• **Original Article**

Spatial assessment of soil contamination by heavy metals from informal electronic waste recycling in Agbogbloshie, Ghana

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Objectives This study examined the spatial distribution and the extent of soil contamination by heavy metals resulting from primitive, unconventional informal electronic waste recycling in the Agbogbloshie e-waste processing site (AEPS) in Ghana.

Methods A total of 132 samples were collected at 100 m intervals, with a handheld global position system used in taking the location data of the soil sample points. Observing all procedural and quality assurance measures, the samples were analyzed for barium (Ba), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb), and zinc (Zn), using X-ray fluorescence. Using environmental risk indices of contamination factor and degree of contamination (C_{deg}), we analyzed the individual contribution of each heavy metal contamination and the overall C_{deg}. We further used geostatistical techniques of spatial autocorrelation and variability to examine spatial distribution and extent of heavy metal contamination.

Results Results from soil analysis showed that heavy metal concentrations were significantly higher than the Canadian Environmental Protection Agency and Dutch environmental standards. In an increasing order, $Pb > Cd > Hg > Cu > Zn > Cr > Co > Ba > Ni$ contributed significantly to the overall C_{deg}. Contamination was highest in the main working areas of burning and dismantling sites, indicating the influence of recycling activities. Geostatistical analysis also revealed that heavy metal contamination spreads beyond the main working areas to residential, recreational, farming, and commercial areas.

Conclusions Our results show that the studied heavy metals are ubiquitous within AEPS and the significantly high concentration of these metals reflect the contamination factor and C_{deg}, indicating soil contamination in AEPS with the nine heavy metals studied.

Keywords Electronic waste, Geostatistical techniques, Heavy metals, Contamination factor, Degree of contamination

Introduction

Changing electrical electronic equipment consumer's lifestyle and the ever growing demand for newer and more efficient technologies are contributing to the increasing rate of obsolesce of electrical materials [1,2]. An estimated 41 million tons of electrical electronic equipment waste, also known as electronic waste (e-waste), were generated globally in 2014 [3]. Fifty to eighty percent of electrical electronic equipment meant for disposal and recycling in developed countries are either legally or illegally shipped to developing countries under the disguise of slightly used or charity materials [4]. Some of these equipment end up in informal recycling sites where primitive recycling methods, such as the open burning of cables, fridge, and television casings, are performed, exposing the environment and entire communities to pollution [5]. Contamination of the envi-

ronment due to uncontrolled e-waste recycling has been reported in Ghana and particularly Ghana's largest e-waste recycling area Agbogbloshie [6-10]. The contamination of the environment is driven by increased quantities of electrical e-waste in Ghana, which have created avenues and opportunities for individuals to make a living by using unconventional, primitive, and crude recycling methods to recover precious and valuable metals from e-waste. The uncontrolled recycling activities of individuals, especially informal recyclers, and the absence of a well-structured management policy or strategy in Ghana have resulted in pollution of the environment through the release of heavy metals from some of the unconventional recycling methods. This present study seeks to determine qualitatively and quantitatively heavy metal contamination from e-waste processing in Agbogbloshie, Ghana, and further through geostatistical techniques and spatial analysis to examine the spatial extent of heavy metal contamination from e-waste processing in Agbogbloshie. The results of this study can be useful in decision making processes for selecting appropriate remediation measures for contaminated environments and protection of affected communities from potential health hazards.

Materials and Methods

Study Area, Sample Collection, and Data Preparation

A total of 132 samples were collected from Agbogbloshie e-waste processing site (AEPS), using a 100 m interval grid based sampling procedure. AEPS is located close to the central business district of Ghana's national capital (Figure S1) and consists of a number of clusters such as burning, dismantling, residential, recreational, commercial, worship, and school sites. Data collection was conducted while observing all standard procedures to avoid cross contamination. The collected samples were air dried at room temperature, sieved through a 100 μm mesh, and pressed into pellets with a diameter of 2.5 cm using a 10-ton hydraulic press. Heavy metal analysis was performed using an X-ray fluorescence (XRF) spectrometer, (at the Department of Geological Survey, Accra, Ghana), at a maximum power of 3000 W (60 Kv and 50 mA). The pelleted samples were placed on a disk which was put on the excitation source of the XRF for a 10-minute irradiation, using a silicon lithium Si (Li) detector with a resolution of 16 V with manganese and potassium alpha peaks throughout the procedure. To validate the procedure and ensure quality, International Atomic Energy Agency (IAEA) standard reference IAEA soil seven was irradiated five times and average values were compared with recommended values before analysis of prepared samples.

