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Biomonitoring of Airborne Heavy Metals Pollution by *Delonix regia* (Boj. ex Hook.) Raf. in Greater Cairo, Egypt

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Delonix regia leaves and bark samples were collected from ten sites along the main motorways from urban and industrial areas of the Greater Cairo (Egypt). Samples were analyzed for the following trace metals by using Inductively Coupled Plasma Mass Spectrometer: Pb, Co, Zn, Fe, Ni, Mn, Cu and Al. Concentrations of the trace metals in leaves were significantly higher (at $p < 0.05$) than in bark and were either within or below the normal ranges. In all the polluted sites, Zn, Co and Ni were the most prevalent and enriched trace metals in the leaves. The Fe concentrations were the highest detected element in the bark in all sites. The ratios of Co, Zn, Ni and Pb concentrations in leaf to bark were high in comparison to the ratios of other metals. The spatial variability of trace metals was only significant (at $p < 0.05$) for Co, Zn and Ni at high probability levels and hangs on the traffic and industrial burden in each site. Our results indicated that *Delonix regia* is an adequate biomonitor of heavy metals pollution in the Greater Cairo, Egypt.

Key Wards: *Delonix regia;* leaves; bark; heavy; metal pollution; biomonitoring; Greater Cairo.

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Introduction

It has been suggested that direct uptake through bark or foliage may be a major pathway by which metals enter trees, particularly in heavily polluted areas (Baes and McLaughlin, 1987). As trees are very efficient in trapping atmospheric particles, which is especially important in urban areas, the use of leaves primarily as biomonitors accumulating trace metals has acquired great ecological importance (Tomas Dvic' *et al.*, 2005; Ukpebor *et al.*, 2010; Al-Khashman *et al.* 2011). Monitoring bark has been found to be a useful bioindicator and biomonitor for airborne pollution tests and supplies low-cost information on the composition and magnitude of pollutant deposition (Kuik and Wolterbeek, 1995; Bohm *et al.*, 1998; Schulz *et al.*, 1999; Bellis *et al.*, 2001; Harju *et al.*, 2002; Migaszewski *et al.*, 2005; Mingorance *et al.*, 2005; Samecka-Cymerman *et al.*, 2009; Ukpebor *et al.*, 2010). The most important anthropogenic sources of metals into the atmosphere are automobile exhaust, industrial emissions, incineration of refuse, agricultural chemicals and inorganic fertilizers (Deboudt *et al.*, 2004).

Delonix regia (Boj. ex Hook.) Raf. (Royal Poinciana in English) is one of the most common leguminous trees in the frost free tropical climates in the world and it is found lining streets in some of the world's most beautiful places (Davison, 2004). The species is now considered the most common street tree in Greater Cairo (GC), Egypt (Hadidi and Boulus, 1989). For this reason, it was easier to represent all the sampling sites in GC using *Delonix regia* more than other tree species due to the difficulty of sampling from other trees (e.g. palms) and/or the infrequent presence of the other tree species in the sampled sites.

GC is the capital of Egypt and is the most overpopulated city in Africa with a population over 1923 million in 2009 (CAPMAS, 2006). Cairo is considered as one of the highly polluted cities in the world due to the increase of urbanization and industrialization processes in and around it. GC houses about more than 1.3 million vehicles, running in the streets (ca. 30 % of the total registered motor- vehicles in Egypt) in addition to many industrial factories and workshops (Shakour *et al.*, 2006; Ministry of State for Environmental Affairs, 2008; Safar and Labib, 2010). Due to the exponential increase in the number of vehicles, the vehicular traffic emissions represent the main source of air pollution in GC (Ministry of State for Environmental Affairs, 2008). In addition, GC has two huge industrial areas in its northern and southern borders (Shoubra El-Kheima and Helwan, respectively) which add more pollution burden on the air quality (Safar and Labib, 2010).

Consequently, this study was designed to assess the atmospheric levels of trace metals in GC by using the most common street tree: *Delonix regia* as biomonitor and to assess the suitability of this species as a biomonitor for a1mospheric metals in GC. Moreover, the spatial variations in the trace elements concentrations in the trees and their relation to the local anthropogenic activities in GC will be assessed.

