Contamination Assessment of Heavy Metals in Road Dust from Selected Roads in Accra, Ghana

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Abstract: This study was conducted to assess the level of contamination of surface dust from paved roads. Dust samples were collected from major and busy roads in the city of Accra Ghana. Elemental analysis of road dust has been carried out in this study using energy dispersive X-Ray fluorescence analysis to identify the possible anthropogenic sources of pollutants in road dust. The elements Ti, V, Cr, Mn, Ni, Cu, Zn, Br, Zr and Pb were among 20 elements identified. The mathematical models; Index of geoaccumulation (Igeo), Enrichment Factors (EF), Contamination factor and degree of contamination were used to identify possible levels of pollution from anthropogenic sources. The results showed that the build of manganese, which is used instead of lead (Pb), as an octane enhancer has not gone above the crustal levels. However, significant Pb contamination was observed in the road dust. All the models agreed well in their results giving moderate to significant levels of pollution for the most part of the selected sites. The results of the Principal Component Analysis gave three major sources contributing to road dust. These are the natural crustal source, vehicular emissions and corroded vehicular parts.

Key words: Energy dispersive x-ray fluorescence, index of geoaccumulation, enrichment factor, degree of contamination, principal component analysis, vehicular emissions

INTRODUCTION

Road dust originates from the interaction of solid, liquid and gaseous materials which are produced from different sources and deposited on a road. The composition and quantity of chemical matrix of road dust are indicators of environmental pollution (Banerjee, 2003). Road dust receives varying inputs of heavy metals from diversity of mobile or stationary sources such as vehicular emission, industrial plants, power generation plants, oil burning, waste incineration, construction and demolition activities as well as resuspension of surrounding contaminated soils (Ahmed and Ishiga, 2006; Al-Khashman, 2007). Lead (Pb), for example is known to come from the use of leaded gasoline whereas Cu, Zn and Cd from tyre abrasion, lubricants, industrial and incinerator emissions (Thorpe and Harrison, 2008; Wileke et al., 1998). The source of Ni and Cr in street dust is believed to be due to corrosion of vehicular parts (Lu et al., 2009; Akhter and Madany, 1993; Ferguson and Kim, 1991) and chrome plating of some motor vehicle parts (Al-Shayep and Seaward, 2001). The phenomenon contributes significantly to the pollution of urban environment. This makes the study of road dust important for determining the origin, distribution and level of heavy metal in urban surface environments.

Over the last decade, many studies have focused on heavy metal concentration, distribution and source identification in road dust while many of these studies have been carried out in developed countries (Sezgin et al., 2003; Charlesworth et al., 2003; Imperato et al., 2003), only limited information is available on heavy metals of road dust for developing countries, including Ghana (Kylander et al., 2003). Elevated levels of heavy metal contents are ubiquitous in urban environment as a result of a wide range of human activities. As a result, the adverse effects of poor environmental conditions on human health are most evident in urban environments, particularly in developing countries where urbanization, industrialization and rapid population growth are taking place on an unprecedented scale. Exposure to heavy metals in road dust can occur by means of ingestion, inhalation and dermal contact. The adverse effects of heavy metals in road dust include respiratory system disorders, nervous system interruptions, endocrine system malfunction, immune system suppression and the risk of cancer in later life (Ferreira-Baptista and Miguel, 2005).

Accra, the capital city of Ghana, has experienced rapid urbanization and industrialization over the last few decades. It is estimated that between 1998 and 2007 a total of five hundred and eighty nine thousand one hundred and eighty two (589, 182) vehicles were registered in Ghana. Out of this number Accra and Tema metropolis alone accounted for about 65% of total number of registered vehicles (DVLA, 2008) which included cars, trucks, buses etc. Most of these vehicles can be described
as “second hand” (used). The rapid growth of industry, population and vehicle exerts a heavy pressure on its urban environment. The main objective of this study was to determine the concentration of heavy metals in road dust samples collected from selected roads in Accra and to assess their levels of contamination. In this study, the road dust contamination was assessed using various indices, including index of geoaccumulation, enrichment factor (EF), contamination factor (degree of contamination) and Principal Component Analysis (PCA).

