 is common, whereas in metal excluder species, translocation factors are typically lower than 1 [26].

# III. Results and Discussion

## *A. Hg concentrations in soil*

The soil samples were collected from Aktepe and Gözeçukuru area in the study area and its surroundings. Hg contents of the soil samples were found out to be between 79 and 62532 ppb (mean: 7057 ppb). Hg concentrations in similar mining areas range from 8.4 to 610 ppm in Lanmuchang, Guizhou (southwestern China) Hg–Tl ore deposits [16]; 5 to 1710 ppm in Almadén mine (Spain) [27]; 12.1 to 100 ppm in Türkönü mercury mine, Turkey [28]; 0.21 to 3.4 ppm in ancient mining area of England [29] and 0.2 to 1.9 ppm in Hg mining of Canada; 2.6 to 2.9 ppm in Hg mining of France; 0.09 to 0.22 ppm in Hg mining of Brazil [30]. This mean value of Hg in the study area is lower than from mercury contents in the soils around these mercury deposits but many times higher than average mercury contents of lithosphere (0.05 ppm) and unpolluted soils (0.03 ppm) [31]. Mercury out of mining are also observed in soils around coal power stations and metallurgic plants (0.4-7.55 ppm), in some chemical works [32] and at former battery recycling facility [33]. This higher Hg concentration can be related to the Ag, Tl, As, and Pb deposits of the Gumuskoy region, because the presence of Hg showed a linear correlation with the occurrence of some heavy metals These linear correlations (r= 0.48-0.54) were observed between Hg and the heavy metals Pb, As, U, Sb, Tl and Ba, whereas weak linear correlations (r= 0.04-0.10) were observed between Hg and the heavy metals Sr, Cd, Ca and P (Table 1). The linear correlations between Hg and heavy metals (Pb, As, U, Sb, Tl and Ba) supported the idea that Hg and heavy metals were geologically transported jointly to the soil of the study area in hydrothermal solutions.

Significant relationships were detected between Hg content in all plant samples and in soils in this study. Hg in roots changes as a $2^{nd}$ degree polynomial regarding in soil. This means that the more Hg in soil the more Hg in root is accumulated. From the Fig. 1, it is also seen clearly Hg content in shoot increases exponentially with a decreasing slope. This indicates that roots function as a barrier for Hg to move upright to the shoots by accumulating more Hg with the increasing to Hg content in soil. The mechanisms for absorption and transport of Hg differed among plants in the study area. These variations in Hg content in different plants and plant tissues might be genetically controlled by the genotype of plants [34].

The highest Hg concentration in all analyzed soils was 62532 and 49871 ppb in the GL-01 and GL-02 samples (Fig. 1), which were collected from a mineralized area. These soils also had high As, Sb, Tl, Pb and Ba concentrations. Hg mobility in different surface conditions is believed to be medium in oxidizing conditions but high in acid and humid environments [35]. In the soils of the Gumuskoy study area, the maximum concentration of Hg was vertically accumulated

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at the surface or bulk close to surface. The soils with maximum Hg concentration like other heavy metals (Pb, Ag, Zn, Se, Sb, Tl) were generally in brown and dark color and contained more clay and organic matter. The Hg content decreased sharply with increasing depth of soil profile, where the soils were generally light brown color and contained more sand and rock parts and less organic matter.

### *B. Hg concentrations in plants*

In the Gumuskoy mining area, 8 plant species were selected for determination of Hg contents. The chosen plants grow indigenously in the mining area and generally live for one year or two years (annual). The Hg contents of plants in the study area varied considerably, but the average Hg concentrations for plant roots and shoot were 571 and 233 ppb, respectively. However, Hg concentrations of forty-one plant samples ranged from a minimum of 2 and 1 ppb for both plant roots and shoots to a maximum of 7328 and 1299 ppb for plant roots and shoots, respectively. Mercury concentrations (ppb dry weight) of plant parts are given in Fig. 1, together with Hg concentrations of the associated soils.

