Accumulation of heavy metals in *Azadiractha indica* from Akungba-Akoko, Nigeria

E. O. Olanipekun[1], P. O. Tedela[2], F. O. Iyiola[3], O. E. Faniyi[3]
& B. A. Falusi[1]
[1]Department of Chemistry, University of Ado-Ekiti, Ado-Ekiti, Nigeria
[2]Department of Plant Science, University of Ado- Ekiti,
Ado-Ekiti, Nigeria
[3]Department of Chemistry & Industrial Chemistry,
Adekunle Ajasin University, Akungba-Akoko, Nigeria

Abstract

Heavy metals from automobile sources in environments with dense road traffic may pose serious toxicological risks to human health. As a preliminary step towards risk assessment, the impact of traffic density on the accumulation of heavy metals in *Azadiractha indica* from Akungba-Akoko, a University town in South West, Nigeria, was investigated. Samples of soil and the vegetative parts (root, stem, leaf) of *A. Indica* were collected at different sites with varying traffic densities. The samples were digested in acids and analyzed for Cd, Co, Cr, Cu, Fe, Mn, Pb, and Zn, using a flame atomic absorption spectrometer. Results obtained show distinct variations in metal concentrations across the sites and accumulation was highest in the soil and lowest in the stem. The relatively higher concentrations of Cd, Cu, Pb, and Zn exhibited by samples collected within the vicinity of the highway suggest an important anthropogenic source. Conversely, the roughly similar contents of Co, Cr, Fe, and Mn in the investigated samples at all sites, regardless of the traffic density or distance from the roadside, are apparent of a natural origin. Despite the fact that the levels of heavy metals (more importantly Cd and Pb) in the vegetative parts were generally within normal literature values, the stem having the lowest accumulation of metals would probably pose less health hazards to the consumers. Given the results of this work and similar ones, it is imperative to monitor regularly the trace/heavy metal contents of roadside ecosystems in high-traffic areas.

Keywords: heavy metals accumulation, soil, Azadiractha indica, road traffic, Akungba-Akoko, Nigeria.

doi:10.2495/ETOX080161

1 Introduction

Contamination of the ecosystem by trace/heavy metals has continued to receive global attention in the recent past mainly because of the toxicological risks posed by such metals to human health in particular, and the environment at large. Automobiles constitute a major source of environmental pollution in roadside soil and vegetation [1–7]. In Nigeria, most of the investigations [8–12] have been largely confined to urban areas, with minimal information about the status of pollution in the rural areas.

Azadiractha indica is an important medicinal tree widely cultivated and consumed in Nigeria, notably for treatment of malaria fever, a killer African disease. In an investigation [12] on some tree barks, *A. indica* was confirmed as a suitable bio-indicator of aerial fallout of heavy metals. Prolonged consumption of medicinal plants and herbs containing heavy metals even at low concentrations may be detrimental to human health [13], hence the need for regular monitoring.

Akungba-Akoko is a rural town in the South Western part of Nigeria, located on an intersection along the ever busy Lagos-Abuja interstate highway. Majority of the local populace of Akungba usually drink extracts from *A. indica,* in preference to orthodox medicine, for prevention against, or as a cure for, malaria fever. Given the dense road traffic situation often witnessed on a daily basis in this University town, it was anticipated that the environment, especially the roadside ecosystems, might have been contaminated with heavy/trace metals commonly associated with automobile sources. Consequently, as a preliminary step towards risk assessment, this study was undertaken with a view to determining the levels of accumulation and possible sources of some heavy metals in soil and the vegetative parts (root, stem, leaf) of *Azadiractha indica* collected at different locations with varying traffic densities in Akungba-Akoko, Nigeria.

2 Materials and methods

Samples of soil (the upper 5cm layer), and the root bark, stem bark, and leaves of A. indica were concurrently collected at two different sites along the Lagos-Abuja highway in Akungba-Akoko: site A was along the Lagos axis while site B was along the Abuja axis, with average traffic densities of 550 and 370 vehicles/hr, respectively. Similar samples were also taken at a control site along Supare road (average traffic density = 15 vehicles/hr).