The index of geoaccumulation (Igeo) is widely used in the assessment of contamination by comparing the levels of heavy metal obtained to a background levels originally used with bottom sediments (Muller, 1969). It can also be applied to the assessment of road dust contamination (Lu et al., 2009; 2010; Gowd et al., 2010). It is calculated using the equation:

$$I_{geo} = \log_2 \left( \frac{C_n}{1.5B_n} \right)$$  \hspace{1cm} (1)

where $C_n$ is the measured concentration of the heavy metal in road dust and $B_n$ is the geochemical background concentration of the heavy metal (crustal average) (Taylor and Meclenan, 1985). The constant 1.5 is introduced to minimize the effect of possible variations in the background values which may be attributed to lithologic variations in the sediments (Lu et al., 2009).

The following classification is given for geoaccumulation index (Huu et al., 2010; Muller, 1969):

- $<0$ = practically unpolluted, $0-1$ = unpolluted to moderately polluted, $1-2$ = moderately polluted, $2-3$ = moderately to strongly polluted, $3-4$ = strongly polluted, $4-5$ = strongly to extremely polluted and $>5$ = extremely polluted.

Enrichment Factor (EF) of an element in the studied samples is based on the standardization of a measured element against a reference element. A reference element is often the one characterized by low occurrence variability. It is used to differentiate heavy metals originating from human activities and those of natural sources. This is determined by the relation:

$$EF_X = \left( \frac{X_s}{E_{S\text{(ref)}}} \right) \left[ \frac{X_C}{E_{C\text{(ref)}}} \right]$$  \hspace{1cm} (2)

where $EF_X$ is the enrichment factor for the element X, $X_s$ is the concentration of element of interest in sample, $E_{S\text{(ref)}}$ is the concentration of the reference element used for normalization in the sample, $X_C$ is the concentration of the element in the crust and $E_{C\text{(ref)}}$ is the concentration of the reference element used for normalization in the crust (Taylor and Meclenan, 1985).

Five contamination categories are recognized on the basis of the enrichment factor: EF<2 states deficiency to minimal enrichment, EF = 2-5 moderate enrichment, EF = 5-20 significant enrichment, EF = 20-40 very high enrichment and EF>40 extremely high enrichment (Yongming et al., 2006; Kartal et al., 2006).

To assess the extent of contamination of heavy metals in road dust, contamination factor and degree of contamination has been used (Rastmanesh et al., 2010).

The $C_f^i$ is the single element index which is determined by the relation:

$$C_f^i = \frac{C_{e-1}^i}{C_n}$$  \hspace{1cm} (3)

where $C_f^i$ is the contamination factor of the element of interest, $C_{e-1}^i$ is the concentration of the element in the sample, $C_n$ is the background concentration in this study the continental crustal average has been used (Taylor and Meclenan, 1985).

$C_f^i$ is defined according to four categories: $<1$ low contamination factor, $1-3$ moderate contamination factor, $3-6$ considerable contamination factor and $>6$ very high contamination factor.

The sum of the contamination factors of all the elements in the sample gives the degree of contamination as indicated in the equation below:

$$C_{deg} = \sum C_f^i$$  \hspace{1cm} (4)

Four categories have been defined for the degree of contamination as follows: $<8$ low degree of contamination, $8-16$ moderate degree of contamination, $16-32$ considerable degree of contamination and $>32$ very high degree of contamination.

Principal Component Analysis (PCA) is one of the common multi-variate statistical methods used in environmental studies. PCA is widely used to reduce large data sets (Loska and Wiechuya, 2003) and to extract a small number of latent factors (principal components, PCs) for analyzing relationships among the observed variables. If large differences exist in the standard deviations of variables, PCA results will vary considerably depending on whether the covariance or correlation matrix is used (Farnham et al., 2003). The concentrations of the heavy metals evaluated in this study vary by different orders of magnitude. PCA was therefore applied to the correlation matrix for this study and each variable is normalized to unit variance and therefore contributes equally. To make the results more easily interpretable, Varimax with Kaiser normalized rotation was also applied, which can maximize the variances of...
Fig. 1: A map showing the sampling points

the factor loadings across variables for each factor. In this study, all principal factors extracted from the variables were retained with eigenvalues > 1.0, as suggested by the Kaiser criterion (Kaiser, 1960).

