AFFORESTATION OF DOLOMITE GRASSLANDS WITH NON-NATIVE *PINUS NIGRA* IN HUNGARY AND ITS EFFECT ON SOIL TRACE ELEMENTS

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Abstract. The non-native *Pinus nigra* has been widely planted on natural dolomite grasslands in Hungary, yet little is known on its influence on soil properties. We compared soil micro-element concentrations in rock grassland (RG) and under *P. nigra* plantation (PP), both grown on north facing slopes of dolomite bedrock. At PP sites, the original vegetation was RG prior to afforestation. For both vegetation types, five sampling sites were selected, and at each site soil samples were taken from three depths (0-5, 5-10 and 10-15 cm). Micro-element concentrations of corresponding soil layers in the two vegetation types were compared. Under the pine plantation, the concentration of a number of soil trace elements was altered compared to the original rock grassland soil, and this effect increased with soil depth. At the deepest layer, significant differences were found for 10 microelements (Al, Fe, Mn, Ba, Cd, Co, Ni, Pb, Sr, Zn), and in each case the concentration was higher in PP than in RG soil. In contrast, the concentration of Cr, Cu, Hg, Mo and Se was not different between the two habitats throughout the soil layers sampled. Values exceeding the Hungarian environmental limits of background concentrations were detected for lead and cadmium. In the RG soil, concentrations of Pb and Cd were highest in the topmost layer, while for the PP sites these elements showed concentrations higher in the 10-15 cm layer than in the 5-10 cm depth. Concentrations of some other micro-elements showed similar inversion at PP sites. The depth inversion of these, often airborne pollutant trace elements was explained as a consequence of afforestation with *P. nigra* and such phytostabilization effect of the pine stands is assumed to increase with stand age.

Keywords: dolomite, heavy metals, *Pinus nigra*, rock grassland, soil pollution

Introduction

Austrian pine (*Pinus nigra* Arn.) has long been used in afforestation within and beyond its native range in the Mediterranean areas of Europe to prevent soil erosion (Barčić et al., 2006; Topić et al., 2008). Recently, *Pinus nigra* was involved in afforestation in other continents as well, owing to its wide ecological tolerance (Liu et al., 2005; Meamarian et al., 2007). In Hungary, where *P. nigra* is not native, its plantations were first established for soil erosion control and landscape protection purposes on steep dolomite slopes of the Hungarian Central Range in the beginning of the 20th century (Tamás, 2003). Later on, the primary goal of afforestations shifted towards wood production.

Recently, *P. nigra* stands cover 67,168 hectares that is 3.75 % of the total forested land of the country (Kottek, 2008). Foresters consider these afforestations as places of previously useless areas that were successfully integrated into wood production. Indeed, most of the stands are found on nutrient poor, low quality soils of either the steep slopes of dolomite hills (intensively grazed during last centuries), or sand dunes of the Danube-Tisza interfluve. Furthermore, *P. nigra* has not become an invasive species, in contrast to other exotic trees applied for afforestation of low quality habitats in Hungary, like *Robinia pseudoacacia* and *Ailanthus altissima* (Keresztesi, 1983; Udvardy, 1998). It was also suspected that *P. nigra* is able to improve soil quality, thus later on its stands could be followed by planting native forests, mainly oak stands (Tamás, 2003; Barčić et al., 2011).

In spite of arguments from the side of foresters, nature conservationists intensively criticize *P. nigra* plantations from two points of view. 1) These plantations destroy biodiversity of the former species rich grasslands that were supporting populations of several relict and endemic species (Zólyomi, 1942, 1987). 2) In practice, *P. nigra* stands were almost never followed by native trees in the rotation cycle, but instead were re-grown as non-native tree stands (Tamás, 2003). These points indeed highlight conflicts, but a society must decide what to prefer: conserving low productivity grasslands or producing wood, and if the latter the case, then should it be made via pine plantations or managing native deciduous forests instead. The decision should not be based merely on scientific arguments, of course. However, if wood production is the goal then it poses a scientific question: how do pine plantations affect the soil?

Surprisingly few works have been published on the soils of the Hungarian dolomite hills. Kovácsné Láng (1966) studied soils of rock grasslands, and Járó (1996) investigated forests of dolomite slopes, including pine plantations. Járó's work reported data on pH, humus- and macro-element contents of soils, but because of the lack of statistical approach his work was inappropriate to judge soil improvement under pine stands. Therefore, our group made a detailed study of soil properties examined by Járó, and concluded that pine plantations did not improve soil quality (Anton et al., 2008).