The mean Hg values in the soil, roots, and shoots of *Alyssum saxatile* (AL), were 5243, 746, and 155 ppb, respectively. The Hg levels in the soil around AL plants were significantly higher than the mean Hg values in AL shoots and roots. The Hg levels for all AL samples ranged between 45 and 2813 ppb for roots, and between 26 and 300 ppb for shoots on a dry weight basis (Fig. 1). The Hg enrichment coefficients (ECR and ECS) for AL roots and shoots are shown in Fig. 2; the mean ECR and ECS values were 0.10 and 0.04, respectively. Translocation factors (TLFs) for Hg in AL were between 0.06 and 2.26 (mean: 0.85) in this study (Fig. 2), which indicates that Hg was only weakly transferred to the shoot following uptake from the soil to root.

Hg concentrations in the soil, roots, and shoots of *Anchusa arvensis* (AN) are given in Fig. 1. Mean Hg values in the soil, roots, and shoots for AN were similar, at 6959, 485, and 500 ppb, respectively, on a dry weight basis (Fig. 1). The enrichment coefficients (ECR and ECS) for Se in the roots and shoots of AN, shown in Fig. 1.The mean values of 0.06 and 0.06, respectively, indicated that Hg taken up from the soil by AN was transferred to the root. The translocation factor (TLFs) of AN was 1.03 for two samples (Fig. 1); which meant that AN translocation factor was bigger than 1. This result indicates that AN has very well transporting capacity for Hg in semi-arid environments or continental climates.

The mean Se concentrations in the soil, roots, and shoots of *Centaurea cyanus* (CE) were 2052, 90 and 187 ppb, respectively (Fig. 1). The mean Hg values in the shoots of two CE samples were higher than the mean Hg values in the roots, but equal in one sample. However, the mean ECR and ECS of all samples were very low but the TLFs of CE samples were higher than 1 for two samples, equal for one sample (Fig. 2). This value indicates that CE is not very good bioaccumulator plant for Hg when growing in a similar environment and climate.

The mean Hg concentrations in the soil, roots, and shoots of *Glaucium flavum* (GL) were 56202, 5245 and 950 ppb, respectively. The ECR, ECS, and TLF values for GL (mean =0,09, 0,02 and 0,25 respectively) were lower than 1 (Fig. 2).

The mean Hg concentrations in the soil, roots, and shoots of *Onosma* (ON) were 6455, 87 and 171 ppb, respectively (Fig. 1). The Hg values in the shoots of all ON samples were higher than the Hg values in the roots, but lower than the Hg values in the soils. Therefore, the ECR and ECS values for ON are lower than 1, but the TLF values are higher than 1. These values indicate that the ON root does not accumulate Hg from the soil, but it efficiently transfers Hg to the shoot.

The Hg contents of the soil, roots, and shoots of *Phlomis* (PH) were examined in four samples. The mean Hg concentration of the soil, roots, and shoots of PH were 1960, 160 and 369 ppb, respectively. The Hg concentrations in shoots of two PH samples were higher than the Hg concentrations in the soil but other two samples were lower than the Hg concentrations in their soils. The mean ECR, ECS, and TLF values for PH were 0.21, 0.56 and 2.05, respectively. The ECS and TLF of PH-03 and PH-04 samples were than higher than 1 (ECS: 1.03 and TLF: 3.39 for PH-03 sample; ECS: 1.02 and TLF: 2.40 for PH-04 sample). These values indicate that PH would be effective at cleaning or rehabilitating the soils in areas contaminated by Hg.

The mean Hg concentrations in the soil, roots, and shoots of *Silene compacta* (SL) were 448, 8 and 14 ppb, respectively (Fig. 1). The Hg values in the soil were higher than in SL roots and shoots. The mean ECR and ECS values for this plant were lower than 1. The TLFs of SL were generally higher than 1 (mean 1.66), except for one sample. This means that Hg was not transferred from the soil to the root or the shoot by this plant (Fig. 2). This indicates that SL cannot act as a Hg bioaccumulator plant.

