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Abstract

The aim of this study was to evaluate the levels of selected heavy metals/metalloids in filling station dust from the Kumasi Metropolis, Ghana. A total of forty (40) dust samples were analysed for Fe, Ti, Zn, Zr, Mn, Sr, Ba, Cr, Pd, Ni, Cu, As and Mo using X-Ray Fluorescence technique. Mean concentrations of Ba, As, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sr, Ti, Zn and Zr were 92.26, 6.20, 70.41, 50.18, 466.22, 163.68, 4.63, 44.05, 46.93, 106.69, 327.51, 280.32 and

182.05 mg/kg, respectively. The pollution index (*PI*) and geo-accumulation (I_{geo}) index values were in the order of Ba < Mn < Sr < Zr < Cu < Cr < Ni < Mo < As < Zn < Pb < Fe < Ti. The pollution load index had a mean of 2.20, signifying moderate pollution. Higher *PI* and I_{geo} value for Pb, Fe, and Ti indicated high pollution. The *PCA* analysis identified anthropogenic inputs and natural origin as the main sources of pollution in filling station dust. The potential ecological risk index decreased as follows: As > Pb > Ni > Cu > Cr > Zn > Mn > Ba. The contribution of hazard quotient *via* ingestion for most of the heavy metals/metalloids were high with 11.83 % for adults and 88.17 % for children. For health risk assessment, non-carcinogenic values were below the threshold values, except hazard index *via* ingestion. The main exposure pathway for both children and adults was ingestion, followed by dermal contact and inhalation.

Keywords: dust, geo-accumulation index, Kumasi, pollution index, X-Ray Fluorescence technique

1. Introduction

Atmospheric pollution constitutes a major challenge in several countries, especially those under rapid development (Schleicher et al. 2011; He et al. 2013; Teng et al. 2014). During the combustion of wood and fossil fuels, waste incineration, high-temperature industrial processes and traffic, dust containing trace metals are released into the atmosphere (Allen et al. 2001). Dust particulates can affect human health (Ruiz-Jimenez et al. 2012), especially the presence of trace metals, which are noxious to humans *via* inhalation or ingestion (Khairy et al. 2011; Lu et al. 2014).

Heavy metals/metalloids pollution in dust is a problem due to their non-biodegradability, wide occurrence, toxicity, as well as their ability to accumulate over time (Dong et al. 2011). Heavy metals including Ni, Cd, Pb, Zn, Cu, Hg, Cr and others refer to metals with densities >5 g/cm^3 (Li et al. 2014). As a result of the similarities in chemical properties and fate in the environmental, arsenic is frequently referred to as a heavy metal (Huamain et al. 1999). Heavy metals/metalloids pollution in dust is an irreversible process and very difficult to remove once it occurs (Zhou and Song 2004). They also have the potential to cause biomagnification and bioaccumulation in the ecosystem (Manahan 2000). Improper disposal of engine oil, brake fluid, transmission oil and leaded gasoline around the vicinity of fuel contributes to the heavy metals/metalloids load (Dauda and Odoh 2012, (Khorshid and Thiele-Bruhn 2016; Luo et al. 2012; Pant and Harrison 2013). Heavy metals/metalloids in dust can affect human health when exposed *via* ingestion, inhalation and dermal contact (Ling et al. 2008; McLaughlin et al. 2000). For example, extreme exposure to Pb can harm the skeletal, nervous, endocrine, circulatory, immune and enzymatic systems (Zhang et al. 2012). In addition, exposure to Cd can cause pulmonary adenocarcinomas, hypertension, kidney dysfunction, lung cancer, prostatic proliferative lesions and bone fractures, while As exposure can cause dermal lesions, skin cancer, peripheral vascular disease, and peripheral neuropathy (Chen et al. 2015).

Source identification of heavy metals/metalloids is crucial for effective dust remediation and pollution control (Guo et al. 2015). To check the potential risk of metals, assessment tools including enrichment factor (EF), geo-accumulation index (I_{geo}), potential ecological risk index (PER), pollution load index (PLI), and human health risk assessment are used to measure the

pollution level (Ma et al. 2016). Multivariate statistical methods have also been used to access the different sources of metals in the dust (Ma et al. 2016).

In relation to several studies, heavy metals/metalloids pollution in the environment mostly originate from anthropogenic sources (Wei and Yang 2010). In urban road dusts, the anthropogenic sources of heavy metals/metalloids include industrial emission (chemical plant, power plants, metallurgical industry, coal combustion, auto repair shop, etc.), traffic emission (brake lining wear particles, weathered street surface particles, vehicle exhaust particles, tire wear particles), pavement surface, weathering of building and domestic emission (Ahmed and Ishiga 2006; Banerjee 2003; Sezgin et al. 2004). Lately, dust is assessed as an analytical model with implications on environmental and human health (Shen et al. 2017; Ying et al. 2016). Therefore, a number of studies on human health risk evaluation of heavy metals/metalloids pollutions in dust are reported (Han et al. 2016; Lin et al. 2015; Qing et al. 2015). Studies on human health risk assessment of heavy metals/metalloids in Ghana have largely focused on food, cosmetic product, soil, wastewater, ground and surface water contamination (Agorku et al. 2016; Akoto et al. 2008; Akoto et al. 2009; Asare-Donkor et al. 2015; Atiemo et al. 2011; Boateng et al. 2015; Eze et al. 2010; Guo et al. 2015; Kwaansa-Ansah et al. 2011; Nkansah et al. 2016; Obiri 2007).

In the past decade, several studies on dust have been conducted on metal concentrations, source identification and distribution (Glorennec et al. 2012; Laidlaw and Taylor 2011; Lu et al. 2010). While there have been some recent studies on road and street dust (Duong and Lee 2011; Li et al. 2001), very few studies have been reported in dusts around oil filling stations (Afrifa et al. 2013; Dauda and Odoh 2012; Ekperusi and Aigbodion 2015; Emmanuel et al. 2014). Despite these serious effects of trace metals on human health, particularly for children and adults, information about pollution levels and health risks of heavy metals/metalloids in the dust around fuel filling stations in Kumasi metropolis are lacking. The metropolis houses several types of highly polluting filling station industries. Thus, the Kumasi metropolis is facing severe threats from contamination induced by the fast growth of filling station activities that adversely can influence human health in the metropolis. This study, therefore, aims: (1) to measure the concentration of Fe, Ti, Zn, Zr, Mn, Sr, Ba, Cr, Pd, Ni, Cu, As and Mo, (2) to ascertain the possible sources and their relationship using multivariate analysis, and (3) to evaluate the health risk and pollution

levels of heavy metals/ metalloids in dust around the vicinity of fuel filling stations from Kumasi Metropolis.

