

Chemical characteristics and health risk assessment of potential toxic elements in atmospheric PM₁₀ around Ashaka cement factory, Gombe, Nigeria

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Keywords: *atmosphere, cement, carcinogenic risk, non-carcinogenic risk, PM₁₀*

The study determined the ambient mass concentrations, chemical composition and health risks associated with PM₁₀ around Ashaka cement factory, Gombe, Nigeria. The samples were collected for the period (2019-2020). A total of 60 PM₁₀ samples were collected and analyzed for seventeen elements using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES). The data collected were analyzed for descriptive and inferential statistics. The health risk was analyzed for hazard quotient (HQ), hazard index (HI) and cancer risk (CR). The average annual PM₁₀ mass concentrations were found to be higher than the annual limit value for air quality standards (40 µgm⁻³). The HI estimated were >1 for children, while CR values of Cd, Ni, As and Cr for children and Cr for adults were higher than the acceptable value 10⁻⁶, indicating children are more probable to develop cancer than adults.

Introduction

Air pollution is the discharge of substances that are capable of infringing the quality of ambient air [1-3]. Air pollution conveys atmospheric particulate matter (PM) incessantly [4,5], which include fine (PM_{2.5}) and coarse (PM_{2.5-10}) particles that are found to be widely associated with health problems [6]. PM_{2.5} and PM₁₀ are emitted from a variety of sources, including combustion processes, cooking, vehicular traffic, industrial emissions, etc [6]. Exposure to fine particulate matter has been reported to have more strong relationships with adverse health outcomes such as respiratory and cardiovascular disease [7].

Environmental pollution with particulate matter (PM) and potential toxic elements (PTEs) is gaining global attention due to their increase caused by rapid urbanization and industrialization in recent years [8-10]. The contamination of the environment with PTEs and PM affects human health, quality of life, and the natural functioning of an ecosystem [11-13]. PTEs are among the most relevant substances emitted during the process of industrial production and are of significant environmental concern due to their wide sources, toxicity, accumulative behaviours and non-biodegradable properties [14,15]. The occurrence of these elements in the environment could be from

natural sources or human activities such as mining, coal combustion, fuel combustion, biomass combustion, cement production, etc [10].

Cement production is one of the major industrial sources of PMs and PTEs, especially zinc, lead, nickel, cadmium, mercury, arsenic and copper. These PTEs are usually generated from combustions of fossil fuels and the processing of the raw materials [16]. Typical raw cement is made up Pb (20 mgkg^{-1}), Cu (21 mgkg^{-1}), Cr (25 mgkg^{-1}) and Zn (53 mgkg^{-1}) [17]. The deposition of the potential toxic elements occurred at different distances in the vicinity of cement factories and is influenced by particle size, wind velocity and stack fumes [17], and are capable of deteriorating air quality. PTEs are an important part of PM since they can have both carcinogenic and non-carcinogenic effects [18].

PM is a global issue with the world's population of about 91% living in regions where air pollution exceeded the World Health Organization (WHO) guidance [19]. According to the WHO, air pollution is responsible for the death of about 3.7 million people in 2012, from heart disease and stroke (29%), lung cancer (16%), chronic-obstructive pulmonary related diseases (11%) and respiratory infections (13%) [5,20]. Risk assessment models are used to estimate the health risk of elements including non-carcinogenic risk and carcinogenic risk [8, 21]. Non-carcinogenic risk was reported to be associated with systemic toxicity (e.g., liver,

kidney, or generalized toxicity), neurotoxicity (brain pathology), reproductive toxicity (e.g., fertility), etc [22], while carcinogenic risk is associated to lung cancer, gastric carcinoma, brain cancer, etc [23]. Thus, there is a need to investigate the levels of PM in the air around cement factories.

Ashaka cement factory is about 9 km Bajoga, Funakaye local government area of Gombe State, northeastern Nigeria. It was established in 1967, with an installed capacity of 500,000 MT [24] to meet the needs of construction works in the Northeastern part of Nigeria [25]. About 90% of the energy requirements of the factory is realised from coal combustion [26]. This study was therefore carried out to determine the mass concentration, chemical characterization of ambient PM_{10} , and health risk associated with potential toxic elements in the samples with a view to provide information on the extent of contamination of the elements in the PM_{10} and health risk to the residents.