The Hg contents of the soil, roots, and shoots were analyzed in five samples of *Verbascum thapsus* (VR). The mean Hg concentrations in the soil, roots, and shoots of VR were 4625, 145, and 139 ppb, respectively. The Hg concentrations in VR shoots and roots were lower than the Hg concentrations in soil but the Hg concentrations in shoots of VR were higher than the Hg concentrations in roots of VR, except for one sample (Fig. 1). The mean ECR, ECS and TLF values for VR were 0.03, 0.06, and 2.47, respectively. The TLF values (2.47) were higher than 1 and this value indicates that VR has ability to transport from roots to shoots for Hg.

## IV. Conclusions

The Hg levels of soils from the Gumuskoy Ag-As mining area varied between 79 and 62532 ppb (mean: 7057 ppb), which are somewhat higher than those of uncontaminated surface soils reported in other countries. The distribution and accumulation of Hg was examined in roots and shoots of 8 different plant species growing naturally in the Gumuskoy soils. The mean concentrations of Hg in roots and shoots of these plants were found 571 ppb and 233 ppb, respectively. These results show that the fraction of available Hg for plants in soils of Gumuskoy mining zones was lower than in their

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soils and but higher than Hg concentrations of plants grown in uncontaminated areas, despite toxic levels of Hg are found in these soils. Hg concentrations in studied plants grown on these soils can be considered as phytotoxic, although no symptoms of Hg toxicity are observed in any of the studied plant species. The main reason for this, the roots of studied plants functioned like a barrier in root preventing that Hg from reaching the aerial parts of plants. However, according to TLF values, *A. arvensis* (AN), *C. cyanus* (CE), *Onosma* sp. (ON), *Phlomis* sp. (PH), *S. compacta* (SL), and *V. thapsus* L. (VR) showed a higher ability in increasing to Hg transport from roots to shoots. These results can make them good candidates for Hg phytoremediation of contaminated soils. *A. saxatile* L. (AL), and *G. flavum* (GL), had a lower ability to reduce Hg transport to shoots from roots because these plants behaved as excluders for Hg, storing the metal mainly in the root.

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About Authors:

Merve SASMAZ
PhD student in the Department of Environmental Engineering, Firat University, Elazig Turkey.
Research interests: Aquatic plants, Accumulation, Phytoremediation, Rehabilition of polluted areas

Dr. Bunyamin Akgul
Ass. Prof, Department of Geological Engineering, Firat University, Elazig Turkey
Specialist in rock and mineral geochemistry
Research interests: Petrology, granitoids, volcanic rocks

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Derya Yıldırım
She has a master level in geology science, Firat University, Elazig Turkey.
Research interests: Plant geochemistry, heavy metal pollution in soil and terrestrial plants

Ahmet SASMAZ
Professor, Head of Geological Engineering, Firat University, Elazig Turkey
Specialist in the field of soil and plant geochemistry
Research interests: Phytoremediation, heavy metal accumulation in the plants grown on the mining area, ore deposits, gas geochemistry