2. Materials and methods

2.1 Study area

Sampling was done from two filling stations in each of the twenty (20) sub-metros in Kumasi. The Metropolis of Kumasi is centrally situated in the Ashanti Region, Ghana with about 270 km north of Accra. Kumasi lies between longitude $1.30^0 – 1.35^0$ and latitude $6.35^0 – 6.40^0$ with an elevation above sea level of 250 – 300 metres. The wet sub-equatorial region is the area where Kumasi Metropolis has a mean maximum and minimum temperature of 30.7 ^oC and 21.5 ^oC, respectively. The average humidity is about 84.16 % and 60.00 %, respectively (Ghana Statistical Services 2010). The double maxima rainfall regime of Kumasi (214.3 mm in June and 165.2 mm in September) has a direct effect on the environment and population growth (Ghana Statistical Services 2010). The major rainy season occurs between March/April to July and September to October as the minor rainy season. Furthermore, between November to early February, Kumasi experiences dry season, which affects small streams to dry up. The land area of the Metropolis is about 254 square kilometres. Kumasi is accessible from all zones of the country due to its unique central position. The administrative capital of Ashanti Region is Kumasi, which is the second largest city of Ghana. Kumasi is a rapidly developing metropolis with about two million people and annual growth rate of more than 5.4 %. The physical structure of Kumasi is global with a centrally located area for commercial activities, such as the Adum shopping centre, the central market and Kejetia Lorry Park. In addition, there are satellite markets, such as Oforikrom market, Asafo market, Atonsu market and Bantama market in the metropolis. Other economic activities include the Anloga wood market, the Kaase/Asokwa industrial area and Suame Magazine. Most industries, which deal in soap making, logging and food processing, are located at Kaase and Asokwa industrial area. According to the Ghana Statistical Service, about 60, 46 and 48 % of the Metropolis are rural, peri-urban and urban, respectively, confirming the rapid rate of urbanisation (Ghana Statistical Services 2010).

2.2 Sampling

Twenty fuel filling stations were chosen randomly, two from each of the twenty sub-metros for study. At the sampling site, the site was zoned out into five parts, namely, the four corners and the Centre. The GPS coordinates of all the sampling sites were taken and are presented in **Figure 1**.

<Figure 1>

Dust samples were sampled from surfaces by sweeping. The dust samples were gathered from these zones into brown paper envelopes using soft touch brush and plastic dustpan. The paper envelopes were labelled according to the name of the filling station and the sub-metro. Three samples were then sampled from the Kwame Nkrumah University of Science and Technology botanical gardens to serve as pristine samples since the dust location is devoid of any contamination from vehicular activity.

2.3 Sample preparation

The dust samples were dried at room temperature. The samples were then sieved with a mesh of pore size of 60 and aperture of 20 micrometres. As a measure of avoiding cross-contamination, the sieves were washed intermittently after each sieving. The sieved samples were homogenised and ground with mortar and pestle and then kept in a desiccator prior to analysis.

2.4 Sample analysis

The heavy metals/metalloids content in the dust samples was determined with Thermo Scientific Niton X-ray Fluorescence (XRF) analyser (NDTr-XL3t-86956). The sample holder was filled halfway with the digested sample and covered with a Mylar film. The cupped sample was then placed in the XRF shroud and scanned for 180 seconds to obtain the desired result. All the samples were treated in the same manner. All the XRF sample analysis was done in triplicate. The equipment was calibrated with reference material of OC USGS SAR-M 180-673. Recovery rates in the range of 90 and 116 % were sufficient for analysis. The limits of detection for As, Cr, Ba, Fe, Cu, Mn, Mo, Pb, Sr, Ni, Ti, Zr, and Zn were 0.01, 0.05, 0.03, 0.82, 0.28, 0.70, 0.56, 1.60, 0.23, 0.35, 0.64, 1.27, and 0.15 mg/kg, respectively; while limits of quantification were 0.04, 3.24, 0.52, 0.58, 2.13, 0.95, 2.62, 5.12, 3.53, 0.86, 1.56, 0.58 and 4.07 mg/kg, respectively.

2.5 Evaluation of heavy metals/metalloids contamination

Pollution levels of heavy metals/metalloids in filling station dust were investigated using *PER*, I_{geo} and *PI* (Pan et al. 2016; Ying et al. 2016). Geoaccumulation index was initially applied to river sediments and has also been used for the evaluation of dust pollution (Salati and Moore 2010). Geoaccumulation index allows the assessment of environmental contamination between pre-industrial and current concentrations. In this study, I_{geo} was evaluated following the Muller (1969) equation:

$$I_{geo} = I_2\left[\frac{C_n}{1.5B_n}\right] \quad (1)$$

where B_n is geochemical background value in dust and C_n is the estimated concentration of heavy metals/metalloids *n*. In this study, the background geochemical compositions by Taylor and McLennan (1995) were used as the background values for calculating the I_{geo} values. The constant 1.5 allows us to analyse the natural variations in the concentration of a given substance in the environment and to detect small anthropogenic effects (Wei and Yang 2010). The I_{geo} classification is given in Table 1 (Chen et al. 2005).

<Table 1>

The pollution index was the concentration of heavy metals/metalloids in the filling station dust relative to the background concentration of the equivalent heavy metals/metalloids (Kamani et al. 2015). To evaluate the pollution status of the filling station dust samples, the *PLI* of each heavy metals/metalloids was calculated according to Eqn. 2:

$$PLI = \sqrt[n]{PL_1 x\ PL_2 x\ PL_2 ... x\ PL_n,} \quad (2)$$

The *PLI* classification of heavy metals/metalloids contamination is given in **Table 1** (Islam et al. 2015).

2.6 Potential ecological risk

To access the heavy metals/metalloids toxicity, the *PER* method by Hakanson (1980) was used to estimate their level of pollution in filling station dust. The *PER* was evaluated according to Eqn. 3-5:

$$PER = \sum_i^n E_r^i \quad (3)$$

$$E_r^i = T_n^i \text{ x } C_r^i \quad (4)$$

$$C_r^i = C^i / C_n^i \quad (5)$$

where C_j^i, C^i, C_n^i, $E_r^i$and T_n^i represent the pollution factor, estimated concentration, background concentration, PER index and toxic response factors for heavy metals/metalloids *n*, respectively. The toxic response factor for Cu, Zn, Pb, Mn, Ni, Cr, As and Ba were 5, 5, 1, 1, 5, 2, 10 and 1, respectively (Hakanson 1980).

As summarised in **Table 1**, the E_r^i and PER classifications defined by Hakanson (1980) were used in this study.

<Table 1>

2.7 Human health risk assessment

Human health risk assessment involves the evaluation of possible human health effect in the contaminated environmental media (Li et al. 2014). The carcinogenic and noncarcinogenic risks through dermal contact, ingestion and inhalation exposure pathways were evaluated using the human health risk assessment (Qing et al. 2015).

2.8 Exposure assessment

The health risk assessment is centred on the exposure factors and guidelines handbook of United States Environmental Protection Agency (USEPA) (USEPA 2002; USEPA 2003). The average daily dose (ADD) *via* inhalation (ADD_{inh}), ingestion (ADD_{ing}), and dermal contact (ADD_{derm}) for both children and adults were evaluated following Eqns. 6–7:

$$\mathrm{ADD_{ing}} = \mathrm{C_{dust}}\, x\, \frac{\mathrm{IngR\ x\ EF\ x\ ED}}{\mathrm{BW\ x\ AT}}\, x\, 10^{-6} \quad (6)$$

$$\mathrm{ADD_{inh}} = \mathrm{C_{dust}}\, x\, \frac{\mathrm{InhR\ x\ EF\ x\ ED}}{\mathrm{PEF\ x\ BW\ x\ AT}} \quad (7)$$

$$\mathrm{ADD_{dermal}} = \mathrm{C_{dust}}\, x\, \frac{\mathrm{SA\ x\ AF\ x\ ABS\ x\ EF\ x\ ED}}{\mathrm{BW\ x\ AT}}\, x\, 10^{-6} \quad (8)$$

The exposure factors and values (Ying et al. 2016) used in the risk assessment are given in **Table 2**.