Material and Methods

a)Study rites description

Ten sampling sites (Fig.I) were located along the main motorways in GC. passing from North to South. The selected sites were located within two areas. The first was an industrial area and characterized by heavy traffic and heavy population and located in Shoubra El-Kheima, Helwan, Matariya and Tura. The second one is an urban area with heavy traffic and located in Rod El-Farag , El-Sahe Ramsis, Attaba, Maadi and Giza.

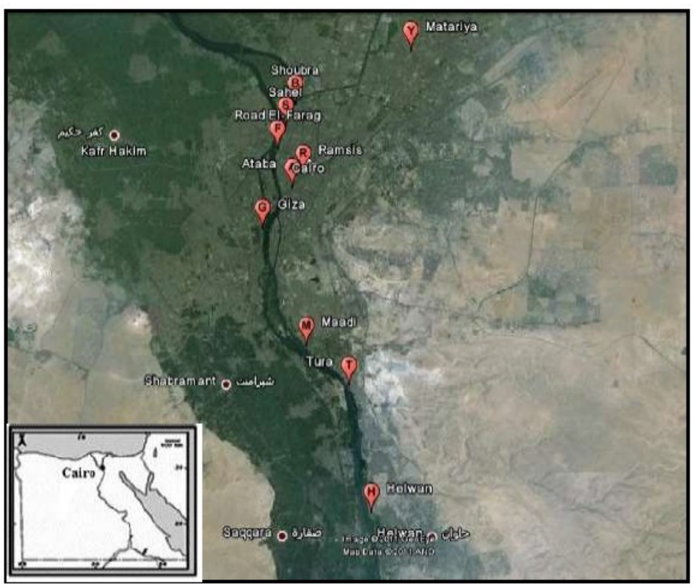

Fig.1. The location map for contaminated sites in Greater Cairo (GC), Egypt

a) Plant sampling and analysis

Sampling and analysis procedures of leaf and bark were according to the methods proposed by Kozlov *et al.* (2000), Oliva and Rautio (2004) and Ukpebor *et al.* (2010) . The samples were collected in August and September 2009. Samples were obtained from *5* individual trees in each site, by collecting leaves from the uppermost branches at the border of the tree, at a height of 2-3 m above ground surface. Each tree was represented by one composite leaf sample with total of *50* samples from all sites. Leaflets were separated from the branches at the laboratory excluding petioles and rachis. From the same trees, bark samples were collected from all sides of the tree trunk. The leaves and bark samples were dried in an oven at 70°C for 48 h and then milled. The samples (0.5 g) were ashed at 550°C in muffle furnace for 3 hours (Kozlov *et al.,* 2000; Oliva and Rautio, 2004; Ukpebor *et al.* 2010) and then digested with 10 ml nitric acid (2.8%) overnight. The sample volume was brought to *50* ml using ultra pure distilled water and stored in *50* ml polypropylene bottles. Diluted solutions (10%) were analyzed for Pb, Co, Zn, Fe, Ni, Mn, Cu and Al using Inductively Coupled Plasma Mass Spectrometry (ICP-MS; Agilent 7500). The ICP multi-element standard solution (Merck, VI CertiPUR®) diluted to concentrations of 10, 100 and 1000 ppb and a 5.00 mMKHiP0$_4$ standard diluted to -100, 1000 and 10000 ppb (K and P) was used to construct a linear calibration graph for the analysed elements. An internal standard solution containing 800 ppb indium was added to the samples during analysis. The results were calculated on dry weight basis. The samples analysis was carried out in the Department of Earth Sciences, University of Gothenburg, Sweden. It is worth mentioning that the determined trace metals of this study have been reported as the most monitored and abundant contaminants in air and particulate matters in Greater Cairo (Shakour *et al.* 2006; Ministry of State for Environmental Affairs, 2008; Safar and Labib, 2010).

b) Statistical analysis

Non-parametric Spearman correlation coefficients between the metal concentrations for leaves and bark were calculated. Differences between sampling sites in terms of concentrations of elements in leaves and bark were evaluated by ANOVA. The t-test was applied to absolute data to compare the spatial variations of metal concentrations in the leaves and bark of the contaminated sites. For each element concentration, the ratio between leaf and bark at each site was calculated. Statistical packages for exploratory and correlation analyses were STATISTICA 7.0 (StatSoft Inc.) and SPSS 12.0 for Windows (SPSS Inc.).

