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Ecological and Health Risk Assessment of Lead Contamination in Soil, Water and Plant grown nearby Lead Acid Battery Industry in Southern Area of Bangladesh

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Abstract

A newly introduced battery-operated 'Easy Bike' in Bangladesh have ascended steadily due to a rise in GDP; resulting in a sharp intensification of lead acid battery (LAB) uses. Reuse and recycle of waste LAB has a cumulative environmental impact in developed countries like Bangladesh, either positive or harmful. If the effluent water from LAB recycling industry is discharged to any drainage channel, canal, stream, river or any other water bodies, it will be contaminated with Lead (Pb). The following paper aims to determine the Pb content in agricultural soil, water and plants nearby battery recycle factory; and possible ecological and health risks of nearby residents from Pb contamination. Soil, water and plant were collected from 13 different sites and all the sampling sites were observed as very strongly polluted according to geochemical index. The consumption of Rice and vegetables grown on the polluted land were considered to be unsafe. The agricultural lands within 2 km radius are found to be extremely contaminated and the inhabitants of the study area consuming rice and vegetables can be exposed to Pb with a lifetime cancer risk as total carcinogenic risk were several times higher than the acceptable risk limit.

Keywords: Lead acid battery, Geochemical index, Carcinogenic risk, Lead contamination.

"1. Introduction"

Lead is a neurotoxic element that harms almost every system in the body [1, 2]. Up to 100 million individuals are thought to be exposed to lead pollution in soil around the world, with people in low and middle-income nations like Bangladesh bearing the brunt of the burden [3, 4]. Bangladesh is one of the fastest growing developing countries in the world due to significant economic growth. With increasing GDP, urbanization and economic development, the use of lead containing materials for battery production to utilize in electronic vehicles and solar systems are also increasing [5]. After using the lead accommodated battery materials, a large portion is sent to recycling industry. Recycling Pb acid battery has been identified the major sources of Pb exposure in the environment through their transportation, separation, dumping of battery waste and discharging of untreated battery waste [6]. During recycling of Pb battery, Pb can easily be spread out to the surrounding environment through disposal of waste, leaching, surface runoff and may find its way into nearby streams and cultivated soils; consequently taken up by food crops into their tissues [7-9]. Thus, Pb accumulation in vegetables and soil can pose serious health and ecological risk [10, 11]. The ecological and health risk assessment are evaluated by different widely accepted index such as transfer factor (TF), contamination factor (CF), geochemical index (I_{geo}) and dietary intake, non-carcinogenic (THQ) and carcinogenic risk (TR) [12, 13]. Elevated blood lead levels are substantially associated with higher rates of cardiovascular morbidity and death as damaged blood vessels and clogged blood arteries can cause hypertension and eventually a fatal heart attack [14]. Excess toxic metals such as Pb accumulation result in chronic illnesses in humans such as cancer, through the food chain interaction, even in low

concentrations throughout the lifetime, can possibly result in a number of carcinogenic and non-carcinogenic health problems including neurological, immunological, cardiovascular, renal, and reproductive disorders [15, 16]. The assessment and associated risk due to the exposure and intake of lead through lead recycling battery industry and smelter have been investigated in all over the world, like in Iran, China, Ireland, Indonesia etc. [17-21]. However, systematic investigation on lead contamination and environmental risks due to recycling activities of lead battery is very limited in the southern area of Bangladesh. The present study was conducted to determine the lead content in agricultural soil, water and plants in the nearby battery recycle industry and to evaluate the possible ecological and potential health risk associated with the lead contamination in the southern area of Bangladesh.