Sampling was done in August 2006, and a minimum of ten soils and five tree parts were randomly collected at distances of 5m, 10m, and 20m from the roadside in sites A and B, and at 150m in the control site. Soil samples were homogenized, dried at 105°C, and digested with a 1:3 solution of $HClO_4$ and HNO_3 [14]. Tree parts were rinsed in deionized water, dried at 105°C, milled, and digested with a 1:2.5 solution of $HClO_4$ and HNO_3 [15]. Digestion was conducted in digester block and separation of extracts from solid residue was done by centrifugation at 3500 rpm. Blank determinations were carried out using the above procedures.

Heavy metals in solutions were analyzed using an air-acetylene flame atomic absorption spectrometer (Buck Scientific Model 210), fitted with a deuterium lamp for continuous background correction. Validation of digestion methods was done using certified reference materials: CRM 141R (soil) and SRM 1547 (peach leaves), and recoveries ranged from 89.5 to 112%, while relative standard deviations (RSDs) were generally below 10%. Calibration curves for metal analysis had a correlation coefficient, $r \geq 0.996$. Data presented were averages of five replicate analyses. The Minitab statistical package (MTB 11) and the Statistical package for the social sciences (SPSS version 13.0) were used for ANOVA and Pearson's correlation coefficient calculations, respectively. The enrichment factor (EF) for a given element x, was calculated as follows:

$$EF = [C(x)/C(Co)]/[C(x,background/C(Co,background)]$$

where C represents the concentration, and cobalt (Co) was taken as the element with minimal anthropogenic influence.

3 Results and discussion

The average concentrations of trace elements in soil samples collected at various sites in Akungba-Akoko are given in table 1. The concentrations of cobalt (Co), chromium (Cr), iron (Fe), and manganese (Mn) in samples from all sites including the control site were roughly similar, regardless of the traffic volume and proximity to highway, thus suggesting that these metals were probably of natural (soil) origins. Conversely, the levels of cadmium (Cd), copper (Cu), lead (Pb), and zinc (Zn) obviously decreased with increasing distance from roadside, and site A with a greater traffic density exhibited higher metal concentrations than site B. Cd and Cu, unlike Pb and Zn, were highly elevated in the vicinity of the road (at distances ≤10m). The differences in pollution patterns at a distance of 20m between Cd and Cu on one hand, and Pb and Zn on the other hand was apparently a reflection of differences in the particle size and vehicle source [16]. Consequently, the short travel distance of Cd and Cu was perhaps due to their larger particle sizes compared to those of Pb and Zn. Thus, deposition of Cd and Cu would be dominated by depositional processes, and the size difference was probably a function of vehicle-associated wear rather than with the finer Pb aerosols emitted by automobile exhaust which would be governed to a greater extent by dispersional processes under normal mixing conditions [17]. The greater dispersion of Pb and Zn from roadside ecosystems was probably responsible for their elevated levels even at a distance of 20m from the road edge.

The foregoing results show that Cd, Cu, Pb, and Zn were apparently from an important anthropogenic source which we suspect to be mainly the automobiles since there were no industrial and mining activities, or power plants in the study area and its immediate environ. The enrichment factors (EF) calculated for Cd, Cu, Pb, and Zn in the soil samples collected at site A (5m) were 2.6, 1.7, 10.4, and 6.6, respectively, implying that the soil was mostly enriched with Pb and least enriched with Cu. The fact that the ER values were <2 for Cu, between 2-5 for Cd, and 5-20 for Pb and Zn, suggested that the soil was polluted, minimally with Cu, moderately with Cd, and significantly with Pb and Zn [6, 7]. Among the polluting elements, statistically relevant correlations were obtained between Cd & Cu (r =0.903, p = 0.014), Cd & Pb (r = 0.942, p = 0.05), Cd & Zn (r = 0.971, p = 0.001), Cu & Pb (r = 0.958, p = 0.003), Cu & Zn (r = 0.855, p = 0.030), and Pb & Zn (r = 0.925, p = 0.008). These high values of correlation coefficients further show that the metals could have probably emanated from common anthropogenic sources. In comparison with data from investigations conducted in other parts of South West (Nigeria), the levels of Cd, Cu, Pb, and Zn obtained in our study were lower than those of Ikeja soils [10] but comparable to those of Osogbo soils [11]. Heavier road traffic coupled with several industrial activities and presence of two airports would probably explain the higher metal burdens in the Ikeja soils. The concentrations of Cd, Cu, Pb, and Zn in the analyzed soils were also lower than the values reported for soils in urban cities like London [18] and Hong Kong [19, 20]. The Cu, Pb, and Zn contents were, however, comparable with the data reported for Aberdeen soils [21].