Results

Concentration of heavy metals in the leaf samples

The values of all determined elements in leaves and bark samples of *Delonix regia* from the contaminated sites are given in Table I and 3.The concentrations of investigated metals in leaves were significantly higher (at p-value < 0.05) than in bark samples. Zn, Co and Ni were the most prevalent and enriched trace elements in plant leaves in contaminated sites. Shoubra El-Kheima site had the highest detected concentrations for all metals. On the contrary, Giza and Ataba sites had the lowest concentrations. The ranges of the elemental concentrations of this study lie within or below the normal ranges reported by other investigators (Alloway, 1995; Oliva and Mingorance, 2006). The Pb concentrations varied from 0.7 to 2.6 mg kg^{-1} in the leaves (Table 1). The concentration of Co varied from 7.9 to 11.9 mg kg^{-1} while this range varied from 33.1 to 79.4 mg kg^{-1} for Zn. The Fe concentrations varied from 0.9 to 2.6 mg kg^{-1} and few samples exceeded 1.8 mg kg^{-1}, while Ni concentrations ranged from 5.3 to 12.1 mg kg^{-1} with values frequency 6.4 – 8.6 mg kg^{-1} for most samples. The Mn and Al concentrations in the leaf samples were more or less comparable with very narrow range which did not exceed 1.1 mg kg^{-1}. Cu was the least detected element in the plant leaves with very low concentrations in all samples (Table 2).

In all the sampled sites, Cu was significantly correlated (p-value < 0.05) with Pb, Mn and Al ($r = 0.8$, 0.6 and 0.7, respectively), while Pb showed high significant correlation with Al ($r = 0.8$) (Table 2).

Table 1.The mean values ± standard deviation for the heavy metals concentrations (mg kg-[1] DW) in leaves samples

Site	Pb	Co	Zn	Fe	Ni	Mn	Cu	Al
Shoubra m-kheima	**2.6±1.4**	11.5±0.8	79.4±22.7	2.6±1.2	**12.1±2.6**	1.1±0.1	0.10±0.01	1.0±0.3
El-Sahel	1.1±0.0	11.3±0.0	39.9±0.l	1.7±1.8	10.7±0.l	0.7±0.2	0.05±0.01	0.6±0.l
Rod El-Farag	1.7±0.5	9.4±4.8	48.9±14.8	1.8±0.4	8.6±2.8	0.7±0.2	0.06±0.01	0.9±0.l
Ramsis	1.1±0.4	11.4±0.8	54.6±8.27	1.4±0.3	8.2±1.1	0.8±0.3	0.10±0.01	0.7±0.1
Ataba	1.3±0.2	7.9±1.3	42.4±8.11	1.0±0.2	5.3±1.1	0.8±0.2	0.08±0.02	0.8±0.l
Matariya	1.6±0.2	11.1±0.l	39.3±8.24	1.0±0.2	8.3±2.5	0.8±0.2	0.10±0.01	1.0±0.2
M""1i	1.1±0.3	9.0±2.9	47.5±11.4	1.2±0.4	6.4±2.8	0.5±0.0	**0.07±0.00**	0.6±0.0
Tura	0.7±0.3	9.9±2.2	**33.1±8.62**	1.1±0.2	6.7±2.7	0.5±0.2	0.05±0.01	0.9±0.2
Helwan	**1.8±1.0**	11.9±1.6	33.3±6.01	1.2±0.l	6.8±0.2	0.7±0.0	**0.11±0.00**	1.0±0.0
Giza	0.7±0.4	9.1±3.9	34.9±17.8	0.9±0.5	7.0±4.5	0.2±0.2	0.04±0.01	0.7±0.0

Table 2. Spearman correlation coefficient between the heavy metals in leaves samEles. Values in bold are si!l!!ificant at E < 0.05