"2. Materials and methods"

Study Area

Khulna, third largest industrial city, is situated in the southeast part of Bangladesh. There are many recycling battery industry in the northeast part of Khulna city. The current study was carried out in the nearby area of a renowned lead acid battery recycling industry, in Khulna city, Bangladesh (89°30' E and 22°55' N). The study area being a subtropical monsoon climate is characterized by a hot, humid summer from March to June; a cool, rainy monsoon season from June to October; and a cool, dry winter from October to March. The annual precipitation and temperature were 156.53 mm and 26.3°C, respectively during the sampling year. Since the beginning of battery recycling process, a large quantities of effluents has been discharged without any treatments into a nearby water body named Beel Dakatia and a river named Bhairav through 2 separate discharge channels. Water from one of this discharge channels (D1) is used for crop cultivation and irrigation and other (D2) for drinking and household activities. In total 13 sampling sites were selected along the discharge channels where eight sampling sites were located in vicinity of the industry to Beel Dakatia, approximately 2 km away from the industry and five sampling sites were located in the industry to Bhairav river, approximately half km away from the industry (Fig. 1). Most of the agriculture fields in this area were distributed on both sides of the discharge channel (D1) and these agricultural fields were irrigated with water from D1. Rice and different vegetables are the primary cultivated crop in this area. Grass that grow nearby the channel is used as fodder. In total eight soil, water and plant samples, two hundred meter distance per sample, were collected along the discharge channel (D1). Another five water samples only, hundred meter distance per sample, were collected along the discharge channel (D2).

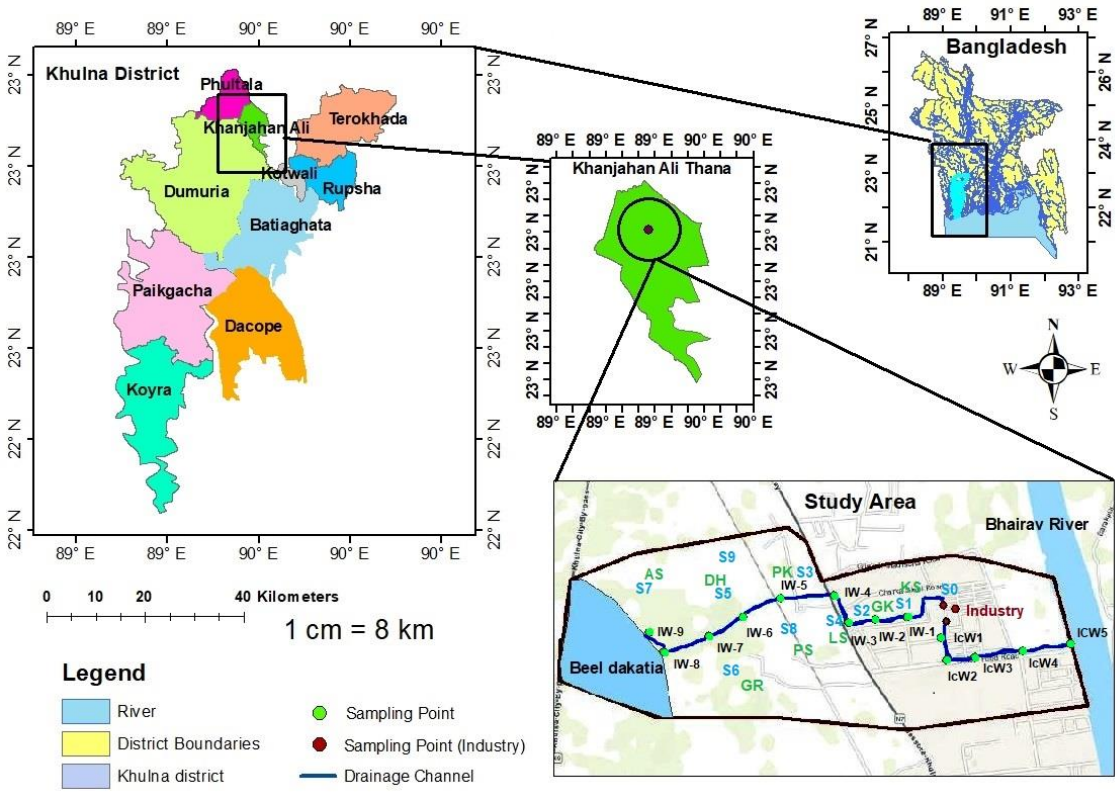


Fig. 1. Study area map

The collected plants samples were Spinach (Spinacia oleracea), Red amaranth (Amaranthus gangeticus), Creeping woodshorrel (Oxalis corniculata), Water spinach (Ipomoea aquatic), Kangkong (Ipomoea reptans), Grass (Cynodon dactylon), Rice (Oryza sativa), Taro (Colocasia esculenta) which were indicated as PS, LS, AS, PK, GK, Gr, DH, KS, respectively.