However, questions about the micro-element contents still remained open, because rock grasslands and pine stands were never studied from this point of view earlier. Since the afforestation of grassland habitats is obviously a major change, we hypothesized that the micro-element contents of the soil is altered during this vegetation transformation. To test this, we compared micro-element contents of soils of *P. nigra* stands and rock grasslands based on standardized soil sampling followed by statistical evaluation. Such comparisons can reveal important effects that should be encountered in the evaluation of the success of pine plantations establishment.

Materials and methods

The study region was the Buda-Hills (near Budapest, Hungary) where rock grasslands and *P. nigra* plantations are found over large areas. The Buda-Hills are formed by Triassic dolomite bedrock that are fragmented by surface erosion into several hills elevating 250-600 m above see level (Pécsi, 1958). Hill-slopes are steep (25°-50°) and are covered by shallow rendzina ("Leptosols" group according to the system of WRB 2006). The climate of the region is semiarid temperate, annual mean temperature is 9.9 °C, yearly precipitation is 575 mm (Marosi & Somogyi, 1990). A dry period of one or two months is common in summer and causes particularly arid conditions on the hill slopes of shallow soil. On the north facing slopes the typical vegetation is closed rock grassland (RG) dominated by *Festuca pallens* and *Bromus erectus*. *Pinus nigra* stands (PP), created by afforestation after World War II, are also found on north facing slopes. The stands selected for sampling were even-aged, 50 years old plantations with canopy cover of 80-95% and tree height of 14-18 m. For each sampling site, the presence of rock grassland prior to afforestation was ascertained based on historical photographs.

For both vegetation types (RG and PP), five stands were selected for sampling (*Table 1*). In each stand, four soil samples were taken at least 4 m apart from each other on 19th March (RG stands) and on 24th April (PP stands) in 2002. Soil samples were taken with soil corer from three depths: 0-5 cm, 5-10 cm and 10-15 cm. Soil samples were air-dried in the laboratory, then skeleton (d >2 mm), roots, twigs and other plant fragments were removed by sieving. The corresponding layers of the four soil cores taken from a stand were combined.

Table 1. *Basic geographical data of the 5-5 sampling sites for closed rock grassland (RG) and for Pinus nigra plantation (PP) in the Buda Hills, Hungary*

Short name	**Slope aspect**	**Slope angle**	**Altitude a.s.l. (m)**	**GPS-coordinates**	
				Latitude	**Longitude**
RG	NW(320°)-N(20°)	25-30°	385-400	1. 47° 35.695’	18° 52.778’
				2. 47° 35.699’	18° 52.779’
				3. 47° 35.709’	18° 52.803’
				4. 47° 35.767’	18° 52.739’
				5. 47° 35.767’	18° 52.727’
PP	N(350°)-N(20°)	20-30°	390-405	1. 47° 35.452’	18° 53.928’
				2. 47° 35.445’	18° 53.915’
				3. 47° 35.440’	18° 53.903’
				4. 47° 35.455’	18° 53.934’
				5. 47° 35.452’	18° 53.918’

The following methods were used for soil analysis: a) pH: standard pH measurements were made in a suspension of 1:5 soil:water; b) Ca^{2+}, Mg^{2+}, Na^{+} and K^{+} concentrations were also determined in 1:5 soil:water suspension using flame photometer; c) organic matter content (expressed in weight ratio), according to Tyurin (1951); d) K_2O: plant available-K was determined with Lakanen-Erviö extraction (NH_4-acetate + EDTA, Lakanen & Erviö, 1971); e) total As-, Ba-, Ca-, Cd-, Co-, Cr-, Cu-, Hg-, Mg-, Mo-, Na-, Ni-, Se-, Sn-, Pb- and Zn-content were analysed by digestion with Aqua regia (1.0 g soil + 4.5 cm^3 HCl + 1.5 cm^3 65 m/m% HNO_3 + 1.0 cm^3 30 m/m% H_2O_2) for 15 minutes in microwave chamber, then the samples were allowed to cool down, transferred quantitatively to a volumetric flask and filled to 50 cm^3 with doubled deionised water. Before measurements, the samples were allowed to settle down. Jobin-Yvon Ultrace 2000 plasma spectrometer was used to determine quantities of trace elements.