Table 1: Average concentrations ± s.d. (mg/kg) of trace elements in Akungba soils.

	Site A*, distance (m)			Site B**, distance (m)			Control site
Metal	5	10	20	5	10	20	
Cd	0.82±0.25	0.58±0.11	0.36±0.06	0.63±0.05	0.42±0.16	0.37±0.11	0.34±0.06
Co	9.35±2.35	10.06±2.77	9.74±2.29	10.22±3.75	10.95±2.01	10.60±1.38	10.17±2.45
Cr	15.20±4.30	17.13±2.31	16.80±4.12	16.91±4.17	16.74±1.91	17.29±4.25	16.53±2.27
Cu	20.42±4.49	14.16±0.96	13.95±2.00	18.90±1.09	13.55±1.79	13.04±0.90	12.86±1.20
Fe	6800±385	7500±246	7075±288	7782±291	8166±355	8007±398	7816±339
Mn	264.2±46.9	321.6±25.0	296.0±35.2	310.2±26.8	278.3±20.5	304.6±17.3	280.3±21.9
Pb	80.34±9.99	28.50±4.21	19.38±3.05	52.61±6.71	30.20±3.16	16.04±2.70	8.64±1.95
Zn	61.07±7.22	36.15±3.58	20.91±2.58	34.76±3.59	22.40±4.33	21.27±5.43	10.12±2.48

∗: average traffic density of 550 vehicles/hr;

∗∗: average traffic density of 370 vehicles/hr.

Tables 2, 3, and 4 present the mean contents of heavy metals in the root, stem and leaves of *A. indica* collected at different sites, respectively. It is interesting to note that all the three parts have medicinal values hence their assessment in this study. Just like for the soils, the concentrations of Co, Cr, Fe and Mn, in the root (table 2), stem (table 3), and leaves (table 4) were roughly similar, regardless of the traffic density or distance from roadside, and were comparable to the background levels at the control site. On the other hand, the levels of Cd, Cu, Pb, and Zn in all the three vegetative parts decreased with increasing distance from the road, with the stem (table 3) being the least contaminated and the leaves (table 4) the most polluted.

The overall mean values (mg/kg) of pollutants, in root (table 2), stem (table 3), and leaves (table 4), for both sites A & B were: Cd (0.16±0.06, 0.10±0.04, 0.20±0.08); Cu (6.18±1.31, 4.63±1.26, 7.26±1.36); Pb (2.60±0.83, 1.51±0.93, 2.95±0.96); and Zn (11.41±0.99, 9.70±1.12, 11.20±2.16), respectively.

Table 2: Average metal concentrations ± s.d. (mg/kg) in root of *A. indica* from Akungba.

Metal	Site A*, distance (m)			Site B**, distance (m)			Control site
	5	10	20	5	10	20	
Cd	0.24±0.07	0.19±0.08	0.10±0.03	0.20±0.10	0.15±0.09	0.10±0.03	0.08±0.02
Co	0.60±0.19	0.55±0.22	0.57±0.20	0.71±0.16	0.76±0.17	0.62±0.14	0.65±0.14
Cr	0.75±0.20	0.72±0.22	0.80±0.24	0.70±0.16	0.83±0.14	0.80±0.19	0.76±0.16
Cu	8.36±1.59	6.58±1.43	5.97±1.27	6.41±1.32	5.06±1.05	4.68±0.67	4.39±0.72
Fe	145.7±32.1	148.3±15.4	150.2±24.2	140.5±17.3	151.2±19.2	132.8±15.1	144.6±16.2
Mn	11.95±2.42	11.80±1.95	12.74±2.41	10.78±2.04	10.90±1.76	10.96±1.77	13.65±2.23
Pb	3.21±1.42	2.79±1.00	2.59±1.01	3.40±1.39	2.53±1.12	1.05±0.39	0.74±0.18
Zn	12.90±2.16	11.32±0.91	10.56±2.00	12.25±1.80	11.09±2.00	10.34±1.75	5.58±1.28

∗: average traffic density of 550 vehicles/hr;

∗∗: average traffic density of 370 vehicles/hr.

