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INVESTIGATION OF THE TOXICITY LEVEL OF HEAVY METALS RESULTED FROM THE MIGRATION OF LEACHATE POLLUTANTS ACROSS THE SOIL PROFILE OF A MUNICIPAL DUMPSITE

***[1]Ahmed Mohammed, [2]Sadiq Umar, [2]Aminu Ismaila, [3]Mohammed Sani Abdulkarim, [2]Abdulsalam Ismaila**

[1] Department of Physics, School of Science Education, Federal College of Education (Technical), Gusau-Nigeria
[2]Department of Physics, Faculty of Physical Sciences, Ahmadu Bello University, Zaria-Nigeria
[3]Department of Applied Sciences, College of Science and Technology Kaduna Polytechnic, Kaduna-Nigeria

*Corresponding Author's Email: ahmedmohammed7984@gmail.com

ABSTRACT

In Nigeria and many developing countries, solid wastes are dumped in open landfills which are not provided with lining system to prevent leachate from infiltrating the soil into the groundwater. This study investigates the contents and concentration of heavy metal soil pollution in one of the dumpsite in the city of Kaduna, Nigeria. Six soil plumes were collected at different depth: 0, 25, 50, 100, 120 and 150 cm down the profiles and were analyzed for metallic elements. The instrumental neutron activation analysis (INAA) has been deployed to investigate the vertical distributions of heavy metals in soil profiles. The analysis revealed the presence of twenty-nine elements which shows a remarkable variation in concentration in all the soil samples. The mean concentrations of most of the detected elements were higher than their background values with the reference elements of Al, Fe, Ti showing the higher concentration in all samples. The mean concentration of Al, Fe, and Ti were 80868.3 ppm, 26260 ppm, and 3532.7 ppm respectively which are generally higher than their background values. Statistical analysis including the Pearson correlation of the elements and clustering analysis revealed a higher significant positive correlation among the samples. Repetitive order was noticed with some group of elements in all samples except A4. A significantly higher relationship displayed by all pair samples suggests a common anthropogenic source. There is a clear heavy polluted hotspot of Al, Fe, Ti, Potassium (K) and sodium (Na) as a result of anthropogenic activities that occurred in the study area.

Keywords: Dumpsite, instrumental neutron activation analysis, soil plume, and solid waste.

INTRODUCTION

Solid waste is defined as waste products and materials that are spreadable. In Nigeria and several African countries, solid wastes are dumped in open dumpsites whichare not provided with a lining system to prevent leachate from infiltrating the soil thereby transferred hazardous/harmful elements into the groundwater. This practice is motivated by the lack of effective legislative measures on waste management (separation, collection, disposal, recycling, and energy conversion). a Literature reports indicates that solid waste if left for a long time will be acted upon by the combined effects of physical, chemical and biological processes and will undergo decomposition (Haque *et al.*, 2013a) leading to emission of leachate and landfill gas, which primarily composed of methane and carbon dioxide. Leachate is described as water-based solution of compounds from solid wastes dumped in a landfill (Oni *et al.*, 2011). The generation of leachate is caused by precipitation and percolation through waste deposited in a landfill. Once in contact with decomposing solid waste, the percolating water becomes contaminated and it then flows out of the waste material (Olarewaju *et al.*, 2012) and this infiltrates into or onto the soil as pollutants. Leachate from decomposed solid waste transmits heavy metallic elements such as Cu, Fe, Cd, Ni, Cr etc. that may get into surface water body or percolate groundwater causing potential contamination of the ground water and affects food chain. Such contamination of water resources may possess substantial risk to the local natural environment (Haque *et al.*, 2013b). Leachate is partially filtered as it passes through soil layers but, the potential movement of the pollutants across the soil profiles varies greatly from one site to another according to number of factors which includes: the permeability of the soil, the waste profile and pollutants concentration in the leachate (Haque, *et al.*, 2013b). There is also inherent factors such as soil texture (percentage of sand, silt, and clay) and type which also affects soil infiltration. Pollutants move more quickly through large pores of sandy soil than it does through small pores of clay soil, especially if it is compacted and has little or no structure. Some clay soils develop shrinkage cracks as they dry which acts as the direct conduits for water entering the soil, causing clay soil to have higher infiltration rates under dry conditions. Where cracks do not occur, clay soil have slow infiltration rates.