	Pb	Co	Zn	Fe	Ni	Mn	Co
Co	0.4						
Zn	0.4	0.1					
Fe	0.5	0.6	0.6				
Ni	-0.03	0.3	0.2	0.3			
Mn	0.6	0.4	0.6	0.4	0.1		
Cu	08	0.6	0.2	0.3	-0.2	06	
Al	08	0.5	0.1	0.2	0.1	0.6	0.7

Concentration of heavy metals in the bark samples

In general, the measured elements showed low concentrations in the bark samples. The Fe concentrations showed the highest concentrations in comparison to other metals analyzed in all sampled sites (fable 3). The Fe content ranged from 3.1 to 5.3 mg kg-[1] which is below the normal range in plants reported by other investigators (Kabata-Pendias and Pendias, 1992; Cicek and Koparal, 2004). The highest concentrations of Pb, Co and Zn were detected in Shoubra El-kheima site while the highest concentrations for Fe and Mn were detected in the samples collected from Ataba site. On the other hand, the concentrations of Ni, Cu and Al were very low in comparison to other analyzed elements (Table 3).

The Fe content in bark samples showed high significant correlation coefficient (p-value < 0.05) with Co (r = 0.9), Ni (r = 0.6), Cu (r = 0.7) and Pb (r = 0.6) (Table 4). On the other hand, Co was significantly correlated with Pb, Ni and Cu, while Al was correlated with Zn and Co. Furthermore, Pb was significantly correlated with Zn while Mn was correlated with Cu (r = 0.6).

The ratio of metal concentration of leaves to bark (RvtJ

The highest values occurred in Co, Ni, Zn and Pb (Table 5). The plants growing in Helwan site showed the highest Rl/b ratios for Co (3983.3), Pb (36.0) and Cu (3.5). On the contrary, plants growing in Shoubra El-Kheima site showed the lowest ratios for Co (82.3) and Pb (3.1) and an intermediate ratio (2.5) for Cu in comparison to other sites.

Table 3. The mean values ± standard deviation for the heavy metals concentrations (mg kg-[1] DW) in bark samples. The concentration values of Pb, Co, Ni and Cu are multiplied by 10-[1]·

Site	Pb	Co	Zn	Fe	Ni	Mn	Cu	Al
Shoubra El-kheima	8.3 ± 0.9	1.4 ± 0.1	0.6 ± 0.4	4.4 ± L0	0.3 ± O.oJ	0.2±0.1	0.4 ± 0.0	0.2 ± 0.1
El-Sahel	0.7 ± 0.3	0.1 ± 0.7	0.2 ± OJ	4.4 ± OJ	1.2 ± 2.6	0.3 ± 0.2	0.3 ± 0.0	0.1 ± 0.0
RodE1-Farag	0.7 ± 0.0	0.1 ± 0.0	0.2 ± 0.0	4.3 ± 0.7	0.3 ± 0.0	0.4 ± 0.2	0.4 ± 0.0	0.2 ± 0.1
Ramsis	0.6 ± 0.0	0.04 ± 0.0	0.2 ± 0.1	3.9 ± 0.5	0.3 ± 0.0	0.5 ± 0.3	0.4 ± 0.0	0.2 ± 0.l
Ataba	0.8 ± 0.0	0.1 ± 0.0	0.3 ± 0.1	5.3 ± 2.7	1.5± 0.3	0.7 ± 0.2	0.6 ± 0.0	0.2 ± 0.1
Matariya	0.7 ± 0.0	0.05 ± 0.0	0.2 ± OJ	4.3 ± 0.8	0.3 ± 0.0	0.4 ± 0.2	0.4 ± 0.0	0.2 ± 0.1
	0.6 ± 0.0	0.03± 0.0	0.2 ± 0.1	3.1± 0.2	0.3 ± 0.0	0.1±0.0	0.3 ± 0.0	OJ ± 0.0
	0.6 ± 0.0	1.4 ± 0.0	0.2 ± 0.1	4.3 ± 1.5	2.6 ± 0.3	0.3 ± 0.2	0.4 ± 0.0	0.2 ± 0.0
Helwan	0.5 ± 0.0	0.03 ± 0.0	0.1 ± OJ	3.4 ± 0.5	0.3 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.2 ± 0.l
Giza	2 ± 0.2	0.04 ± 0.0	0.2 ± 0.2	3.8 ± 1.5	0.2 ± 0.0	0.3 ± 0.2	0.3 ± 0.0	0.2 ± 0.l