In the statistical analyses, values of soil properties were compared by Student's *t*-tests between corresponding soil layers of rock grassland and pine plantation sites. Gaussian distributions (normality) of data was tested using the method of Kolmogorov and Smirnov. The F-test was used to determine if the standard deviations of the compared data sets were equal. Differences were considered significant at $p<0.05$ probability level. For the analyses the SAS ver. 6.4 (SAS, 1989) and the Graphpad Instat (InStat, 2003) softwares were used.

Table 2. *Comparisons of trace element contents in the 0-5 cm soil layers of the closed dolomite rock grassland (RG) and the Pinus nigra plantation (PP), based on t-test (n= 5)*

Digestion method	**Soil properties**	**RG**		**PP**		
		mean	**S.d.**	**mean**	**S.d.**	**sign.**
1:5 soil: water extract	Organic matter, w%	24.9	6.5	20.5	0.7	n.s.
	pH (H_2O)	7.4	0.1	7.1	0.1	++
	Ca^{2+}, mg/kg	271	31.1	161	14.8	++
	Mg^{2+}, mg/kg	105	13.7	70.4	8.5	++
	Na^{+}, mg/kg	5.7	1.1	6.0	2.4	n.s.
	K^{+}, mg/kg	19.5	2.7	2.8	3.8	++
LE-method	K_2O, mg/kg	186	41.5	185	18.0	n.s.
extracted by aqua regia	Ba, mg/kg	9.6	0.9	11.1	1.2	+
	As, mg/kg	0.4	0.2	0.4	0.1	n.s.
	Hg, mg/kg	0.1	0.1	0.0	0.0	n.s.
	Se, mg/kg	0.0	0.0	0.0	0.1	n.s.
	Mo, mg/kg	0.0	0.0	0.0	0.0	n.s.
	B, mg/kg	4.6	0.6	3.8	0.5	+
	Zn, mg/kg	54.3	15.8	30.7	4.6	+
	Pb, mg/kg	46.2(!)	17.3	30.3(!)	4.9	n.s.
	Co, mg/kg	0.7	0.1	1.4	0.4	+
	Cd, mg/kg	0.8(!)	0.2	0.8(!)	0.0	n.s.
	Ni, mg/kg	1.5	0.3	2.5	0.3	++
	Fe, mg/kg	529.2	100.6	474.0	39.9	n.s.
	Cr, mg/kg	0.4	0.6	0.2	0.0	n.s.
	Mn, mg/kg	218.6	39.5	304.6	88.3	+
	Cu, mg/kg	10.1	10.3	4.8	0.3	n.s.
	Al, mg/kg	153.0	38.9	192.8	35.9	n.s.
	Sr, mg/kg	6.9	0.8	6.2	0.4	n.s.
	<2mm fraction, w%	55.3	16.4	100.0	0.0	

Abbreviations: ++ = significant differences at p< 0.01 level; + = significant difference at p< 0.05 level; n.s. = not significant (p> 0.05); sign.= level of significance; * = components showing considerable differences, but their significance were not tested (because some "0" values in the data set); ! = concentrations above the limit of Hungarian environment protection rules; LE = Lakanen and Erviö; w% = weight percentage. Concentrations below the lower detection limit were uniformly replaced by zero values.

Table 3. *Comparisons of trace element contents in the 5-10 cm soil layers of the closed dolomite rock grassland (RG) and the Pinus nigra plantation (PP), based on t-test (n= 5). Abbreviations are the same as listed for Table 2.*