Pollutants found in municipal solid waste landfill leachate fall broadly into four categories: dissolved organic matter, inorganic macro components, heavy metals, and a xenobiotic organic compound found in household and industrial chemicals. The severe problems of these pollutants are their infiltration into the soil structure (Kumar *et al.*, 2002). Infiltration is the process by which a fluid passes through or into another substance traveling through pores and interstices and soil infiltration refers to the soils ability to allow pollutants movement into and through the soil profile (Farzad *et al.*, 2015). The factors influencing the infiltration rate have a direct effect on the soil structure namely the soil porosity. The average pore size of the soil, distribution of pore sizes and connectivity of pores are of greater importance. The soil pores must be large enough and offer sufficient continuity in order for infiltration to occur.

This study aims to investigate the toxicity level of heavy metals resulted from the migration of leachate pollutants across the soil layers of a municipal dumpsite. The prime focus is to predict the soil and ground water pollution and justify the necessity of solid waste stabilization and re-use. A nondestructive technique called INAA was employed in determining the elemental composition, and their concentration from the soil samples.

METHODOLOGY

Sample collection

A pit was dug around the dumpsite of Ahmadu Bello road at unguwar dosa area in Kaduna north where soil samples were collected at different depth: 0, 25, 50, 100, 120 and 150 cm respectively. The collected samples were emptied into a clean double polyethylene bags and were labeled according to their respective depths with A1 representing 0 cm and A6 represents 150 cm respectively. This is done to prevent interference or mix-up of samples. The six soil samples were air and oven dried and then subjected to INAA using Nigeria Nuclear Research Reactor 1 (NIRR-1) facility located at the center for energy research and training (CERT), Ahmadu Bello University Zaria.

Sample analysis

Samples were first dry through an open air in a room and then dried in an oven for some days at a temperature of 60 ^{o}C. They were then crushed into fine powders of microns' size particles in an agate mortar to make them homogeneous. About…. mg of each sample was put in a polyethylene bags labeled A1 - A6. The bags together with vials are washed in distilled water three times and taken for analysis at CERT, Zaria, Nigeria. For long irradiation, samples and standards were packed together in an irradiation vial and irradiated simultaneously for six hours with a research reactor operated at 15 kW and at a thermal neutron flux of 5 x 10^{11} neutron per second in accordance with the (Mohammed *et al.*, 2019). The counting conditions are summarized in Table 1 and the concentration of the irradiated samples and standards are presented in Table 2.

Table 1: Irradiation parameters

Irradiation procedure	Irradiation period (min.)	Neutron flux (ncm^{2}s^{-1})	Decay time		Counting time (s)	
					1st count	2nd count
Long irradiation	360	5 x 10^{11}	3 days after irradiation (1st count)	7 days after 1st count (2nd count)	1800	3600
Short irradiation	1	2.5 x 10^{11}	Immediately after irradiation (1st count)	3 hrs after 1st count (2nd count)	600	600

Theory

Neutron Activation Analysis (NAA) was discovered in 1936 when Hevsy and Levi (Hevesy and Levi, 1936) found that samples containing certain earth elements become highly radioactive after exposure to a source of neutrons. This analytical technique is powerful in identifying many elements present in samples of unknown composition performing both quantitative and qualitative multi-element analysis of major, minor and trace elements in samples (Chaubey *et al.*, 2011).