Experimental part

Material and Methods

Study area

The study area is Ashaka Cement Factory, located in Ashaka town, Bajoga, Funakaye Local Government Area, Gombe State, Northeast Nigeria. Ashaka Cement factory is 9 km north of Bajoga. The factory lies between latitudes $10^{\circ}55'30''\text{N}$ and $11^{\circ}56'30''\text{N}$, and longitudes $11^{\circ}28'30''\text{E}$ and $11^{\circ}29'30''\text{E}$ (**Figure**

1). The climate of the study area is tropical with dry/harmattan season (November-April) and wet season (May-October) with an average temperature of 25 °C. The maximum annual rainfall of 800 to 900 mm during August and a minimum of 250 to 350 mm during the early and the later period of the wet season, with a relative humidity range from 60 to 80%.

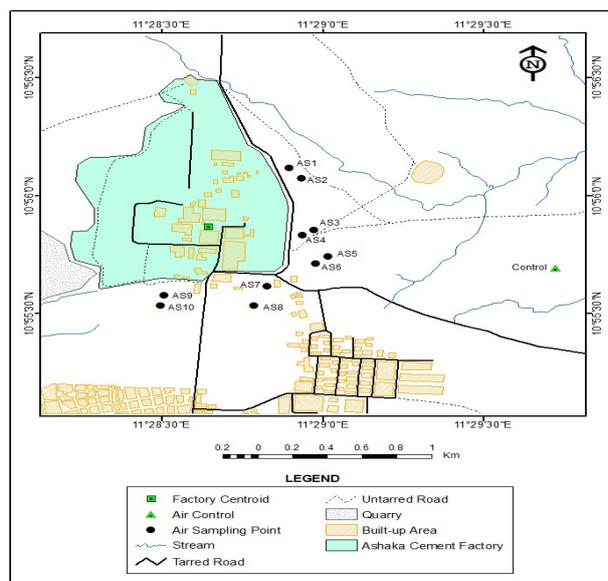


Figure 1. Map of the study area showing the sampling points

Sampling and analysis

Sampling was carried out from June 2019 to May 2020, around the Ashaka cement factory. The air samples were collected using a high-volume air sampler (HiVol 15000) capable of measuring particles under 10 μm in size. Samples were collected in both dry and wet seasons at each sampling location. Filter masses were measured on a microbalance before and after samples collection. The calibration of the sampler was carried out as reported elsewhere [27]. Each retrieved filter was cut into fragments, put into Teflon cups and digested in 20 mL

freshly prepared aqua-regia ($\text{HNO}_3\text{:HCl}$; 1:3). The resulting solutions were analyzed to determine the contents of the potential toxic elements using inductively coupled optical emission spectrometry (ICP-OES Agilent 720-ES).

Quality control

The ICP-OES (Agilent 720-ES) was calibrated with Accu standard (QCSTD-27) multi-element standard solution. The samples and blank filter were digested with aqua-regia, and the volume was made to 50 mL with distilled water for the quantitative determination of the concentrations in unknown samples, which were estimated by standard internal method. In addition, the mean recovery percentages of the metals ranged from 86.5% to 121%. The limits of detection for As, Ba, Cd, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb, Sc, Se, Sr, Ti, V and Zn were 0.003, 0.004, 0.0001, 0.0004, 0.0008, 0.01, 0.0006, 0.0001, 0.005, 0.001, 0.0001, 0.0001, 0.003, 0.003, 0.003, 0.009 and 0.0003 $\mu\text{g}\text{m}^{-3}$, respectively.

Data analysis

The data obtained were subjected to descriptive analysis (mean and standard deviation) and inferential (Pearson correlation, t-test and principal component analysis) statistics using SPSS software version 25. Pearson correlation and principal component analysis were adopted to determine the origins of the collected particles, while t-test was used to

determine seasonal significance at $p < 0.05$ [28, 29].