Table 3: Average metal concentrations ± s.d. (mg/kg) in stem of *A. indica* from Akungba.

Metal	Site A*, distance (m)			Site B**, distance (m)			Control site
	5	10	20	5	10	20	
Cd	0.15±0.06	0.11±0.03	0.04±0.02	0.12±0.02	0.10±0.02	0.06±0.03	0.04±0.02
Co	0.62±0.16	0.58±0.12	0.60±0.13	0.68±0.14	0.71±0.28	0.65±0.21	0.57±0.12
Cr	0.64±0.17	0.67±0.15	0.65±0.14	070±0.21	0.76±0.22	0.76±0.27	0.60±0.14
Cu	6.33±1.28	4.40±1.24	4.13±0.91	5.98±1.20	3.73±0.95	3.20±0.78	2.85±0.96
Fe	156.2±13.7	150.4±16.4	153.6±17.1	147.4±10.6	149.7±8.5	158.2±14.9	151.2±21.7
Mn	10.49±1.63	10.07±1.79	11.20±2.32	12.35±1.81	13.09±1.77	12.83±2.05	10.56±1.82
Pb	2.23±0.72	1.57±0.60	0.45±0.12	2.69±0.86	1.74±0.73	0.40±0.11	0.18±0.06
Zn	11.05±1.60	10.76±1.79	8.94±1.02	10.07±1.21	9.20±1.24	8.15±1.64	4.25±1.12

∗: average traffic density of 550 vehicles/hr;

∗∗: average traffic density of 370 vehicles/hr.

Table 4: Average metal concentrations ± s.d. (mg/kg) in leaves of *A. indica* from Akungba.

Metal	Site A*, distance (m)			Site B**, distance (m)			Control site
	5	10	20	5	10	20	
Cd	0.31±0.08	0.20±0.04	0.12±0.03	0.26.±0.08	0.21±0.08	0.10±0.04	0.08±0.03
Co	0.56±0.08	0.52±0.07	0.60±0.10	0.69±0.12	0.65±0.09	0.72±0.06	0.54±0.07
Cr	0.78±0.08	0.80±0.10	0.72±0.12	0.70±0.10	0.83±0.08	0.81±0.12	0.70±0.12
Cu	9.50±2.13	7.25±1.65	7.06±0.88	7.94±1.68	6.04±1.33	5.75±1.13	5.10±0.97
Fe	126.3±12.4	118.6±18.1	130.4±16.5	114.8±16.1	109.2±12.3	120.3±18.9	135.5±14.2
Mn	11.67±3.00	12.92±1.61	12.75±1.06	12.39±1.52	10.99±1.63	11.70±1.36	14.03±1.01
Pb	3.91±1.02	2.74±1.08	2.80±1.16	4.26±1.02	2.13±0.46	`1.85±0.53	0.62±0.14
Zn	14.53±1.92	10.80±1.61	10.25±1.58	12.94±2.52	10.13±1.50	8.57±1.37	6.04±1.58

∗: average traffic density of 550 vehicles/hr;

∗∗: average traffic density of 370 vehicles/hr.

Except for Zn, these values differ significantly at $p<0.05$ for the three investigated tree parts and were higher than the corresponding background levels for: Cd (0.08±0.02, 0.04 ± 0.02, 0.08 ± 0.03); Cu (4.39 ± 0.72, 2.85 ± 0.96, 5.10 ± 0.97); Pb (0.74 ± 0.18, 0.18 ± 0.06, 0.62 ± 0.14); and Zn (5.58 ± 1.28, 4.25 ± 1.12, 6.04 ± 1.58). Given the comparatively low levels of trace metals in the stem especially the toxic ones (Cd and Pb), it may be much safer to utilize the stem for therapeutic purposes. The concentrations of Cu, Pb, and Zn in the stem were lower than the data reported elsewhere [12].