An isotope of atomic mass A and atomic number Z when placed in a neutron flux will, in general, absorb a neutron to become the isotope (A+1, Z). Some resulting isotopes, being unstable, will decay, emitting one or more gamma-rays with energies and half-life characteristic of the particular isotope. During irradiation, the naturally occurring stable isotopes of most elements that constitute alluvial samples are transformed into radioactive isotope by neutron capture. The number of radionuclides produced in activation of a natural isotope is given by the expression:

$$N_{(A+1)} = \frac{\emptyset \sigma_{aA} N_{oA}}{\lambda_{(A+1)}} \left(1 - e^{-\lambda(A+1)T}\right) e^{\lambda(A+1)l} \qquad 1$$

The activity, A is given by:

$$A = \frac{dN_{(A+1)}}{dt} \qquad 2$$

where ϕ is the neutron flux, σ_{aA} is the neutron absorption cross-section of the original isotope, N_{oA} is the number of nuclei of element under consideration of isotope A, $\lambda_{(A+1)}$ is the decay constant of the created isotope, T is the irradiation time and dt is the time between the stop of the activity and start of the counting process.

The basic essentials required to carry out an analysis of soil samples by NAA are a source of neutrons, higher purity germanium (HPGE) detector coupled with a multi-channel analyser (MCA) and a printer (Chaubey, *et al.*, 2011).

The flow of a liquid through a porous media is governed by Darcy's law given by Equation 3.

$$Q = -KA\frac{dh}{dl} \qquad 3$$

where A is the cross-sectional area through which flow can occur, $\frac{dh}{dl}$ is hydraulic gradient and K is a constant. The negative sign indicates that water flowing from highest to lowest hydraulic head.

RESULTS AND DISCUSSIONS

Descriptive statistical results of the vertical distribution of elements in soil Profiles

Data in Table 1 shows the concertation and presence of twenty-nine elements consisting of heavy metals, reference elements (Al, Fe, Ti) and naturally occurring radioactive materials (Th and U) evenly distributed across the soil profiles for each of the six soil plumes. The analysis indicates that concentrations of reference elements were measured higher in all the soil samples. Elemental K also gives a higher concentration in all the sample with a mean value of 10175 ppm. Of these, Al shows a maximum value of 39820 ppm in sample A6 and a minimum value of 29900 ppm in sample A5. On the other hand, a peak value of 28690 ppm in sample A6 and a minimum value of 23490 ppm in sample A3 is recorded for Fe. The measurements for Ti indicates a higher level of 4600 ppm in sample A2 and the lowest values of 2806 ppm in A4. The higher concentration of these elements can be explained by the presence of ore in the top layer which had undergone strong dissolution due to the action of physical, chemical and biological processes coupled with the action of water during the raining season in the dumpsite (Matini *et al.*, 2011). Evidence of a higher level of Fe at the topsoil is being reported in the geology of the present study site (Abdullahi, 2009). The soil geology varies in composition, color, and texture and in most places they are predominantly laterite and quartz grains deep brown or reddish brown. Therefore, the higher concentration at the topsoil is evidence of strong dissolution of materials containing Fe thereby raising the PH value and making the soil acidic. The acidic nature of the topsoil induces more dissolution of heavy metals (Matini, *et al.*, 2011). The present authors are on the opinion that the detected elements (shown in Table 1) down the profiles is evidence of leaching processes. This assertion is supported by the research findings reported by Kirpichtchikova *et al.* (2006) and Li *et al.* (2013) that soils are the major sink for elements released into the environment. McLean and Bledsoe (2002) noted that when metals are introduced at the soil surface, downward transportation does not occur to any great extent unless the metal retention capacity of the soil is overloaded. In addition, metal interaction with the associated waste matrix enhances mobility. In this study, it is opined that the fluctuation in concentration of the detected elements from topsoil down the profile is due to variations in soil texture and properties in agreement with the previous studies (Abdullahi, 2009; Ahmed *et al.*, 2017). The finding of this study is in contradiction with the earlier report (Kaminski and Landsberger, 2000) that, metal displayed a systematic drop in concentration with depth. The experimental study by Adriano (2003) reported that the concentration of an element in soils persists for a long time after their introduction. The inference between the two authors could go like this, as for Kaminski and Landsberger, 2000, it is in line with the fact that soil is naturally a filter to any particulate matter through it and for Adriano (2003), the soil texture of the his research site could had supported his result especially if the texture is predominantly uniform through out the samples scope.