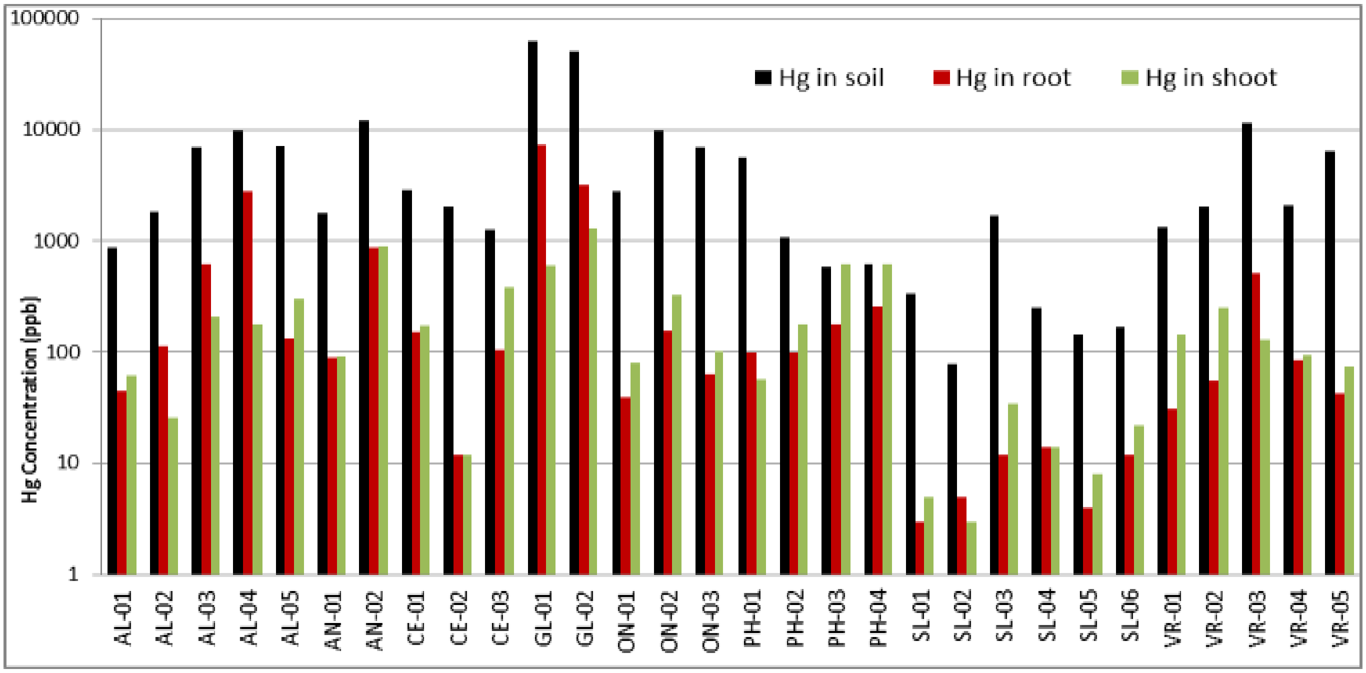


**Figure 1**: The Hg concentrations of soils, roots and shoots of 12 plant species.

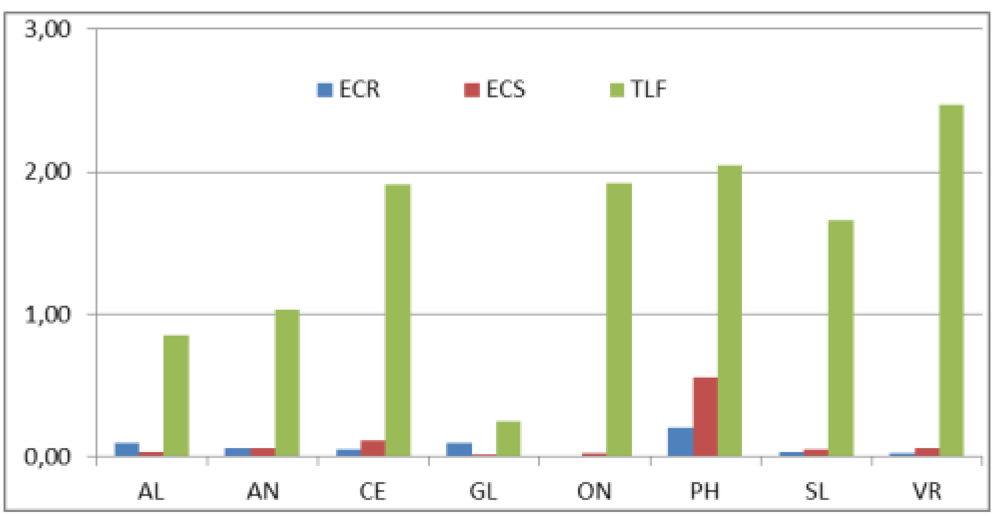


**Figure 2.** Mean translocation factors (TLF) and enrichment coefficients for roots (ECR) and shoots (ECS) of plants in the study area.