MATERIALS AND METHODS

Four road dust sampling sites were selected in Accra (Fig. 1) based purely on traffic density. The sites have an average traffic density of 5000 vehicles/day.

The sites chosen were Tema Motorway (TM) which is the main link between the port city of Tema and Accra, the capital city. It is well noted for the high speed vehicular movements for most part of the day. The Tetteh Quarshie Interchange (TQ) was chosen because most vehicles entering Accra from the eastern corridor pass through the interchange. It is characterized by heavy vehicular traffic during the morning and evening rush hours. The John Teye-Pokuase highway is the major link between Accra and northern part of Ghana. The Mallam Junction-Weija road is the main link between Accra and the western corridor and it is part of the Trans-ECOWAS highway. At the sampling sites, about 500 g of road dust composite sample was collected by sweeping using soft
The analytical procedure. A standard reference material was used for the validation of beam and irradiation for 600 sec. The IAEA SOIL 7 Sample pellets were placed at an angle 45º to the primary Motorway. With the exception of JT all the values 117.45 mg/kg with highest value recorded at the Tema (Pb) in road dust ranging between 33.64 mg/kg and indicating the validity of the method. The results gave ratios of 10% or less calculating the ratio of experimental values to certified from SOIL 7 was compared with the certified values by laboratories of the National Nuclear Research Institute of polyethylene bags, carefully labeled and taken to the laboratories for 1 week and sieved through a 200 m mesh nylon sieve. 10.0 g of the pulverized road dust sample was pressed into a 2.5 cm diameter pellet under 10 tons pressure to obtain a thick sample pellet. All procedures of handling the samples were carried out ensuring the avoidance of any possible potential cross-contamination.

The elemental concentrations in the samples were measured using energy dispersive X-ray fluorescence (EDXRF). EDXRF provides a rapid and non-destructive method for the analysis of trace and major elements in road dust samples (Yeung et al., 2003). The Compact 3K5 X-ray Generator EDXRF spectrometer which was used for the elemental analysis has a Mo anode and operated at 800 W (40 kV and 20 mA). The irradiation was done using a Mo secondary target arrangement coupled to a liquid- nitrogen-cooled Si(Li) detector with a beryllium. The detector has a resolution of 160 eV for Mn Kα peak. Sample pellets were placed at an angle 45º to the primary beam and irradiation for 600 sec. The IAEA SOIL 7 standard reference material was used for the validation of the analytical procedure.

### RESULTS AND DISCUSSION

The mean concentration of the elements obtained from SOIL 7 was compared with the certified values by calculating the ratio of experimental values to certified reference values. The results gave ratios of 10% or less indicating the validity of the method.

The results in Table 1 show the concentration of lead (Pb) in road dust ranging between 33.64 mg/kg and 117.45 mg/kg with highest value recorded at the Tema Motorway. With the exception of JT all the values recorded from the other three sites exceeded the alert value. The mean Pb level in the road dust on the TM exceeded the intervention level of 100 mg/kg (Lacatusu et al., 2009) which could pose potential threat to humans and critical environmental media such as water bodies. Significantly high levels of Cr were measured from all sites which were above the recommended alert levels. The Tetteh Quarshie Interchange recorded the highest concentration of Cr (220.37) which is more than twice the alert levels. Cr is a known carcinogen hence such high levels should be a source of concern for human health particularly children and the vulnerable aged living close to these busy roads. The levels of V obtained in this study ranged from 28.14 to 197.00 mg/kg. The levels observed on the Tema Motorway and the Interchange were more than double the alert levels. Zn and Cu concentrations in the road dust from the sites were significantly high. The highest concentration of Zn and Cu at 371.66 and 76.53 mg/kg, respectively, were obtained from the Interchange. Zinc and Cu are known to come from tyre wear and brake wear respectively. As stated earlier, the Interchange is characterized by heavy traffic during the morning and evening rush hours leading to intermittent braking of vehicles. The element Br, Zr, Ni, Ti and Mn were obtained in significant concentrations suggesting that sources other than natural are contributing to their levels.