<Table 2>

2.9 Non-carcinogenic risk assessment

The carcinogenic and noncarcinogenic adverse effects were evaluated with the hazard quotient (*HQ*), carcinogenic risk (*RI*), and hazard index (*HI*) approaches (Ying et al. 2016). According to (USEPA 1989), the hazard quotient is the average daily dose of heavy metals/metalloids with reference to its reference dose (RfD):

$$\mathrm{HQ} = \frac{\mathrm{ADD}}{\mathrm{RfD}} \quad (9)$$

If $HQ < 1$, signifies no adverse effects, whereas $HQ > 1$, signifies adverse effects (USEPA 2011).

To assess the overall adverse effects of noncarcinogenic risk, the hazard index approach was applied (USEPA 1986). The *HI* is the sum of *HQ* through the three exposure pathways for heavy metals/metalloids. For a mixture of contaminations, the *HI* was evaluated according to Eqn. 10 (USEPA 1989):

$$HI = \sum HQ_i \quad (10)$$

$HI < 1$ denotes noncarcinogenic effects, whereas $HI > 1$ signifies adverse effects.

2.10 Carcinogenic risk assessment

The carcinogenic risk is the possibility of an individual to develop cancer during the lifetime exposure to the carcinogenic threats (Li et al. 2014). According to (USEPA 1989), the slope factor (*SF*) directly convert the *ADD* of contaminant exposed over a lifetime risk of a cancer patient:

$$\mathrm{Risk} = \mathrm{ADD\ x\ SF}$$

The values for *SF* in mg/kg day, *RfD* and other calculated parameters are presented in **Table 3**.

<Table 3>

Risk value < 10^{-6} represents no carcinogenic risk to health from the dust, while a risk value > 1 x 10^{-4} denotes high risk of developing cancer. A risk value ranging from 1 x 10^{-6} - 1 x 10^{-4} signify an acceptable risk to human health (Hu et al. 2012).

2.11 Statistical analysis

Descriptive statistics and multivariate analysis were calculated using IBM Statistical Package for the Social Sciences (SPSS) *version* 20. The normality tests were analysed using the Shapiro-Wilk test. The Shapiro-Wilk test is based on the correlation between the heavy metals/metalloids concentration and the corresponding normal scores (Peat and Barton 2008) and offers better power than the Kolmogorov-Smirnov test even after the Lilliefors correction (Steinskog et al. 2007). Principal component and Pearson's correlation analysis were used to evaluate and ascertain the possible source of heavy metals/metalloids contamination in the filling station dust. According to Lee et al. (2006), the principal component is transformed from the original variable *via* eigen analysis. Herein, varimax rotation with Kaiser normalisation was employed to extract components with eigenvalue >1.

3. Results and Discussion

3.1 Heavy metals/metalloids concentration in filling station dust

The descriptive statistics of heavy metals/metalloids concentration in the filling station dust sampled from the Kumasi Metropolis are presented in **Table 4**.

<Table 4>

The filling station dust exhibited distinct variations in the levels of heavy metals/metalloids; with concentration ranges of 2.58-13.28, 33.72-142.63, 21.81-150.04, 89.85-681.87, 14.55-248.58, 2.77-8.93, 56.08-327.06, 6.48-129.87, 9.42-183.94, 347.00-585.53, 12.95-520.68, 21.30-59.91, and 92.02-250.38 mg/kg, for As, Ba, Cr, Fe, Cu, Mo, Mn, Pb, Sr, Ti, Ni, Zn and Zr respectively. The mean concentrations of As, Ba, Cr, Fe, Cu, Mn, Mo, Pb, Sr, Ti, Zn, Ni, and Zr were 6.20, 92.26, 70.41, 466.22, 50.18, 163.68, 4.63, 46.93, 106.69, 327.51, 280.32, 44.05, and

182.05 mg/kg, respectively. The mean concentrations of As, Cr, Cu, Fe, Mn, Mo, Pd, Ti, Zn and Zr were above the corresponding reference value and this indicates pollution of these metals in the filling station dust. Considering the mean concentration, the heavy metals/metalloids in the filling station dust were in the increasing order: Fe > Ti > Zn > Zr > Mn > Sr > Ba > Cr > Cu > Pd > Ni > As > Mo. The normality distributions of heavy metals/metalloids concentrations were checked by the one-sample Shapiro-Wilk normal test. The results showed that the concentrations of Fe, Ti, Zn, Zr, Mn, Sr, Ba, Cr, Cu, Pd, Ni and Mo were normally distributed ($p > 0.05$) in the collected filling station dust (Wang et al. 2012), while As concentration showed non-normal distribution due to ($p < 0.05$) (Zhou et al. 2014). According to Nezhad et al. (2015), the coefficient of variation, which shows the variability degree of heavy metals/metalloids concentration are classified as low variability with coefficient of variation ≤ 20 %, moderate variability (21 % < coefficient of variation ≤ 50 %), and high variability (50 % < coefficient of variation ≤ 100 %) (Qing et al. 2015). Based on this classification, the heavy metals/metalloids in the filling station dusts decreases as: Cu (99.74 %) > Pb (68.68 %) > Cr (52.24 %) > As (43.87 %) > Zn (42.74 %) > Ba (41.66 %) > Mn (40.15 %) > Ni (36.98 %) > Mo (36.07 %) > Sr (34.74 %) > Zr (22.18 %) > Ti (7.33 %) > Fe (6.75 %). The coefficient of variation of Pb and Cr specified a moderate degree of variability, which reveals no homogeneous levels. A large coefficient of variation of >70 % was found for Cu, an indication that Cu concentration differed greatly in the study region. The skewness values of Cu, As, Mo and Pb were >1, indicated that they were positively skewed towards low concentration. Moreover, while the negative kurtosis values of Ba, Cr, Ni, Sr, Zn and Zr, indicated that their distribution in the filling station dust was less steep than normal (Chen et al. 2012).

3.2 Comparison with other heavy metals/metalloids dust studies

In order to compare the examined heavy metals/metalloids concentration with studies from other countries, their concentrations in filling station dust of Kumasi Metropolis were collated (**Table 5**).

<Table 5>

In **Table 5**, the mean concentrations of Cr measured agreed with data from Guangzhou and Xiandao, both from China, but lower than those reported in other cities, such as Madrid, Ottawa,

Hangzhou, Urumqi. The mean concentrations of Ni measured was comparable with those reported in Oslo, Madrid, Baoji and Urumqi, but was higher in other compared cities except for Shanghai, China. The mean concentration of Cu was lower than cites except for Xiandao. The Zn concentration is similar to Urumqi, while lower than other cities except for Ottawa and Xiandao. The mean concentrations of As and Pb were lower than other compared cites except for Ottawa. However, the mean concentrations of Mn, Fe, Sr, Zr, Ti and Ba were higher in all the compared cities (**Table 5**).