Statistically significant correlations were obtained between the following pairs of metals in leaves: Cd and Zn ($r = 0.924$, $p = 0.009$), Cu and Pb ($r = 0.879$, $p = 0.021$), Cu and Zn ($r =0.961$, $p = 0.002$), Pb and Zn ($r = 0.918$, $p = 0.010$), Cu and Zn ($r = 0.961$, $p = 0.002$), and Pb and Zn ($r = 0.918$, $p=0.010$); in stem: Cd and Pb ($r = 0.897$, $p = 0.015$), and Cd and Zn ($r = 0.852$, $p = 0.031$); and in root: Cd and Cu ($r = 0.835$, $p = 0.039$), Cd and Zn ($r = 0.955$, $p = 0.003$), and Cu and Zn ($r = 0.859$, $p = 0.029$). Significant high correlation coefficients were also exhibited between metals in soil and root: Cd ($r = 0.967$ $p = 0.002$), Cu ($r = 0.835$, $p = 0.038$), and Zn ($r = 0.904$, $p = 0.013$); in root and stem: Cd ($r = 0.975$, $p = 0.001$), Cu ($r = 0.877$, $p = 0.022$), and Zn ($r = 0.843$, $p = 0.035$); and finally in leaf and stem: Cd ($r = 0.958$, $p = 0.003$), Cu ($r = 0.946$, $p = 0.004$), and Zn ($r = 0.830$, $p = 0.041$). These high r values suggest that the metals could have originated from common sources.

4 Conclusions

The soils and the vegetative parts of *A. indica* collected within the vicinity of the highway in Akungba-Akoko, Nigeria were mainly polluted with Cd, Cu, Pb, and Zn, which apparently originated from automobile sources. However, the concentrations of these chemical elements in the investigated soils and medicinal tree parts were generally within the permissible limits. The normal concentration ranges (mg/kg) in soils and plants, respectively, are for: Cd (0.01 - 2.0, 0.1 - 2.4); Cu (2 - 25, 5 - 20); Pb (2 - 300, 0.2 - 20); and Zn (1 - 900, 1 – 400) [22, 23].

Heavy metals have natural tendencies of accumulating in organisms during the lifetime and may interact with each other and with trace elements vital for body functions [3]. Consequently, prolonged consumption of medicinal plants grown along busy highways should, as much as possible, be avoided due to the potential health hazards often posed by trace metals commonly associated with vehicle sources.

Finally, given the results of our present study and similar ones, it is imperative to monitor regularly the trace/heavy metal contents of roadside ecosystems in high traffic areas.

References

[1] Singh, N. Yunus, M., Srivastava, K. Singh, S. N., Pandey, V., Misra, J. & Ahmad, K.J., Monitoring of autoexhaust pollution by roadside plants. *Environmental Monitoring and Assessment*, **34(1),** pp. 13–25, 1995.

[2] Caselles, J., Colliga, C. & Zornoza, R., Evaluation of trace element pollution from vehicle emissions in petunia plants. *Water, Air, and Soil Pollution,* **136(1-4)**, pp. 1–9, 2002.

[3] D'Elia, V., Lodi, V., Naldi, M., Gherarardi, G., Violante, F.S., Badiello, R. & Raffi, G.B., Gardening work and heavy metals in urban environment. *Arh hig rada toksikol,* **50(2)**, pp. 163–170, 1999.

[4] Jaradat, Q. M. & Momani, K. A., Contamination of roadside soil, plants, and air with heavy metals in Jordan, a comparative study. *Turkish Journal of Chemistry,* **23**, pp. 209–220, 1999.

[5] Tokalioglu, S., Kartal, S., & Birol, G., Application of a three-stage sequential extraction procedure for the determination of extractable metal contents in highway soils. *Turkish Journal of Chemistry*, **27**, pp. 333–346, 2003.