Analysis of results in Table 1 indicates that concentration of elements such as Sb, Mg and Ca in some samples are below the detection limit of the apparatus used with the Sb not detected down the depth profile except only with sample A4 which shows concentration less than unity. In general, the average natural levels of heavy metals recorded particularly in the case of Al, Fe, K and Ti are above the global average. This suggests that the risk of contamination of underground water via a leaching process is higher. However, the decision of relocating the present dumping site to a more relatively safer area outside the residential regions depends on political, social and economic issues and in-depth risk analyses considering all routes of exposure to these elements.

Heavy metals are naturally occurring elements that are found in the earth's crust which have many economic importance but in their higher concentration present both health and environmental ramifications. Heavy metal-induced carcinogenic and toxicological effects involves many mechanistic aspects, some of which are not clearly understood and addressed scientifically. However, each metal is known to have unique features and physic-chemical properties that confer to its specific toxicological mechanisms of action (Tchounwou *et al.*, 2012). For example metallic lead interferes with a variety of body processes and is toxic to many organs and tissues including the bones, heart, intestines, kidneys, and reproductive and nervous systems. It is found to interferes with the development of the nervous system thereby causing a permanent learning and behavior disorders (Rajeswari and Namburu, 2014). Aluminium in its pure form has no known biological role and its classification into toxic metals is controversial. However the compound of aluminum such as Aluminium Phosphide (AlP) is highly toxic and has a fatal dose of between 0.15 and 0.5 grams (0.0053 and 0.0176 oz) (Rajeswari and Namburu, 2014). The death from this compound is as a results of profound shock, myocarditis and multi-organ failure. Aluminum is one of several metals known to affect the neurological system as reported by the Agency for Toxic Substances and Disease Registry, part of the U.S. Department of Health and Human Services. Excess iron leads to its accumulation in vital organs such as heart, liver, pancreas, and endocrine glands. This stimulates the formation of free radicals, which damage proteins, DNA, membranes, and can lead to death of the body cells. The death of the cells could lead to serious health issues, including: Cirrhosis liver cancer, cardiac arrhythmias, diabetes, Alzheimer's disease, bacterial and viral infections (Piperno, 1998). Other heavy metals such as K and Ti are reported to have binding effects on vital cellular components and interfere with their functioning. The metallic Cr, As and Pb can induce multiple organ damage, even at lower levels of exposure (Tchounwou, *et al.*, 2012).

Clustering analysis

Repetitive observable behavior is noticed in some group of elements. The group shows an order of increasing magnitude of concentrations down the profiles where concentrations of Mg<Ca<Sb<Lu<As<Br<Ta<Eu<Cs in A2 and A5, K<Fe<Al and Ti<k<Fe<Al in all samples with exception of sample A2. Furthermore within the samples A1 and A3, concentrations of Mn<Ba<Na <Ti<K<Fe<Al with Ca and Mg in both samples not following the trend. Also the concentrations of Ba<Na<Mg<Ti<K<Fe<Al for samples A3 and A6 with the little exception in A6 and for samples A2 and A5, concentrations of Ba<Mn<Na<Ti<V<K<Fe<Al.

Pearson correlation analysis of elemental concentrations in each soil profiles

Table 2 below displays the inter-elements correlations matrix in the soil profiles of twenty-nine elements detected from each of the six soil samples. It is clearly seen that there is significantly higher positive correlations amongst the group of elements from different soil profiles. The correlation is significant at 0.01 levels 2-tailed except for A2 and A3 where the elements show a little slight positive correlation with the significant value of 0.05 levels 2-tailed. The similarity in a relationship in this group of elements indicates a common anthropogenic source.