Digestion method	**Soil properties**	**RG**		**PP**		
		mean	**S.d.**	**mean**	**S.d.**	**sign.**
Tyurin	Organic matter, w%	18.3	2.5	11.2	1.9	++
1:5 soil: water extract	pH (H_2O)	7.4	0.0	7.4	0.1	n.s.
	Ca^{2+}, mg/kg	226	8.2	169	10.9	++
	Mg^{2+}, mg/kg	87.1	3.9	62.4	7.3	++
	Na^+, mg/kg	5.6	0.7	0.5	0.9	++
	K^+, mg/kg	14.9	2.6	0.4	0.8	++
LE-method	K_2O, mg/kg	124	8.7	171	14.4	++
extracted by aqua regia	Ba, mg/kg	7.3	1.0	9.8	1.6	++
	As, mg/kg	0.3	0.2	0.1	0.1	+
	Hg, mg/kg	0.1	0.1	0.0	0.0	n.s.
	Se, mg/kg	0.0	0.0	0.0	0.0	n.s.
	Mo, mg/kg	0.0	0.0	0.0	0.0	n.s.
	B, mg/kg	4.8	0.4	4.3	0.5	+
	Zn, mg/kg	38.3	37.0	10.1	2.5	n.s.
	Pb, mg/kg	29.7(!)	9.7	12.4	1.3	++
	Co, mg/kg	0.6	0.2	1.5	0.4	++
	Cd, mg/kg	0.5	0.1	0.6(!)	0.1	+
	Ni, mg/kg	0.9	0.1	1.5	0.4	++
	Fe, mg/kg	383.2	53.9	280.6	38.2	++
	Cr, mg/kg	0.1	0.0	0.1	0.0	n.s.
	Mn, mg/kg	186.8	41.0	354.4	94.5	++
	Cu, mg/kg	3.1	0.5	2.8	0.4	n.s.
	Al, mg/kg	88.2	16.9	89.7	24.9	n.s.
	Sr, mg/kg	6.1	0.4	5.8	0.6	n.s.
	<2mm fraction, w%	48.3	13.8	100.0	0.0	

Results

Results are focused on remarkably significant differences of micro-element contents of the studied soils. Special attention is paid to pollutant trace elements, considering the pollution threshold limits in Hungary (KÖM, 2000). Tables also contain data on pH, organic matter content and the main macro-element concentrations of the soil to better inform readers, although a detailed evaluation of these data was given earlier (Anton et al., 2008).

In the upper 5 cm soil layer, concentrations differed significantly for six trace elements between samples taken from the rock grasslands and the pine plantation (Table 2). Concentrations of barium (Ba), cobalt (Co), nickel (Ni) and manganese (Mn) were higher in the soils of pine plantations, whereas that of boron (B) and zinc (Zn) were higher in the rock grasslands' soils. Actual concentrations of lead (Pb) and cadmium (Cd), two toxic heavy metals, exceeded the reference limits of background concentrations at both sites with more or less the same extent.

In the 5-10 cm soil layer, differences of trace element concentrations increased between the two vegetation types, and were significant for nine elements (*Table 3*). In the soils of pine plantations, similarly to the 0-5 cm layer, higher concentrations were detected for Ba, Co, Ni and Mn. Furthermore, the concentration of cadmium (Cd) was also significantly greater here than in the rock grasslands' soils. At the same time, concentrations of arsenic (As), boron (B), lead (Pb) and iron (Fe) were significantly higher in the soil of closed rock grasslands than that of pine plantations. Concentrations exceeding the limit of corresponding background values were found in this depth for two micro-elements only: lead (Pb) was above limits in the rock grasslands, while cadmium (Cd) was so in the pine plantations.

Table 4. *Comparisons of trace element contents in the 10-15 cm soil layers of the closed dolomite rock grassland (RG) and the Pinus nigra plantation (PP), based on t-test (n= 5). Abbreviations are the same as listed for Table 2.*