Health risk assessment

The exposure of PTEs determined in PM_{10} was assessed for human health effect using the model designed by the United States Environmental Protection Agency (USEPA) and other relative studies [8,21,30]. The three potential exposure pathways of PTEs in PMs are inhalation, ingestion and dermal contact [8,21, 30, 31-33], according to equations 1-6.

$$ADD_{inh} = \frac{C \times InhR \times EF \times ED}{PEP \times BW \times AT} \quad (1)$$

$$ADD_{derm} = \frac{C \times SA \times AF \times ABS \times EF \times ED \times CF}{BW \times AT} \quad (2)$$

$$ADD_{ing} = \frac{C \times IngR \times EF \times ED \times CF}{BW \times AT} \quad (3)$$

Non-carcinogenic risk assessment

The hazard quotient (HQ) was used to estimate the non-carcinogenic effect of PTEs in PM and was evaluated using the expression below.

$$HQ = \frac{ADD}{RfD} \quad (4)$$

Hazard index (HI) is the sum of multiple-route HQ (US EPA) [34]. A HI value >1 implies that non-carcinogenic effects may occur, the higher the value of HI, the higher the likelihood of non-carcinogenic effects (USEPA) [30].

$$HI = \sum_n^i HQ_i \quad (5)$$

Carcinogenic risk assessment

Carcinogens risk was also estimated using equation 6

$$CR = ADD_{inh} \times CSF_{inh} \quad (6)$$

Where; ADD = average daily exposure dose of elements in PM ($\mu\text{gm}^{-3}\text{day}^{-1}$), RfD = reference dose, and CSF = cancer slope factor ($\mu\text{gm}^{-3}\text{day}^{-1}$) [20, 33, 35, 36, 37]. The values of RfD and CSF are presented in **Table 1** (All Tables are presented on Supplementary Information).

The CR values below 1×10^{-6} are considered insignificant, while the CR values above 1×10^{-4} indicate harmful effects to human beings [38]. The parameters and input assumptions for exposure assessment are presented in **Table 2**.

Results and discussion

PM₁₀ mass concentrations

The seasonal variations and average annual mass concentrations of PM_{10} during the study period are presented in **Figure 2**. The average PM_{10} concentrations were ($802 \pm 23.50 \mu\text{gm}^{-3}$) in the dry season and ($587 \pm 13.65 \mu\text{gm}^{-3}$) in the wet season with the annual concentration of $1389 \mu\text{gm}^{-3}$. The PM_{10} mass concentrations in the dry season are significantly higher than those in the wet season; this is mainly related to harmattan period in the dry season, which leads to increasing the levels of dust concentration in the atmosphere between December to February. The annual average mass concentration of $115.75 \mu\text{gm}^{-3}$ obtained in this study exceeded the PM_{10} average annual concentration for air quality standards (20 and $40 \mu\text{gm}^{-3}$) by 478.75% and 189.38% [41,42], respectively. However, the annual mass concentrations of PM_{10} in this study was lower than the $1552 \mu\text{gm}^{-3}$ reported around a

cement factory in Hattar, Huripur, Pakistan [43] and the 3200 $\mu\text{g}\cdot\text{m}^{-3}$ around a cement plant in Changping, China [9], but higher than the annual concentrations of 227.60 $\mu\text{g}\cdot\text{m}^{-3}$ in Accra metropolis of Ghana [44] and 502.30 $\mu\text{g}\cdot\text{m}^{-3}$ in Ibadan, Nigeria [27].

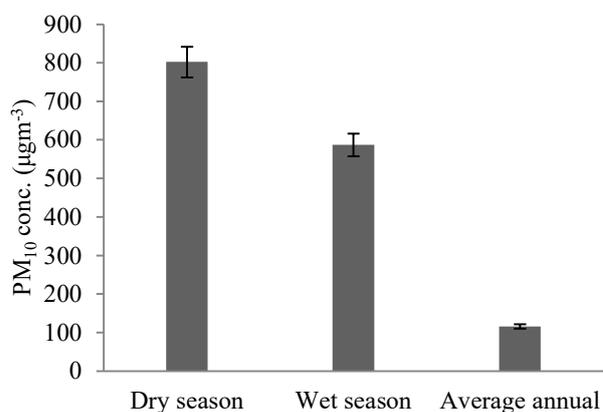


Figure 2. PM₁₀ seasonal and average annual mass concentrations during study period

Seasonal variation in concentrations of potential toxic elements in PM₁₀

Average concentrations of PTEs in PM₁₀ samples are presented (Tables 9 and 10) in dry and wet seasons. The mean concentrations of the 17 potential toxic elements in the different seasons are presented in Table 3. The results showed a trend of the variation in the concentrations of the elements. The concentrations of elements in the dry season are significantly higher than those in the wet season, except Mn. That may be due to the impact of production activities and dust episode of harmattan in the dry season. The distributions of elements in PM₁₀ during dry season were in the descending order Fe > Zn > Mn > Ti > Sr > Cu > Ba > Pb > Cr > V > Ni > As > Sb > Sc > Cd > Se

> Mo, while the concentrations in wet season followed the order Fe > Zn > Mn > Ti > Sr > Ba > Cr > V > Pb > Cu > Ni > As > Sb > Cd > Mo > Sc > Se. The results revealed a significant difference ($p > 0.05$) among the elemental concentrations of As, Cr, Pb and Sc during the dry and wet seasons. The seasonal variations of elements in atmospheric PMs might be influenced by local emission sources, wind direction and meteorological conditions [45,46].