Statistical and Geostatistical Analysis

The study adopted the steps as shown in Figure S2 for the statistical and geostatistical analysis of the heavy metals barium (Ba), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb), and zinc (Zn) identified in the soil samples. The selection of heavy metals used in our study was based on the criteria defined by the Dutch soil quality and guidance standards for contaminated lands and environmental remediation [11].

Data Preparation and Exploratory Analysis

Heavy metal concentrations values together with the coordinates were listed in Microsoft Excel 2013 (Microsoft Co., Redmond, WA, USA) and exploratory analysis of descriptive variables, normality, and correlation between heavy metals were performed using R.3.2.1 software.

Trend Analysis and Variogram Construction

Trend analysis was performed using ArcGIS 10.1 to identify global trends or patterns which have to be taken into account before applying geostatistical interpolation. Variogram models and the global Moran's index were used to examine spatial autocorrelation and spatial variability of heavy metals. Spatial variability of the data was also assessed for each heavy metal using semivariogram or entropy voronoi maps before prediction or kriging of the data [12]. Exponential, spherical, and K-Bessel models were chosen as they obtained the best fit in the assessment of spatial autocorrelation for the heavy metal variables. The sill, nugget, and range for each of the models (exponential, K-Bessel and spherical) were also explored to ascertain variability and spatial dependency of the heavy metal dataset.

Kriging and Cross Validation

The final step in the geostatistical data analysis was kriging. With data not indicating any direction and no local drift or trend in the data set, anisotropic kriging, universal kriging, and co-kriging procedures were avoided in the prediction. The ordinary kriging process was performed on the degree of heavy metal contamination.

Contamination Assessment

There are three categories of heavy metal assessment indices [13], which include (a) contamination indices [14] and metal enrichment index, (b) background enrichment indices [15]; index of geoaccumulation [16], contamination factor (CF) and degree of contamination (C_{deg}) [17], and (c) ecological risk indices [18]. For the purpose and objectives of this study, the CF and C_{deg} were chosen over the other indices for the following

reasons:

CF and C_{deg} indices overcome the requirement of using a suitable location for background concentration value by using continental crust average as specified in [17].

The C_{deg} complimented by the CF provides a comprehensive picture of a particular site by aggregating individual heavy metal toxicity as single contamination index while pollution, metal enrichment, enrichment indices, and index of geoaccumulation are only suitable for evaluating single elements.

The CF and the C_{deg} have been used over the years to assess or ascertain the extent of heavy metal contamination of soils by comparing the results for the contaminants with different reference or background levels [7,19–21]. The CF which evaluates environmental pollution by single substances is expressed as:

$$C_f^i = \frac{c_{0-1}^i}{c_n^i}$$

where C_f^i is the CF of the element of interest; c_{0-1}^i is the concentration of the element in the sample; $\overline{c_n^i}$ is the background concentration or the continental crustal average as was used by Muller [17]. The classification for contamination ranges from CF < 1 as low CF; 1 < CF ≤ 3 as moderate CF; 3 < CF ≤ 6 as considerable CF and CF > 6 very high CF. Complementing the CF is the C_{deg}, which is the sum of CFs and describes the contamination of the environment by all examined substances, further defining the quality of the environment. The C_{deg} is expressed as:

$$C_{deg} = \sum C_f^i$$

The C_{deg} is useful to identify hot spots within the sampling location. It is also categorized into four stages, such as $C_{deg} < 8$ implying a low, C_{deg} $8 \leq C_{deg} < 16$ indicating a moderate C_{deg}, $16 \leq C_{deg} < 32$ indicating a considerable C_{deg}, and $C_{deg} \geq 32$ indicating a very high C_{deg}.