Enrichment factors of heavy metals were calculated (Table 2) for the road dust sample using the continental crust average where Fe was used as reference element for normalisation. The results of the enrichment factor calculations reveal the enrichment of some the element in the road dust in this study. The enrichment factors for Pb obtained from all the sites ranged from 8 to 18.5 which indicate that, the element is significantly enriched in the road dust. Pb was for a long time used as an octane enhancer until it was phased out in 2004. This could be a source of lead in the road dust. Zn showed moderate to significant (4.9-8.5) enrichment from all sites. The lowest enrichment for Zn was observed on TM whereas the highest observed at TQ. The highest enrichment of Zn at TQ could result from traffic congestion and significant tyre abrasion during rush hours (Liu et al., 2003). The enrichment of Zr was significant (6.3-14.1). The high enrichments obtained at all the four sites (JT, MJ, TM and

<table>
<thead>
<tr>
<th>Element</th>
<th>JT Mean</th>
<th>MJ Mean</th>
<th>TM Mean</th>
<th>TQ Mean</th>
<th>Alert value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti</td>
<td>2206.76</td>
<td>1885.14</td>
<td>3489.70</td>
<td>3003.15</td>
<td>50</td>
</tr>
<tr>
<td>V</td>
<td>28.14</td>
<td>61.11</td>
<td>197.00</td>
<td>112.53</td>
<td>100</td>
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<tr>
<td>Cr</td>
<td>123.75</td>
<td>166.80</td>
<td>152.25</td>
<td>220.37</td>
<td></td>
</tr>
<tr>
<td>Mn</td>
<td>275.39</td>
<td>235.93</td>
<td>355.82</td>
<td>379.63</td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td>19782.00</td>
<td>23786.25</td>
<td>35871.92</td>
<td>36630.34</td>
<td></td>
</tr>
<tr>
<td>Ni</td>
<td>9.39</td>
<td>15.88</td>
<td>6.46</td>
<td>13.44</td>
<td>75</td>
</tr>
<tr>
<td>Cu</td>
<td>29.01</td>
<td>48.25</td>
<td>43.53</td>
<td>76.53</td>
<td>100</td>
</tr>
<tr>
<td>Zn</td>
<td>124.52</td>
<td>161.43</td>
<td>213.28</td>
<td>371.66</td>
<td>300</td>
</tr>
<tr>
<td>Br</td>
<td>0.58</td>
<td>1.75</td>
<td>7.07</td>
<td>4.10</td>
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</tr>
<tr>
<td>Zr</td>
<td>712.58</td>
<td>975.49</td>
<td>662.75</td>
<td>1003.95</td>
<td></td>
</tr>
<tr>
<td>Pb</td>
<td>33.64</td>
<td>58.66</td>
<td>117.45</td>
<td>92.33</td>
<td>50</td>
</tr>
</tbody>
</table>

The calculatd results of \( I_{\text{geo}} \) values for Ti, V, Cr, Mn, Fe and Ni were all lower than 1 indicating unpolluted to moderately polluted. As confirmed by the enrichment factor calculations, the highest point of pollution was at TM which gave a high \( I_{\text{geo}} \) value of 2.65 followed by TQ which gave 2.30. The contamination factor for each element was computed and results presented in Table 4. The results reveal that the elements Ti, V, Mn, Fe, Cu and Br gave values less than 1 signifying low contamination from anthropogenic sources. Moderate contamination was obtained for V at (TM), Cr, Cu at (TQ), Zn at (JT and MJ), Br at (TQ and TM) and Pb at (JT). Zn at (TQ and TM), Zr and Pb at (MJ) revealed considerable contamination factor values. As confirmed by both EF and \( I_{\text{geo}} \) values the highest cases of contamination of Zn was obtained at TQ. This attests to the fact that slow movement of vehicles and intermittent braking during rush hours lead to increasing emissions of Cu and Zn from tyre and brake abrasions. Very high contamination was obtained for Pb at (TM and TQ). Even though there is a ban on the use of leaded gasoline in Ghana, Pb is a stable isotope in soil hence it is likely to take a considerable length of time before an appreciable depletion of the element can take place. Other sources like tyre, brake dust and other vehicular abrasions contribute to the levels of Pb in road dust (Thorpe and Harrison, 2008).