3.3 Multivariate analysis

Source of heavy metals/metalloids pollution

To check the level of heavy metals/metalloids pollution and identify their possible source in the filling station dust, principal component and correlation coefficient analyses were performed (Facchinelli et al. 2001). The relationships among the estimated heavy metals/metalloids were carried out using Pearson's correlation analysis. **Table 6** demonstrates that Mn, Cr, Ti, and Zr were significantly correlated ($p < 0.01$) with each other, signifying similar sources of these metals.

<Table 6>

Pb was positively correlated with Zn, Cu, and Fe; Zn was positively correlated with Mn, Cu, Fe, and Cr, but negative with Ni. There existed a significant positive correlation between Cu with Fe and Mn. A similar trend was observed for Fe with Mn and Cr; and for Cr with Ti and Zr. In the case of Ti, there was significant positive correlation with Sr, but negative with Zr. Though the significant positive relationship cannot always be attributed to a similar source, this correlation analysis can still offer interesting information on the pathway and source of heavy metals/ metalloids (Lu et al. 2010). For instance, Zn, Cu, and Fe, and Pd with significant positive correlation could be originating from anthropogenic sources, such as fossil fuel combustion *via* atmospheric deposition and leaded fuel. Mo, Ba and As might have originated from different sources with reference to the other heavy metals/metalloids.

In addition to the principal component analysis, Bartlett's and KMO test were employed to examine the sampling sphericity and adequacy. The PCA results of heavy metals/metalloids are given in **Table 7**.

<Table 7>

Three components with eigenvalues >1 were obtained, which account for 97.80 % of the extracted variance. The first component with positive loading for Mo, As, Mn, Ti, Ba, Zr and Sr, whose coefficients were 0.529, 0.945, 0.534, 0.946, 0.524, 0.734 and 0.928, respectively explained 45.25 % of the total variance. The Mo, As, Mn, Ti, Ba, Zr and Sr in PC1 were strongly correlated, signifying their similar source. PC1 consisting of As, Mn, and Ba were mainly influenced by natural sources (Lu et al. 2010). The second component showed strong positive loading (>0.75) for Pb, Zn and Cu, whose coefficients were 0.987, 0.999 and 0.862, respectively explained 33.30 % of the total variance. PC2 including Pb, Zn and Cu could be associated with anthropogenic inputs, such as industrial and traffic pollution (Chen et al. 2014). Moreover, improper disposal of engine oil and a significant amount of emissions influences Pb, Cu, and Zn pollution in the filling station dust, as shown in PC2. The third component showed strong loading (>0.75) for Mo (0.830) and Ba (0.851) with 19.25 % of the extracted variance. The heavy metals/metalloids in the three PC's demonstrate their different sources. The correlation between Mo and Ba in PC1 and PC3, suggests their origin from dust as the parent material. The poor loadings of Ni, Fe, and Cr can be attributed to differences in sources of materials and geochemical behaviour of parameters (Edet et al. 2003), as well as quasi-independent behaviour within the group (Lu et al. 2010).

3.4 Heavy metals/metalloids pollution assessment

The estimated I_{geo} result of heavy metals/metalloids in filling station dust is given in **Figure 2**.

<Figure 2>

The mean values of I_{geo} increased following the order: Ba < Mn < Sr < Zr < Cu < Cr < Ni < Mo < As < Zn < Pb < Fe < Ti. The mean I_{geo} of Mo, Zr, Sr, Cu, Mn, Cr, Ba and Ni showed that dust from filling stations was practically unpolluted. The mean I_{geo} of As and Zn signify moderately polluted, whereas the mean I_{geo} for Pb, Fe, and Ti designate very highly polluted. In the study

area, the I_{geo} values signify 'unpolluted' to 'highly polluted' of the investigated heavy metals/metalloids. Among the estimated heavy metals/metalloids, Ti and Fe were significantly accumulated in the filling station dust, as showed by their corresponding mean geochemical accumulation values of 12.46 ± 0.41 and 12.06 ± 0.64. The presence of Zn and As in the filling station dust poses an adverse environmental concern. Similarly, Wei and Yang (2010) deduced that for heavy metals in urban soils in China, Cr and Ni appeared to cause the least contamination in the selected cities, while Cu, Pb, Zn, and Cd showed the highest I_{geo} values. Cr and Ni appear to be the least contaminated elements in all the cities, while Cu, Pb, Zn and Cd show the highest I_{geo} values for most cities (Wei and Yang 2010). The PIs results for heavy metals/metalloids in the filling station dust varied significantly across the different heavy metals/metalloids (**Figure 3**).

<Figure 3>

The mean PIs of Fe, Pb and Ti were much higher, varying from 16.51 to 97.02, 0.64 to 9.97 and 29.33 to 88.24, with a values of 5.75, 46.67 and 58.45, respectively. This showed that Mo, Zn, As, Cu, Cr and Ni of filling station dust were classified as middle PI. Zr, Sr, Mn and Ba exhibited lower mean values and were classified as low PI. The PI value for the calculated heavy metals/metalloids was in the decreasing order: Ba (0.11) < Mn (0.19) < Sr (0.22) < Zr (2.06) < Cr (1.30) < Cu (1.36) < Ni (1.47) < Mo (2.06) < As (2.82) < Zn (2.85) < Pb (5.75) < Fe (6.54) ≤ Ti (6.54). Therefore, Pb, Fe, and Ti were the main contaminants of the study area reaching a very strong contamination level. The high level of Pb in filling station dust samples can be credited to leaded fuels, emissions from tyre wear, bearing wear and lubricating oils (Kamani et al. 2015). The PLIs in all filling station dust samples varied between 0.10 and 6.32 with a mean value of 2.20, signifying that the filling station dust was 'moderately polluted' in the Kumasi Metropolis.

3.5 Ecological risk index

The *PER* is a measure of the sensitivity of several biological systems to deadly substances and elucidates the potential risks induced by heavy metals/metalloids (Qing et al. 2015; Ying et al. 2016). The *PER* results are presented in **Table 8**.

<Table 8>

The mean E_r^i value for all the heavy metals/metalloids except arsenic were below 40, thus indicating low *PER*. This signifies that arsenic has a moderate risk to the environment and human health. The *PER* index showed the order: As > Pb > Ni > Cu > Cr > Zn > Mn > Ba. To access the overall PER of the observed heavy metals/ metalloids in the filling station dust in the Kumasi metropolis, *PER* was evaluated as the summation of the risk factors. The contribution to the overall *PER* displays that arsenic contributed 32 % of the total *PER*. The *PER* value for all the sampling sites ranged from 1.34 to 784.73 with a mean value of 187.72, indicating low to very high potential ecological risk. Nevertheless, the mean *RI* shows a considerably high risk (*PER* > 100) in the Kumasi Metropolis, largely due to arsenic contamination. This should be remediated to prevent the possible ecological adverse effect.