Results

Descriptive Parameters of Heavy Metals Within Study Area

To evaluate the raw data set, we calculated the descriptive statistics of mean, median, standard deviation, skewness, and coefficient of variation (CV) (Table 1). The concentrations of the heavy metals Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb, and Zn under investigation were extended over several orders of magnitudes. Compared with the Dutch and the Canadian Environmental Quality Standards for Soils, the mean values of Ba, Cu, Pb, and Zn were above both the optimal and action values of the Dutch and Canadian Soil Quality Guidance Values (SQGV) while those of Cd, Co, Hg, and Ni had mean values below action required values of SQGV. The skewness values for Co, Cd, Cr, and Hg were relatively low, however, those for Ba, Pb, Zn, Cu, and Ni were high, indicating non-normality of the data set for these heavy metals. Further, CV values of Ni, Pb, Cu, and Zn were 6.05, 1.82, 1.81, and 1.40, respectively, and higher than those of Ba, Co, Cr, Cd, and Hg, suggesting that Ni, Pb, Cu, and Zn had greater variation among soils within the study area.

Table 1 also shows the mean CF for each of the heavy metals, indicating toxicity contribution. CFs of Ba, Co, Cr and Ni were within the moderate CFs while that of Cd, Cu, Hg, Pb and Zn were approximately 5, 4, 5, 11 and 3 times respectively within the high CF classification .

Correlation Between Heavy Metals

Table 2 shows the correlations between heavy metals. Our results indicate significant correlation at $p < 0.05$ of Ba with all heavy metals apart from Ni and Pb. Cd also showed significant correlation at $p < 0.05$ with all other heavy metals apart from Cr and Ni, while Co showed significant correlation at $p < 0.05$ with Cr, Cu, Hg, and Zn, but weakly correlated with Ni and Pb. Table 2 shows

Table 1. Summary statistics of heavy metal concentrations in soil

| Heavy metal | Mean | Median | Minimum | Maximum | SD | CV | Skewness | Mean CF | Dutch SQGV | | Canadian SQGV |
									Optimal	Action	
Ba	627.7	574.0	120.6	2635	339.1	0.54	1.88	1.46	200	625	500
Cd	6.6	3.9	0.4	26.5	6.8	1.04	1.35	32.80	1	12	10
Co	46.6	35.3	8.8	153.7	29.2	0.63	1.12	1.86	20	240	-
Cr	296.6	197.3	21.1	1332	273.3	0.92	1.57	2.94	100	380	64
Cu	1387.9	290.7	9.4	18285	2507.2	1.81	3.70	23.11	36	190	63
Hg	2.7	1.4	0.4	13.4	2.9	1.07	2.03	32.61	0.3	10	7
Ni	61.7	13.9	0.6	4003	373.4	6.05	10.59	0.37	35	210	50
Pb	953.2	290.9	14.2	10280	1734.8	1.82	3.29	70.25	85	530	140
Zn	1371.1	576.6	41.9	12907.5	1923.8	1.40	3.07	17.85	140	720	200

Concentration values are measured in ppm.
SD, standard deviation; CV, coefficient of variation; CF, contamination factor; SQGV, soil quality and guidance values; Ba, barium; Cd, cadmium; Co, cobalt; Cr, chromium; Cu, copper; Hg, mercury; Ni, nickel; Pb, lead; Zn, zinc.

Table 2. Correlation between heavy metal concentrations in soil at AEPS

Heavy metal	Ba	Cd	Co	Cr	Cu	Hg	Ni	Pb	Zn
Ba	1								
Cd	0.712*	1							
Co	0.748*	0.599*	1						
Cr	0.604*	0.356	0.549*	1					
Cu	0.449*	0.761*	0.435*	0.042	1				
Hg	0.602*	0.804*	0.563*	0.190	0.855*	1			
Ni	0.115	0.050	0.328	0.001	0.181	0.472*	1		
Pb	0.341	0.724*	0.266	-0.139	0.903*	0.874*	0.250	1	
Zn	0.549*	0.767*	0.598*	0.252	0.856*	0.778*	0.140	0.775*	1

AEPS, Agbogbloshie electronic waste processing site; Ba, barium; Cd, cadmium; Co, cobalt; Cr, chromium; Cu, copper; Hg, mercury; Ni, nickel; Pb, lead; Zn, zinc.
*$p<0.05$.