Table 4. Speannan correlation coefficients between the heavy metals in bark samEles. Values in bold are significant at E < 0.05

	Pb	Co	Zn	Fe	Ni	Mn	Cu
Co	0.5						
Zn	0.9	0.3					
Fe	0.6	0.9	0.4				
Ni	-0.1	0.5	-0.2	0.6			
Mn	0.1	0.0	0.2	0.2	0.1		
Cu	0.3	0.6	0.4	0.7	0.4	0.6	
Al	0.5	0.4	0.6	0.4	-0.1	0.4	0.6

Table 5. Comparison between the different sites for the ratios of heavy metals concentrations ofleaves to bark samEles (Rllb}·

	Pb	Co	Zn	Fe	Ni	Mn	Cu	Al
Shoubra El-kheima	3.1	82.3	128.1	0.6	403.3	4.7	2.5	4.3
El-Sahel	15.3	1135.0	234.8	0.4	89.6	2.6	1.5	4.3
Rod El-Farag	24.6	937.6	257.4	0.4	286.7	1.9	1.6	4.8
Ramsis	18.0	2860.0	303.1	0.4	273.3	1.6	2.1	4.3
Ataba	16.5	788.0	160.6	0.2	35.6	1.1	1.3	4.1
Matariya	22.3	2226.0	187.1	0.2	237.1	2.4	2.6	4.3
Maadi	18.5	3006.7	250.1	0.4	214.7	3.7	2.4	5.1
Tura	12.7	711.4	207.1	0.2	25.8	2.0	1.3	5.9
Helwan	36.0	3983.3	256.5	0.4	228.3	2.3	3.5	5.8
Giza	3.6	2280.0	145.4	0.2	350.0	0.6	1.2	4.1

Spatial variations in metal concentrations

The t-test showed that spatial variability of the trace metals in leaves samples at different contaminated sites was only significant for Co ($t = 2.24$, $p < 0.05$), Zn ($t = 7.83$, $p < 0.001$) and Ni ($t = 2.20$, $p < 0.05$). On the other hand, these variations were not significantly different (at $p < 0.05$) for bark samples from all sites (data not shown).

Discussion

The sampling sites in our study were subjected to heavy traffic density, in addition to huge residential and industrial activities (Rizk and Khoder, 2001; Ministry State for Environmental Affairs, 2008; Safar and Labib, 2010). Our results revealed that the accumulation of the heavy metals in the leaves and bark of *D. regia* lie within or below the normal range reported for plants (Kabata-Pendias and Pendias, 1992). The prevalence of Zn, Co and Ni in the leaves of plants growing in the polluted sites could be ascribed to the presence of these heavy elements in the roadway environment (Campo *et al.* 1996; Denier van der Gon *et al.,* 2007). Since leaded petrol has been phased out in GC, Zn has been proposed to be a significant reliable tracer of motor vehicle emissions (Rizk and Khoder, 2001; Oliva and Rautio, 2004; Shakour *et al.,* 2006). It is well known that the significant natural source of atmospheric Ni is wind borne dust particles derived from the weathering of rocks and soils and from volcanic eruptions (US EPA, 1984). Although Ni occurs throughout the environment, industrialization has increased its flux. Khillare *et al.* (2004) concluded that the anthropogenic stationary sources that release Ni into ambient air include combustion and incineration sources and high temperature metallurgical operations. The mobile source contribution to Ni emission inventories is small and derived primarily from engine wear and impurities in engine oil and fuel additives (US EPA, 2002). In GC, the particulate matters emitted from gasoline powered vehicles are the main contributor to Ni in the air (Shakour *et al.,* 2001). In some sites in GC, Shakour *et al.* (2006) found that the annual mean concentration of Ni was approximately 6 times more than the guide value (0.02 µg Nim-^{3}) which was proposed by World Health organization (WHO, 2000).The enrichment of Co in the leaves of *D. regia* was higher than many other elements in the same sample. As we know, Co naturally occurs in the earth's crust as cobaltite [CoAsS] , erythrite [C03(As04)2] and smaltite [CoAsi] (Barceloux, 1999). But, increasing in Co concentration of soils can be caused by deposition from the burning of fossil fuels, wearing of Co containing alloys and spreading of sewage sludge and manure (Barceloux, 1999). The uptake and distribution of Co in plants is species-dependent and controlled by different mechanisms (Kukier *et al.,* 2004; Li *et al.,* 2004; Bakkaus *et al.,* 2005).