[6] Krolak, E., Heavy metal content in falling dusts, soil and dandelion (*Taraxacum officinale* webb.) in Southern Podlasie Lowland. *Electronic Journal of Polish Agricultural Universities*, **4(1)**, pp. 1–13, 2001.

[7] Sutherland, R.A., Depth variation in copper, lead, and zinc concentrations and mass enrichment ratios in soils of an urban watershed. *Journal of Environmental Quality,* **29**, pp. 1414–1422, 2000.

[8] Iwegbue, C.M.A., Egobueze, F. E. & Opuene, K., Preliminary assessment of heavy metal levels of soils of an oilfield in the Niger Delta, Nigeria. *International Journal of Environmental Science and Technology*, **3(2)**, pp. 167–172, 2006.

[9] Kakulu, S. E., Trace metal concentration in roadside surface soil and tree bark: a measurement of local atmospheric pollution in Abuja, Nigeria. *Environmental Monitoring and Assessment,* **89(3**), pp. 233–242, 2003.

[10] Fakayode, S. & Onianwa, P., Heavy metal contamination of soil and bioaccumulation in Guinea grass (*Panicum maximum*) around Ikeja Industrial Estate, Lagos, Nigeria. *Environmental Geology*, **43(1–2)**, pp. 145–150, 2002.

[11] Fakayode, S. O. & Olu-Owolabi, B. I., Heavy metals contamination of roadside topsoil in Osogbo, Nigeria: its relationship to traffic density and proximity to highways. *Environmental Geology*, **44(2)**, pp. 150–15, 2004.

[12] Odukoya, O.O., Arowolo, T.A. & Bamgbose, O., Pb, Zn, and Cu levels in tree barks as indicator of atmospheric pollution. *Environment International*, **26 (1–2)**, pp. 11–16, 2000.

[13] Dwivedi, S. K. & Dey, S., Medicinal herbs: a potential source of toxic metal exposure for man and animals in India. *Archives of Environmental Health*, **57(3)** pp. 229–231, 2002.

[14] MAFF, *The Analysis of Agricultural Materials, Reference Book 427*, Ministry of Agriculture, Fisheries, and Food, London, 1986.

[15] Awofolu, O.R., A survey of trace metals in vegetation, soil, and lower animals along some selected major roads in Metropolitan City of Lagos. *Environmental Monitoring and Assessment*, **105**, pp. 431–447, 2005.

[16] Sutherland, R.A. & Tolosa, C.A., Variation in total and extractable elements with distance from roads in an urban watershed. *Water, Air, and Soil Pollution*, **127**, pp. 315–338, 2001.

[17] Harrison, R.M. & Johnston, W.R., Deposition fluxes of lead, cadmium, copper and polynuclear aromatic hydrocarbons (PAH) on the verges of a major highway. *Science of the Total Environment*, **46**, pp. 121–135, 1985.

[18] Thornton, I., Metal contamination of soils in urban area. *Soil in the Urban Environment*, eds. P. Bullock & P.J. Gregory, Blackwell: Oxford, pp. 47–75, 1991.

[19] Wong, M.H., Chen, T.B. & Wong, J.W.C., Trace metal contamination of the Hong Kong soil environment: a review. *Contaminants and the Soil Environments in the Australia-Pacific Region*, Kluwer Academic Publisher: Dordrecht, pp. 501–511, 1996.

[20] Li, X.D., Poon, C.S. & Pui, S.L., Heavy metal contamination of urban soils and street dusts in Hong Kong. *Applied Geochemistry*, **16**, pp. 1361–1368, 2001.

[21] Paterson, E., Sanka, M. & Clark, L., Urban soils as pollutant sink – a case study from Aberdeen, Scotland. *Applied Geochemistry*, **11**, pp. 129–131, 1996.

[22] Alloway, B.J., *Heavy Metals in Soils*, Blackie and Sons Ltd.: Glasgow and London, pp. 177–196, 1990.

[23] Pendias, A.K. & Pendias, H., *Trace Elements in Soil and Plants. V. In: Lead*, 2nd edition, Boca Raton Arbor: London, pp. 187–198, 1992.