Table 3. Summary statistics and geostatistical parameters for degree of contamination

Contamination assessment								
Summary statistics	Mean	Median	SD	Minimum	Maximum	CV	Skewness	Kurtosis
	158.68	63.21	222.42	3.36	968.01	1.41	2.13	4.14
Spatial autocorrelation	MI	EV	Variance	Z-score	p-value	DT		
	0.175	-0.008	0.006	2.362	0.02	500		
Geostatistical characteristics	Model	Nugget (Co)	Sill (C1)	Co/C1	Range			
	Spherical	0.219	1.022	0.214	851.49			
	Exponential	0.288	0.911	0.317	1057.40			
	Gaussian	0.350	0.898	0.390	712.01			
	K-Bessel	0.344	0.916	0.375	753.10			

SD, standard deviation; CV, coefficient of variation; MI, Moran index; EV, error variance; DT, distance threshold.

that Ni only correlated significantly with Hg and weakly with all other heavy metals. The weak correlation of Cr and Ni with other heavy metals indicates sources of Cr and Ni independent of the other heavy metals, while the closely significant correlation between Ba, Cd, Co, Cu, Pb, and Zn indicates a source of similar origin or activity (such as Pb in printed circuit boards, Ba, Cd, Pb, Zn in cathode ray tube and batteries, Cu in cables and transformer coils, which are the main devices processed at AEPS).

Contamination Assessment

To describe the extent of contamination and to examine the toxicity of the metals under investigation, we calculated the C_{deg} as the sum of all CFs for each element present at a site or location. The mean C_{deg} was 158.68, indicating a very high contamination. The calculation of the C_{deg} also revealed a 1.41 CV, indicating high variability in the C_{deg}, and a skewness value of 2.13, showing a non-normal distribution and positively skewed data of the C_{deg} (Table 3).

Geostatistical and Spatial Distribution of Degree of Contamination

Geostatistical and spatial distribution assessment revealed spatial autocorrelation with a Moran's index at a z-score of 2.362
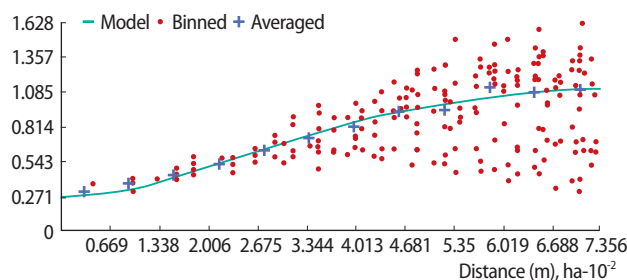
Figure 1. Isotropic semivariogam fitted with spherical model.

and a p-value less than 0.05 (Table 3), indicating that the dataset was spatially related and that both human induced and natural activities contributed to the high C_{deg}.

Spatial Structure of Degree of Contamination

The spatial structure and spatial variation of the C_{deg} were further revealed by the variogram cloud (Figure S3A) and the entropy voronoi map (Figure S3B). The variogram cloud showed the best correlation in the northeast and southwest directions, indicating an omnidirectional orientation and thus isotropy in the C_{deg}. Figure S3B shows the spatial variation in the C_{deg}. Dark and light green areas indicate little variation, while orange and dark red indicate greater variation. The map roughly indicates stationarity in the C_{deg}.

Table 4. Degree of contamination (C_{deg}) and mean concentration of heavy metals per site in AEPS

Site	Ba	Cd	Co	Cr	Cu	Hg	Ni	Pb	Zn	C_{deg}
Burning	641.5	10.1	43.8	171.5	2967.8	3.5	21.8	2666.4	1887.2	393.4
Dismantling	785.7	7.8	66.1	419.2	1643.5	1.6	44.6	846.8	1939.2	193.5
Residential	658.1	2.6	51.5	153.9	1354.6	1.6	27.2	896.1	1170.4	152.2
Recreational	443.7	1.9	36.5	338.6	762.9	0.5	14.8	355.2	700.3	74.2
Commercial area	493.6	0.6	29.6	290.9	157.7	0.3	6.2	163.9	516.9	36.2
Worship	783.9	0.3	30.5	184.6	118.2	0.4	4.9	117.0	419.9	29.4
Farm	315.4	0.2	23.7	319.2	91.4	0.5	4.9	143.5	271.2	29.1
School	394.3	0.3	42.3	118.8	47.6	0.6	2.2	111.3	293.2	25.9
Clinic	581.1	-	28.5	119.3	40.7	-	-	55.8	220.5	12.0