3.6 Human health risk assessment

Exposure assessment

The calculated mean daily intake of heavy metals/metalloids for adult and children in the filling station dust *via* inhalation, dermal contact and ingestion are presented in **Tables 9**.

<Table 9>

The trends of *ADDs* for heavy metals/metalloids *via* dermal contact, inhalation, and ingestion were in the order: Fe > Ti > Zn > Zr > Mn > Sr > Ba > Cr > Cu > Pb > Ni > As > Mo. The exposure route, which on the average brings about the highest risk for adults and children was ingestion followed by dermal contact with inhalation being the lowest. Qing et al. (2015) observed the same order of daily intake of heavy metals.

The health risks assessment results are given in **Table 10**.

<Table 10>

The *HQ* results for all the heavy metals/metalloids were <1, signifying no risk for both adult and children. The mean *HQ* values for adults and children *via* ingestion were as follows: Zr > Cr > As > Pb > Ni > Cu > Mn > Mo ≥ Zn > Fe > Ba > Sr > Ti, while that *via* inhalation were Cr > As > Pb > Zn > Ni > Cu > Mn > Mo > Fe > Ba > Sr > Ti > Zr, and *via* dermal contact were Ti > Fe > Zn > Sr > Mn > Ba > Cu > Ni > Cr > Pb > Zr > Mo > As. In summary, the mean exposure

route of heavy metals/metalloids for adults and children decreased according to the following order: inhalation < dermal contact < ingestion. In the present study, cutaneous and oral exposure are more important than inhalation. The findings in this study agree well with other studies (Benhaddya et al. 2016), which also observe ingestion to be the highest exposure pathway to health risk. The contribution of HQ_{ing} for most of the heavy metals/metalloids were the highest with 11.83 % for adults and 88.17 % for children. This confirms ingestion as the major exposure route that can adversely affect human health. The *HI* results *via* inhalation and dermal contact with all the heavy metals/metalloids were <1, except for ingestion in adult, signifying no non-carcinogenic risk. The *HI* values in children were observed to develop more non-carcinogenic risk in the filling station dust compared to adults in the Kumasi Metropolis. This can be attributed to the fact that children tend to have significant contact with soil during their outdoor play activities and are more likely to have a direct hand-to-mouth exposure of dust (Luo et al. 2012). Similar findings can be seen from other studies including Qu et al. (2012), Zota et al. (2011), and Man et al. (2010). Owing to lack of carcinogenic slope factor for Cu, Ba, Mn, Fe, Ni, Ti, Mo, Zn, Sr and Zr, only the slope factors for As, Cr and Pb were given in the literature. The *RI* values in the filling station dusts of Kumasi Metropolis were 8.74×10^{-8} (As), 3.31×10^{-7} (Cr), and 3.75×10^{-9} (Pb) for children and 1.17×10^{-7} (As), 4.44×10^{-10}(Cr), and 5.03×10^{-9} (Pb) for adults (Table 9). The risk index of Cr, As and Pb for both adults and children were $<10^{-6}$, showing no plausible carcinogenic risk. However, the HI level *via* ingestion indicates that the carcinogenic risks of the observed heavy metals/ metalloids cannot be overlooked.

4. Conclusion

The concentration, pollution level, and health risks assessment of heavy metals/metalloids in dust from filling stations within the Kumasi Metropolis, Ghana were evaluated in this study. This study offers significant information on the concentrations of Fe, Ti, Zr, Mn, Sr, Ba, Zn, Cr, Cu, Pd, As, Ni and Mo in dust impacted by fuel and other related emissions. ~~The average concentration of Ba, Cu, As, Cr, Fe, Mn, Mo, Ni, Sr, Pb, Ti, Zr, and Zn were 92.26, 50.18, 6.20, 70.41, 466.22, 163.68, 4.63, 44.05, 106.69, 46.93, 327.51, 182.05, and 280.32 mg/kg, respectively.~~ The PCA analysis identified anthropogenic inputs and natural origin as the main sources of heavy metals/metalloids in the filling station dust of Kumasi Metropolis. The *PI* and I_{geo} revealed the order of Ba < Mn < Sr < Zr < Cu < Cr < Ni < Mo < As < Zn < Pb < Fe < Ti for

both I_{geo} and *PI*. The higher *PI* values for Pb, Fe, and Ti in the filling station dust indicated a very strong pollution level, which could be attributed to leaded fuels and emissions from bearing wear, lubricating oils and tyre wear. The *PLIs* in all filling station dust samples varied between 0.10 and 6.32 with an average value of 2.20, signifying moderately pollution in the Kumasi Metropolis. The mean value of E_r^i for all the heavy metals/metalloids except As were below 40, indicating low risk. The *PER* value for all the sampling sites ranged from 1.34 to 784.73 with a mean value of 187.72, ~~indicating low to very high *PER*. The *PER* index revealed that the heavy metals/metalloids pollution decreases as: As > Pb > Ni > Cu > Cr > Zn > Mn > Ba.~~ The *HQ* results indicate no risk for both adult and children, as the *HQ* values were lower than safe limit. The contributions of HQ_{ing} for most of the heavy metals/metalloids were the highest with 11.83 % for adults and 88.17 % for children. This signifies ingestion as the major exposure route that can adversely affect human health. The *HI* values were <1, except for *HI* value for ingestion in adult, signifying no non-carcinogenic risk. Nonetheless, the carcinogenic risks values ~~of 8.74 x 10^{-8} (As), 3.31 x 10^{-7} (Cr), and 3.75 x 10^{-9} (Pb) for children and 1.17 x 10^{-7} (As), 4.44 x 10^{-10} (Cr), and 5.03 x 10^{-9} (Pb)~~ for adults were lower than the threshold value of 1×10^{-6}, showing no carcinogenic risk. The main exposure pathway for both children and adults is ingestion, followed by dermal contact and inhalation. The health risk assessment revealed ingestion as the major exposure route with children being the most exposed to heavy metals/metalloids pollution compared to adults in the Kumasi Metropolis, Ghana. Therefore, the results in this study will be beneficial for environmental planning authorities in the Kumasi Metropolis to manage and control further heavy metals/metalloids pollution at the various filling station sites in the Metropolis.

Compliance with ethical standards

Yes

Conflict of interest

The authors declare that they have no conflict of interest.

Authors' contributions

All the authors contributed equally to the preparation of this manuscript. All authors read and approved the final manuscript.