The highest concentrations ($\mu\text{g}\cdot\text{m}^{-3}$) of As (0.02), Cr (0.073), Ni (0.10), Pb (0.16), Cd (0.19) and Mn (0.63) obtained were above the maximum tolerance limits of 0.006, 0.00025, 0.02, 0.0005, 0.005 and 0.15 $\mu\text{g}\cdot\text{m}^{-3}$, respectively [41,47]. The high concentrations of these elements in PM₁₀ obtained around the vicinity of the cement factory suggest that the cement production is the major source of these elements in study area (combustion of coal and processing the raw materials). The anthropogenic activities are the key sources of Cr, Mn, Pb and Zn [48, 49]. Calcium is the major fingerprint for cement; unfortunately, this element has not been measured in the present study. Among the elements present in PM₁₀, Cu, Ni, Fe, Cr and Zn have been the main toxic elements based on their ability to support electron exchange [50-52] and create reactive oxygen species [53,54]. Elements such as Cd, Ni, Cr and Pb were established to be in significant amounts in the PM₁₀ and therefore, they may pose a severe risk to human health [52,55].

Table 4 presents the elemental constituents of PM₁₀ reported elsewhere in the world, e.g., Jeddah, Saudi Arabia [49], Beijing, China [56], Agra, India [57], Changping, China [9], Ibadan, Nigeria [27], Changhua, Taiwan [58], Accra, Ghana [44], and Lahore, Pakistan [59]. In comparison, the mean concentrations of Mo, Pb, Cu, Mn, Se and Ba in Beijing, China [56], were above the values obtained in this study, while Cu, Fe and Zn in this study were higher than the concentrations reported in Agra, India [57] and Cr, Ni, Cu, Sr, Zn, As and Fe in Jeddah, Saudi Arabia [49]. The high Ba concentration observed in the study may be due to its nature as witherite and baryte accumulate in cement production [60, 61].

Pearson correlation analysis

Pearson correlation analysis for element concentration in the PM₁₀ samples was presented in **Table 5**. The correlation analysis showed that the majority of element pairs have strong positive correlations, $r > 0.6$ ($p < 0.01$) for Fe/As, Mn/Ba, Ni/Ba, Pb/Cd, Pb/Cu, Sb/Cd, Sb/Cu, Sb/Pb, Sr/Cr, Sr/Mn, Sr/Ni, Ti/Ba, Ti/Mn, Ti/Ni, Ti/Sr, V/Ni and Zn/Pb, and $r > 0.4$ ($p < 0.05$) for Cr/As, Fe/Cr, Ni/As, Sr/As, Sr/Fe, Ti/Cr, Ti/Mo, V/Ba, V/Sr, V/Ti, Zn/As and Zn/Cd. However, Se/Mo had strong negative correlation ($r = -0.6$; $p < 0.01$). Most element pairs demonstrated strong significance ($p < 0.05$ and $p < 0.01$), indicating their simultaneous release from the production activities. The anti-correlation of Se with Mo

might indicate different emission sources aside from cement production activities.

Principal component analysis

The results of the principal component analysis (PCA) are presented in **Table 6**. Four factors were identified that explained a cumulative variance of 77% of the total PM₁₀ data set. As, Ba, Mn, Ni, Pb, Sr, Ti and V were loaded in factor 1 with 26% variance. This factor could emanate from anthropogenic sources, such as production and combustion of fossil fuel. Factor 2 was characterized by Cr, Fe, Ni, Sr and Zn with 23% of the variance identified, which could probably originate from emissions of machines and vehicles used in the vicinity of the factory. Factor 3 has high loadings for Cr, Cu, Sb and Zn, having a total variance 19% identified, while factor 4 was significant for only Mo with 9% variance identified. This component may indicate contributions from lithogenic and paedogenic origins.