Concentration values are measured in ppm.
AEPS, Agbogbloshie electronic waste processing site; Ba, barium; Cd, cadmium; Co, cobalt; Cr, chromium; Cu, copper; Hg, mercury; Ni, nickel; Pb, lead; Zn, zinc.
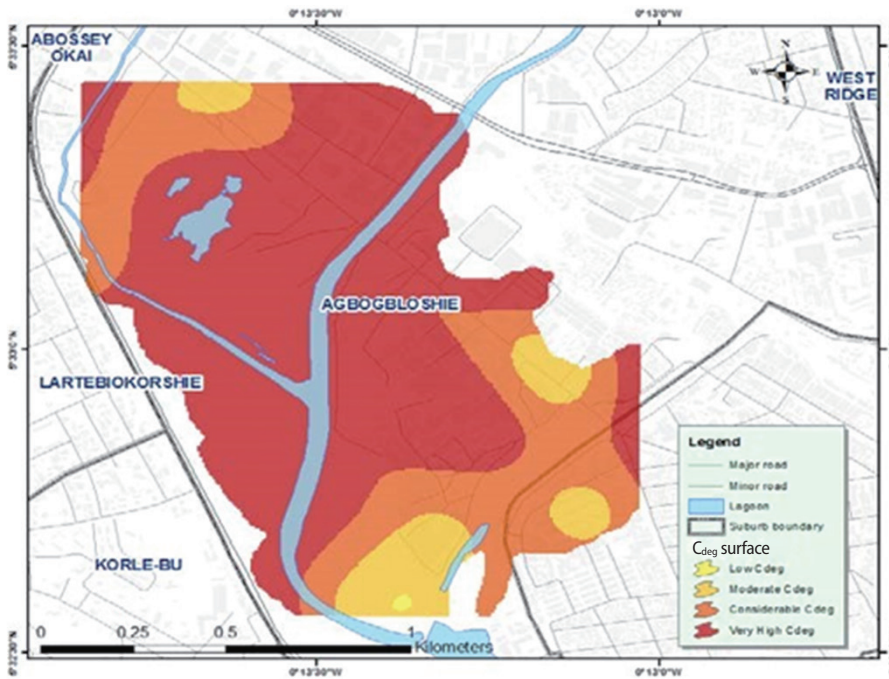
Figure 2. Spatial distribution map for degree of contamination (C_{deg}).

The isotropic semivariogram model for the C_{deg} exhibited a very good structure, which was best fitted with a spherical model in ArcGIS 10.1 (Figure 1). The model resulted in the following model parameters: a nugget value of 0.219, a sill of 1.022, and a range of 851.49 meters (Table 3). While a smaller nugget value reveals that the sample density is adequate to a good spatial structure, the nugget to sill ratio, which if less than 25%, reveals strong spatial dependence of the variable; values between 25% and 75% indicate moderate spatial dependence and at values greater than 75%, the C_{deg} variable shows a nugget to sill ratio of 0.214 (that is 21.4%, indicating moderate spatial dependence of the C_{deg} dataset). Furthermore, a range of 851.49 also indicated that the length at which the data maintain spatial autocorrelation was longer than the general sampling interval of 100 meters.

Spatial Distribution Map of Degree of Contamination

With the data on the C_{deg} exhibiting spatial autocorrelation, being stationary, omnidirectional (that is isotropic), and showing moderate spatial dependency, simple kriging was used to interpolate the surface and was subsequently classified according to the C_{deg} categories. Figure 2 shows the spatial distribution and extent at which AEPS is contaminated. For the purpose of examining the C_{deg}, the kriged surface was reclassified according to the C_{deg} set by Meza-Figueroa et al. [16]. The red areas in Figure 2 indicate highly contaminated sites within the study area.