The degree of contamination (Cdeg), computed for each site is shown in Table 4. JT gave moderate degree of contamination. The sites MJ, TM and TQ gave Cdeg values of 17.95, 24.22 and 26.05, respectively. This shows considerable degree of contamination at the three sampling sites. TQ gave the highest degree of contamination which is worrying because a lot of hawkings activities take place at this site during rush hours of the day. Long term exposure within the neighbourhood can lead to adverse health effects particularly on children, pregnant women and the aged who are all known to be more vulnerable.

Pearson’s correlation coefficients of heavy metal elements in road dust from the four sites are summarized in Table 5. From the result, it is observed that there is significant positive correlation between Ti, Mn, Br, Zr and Pb with \( p<0.001 \) which may suggest a common source possibly crustal with traces of anthropogenic influence. A good correlation was observed between Cr and Ni (\( r = 0.646 \, p<0.0001 \)). Ni correlated negatively (\( r = -0.353 \) to \(-205 \, p = 0.0001 \) to 0.31) with all the elements while Cr correlated weakly (\( r = 0.316 \) to 0.181 \( p = 0.006 \) to 0.026) which show that independent sources are contributing to Cr and Ni in the road dust. A similar correlation was observed between Zr and Pb (\( r = 0.71 \, p<0.0001 \)). Zn correlated with Cr (\( r = 0.64 \, p<0.0001 \)) and Pb (\( r = 0.51 \, p = 0.0006 \)).

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Table 5: Correlation coefficient analysis

<table>
<thead>
<tr>
<th>Component</th>
<th>Ti</th>
<th>V</th>
<th>Cr</th>
<th>Mn</th>
<th>Ni</th>
<th>Cu</th>
<th>Zn</th>
<th>Br</th>
<th>Zr</th>
<th>Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Correlation</td>
<td>1.000</td>
<td>0.770</td>
<td>0.216</td>
<td>0.086</td>
<td>-0.353</td>
<td>0.778</td>
<td>0.768</td>
<td>0.715</td>
<td>0.682</td>
<td>0.697</td>
</tr>
<tr>
<td>Ti</td>
<td>0.626</td>
<td>0.725</td>
<td>-0.066</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>V</td>
<td>0.206</td>
<td>0.920</td>
<td>-0.01</td>
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<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Cr</td>
<td>0.235</td>
<td>0.183</td>
<td>0.910</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Mn</td>
<td>0.731</td>
<td>0.487</td>
<td>-0.015</td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ni</td>
<td>-0.236</td>
<td>-0.214</td>
<td>0.903</td>
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<td></td>
<td></td>
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<td></td>
<td></td>
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<tr>
<td>Cu</td>
<td>0.716</td>
<td>0.697</td>
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<td></td>
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<td></td>
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<tr>
<td>Zn</td>
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<td>0.794</td>
<td>0.032</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Br</td>
<td>0.465</td>
<td>0.650</td>
<td>0.028</td>
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<td></td>
<td></td>
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<td></td>
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</tr>
<tr>
<td>Zr</td>
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<td>0.205</td>
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<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Pb</td>
<td>0.697</td>
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<td>0.688</td>
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<td>1.000</td>
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Table 6: Rotated component matrix

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<tr>
<th>Component</th>
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<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti</td>
<td>0.626</td>
<td>0.725</td>
<td>-0.066</td>
</tr>
<tr>
<td>V</td>
<td>0.206</td>
<td>0.920</td>
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</tr>
<tr>
<td>Cr</td>
<td>0.235</td>
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</tr>
<tr>
<td>Mn</td>
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<td>0.487</td>
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<tr>
<td>Ni</td>
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<td>Zn</td>
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<tr>
<td>Pb</td>
<td>0.697</td>
<td>0.519</td>
<td>0.181</td>
</tr>
</tbody>
</table>

Extraction method: Principal component analysis

observation was made by Christoforidis and Stamatis (2009). Another group is based on the strong correlation between Cu and Zn (r = 0.938 p<0.0001). Although Zn and Cu correlate well with almost all the elements, their correlation coefficient of almost unity suggests strong association and a common source.