Health risk assessment

Non-carcinogenic risk assessment of elements

The results of average daily exposure dose (ADD) are presented in **Table 11**. The results of hazard quotient (HQ), Hazard index (HI) and Cancer risk (CR) of the elements for both children and adults from three routes of exposure; (inhalation, ingestion and dermal contact) are presented in **Tables 7** and **8**. The results for HQ_{inh} were in the order Mn > Ni > As > Cr > Ba > Pb > Sb > Cu > Cd > Se > Zn for

children, and Mn > Ni > As > Cr > Ba > Se > Pb > Sb > Cu > Cd > Zn for adults in dry season, while in wet season it was Zn > Mn > Ni > As > Cr > Ba > Cu > Cd > Sb > Pb > Se for children, and Zn > Mn > Ni > Cr > Ba > Cd > Cu > As > Sb > Pb > Se for adults. HQ_{ing} was in the order As > Pb > Cr > Sb > Cd > Sr > Cu > Ni > Se > Zn > Mn > Ba > Mo for children and Pb > Cr > Sb > Cd > Ni > Cu > Se > Zn > Mn > As > Ba > Mo > Sr for adults in dry season, while in wet season it was As > Cr > Sb > Pb > Se > Mn > Cd > Ni > Zn > Ba > Cu > Mo > Sr for children, and Cr > Sb > Pb > Se > Mn > Cd > Ni > Zn > Ba > Cu > As > Mo > Sr for adults. HQ_{derm} for both children and adults were in the order Mn > Cr > Zn > Pb > Ni > Cd > Cu in dry and wet seasons. The calculated HQ for the three main exposure routes showed that ingestion of particulate matter pose greater health risk effect followed by inhalation and dermal contact pathways. This order is in conformity with previously reported studies [56, 62,63]. The values of the total hazard index (HI) revealed that children and adults were less than one for inhalation and dermal contact, suggesting no non-carcinogenic adverse health effects. However, the HI of ingestion was greater than 1 for children, but less than 1 for adults, indicating deleterious effects for children.

Carcinogenic risk assessment of elements

The carcinogenic risks (CR) of the elements are presented in **Table 8**. Cancer risk by inhalation are the highest. The values obtained were Cr (1.39×10^{-4}), Ni (6.70×10^{-6}) for

children, and Cr (1.50×10^{-5}) for adults in the dry season, while the values were Cr (6.43×10^{-5}) Cd (3.04×10^{-6}) and Ni (2.61×10^{-6}) for children and Cr (7.64×10^{-6}) for adults in the wet season. The CR values of Cr and Ni for children, and Cr for adults in the dry season were above 10^{-6} , while CR values of Cd, Cr and Ni for children and Cr adults were above 10^{-6} , which exceeded the threshold and suggested a possible cancer development by residents. The ingestion exposure pathways present the highest CR values for As (3.07×10^{-6}) and As (1.42×10^{-6}) for children in dry and wet seasons respectively. As have been reported to be mainly produced during coal burning [64]. Dermal contact revealed the highest CR value for Ni (1.88×10^{-6}) for children in the dry season. Results revealed that children are more vulnerable to health risks than adults. The inhalation of PMs in the study may pose a more carcinogenic risk than ingestion and dermal contact pathways. The cancer risk values obtained in the present study are similar to the values reported in previous studies [8,65-67].

Conclusion

This study assessed the concentrations of potential toxic elements and health risk in PM₁₀ around the Ashaka factory in Gombe, Nigeria. The concentrations of As, Cd, Cr, Mn, Ni and Pb were above the tolerance limits. The multivariate statistical analysis of PCA showed four factors that are mostly anthropogenic in nature, establishing the contribution of cement production activities. The health risk associated

elements in PM₁₀ samples indicated that the exposure pathways for HQs were in the distribution pattern of ingestion > inhalation > dermal, while the HI for children was greater than 1, indicating adverse health effects. The cancer risk value of elements to children was higher than the acceptable value 10⁻⁶, thereby suggesting probable cancer development. The health risk assessment revealed that children were more vulnerable than adults.

Acknowledgment

The authors are grateful to Professor Mohd Talib Latif, Department of Earth Sciences and Environment, Universiti Kebangsaan Malaysia, for his contribution.

Disclosure statement

The authors declare that there exist no conflict of interest.

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