Based on the cluster of areas such as burning, dismantling, recreational, residential, etc., an assessment of the C_{deg} within AEPS is presented in Table 4. Table 4 shows mean heavy metal concentrations in ppm and the C_{deg} per site. With reference to the C_{deg} caused by heavy metals per site, the result indicates burn-

ing > dismantling > residential > recreational > commercial area > worship > farm > school > clinic in a descending order of degree or extent of heavy metal contamination. The extremely high values recorded for the burning and dismantling sites could be due to the intensity of activities by informal e-waste recyclers in these areas; a similar trend was reported by previous studies [22–25]. The remaining sequence/order is residential > recreational > commercial > worship > farm > school > clinic could be attributed to the proximity of these areas to the main burning and dismantling sites of informal recyclers.

Discussion

The CF shows the contribution of each of the heavy metals to the C_{deg}; it shows Zn, Cu, Hg, Cd, and Pb in an increasing order as contributing immensely to the C_{deg}. The kriged map for the C_{deg} estimated on the basis of the CF and reference or background values is consistent with similar studies [7,16,18,26]. The C_{deg} map showed most of the areas within AEPS, which consists of burning, dismantling, residential, recreational, commercial, worship centers, and school were highly or severely contaminated with the studied heavy metals. A further assessment of the spatial distribution map revealed 66%, representing 110 ha of the land area, classified as severely contaminated, while 25% (42 ha), 8% (14 ha), and less than 1% classified as considerably, moderately, and slightly contaminated, respectively. The significantly high C_{deg}, spatial autocorrelation, and spatial random variance in the C_{deg} indicate anthropogenic influence, i.e., e-waste recycling activities. Hereby, school, residential, market, farm, and worship areas are of particular concern as children spend a considerable amount of time there. Heavy metals, in particular Pb and Cd, can cross the blood brain barriers of children, exerting toxic or hazardous effects which might result in a low IQ and developmental disorders; in children and adults, heavy metals can also cause cancer [7,27,28].

The spatial assessment of soil contamination from an informal e-waste recycling site in Agbogbloshie was undertaken by examining levels of heavy metals and the spatial extent of heavy metal contamination as possible environmental impacts of e-waste recycling activities. The study concludes that nine heavy metals, namely Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb, and Zn were ubiquitous within AEPS and that heavy metal concentrations in soils from the study exceeded the optimal and action requiring limits of both the Dutch and Canadian SQGV by 10 to 1000 times. These high heavy metal concentrations indicate pollution of AEPS with these nine heavy metals, in particular with Cd, Cu, Hg, Pb, and Zn. In addition, kriging requirements of normality, spatial variation, and autocorrelation were met before predic-

tion of spatial distribution maps, which revealed the contamination of the study area to an extent beyond just the main working areas of dismantling and burning sites to areas close to schools, the clinic, residential, and worship premises. The concentrations of these heavy metals and the spatial extent of their distribution poses an ecological risk for humans and other terrestrial and aquatic species at AEPS; therefore, further research of potential ecological and human health risks of informal e-recycling is needed.

Acknowledgements

Conflict of Interest

The authors have no conflicts of interest associated with the material presented in this paper.