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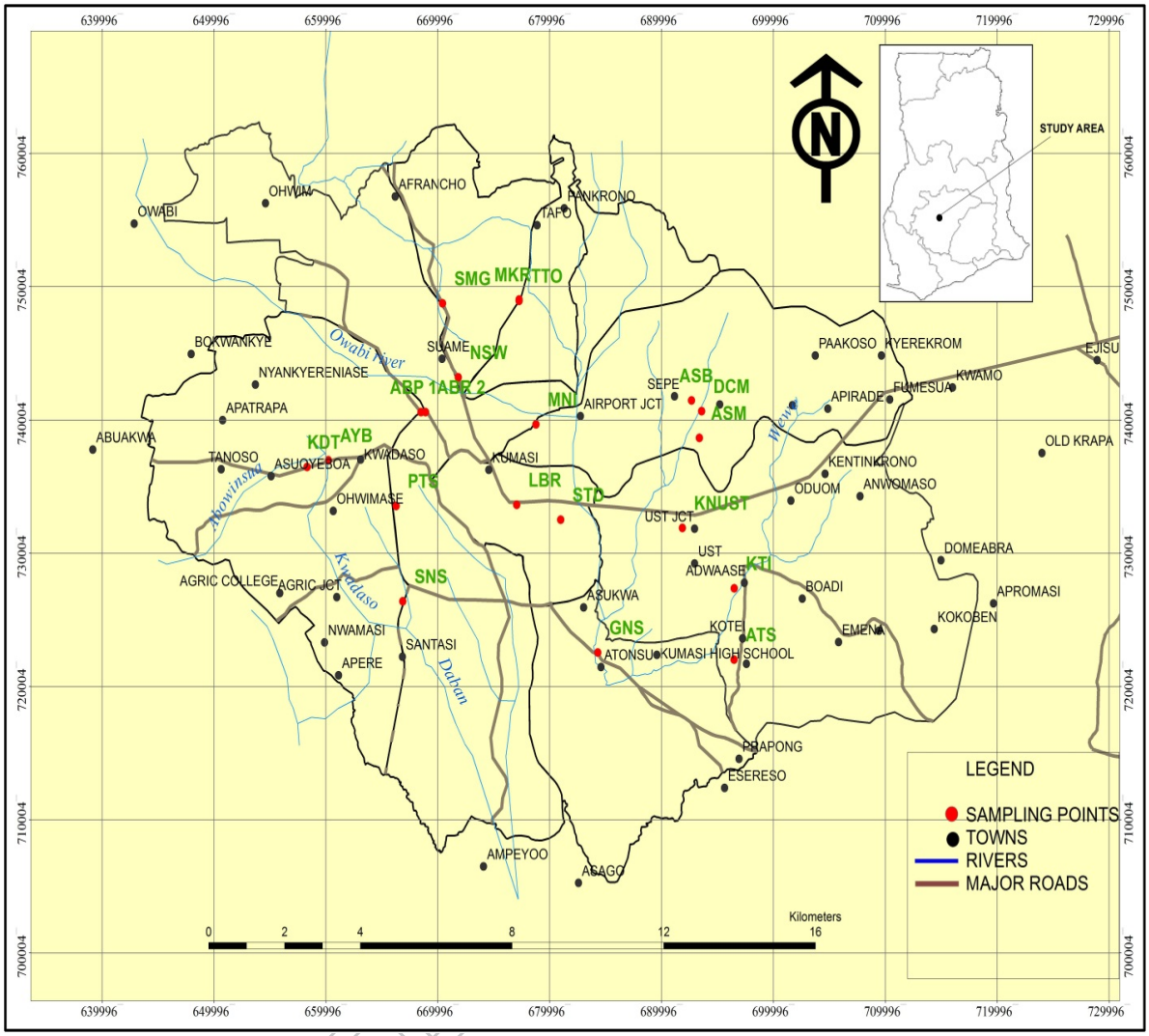

Figure 1. Map of Kumasi displaying the sampling areas

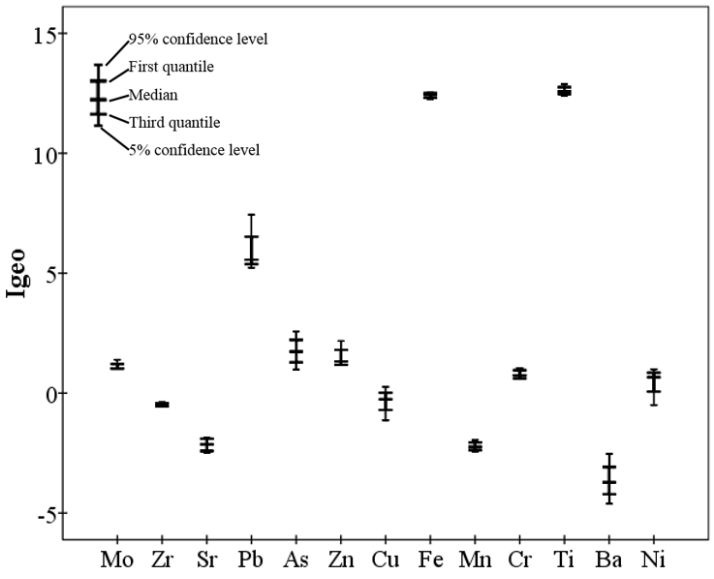

Figure 2. Box-plots of the I_{geo} for heavy metals/metalloids in the filling station dust of Kumasi Metropolis.

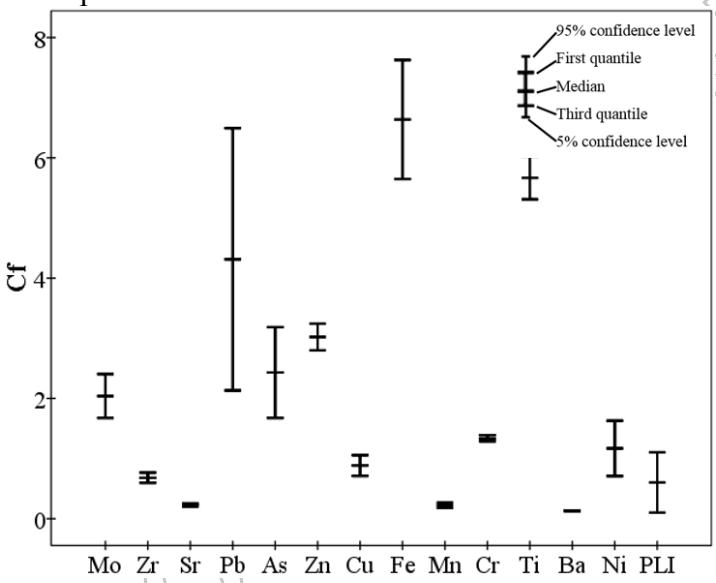

Figure 3. Box-plots of pollution load index and pollution index for heavy metals/metalloids in the filling station dust of Kumasi Metropolis.