Table 2: Analytical results of INAA for six geological samples

	Concentration (ppm)						
Element	A1	A2	A3	A4	A5	A6	Mean
Mg	BDL	BDL	2141	1543	BDL	1948	1877.3
Al	32990	31300	319780	31420	29900	39820	80868.3
Ca	1815	BDL	BDL	1462	BDL	2010	1762.3
Ti	4600	4617	2888	2806	3178	3107	3532.7
V	45	4617	45	42	47	58	809
Mn	368	427	270	383	491	426	394.2
Dy	11.4	11.2	5.6	9.6	15.2	14.2	11.2
Na	1009	801	941	854	700	1054	893
K	11320	10230	9896	9643	8551	11410	10175
As	1.1	1.5	1.0	2.8	1.2	6.03	2.27
Br	1.9	1.6	0.53	0.9	1.2	0.5	1.1
La	80.8	128.5	45.8	80.8	63.4	60.4	76.6
Sm	12.25	23.3	8.58	13.68	9.47	11.95	13.2
U	9.5	14.9	7.26	9.1	7.3	9.5	9.59
Sc	7.5	7.2	7.03	7.1	7.0	7.82	7.28
Cr	67	69	57.4	70	74.4	129	77.8
Fe	24110	25670	23490	28300	27300	28690	26260
Co	10.4	9.5	7.2	9.9	12.8	11.3	10.18
Zn	53	50	38.6	46	44	54	47.6
Rb	91	88	83	74	76	93	84.2
Sb	BDL	BDL	BDL	0.72	BDL	BDL	0.72
Cs	3.7	4.0	3.3	3.6	3.8	4.4	3.80
Ba	416	360	386	406	289	487	390.67
Eu	2.0	2.1	1.1	2.0	1.8	1.9	1.82
Yb	8.1	9.6	4.8	4.3	4.3	3.7	5.8
Lu	1.18	1.49	0.72	0.79	0.06	0.65	0.92
Hf	17.9	16.2	13.7	25.7	13.01	13.2	16.62
Ta	1.4	1.8	1.3	2.0	1.4	2.8	1.78
Th	37.1	104.0	21.3	37.7	30.2	27.2	42.92

BDL: Below Detection Limit

Table 2: Pearson correlation analysis of total contaminant concentrations between soil profiles

	A1	A2	A3	A4	A5	A6
A1	1	0.530**	0.814**	0.992**	0.991**	0.997**
A2	0.530**	1	0.385*	0.503**	0.513**	0.502**
A3	0.814**	0.385*	1	0.764**	0.761**	0.829**
A4	0.992**	0.503**	0.764**	1	0.999**	0.994**
A5	0.991**	0.513**	0.761**	0.999**	1	0.992**
A6	0.997**	0.502**	0.919**	0.994**	0.992**	1

**. Correlation is significant at the 0.01 level (2-tailed).

*. Correlation is significant at the 0.05 level (2-tailed).

In Table 3, a pair of samples correlation analyses between elements from adjacent soil layers is presented. The aim of this statistical analysis is to elucidate if the soil samples from which the elements presented in Table 1 are of the same textural classes. The result indicates a positive correlation between elements from adjacent soil layers. The analyses indicate a strong relationship particularly between pair 2 and pair 3 as indicated in the table. A significantly higher relationship displayed by all pair samples suggests that the soil samples are of the same texture as discussed earlier.

Table 3: Pair samples correlation analysis

		N	Correlation	Significance
Pair 1	A1 & A2	27	0.530	0.004
Pair 2	A3 & A4	27	0.764	0.000
Pair 3	A5 & A6	27	0.992	0.000

CONCLUSION

An initial analysis of soil plumes revealed a total of twenty nine elements with varying concentrations from top surface down the profiles. The metallic Al, Fe, K, Ti recorded the highest concentration in all the profiles, while Lu, Br and Ta recorded the least concentrations in all the profiles. The concentration of Sb is below detection limit in all samples except in sample A4 and Ca element is not detected in A2, A3 and A5. The statistical analysis of the elements amongst themselves shows a positive correlation. The authors recommend that further research should be conducted with a soil plumes taken at a relatively higher depth above 150 cm in order to understand the extent of seepage of elements in the soil matrix.

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CONFLICTS OF INTEREST

The authors declare that they have no conflicts of interest regarding the publication of this paper.

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