Digestion method	**Soil properties**	**RG**		**PP**		
		mean	**S.d.**	**mean**	**S.d.**	**sign.**
	Organic matter, w%	12.0	4.8	12.9	2.7	n.s.
1:5 soil: water extract	pH (H$_2$O)	7.5	0.1	7.3	0.1	n.s.
	Ca^{2+}, mg/kg	200	14.4	165	9.8	++
	Mg^{2+}, mg/kg	65.7	25.2	64.1	6.3	n.s.
	Na^{+}, mg/kg	4.4	2.7	0	0	*
	K^{+}, mg/kg	12.4	2.8	0	0	*
LE-method	K$_2$O, mg/kg	91.2	19.1	218	6.5	++
extracted by aqua regia	Ba, mg/kg	5.0	1.6	10.8	1.2	++
	As, mg/kg	0.2	0.1	0.2	0.1	n.s.
	Hg, mg/kg	0.1	0.1	0.0	0.0	n.s.
	Se, mg/kg	0.0	0.0	0.0	0.0	n.s.
	Mo, mg/kg	0.0	0.1	0.0	0.0	n.s.
	B, mg/kg	3.9	0.6	4.3	0.6	n.s.
	Zn, mg/kg	11.7	6.9	19.2	3.7	+
	Pb, mg/kg	12.7	1.5	18.6	2.1	++
	Co, mg/kg	0.5	0.2	1.6	0.4	++
	Cd, mg/kg	0.3	0.1	0.7(!)	0.1	++
	Ni, mg/kg	0.5	0.1	1.9	0.3	++
	Fe, mg/kg	261.2	44.6	339.2	41.9	+
	Cr, mg/kg	0.4	0.7	0.2	0.0	n.s.
	Mn, mg/kg	135.2	57.3	350.0	90.9	++
	Cu, mg/kg	6.6	11.0	3.7	0.4	n.s.
	Al, mg/kg	68.0	34.0	122.7	27.3	+
	Sr, mg/kg	5.0	0.3	6.3	0.6	++
	<2mm fraction, w%	37.8	17.1	>90		

In the 10-15 cm soil layer, further increase was observed in the number of trace elements expressing significant concentration differences between RG and PP sites. Noteworthy, that the 10 trace elements showing significant differences (Ba, Zn, Pb, Co, Cd, Ni, Fe, Mn, Al and Sr) always reached higher concentrations in the soils of PP than that of RG (Table 4). In this depth, cadmium (Cd) was the only trace element that surpassed the corresponding limits of background concentrations, as 0.7 (+/-0.1) mg/kg was measured in the pine plantations. It was also remarkable that lead (Pb) concentration was significantly higher under pine stands than at the grassland sites (as opposed to the two upper layers), although it remained within the permissible background limits. In the soils of pine stands, an additional feature of lead concentration was observed: it reached higher value in the 10-15 cm layer than in the 5-10 cm depth. Further two trace elements, zinc and cadmium, displayed a similar concentration inversion: both had significantly higher concentrations in the lowermost soil layer than in the intermediate soil layer of pine stands. In contrast to pine stand soils, lead-, zinc- and cadmium concentrations in the rock grassland soils always showed monotonic decrease from upper to lower layers.

Discussion

Our results showed that under the pine plantation the concentration of a number of soil micro-elements is altered compared to the original rock grassland soil, and this effect increased with soil depth. While in the 0-5 cm and 5-10 cm depths the direction of change varied depending on the microelement, in the deepest soil layer (10-15 cm) the pine stand soil exceeded the rock grassland soil in the concentration of each trace element which differed between the two habitats. Only few trace elements displayed differences consistent across all soil layers studied: Ba, Co, Mn and Ni concentrations were higher in the PP than in the RG throughout the 0-15 cm soil depth. The lack of difference between the PP and RG soils were consistent in each soil layer for the elements Cr, Cu, Hg, Mo and Se. The rest of the micro-elements investigated (Al, As, B, Cd, Fe, Pb, Sr and Zn) displayed depth-specific patterns between the two habitats. For the pine plantation, it was also documented that the concentration of certain trace elements were significantly higher in the 10-15 cm than in the 5-10 cm soil layer.

In our study, the two trace elements (Pb and Cd) that were detected in concentrations above the permissible background limits are typically known as airborne pollutants (Migon et al., 1991; Cizmecioglu & Muezzinoglu, 2008). Today, the study area is a nature reserve with no inhabitants, and historically it was used for sheep grazing. Therefore, there is certainly no local source of metal pollution. Instead, the Pb and Cd detected in the soil should originate from anthropogenic activities in the neighborhood.

In this respect, three different emission sources can be considered. The first one is a main road that connects Budapest with Dorog Industrial Region. Traffic roads are well known as heavy metal pollution sources, especially during the middle and late 20th century, prior to the introduction of led-free gasoline in the 1990’s (Harrison & Laxen, 1981; Zupančič, 1999; Enayatzamir, 2008).

The second possible source is the open air dolomite quarry at Pilisvörösvár, which was first mined at the end of the 19th century and operates continuously since then. According to literature data, limestone and dolomite quarries, together with related stone crushers can be responsible for heavy-metal emission (Connor & Shacklette, 1975; Durn et al., 1993).