The presence of low concentrations of the trace heavy elements detected in this study may be attributed at first blush to a weak bioaccumulation potentiality of *D. regia* to elements. But, this might be an invalid conclusion when one compares the data of this work with trace elements contents of *D. regia* from polluted sites in Benin City, Nigeria (Ukpebor *et al.,* 2010). They reported that *D. regia* can accumulate high concentrations of Pb and Zn in leaves and bark. For instance, previous studies have indicated that the bark of the trees was able to accumulate Pb with range of 20--70 µg g^{-1}, exceeded the normal plant Pb concentration of 0.2-20.0 µg g^{-1} (Ukpebor *et al.*, 2010). Accordingly, we might ascribe the presence of low concentrations of trace metals in bark and leaves of the plants basically to the low concentrations of these elements in the air as reported recently by many investigators on the improvements of air quality in GC, particularly after application of unleaded gasoline and natural gas as automobile fuel since 1991 (Rizk and Khoder, 2001; Shakour *et al.,* 2001, Ministry of State for Environmental Affairs, 2008; Safar and Labib, 2010). For example, the annual average air concentration of Pb in Shoubra El-Kheima area (an industrialized environment containing several lead smelters) was 23.09 µg m^{-3} in 1999 and declined to 4.lµg m^{-3} in 2001 (Shakour *et al.*, 2001; Safar and Labib, 2010). This was confirmed with measuring Pb concentrations in particulate matters which dropped from 3.6 µg m^{-3} in 1998 to 0.2 µg m^{-3} in 2007 (Safar and Labib, 2010).

The dominance of Fe in bark samples could represent soil resuspension pollution since this element is a typical soil constituent (Oliva and Mingorance, 2006) while the strong presence of other elements, e.g. Pb was relevant to site conditions. The weak presence of Ni, Cu and Al in bark could be due to the smooth surfaces of *D. regia* bark which trap fewer pollutants. The reported correlations between elements concentrations in either leaves and bark samples could be due to its origin and local site conditions.

The findings of Rut, showed great variation between bark and leaves of *D. regia* in deposition of the metals and showed more efficiency of the leaves over the bark. The high ratio appeared is possibly attributed to the more traffic and industry related metals (i.e. Co, Zn, Ni and Pb) while the low ratio which were observed for Fe and Al are more related to the soil factor and/or due to the low quantities of metals produced during fuel combustions (Cu and Mn). The efficiency of leaves to accumulate metals more than bark is inconsistent with that reported for *D. regia* in Nigeria (Ukpebor *et al.*, 2010) and many other investigated species (e.g. Oliva and Mingorance, 2006). This might be ascribed to dry climatic conditions in GC and low rainfall (Anonymous, 2005) which enable the deposited pollutants to persist long time on the leaf surfaces of the trees. The spatial variations of metals were more related to the density of traffic and industrial activities rather than to the high density of population in the sites.

The presence of high variations in *RiA*, ratio for heavy metals among sites can be explained by the differences in anthropogenic activities in each site and the accumulation capability of leaves to bark samples. The significant spatial variability of Co, Zn and Ni among sites might confrrm the continuity of these pollutants production in the atmosphere from many workshops in Cairo which recently monitored by Egyptian Environmental Affairs Agency (Egypt state of the environment report, 2009).

Conclusions

This study can be outlined in the following points: 1- Our results indicated *Delonix regia* is an adequate biomonitor of heavy metals pollution in Greater Cairo, 2- leaves of *D. regia* accumulated many folds of metals more than the bark, particularly for Co, Zn and Ni, 3- most of the metals in GC lie within or below the normal range reported for non-polluted areas, and 4- the spatial distributions of metals in sampled plants were only significant for Co, Zn and Ni.

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