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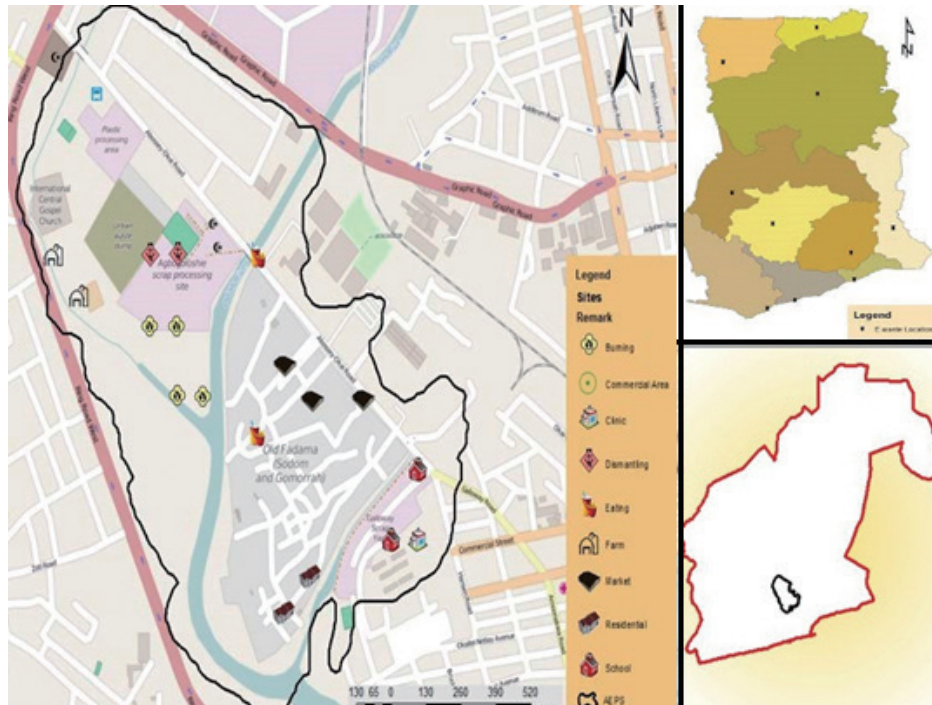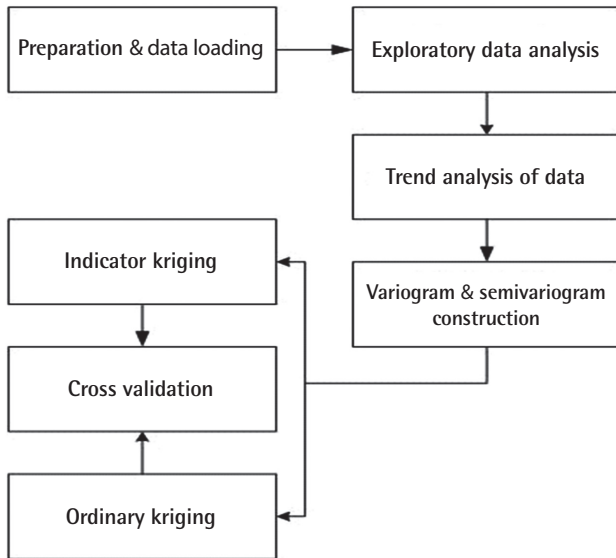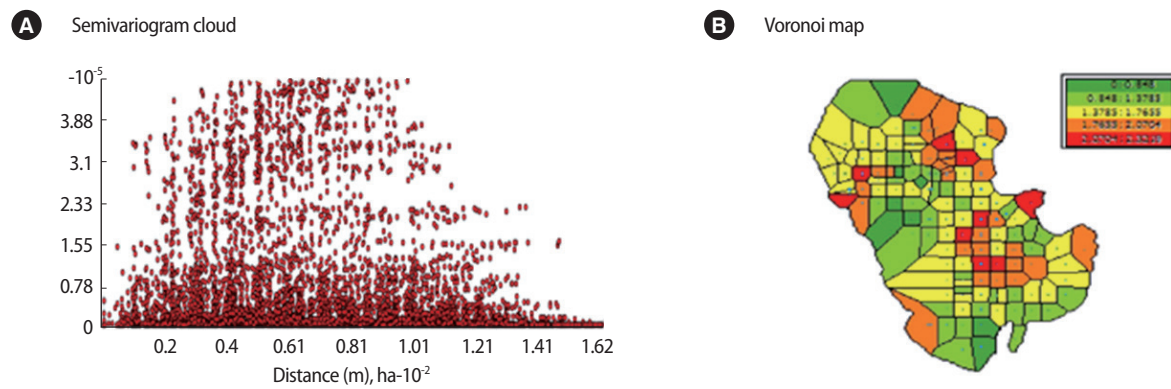**Figure S1.** Location map of Agbogbloshie processing site.

Figure S2. Steps involved in statistical and geostatistical data analysis.

A   Semivariogram cloud

B   Voronoi map

Figure S3. Semivariogram cloud (A) and voronoi map of degree of contamination (B).