The rotated component matrix for the Principal Component Analysis is presented in Table 6. The Varimax rotation with Kaiser Normalisation gave three components accounting for 83.61% of the cumulative variance.

Component 1 has high factor loadings for Ti, Mn, Cu, Zn, Zr and Pb accounting for 41.33 % of total variance. This component is likely to be of natural crustal origin with traces of anthropogenic influence. Ti, Mn and Cu showed low to moderate enrichment giving an indication of crustal origin. Zn gave moderate to significant enrichment value which could suggest strong anthropogenic influence however its strong correlation with other elements gives indication that the level could partly be contributed by natural sources. Pb gave significant enrichment values and correlated well with the other elements except Cr and Ni. Hence its occurrence in component 1 can be attributed partly to complete mixing with natural soil as fresh inputs are limited to non exhaust emission (Thorpe and Harrison, 2008; Yay et al., 2008).

Component 2 has high factor loadings for Ti, V, Cu, Zn, Br and Pb which accounted for 25.71% of the total variance. Ti gave low to moderate values for EF, Igeo and CI, but correlated strongly with V, which is a product of fuel combustion (Lu et al., 2010), showing that they may be coming from a common source. The values obtained for the various indices for Zn, Br and Pb suggest anthropogenic influence to the pollutant load. Zn is a product of tyre abrasion whereas Cu and Ti are products of brake wear. According to Harrison and Thorpe (2008), both tyre and brake abrasions contribute significant amounts of Pb into the environment. This component can therefore be attributed to vehicular emission; both exhaust and non exhaust emissions.

The third component which account for 16.57% of the total variance has Cr and Ni as the major components. The EF and CF obtained for Cr gives an indication of anthropogenic origin. Although Ni gave low values in both cases the correlation analysis shows that the two elements correlated strongly with each other but poorly with the other elements. According to Lu et al (2009), Al-Shayep and Seaward, (2001), Akhter and Madany (1993) and Ferguson and Kim, (1991), the source of Ni and Cr in street dust is believed to have come from corrosion of vehicular parts. This component can therefore be attributed to corrosion of motor vehicle parts. The high rate of corrosion and wear from old vehicles (as a result of the high patronage in imported used cars) plying these roads could have accounted for the significant levels of anthropogenic contributions of Cr and Ni in the road dust.

**CONCLUSION**

Heavy metal contents and their possible sources in (The concentrations and sources of the heavy metals) road dust samples collected from selected major roads in Accra have been studied in this study. Ti, V, Cr, Mn, Ni, Cu, Zn, Br Zr and Pb were among other elements identified in the sampled road dust using Energy Dispersive X-Ray Fluorescence Analysis. The mean concentrations of V, Cr Zn and Pb were found to be higher than the alert values in some cases. Pb at TQ interchange was found to be more than twice the alert levels. The enrichment factor calculated for the elements showed that Cr, V, Cu, Zn, Br, Zr and Pb gave moderate to significant enrichment of the elements in road dust. The computed Index of geoaccumulation gave values in the range of unpolluted to moderately polluted indicating the accumulation of heavy metals from anthropogenic sources. The
contamination factors evaluated for the element further revealed that Cr, V, Cu, Zn, Br, Zr and Pb gave factors ranging from low contamination to very high contamination. The results of the degree of contamination show that TQ interchange is the most contaminated site followed by TM, MJ and JT in that order. Both correlation analysis and Principal Component Analysis (PCA) was used to determine the sources of the heavy metals. The result gave three components which are natural crust, vehicular emission, corrosion and wear of vehicle parts. The results of all the models employed agreed well in explaining the levels and sources of the elements present in the road dust samples.

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