Table 1. Potential ecological risks, risk category indices and the related classifications of heavy metals/metalloids pollution

I_{geo}	Risk category	*PLI*	Risk category	E_r^i value	Risk category	*PER* value	Risk category
$I_{geo} \leq 0$	practically unpolluted	$PLI \leq 1$	unpolluted	<40	Low risk	≤50	Low risk
$0 \leq I_{geo} \leq 1$	unpolluted to moderately polluted	$1 \leq PLI \leq 2$	unpolluted to moderately polluted	40-80	Moderate risk	$50 < PER \leq 100$	Moderate risk
$1 \leq I_{geo} \leq 2$	moderately polluted	$2 \leq PLI \leq 3$	moderately polluted	80-160	Considerable risk	$100 < PER \leq 200$	Considerable risk
$2 \leq I_{geo} \leq 3$	moderately to strongly polluted	$3 \leq PLI \leq 4$	moderately to highly polluted	160-320	High risk	$PER > 200$	High risk
$3 \leq I_{geo} \leq 4$	strongly polluted	$4 \leq PLI \leq 5$	highly polluted	≥320	Significantly risk		
$4 \leq I_{geo} \leq 5$	strongly to extremely polluted	$PLI > 5$	very highly polluted				
$I_{geo} > 5$	extremely polluted						

Table 2. Exposure factors and reference value of parameters used for the human health risk evaluation of heavy metals/metalloids in filling station dust

Factor	Definition	Unit	Value		Reference
			Children	Adults	
C_{dust}	Heavy metal concentration in dust	mg/kg			This study
IngR	Ingestion rate	mg/day	200	100	USEPA (2011
EF	Exposure frequency	days/year	350	350	Environmental site assessment guideline (2009
ED	Exposure duration	years	6	24	USEPA (2011
BW	Body weight	kg	15	55.9	Environmental site assessment guideline (2009
AT	Average time	days	365 x ED	365 x ED	USEPA (1989
InhR	Inhalation rate	m^3/day	7.63	12.8	Li et al. (2001
PEF	Particle emission factor	m^3/kg	1.36 x 10^9	1.36 x 10^9	USEPA (2011
SA	Exposure skin surface area	cm^2	1600	4350	Environmental site assessment guideline (2009
AF	Skin adherence factor	mg/cm day	0.2	0.7	USEPA (1993
ABF	Dermal absorption factor	no unit	0.001	0.001	Chabukdhara and Nema (2013

Table 3. The toxicity response to heavy metals/metalloids as the oral slope factor and oral reference dose

Heavy metals	Oral RfD (mg/kg/day)	Oral SF[a] $(mg/kg/day)^{-1}$
As	3.0×10^{-4}	1.5
Ba	2.0×10^{-1}	nd.
Cr	3.0×10^{-3}	0.5
Cu	4.0×10^{-2}	nd.
Fe	7.0×10^{-1}	nd.
Mn	1.4×10^{-1}	nd.
Mo	5.0×10^{-3}	nd.
Ni	2.0×10^{-2}	nd.
Pb	3.5×10^{-3}	8.5×10^{-3}
Sr	6.0×10^{-1}	nd.
Ti	3.0	nd.
Zn	3.0×10^{-1}	nd.
Zr	4.0×10^{-4}	nd.

nd. not determined
[a](USEPA, 2011)

Table 4. Descriptive data of heavy metals/metalloids concentration (mg/kg) in filling station dust collected from the Kumasi Metropolis and reference value (Taylor and McLennan 1995)

	Min	Max	Mean	Std. Deviation	Coefficient of variation (%)	Skewness	Kurtosis	Reference value
As	2.58	13.28	6.20	2.72	43.87	1.03	1.22	1.5
Ba	33.72	142.63	92.26	38.44	41.66	-0.09	-1.10	550
Cr	21.81	150.04	70.41	36.78	52.24	0.51	-0.25	35
Cu	14.55	248.58	50.18	50.05	99.74	3.23	12.31	25
Fe	89.85	681.87	466.22	31.46	6.75	0.63	0.68	3.5
Mn	56.08	327.06	163.68	65.72	40.15	0.51	0.14	1.5
Mo	2.77	8.93	4.63	1.67	36.07	1.16	1.28	1.5
Ni	21.30	59.91	44.05	16.29	36.98	-1.18	2.18	20
Pb	6.48	129.87	46.93	32.23	68.68	1.30	1.17	20
Sr	9.42	183.94	106.69	37.06	34.74	-1.07	2.82	350
Ti	347.00	585.53	327.51	24.02	7.33	0.33	-0.24	0.3
Zn	12.95	520.68	280.32	119.81	42.74	-0.43	0.78	71
Zr	92.02	250.38	182.05	40.37	22.18	-0.26	-0.58	190

Table 5. Comparison of heavy metals/metalloids concentrations (mg/kg) observed in this study with those found in other heavy metal dust studies

	Ti	Ba	Cr	Mn	Fe	Ni	Cu	Zn	As	Pb	Sr	Zr	Reference
Hong Kong, China (Street dusts)	2370.00	253.00	124.00	594.00	14100.00	28.60	110.00	3840.00	66.80	120.00	121.00	378.00	(Yeung et al. 2003)
Hong kong, China (65 urban street dusts)	–	–	–	–	–	–	173.00	1450.00	–	181.00	–	–	(Li et al. 2001)
London (11 street dust)	1465.00	–	112.00	379.00	22800.00	–	191.00	1176.00	27.00	2008.00	–	–	(Fergusson and Ryan 1984)
Christchurch, New Zealand (12 street dust)	2117.00	–	103.00	313.00	20900.00	–	90.80	716.00	14.50	1223.00	–	–	(Fergusson et al. 1986)
Oslo, Norway(16 street dust)	7452.00	526.00	–	833.00	51452.00	41.00	123.00	412.00	–	180.00	344.00	–	(de Miguel et al. 1997)
Madrid, Spain (16 street dust)	1100.00	–	61.00	362.00	19300.00	44.00	188.00	476.00	–	1927.00	–	–	(de Miguel et al. 1997)
Ottawa, Canada (50 street dust)	–	576.00	43.30	431.50	18948.00	15.20	65.84	112.50	1.30	39.05	459.00	–	(Rasmussen et al. 2001)
Xi'an, China(157 campus dust)	–	958.90	154.20	546.20	–	32.20	62.10	390.70	11.50	151.60	–	–	(Chen et al. 2014)
Xiandao (51 road	–	–	71.60	–	–	–	43.90	171.00	–	66.60	–	–	(Li et al. 2016)

dust)													
Xiandao (51 street dust)	–	–	–	765.19	–	30.74	–	–	24.47	–	–	–	(Li et al. 2015)
Xiandao, China (51 road dust)	–	–	80.66	–	–	–	43.90	66.58	–	241.92	–	–	(Huang et al. 2016)
Kavala, Greece (26 street dust)	–	–	196.00	–	–	57.50	123.90	271.60	16.70	300.90	–	–	(Christoforidis et al. 2009)
Hangzhou, China (25 urban dusts)	–	–	51.29	–	–	25.88	116.04	321.40	–	202.16	–	–	(Zhang and Hao 2009)
Shanghai, China (273 dusts)	–	–	159.30	–	–	83.98	196.80	733.8	–	294.90	–	–	(Shi et al. 2008)
Guangzhou, China (30 urban dusts)	–	–	78.80	–	–	23.00	176.00	586.00	–	240.00	–	–	(Duzgoren-Aydin et al. 2006)
Guwahati, India (25 street dusts)	–	–	0.01	0.3	10.8	0.01	0.01	0.03	–	0.01	–	-	(Hussain et al. 2015)
Urumqi, China (169 urban road dusts)	–	–	54.28	–	–	43.28	94.54	294.47	–	53.53	–	–	(Wei et al. 2009)
Isfahan, Iran (24 road dusts)	–	–	–	–	–	66.63	182.26	707.19	22.15	393.33	–	–	(Soltani et al. 2015)