Both the main road and the quarry are about 3-3.5 km distance from the sampling sites located under the main wind direction (N-NW) of the region. Therefore, these pollution sources most probably contribute to the trace element content of the studied soils.

The third pollutant factor is likely the capital, Budapest. Its population of about 2 million inhabitants with the associated traffic and other anthropogenic activities make it a serious source of heavy metals. Although Budapest is situated 10 km South-East from the study area, under certain weather conditions its emission can reach the region. Other big cities have been reported as sources of considerable heavy metal pollution on their whole surroundings (McDonnell et al., 1997; Li et al., 2011).

The detection of inverse depth distribution for Cd, Pb, and Zn is an unexpected finding of our study, thus it requires discussion. These trace elements are typically airborne pollutants, thus their concentration is usually highest in the uppermost soil layer and then decreases with depth (Adriano, 1986; Davies, 1990; Sipos et al., 2011). Such typical depth distribution was found in the soil of the studied rock grasslands (*Tables 2-4*). However, in the soil of the pine plantation concentrations of Zn, Pb and Cd were significantly higher at depth of 10-15 cm than at depth of 5-10 cm (Tables 3 and 4). In addition, several other trace elements showed similar inversion in concentration distribution although these differences were not significant.

A possible cause of the detected inversion phenomenon in the pine plantation could be associated with the biological effect of *P. nigra* itself. Pine species – as well as other conifers – are known to reduce soil pH beneath their stands (Pallant and Riha, 1990; Thelin et al., 1998). Also for the studied stands, a moderate shift of pH from 7.4 to 7.1 was detected in the upper soil layer (*Table 2*). It is known that soil acidification enhances solubility of metal compounds, thus increases concentration and mobility of metal ions in the soil solution (Strom, 1997; Szalai et al., 2010).

Plant roots (by root exuded organic acids) and also their rhizosphere play active role in soil acidification, therefore, mobility of heavy metal ions can be supported by pine roots towards deeper soil layers. This process could partly be responsible for the accumulation of certain trace elements in deeper soil layers of the pine stands, since pine roots reach much deeper soil horizons than the root system of perennial grasses and herbs that forms the vegetation of the rock grasslands. However, dissolubility studies showed that Pb appears to be strongly bound to the soil matrix, due to complexation and specific adsorption processes (Abumaizar and Smith, 1999; Sipos et al., 2008). Sauve et al. (1998) reported that the solubility of Pb shows a linear decrease from pH 3 to 6.5 and is independent of soil organic matter in that pH range. From pH 6.5 to 8, higher pH promotes the formation and dissolution of organo-Pb complexes, which increase Pb solubility. In this pH range, higher organic matter content results in higher concentrations of dissolved and labile Pb.

In general, cadmium expresses higher mobility than lead, and Cd should be fairly mobile in soils of pH 4.6 to 6.6. However, above pH 7 cadmium shows reduced mobility and in case of soils with high calcium carbonate concentration its co-precipitation with $CaCO_3$ can occur (Filep, 1999). Conclusively, soil acidification due to pine plantation might have a role in the accumulation of polluting trace elements in deeper soil layers, but these movements of metals could be considerable only adjacent to pine roots where the calcareous soil particles are unable to compensate pH decrease caused by the root exuded acids.

A further anthropogenic factor may also be involved in the detected concentration inversion in the pine stands' soil. It is known that the studied pine stands were planted around 1950, within the frame of the so called "barren land afforestation" programme (Tamás, 2003). Prior to bedding out the pine seedlings, narrow terraces (for strip cultivation) were prepared on the dolomite hill-slopes using traditional hand-tools (spade, hoe and pickaxe). This operation obviously resulted in turning over the soil layers, and it could contribute to the movement of polluting heavy metals to deeper layers.

Our results indicate that the establishment of *P. nigra* plantations can be considered as a possible way for reducing concentrations of heavy metals in the upper soil layer, because during this operation soil surface contaminants are placed to deeper soil layers either by immediate turning of soil layers, or later transportation processes along roots. Consequently, uptake of toxic heavy metals by ground layer vegetation of the polluted soils is limited, thus their further passage to food-web is practically prevented. The phytostabilization effect of these plantations increase further with aging (more closed) stands by slowing down near-surface wind velocity, thus reducing concentrations of heavy metal contaminants in the wind-blown dust almost to zero.

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