Table 6. Pearson's correlation matrix for heavy metals/metalloids concentration

	Mo	Pb	As	Zn	Cu	Ni	Fe	Mn	Cr	Ti	Ba	Zr	Sr
Mo	1												
Pb	0.288	1											
As	0.011	0.028	1										
Zn	0.238	0.803[a]	-0.018	1									
Cu	0.357	0.550[a]	-0.027	0.575[a]	1								
Ni	0.201	-0.937	-0.254	-0.973[b]	-0.722	1							
Fe	-0.049	0.488[b]	-0.121	0.655[a]	0.457[b]	0.056	1						
Mn	0.067	0.347	0.134	0.615[a]	0.426[b]	-0.283	0.889[a]	1					
Cr	-0.118	0.374	-0.125	0.455[b]	0.289	0.215	0.940[a]	0.775[a]	1				
Ti	0.025	0.127	0.007	-0.098	0.268	0.037	0.237	0.639[a]	0.714[a]	1			
Ba	0.438	0.075	0.450	0.202	0.460	0.074	0.219	0.612	-0.002	0.204	1		
Zr	-0.296	-0.174	0.193	-0.193	-0.098	0.523	0.320	0.710[a]	0.731[a]	0.723[a]	-0.210	1	
Sr	-0.170	0.032	0.357	0.396	-0.344	0.032	0.338	0.117	-0.030	-0.424[b]	0.265	-0.264	1

[a]Correlation is significant at the 0.01 level.
[b]Correlation is significant at the 0.05 level.

Table 7. Varimax factor loading for heavy metals/metalloids in the filling station dust

	Component		
	PC1	PC2	PC3
Mo	**0.529**	-0.174	**0.830**
Pb	-0.153	**0.987**	-0.058
As	**0.945**	0.129	0.301
Zn	0.037	**0.999**	0.017
Cu	-0.224	**0.862**	0.455
Ni	-0.187	-0.969	0.159
Fe	-0.906	0.049	-0.421
Mn	**0.534**	0.317	0.784
Cr	-0.962	-0.079	-0.262
Ti	**0.946**	-0.177	0.272
Ba	**0.524**	-0.039	**0.851**
Zr	**0.734**	-0.679	0.016
Sr	**0.928**	-0.158	0.337
Eigen value	5.883	4.329	2.788
% of Variance	45.252	33.300	19.248
Cumulative %	45.252	78.552	97.800

Bold values represent positive loadings ≥ 0.5

Table 8. Potential ecological risks of heavy metals/metalloids and their ecological index in the filling station dust

Heavy metal	Minimum	Maximum	Mean
Pb	1.62	32.47	11.73
As	17.20	88.53	41.30
Zn	0.18	7.33	3.95
Cu	2.91	18.19	8.22
Ni	5.33	14.98	11.01
Mn	0.09	0.39	0.27
Cr	1.25	8.57	4.02
Ba	0.06	0.26	0.17
PER	1.34	784.73	187.72

Table 9. Exposure duration (mg/kg-day) for filling station dust in Kumasi Metropolis.

	ADD_{ing}		ADD_{inh}		ADD_{dermal}	
	Children	Adult	Children	Adult	Children	Adult
As	7.93E-05	1.06E-05	2.22E-09	1.00E-09	1.27E-07	3.24E-07
Ba	1.18E-03	1.58E-04	3.31E-08	1.49E-08	1.89E-06	4.82E-06
Cr	9.00E-04	1.21E-04	2.53E-08	1.14E-08	1.44E-06	3.68E-06
Cu	6.42E-04	8.61E-05	1.79E-08	8.10E-09	1.03E-06	2.62E-06
Fe	5.96E-03	7.99E-04	1.67E-07	7.53E-08	9.54E-06	2.44E-05
Mn	2.09E-03	2.81E-04	5.87E-08	2.64E-08	3.35E-06	8.55E-06
Mo	5.92E-05	7.95E-06	1.66E-09	7.48E-10	9.47E-08	2.42E-07
Ni	5.63E-04	7.56E-05	1.58E-08	7.11E-09	9.01E-07	2.30E-06
Pb	6.00E-04	8.05E-05	1.68E-08	7.58E-09	9.60E-07	2.45E-06
Sr	1.36E-03	1.83E-04	3.83E-08	1.72E-08	2.18E-06	5.57E-06
Ti	4.19E-03	5.62E-04	1.17E-07	5.29E-08	6.70E-06	1.71E-05
Zn	3.58E-03	4.81E-04	1.01E-07	4.53E-08	5.73E-06	1.46E-05
Zr	2.33E-03	3.12E-04	6.53E-08	2.94E-08	3.72E-06	9.51E-06

Table 10. Non-carcinogenic risk, overall toxic risk and carcinogenic risk (ADD_{life} ×SF).

	HQ_{ing}		HQ_{inh}		HQ_{dermal}		ADD_{life} ×SF	
	Children	Adult	Children	Adult	Children	Adult	Children	Adult
As	0.264	0.035	7.41E-06	3.34E-06	3.81E-11	9.72E-11	8.74E-08	1.17E-08
Ba	0.006	0.001	1.65E-07	7.45E-08	3.77E-07	9.64E-07	–	–
Cr	0.300	0.040	8.42E-06	3.79E-06	4.32E-09	1.10E-08	3.31E-07	4.44E-08
Cu	0.016	0.002	4.50E-07	2.03E-07	4.11E-08	1.05E-07	–	–
Fe	0.009	0.001	2.39E-07	1.08E-07	6.68E-06	1.70E-05	–	–
Mn	0.015	0.002	4.19E-07	1.89E-07	4.69E-07	1.19E-06	–	–
Mo	0.012	0.002	3.32E-07	1.49E-07	4.74E-10	1.21E-09	–	–
Ni	0.028	0.004	7.89E-07	3.56E-07	1.80E-08	4.60E-08	–	–
Pb	0.171	0.023	4.81E-06	2.17E-06	3.36E-09	8.57E-09	3.75E-09	5.03E-10
Sr	0.002	0.000	6.38E-08	2.87E-08	1.31E-06	3.34E-06	–	–
Ti	0.001	0.000	3.92E-08	1.76E-08	2.01E-05	5.13E-05	–	–
Zn	0.012	0.002	3.35E-06	1.51E-06	1.72E-06	4.39E-06	–	–
Zr	5.819	0.781	1.63E-08	7.35E-05	1.49E-09	3.80E-09	–	–
HI	6.656	0.893	1.89E-08	8.54E-05	3.07E-05	7.84E-05	–	–

Public Interest Statement

The study looks at the possibility of dust blown from a fuel station causing harm to the health of humans. The likely groups of people to this potential harm are fuel station workers, patrons and residents within the vicinity of these stations. The presence of toxic chemicals in spilled fuel, oils, lubricants and exhaust emissions from vehicles that plough the station have the tendency to pollute the dust in and around these stations. This study gives an overview of the potential health implications of fuel stations on the general public if they come into contact with polluted dust.

About the Authors

The group is involved on the determination of levels and distribution of heavy metals in the environment. The current study on fuel stations forms part of the country wide study to determine the effects of human activities on heavy metals and the potential health associated with their exposure.