Soil, water and plant samples analysis

The collected soil samples were air dried, ground 2 mm sieve and stored for laboratory analysis. The collected plant samples were oven dried at 75℃ temperature for 48 hours. The dried plant samples were blended and sieved then stored for further analysis. The soil and plant samples were digested by using HNO_3 and $HClO_4$ digestion mixture. Shortly, 0.5 g sample were digested by concentrated HNO_3 until the mixture almost dry. Then the samples were kept for cooling and the concentrated $HClO_4$ was added. The mixtures were heated until the sample become clear. The remaining excess acids were evaporated by heating then washed, filtered, made volume and stored as digest [22]. The dissolve oxygen (DO) of the collected water samples were determined by using DO Meter (HI98193). The biological oxygen demand (BOD) was measured by incubation in the dark condition for 5 days at 20°C [23]. COD was determined by boiling the content in water bath until the contents were evaporated to dryness and adding 10 ml 0.4 N $K_2Cr_2O_7$ to titrate against 0.1 N Mohr's salt solution [23]. The concentration of lead in the water, soil and plant samples were measured using Atomic Absorption Spectrophotometer [24]. All data were analyzed by using IBM SPSS software. The study area maps were created using ArcGIS-version 10.3 software.

"3. Results and discussion"

Physio-chemical properties and lead content in water, soil and plants

Discharged waste water from battery industry through drainage channel 1, D1 and drainage channel 2, D2 both were observed and chemical analysis were carried out in this study. The pH values were within 5.2 to 7.2 for D1 which were considered as acidic to neutral and within 8.2 to 7 from D2 which were measured as alkaline to neutral. The electric conductivities (EC) of the samples were observed 1075 to 1885 μScm^{-1} from D1 and 8330 to 8690 μScm^{-1} from D2. According to the standard level of EC for irrigation purposes are 750 μScm^{-1}, while the EC values were higher than the standard level, which indicates the presence of ionic compounds in water [25]. Alkaline water and high amount of EC found from sampling site were close to the industry outlet. The TDS and TSS concentration being <100 mgL^{-1} and <450 mgL^{-1} is considered safe for using as irrigation water [26]. As the TDS concentration was 687 to 1470 gL^{-1} from D1 and 780 to 1286 gL^{-1} from D2, so the TDS range of 450-2000 mgL^{-1} is depicted as slight to moderate restricted for use as irrigation water [27]. With a decreasing tendency of TSS concentration 600 to 1800 mgL^{-1} from D1 and 600 to 200 mgL^{-1} from D2, has fallen under severely restricted for irrigation use [27]. Adequate DO is necessary for good water quality, endurance of aquatic organism and decomposition of waste by microorganism [28]. The DO was found in an order: IW-3<IW-1<IW-2<IW-5<IW-6<IW-7<IW-8<IW-9 from D1 and 1 to 1.63 mgL^{-1} from D2, which were below the recommended value 6.25 mgL^{-1} DoE [29]. The BOD was found from 23.75 to 330.15 mgL^{-1} for D1, where higher concentration found in sample IW-1 to IW-7and from10.3 to53.3 mgL^{-1} for D2, where IcW3 was found greater concentration than standard level. COD was observed 1170 to 169.6 mgL-1 from D1, where all the samples were having higher concentration than standard level and 170 to 128.6 mgL-1 from D2 which were within standard level. The DoE standard concentration of BOD and COD is 50 and 200 mgL^{-1} respectively for irrigation water [29]. The study found higher concentration of BOD and COD from D1, which indicated that the water is poisonous to aquatic life in the vicinity and might be the source of numerous disruption on ecology [30]. As the highest value of Pb (68.32 mgL^{-1}) was observed in IW-1 and the value was decreasing with distance that up to IW-5 site, the irrigation water is recommended risky because the concentration is higher than the maximum allowable limit of lead in irrigation water which is 5 mgL^{-1} as recommended by the FAO [27]. From IW-5 to IW-9 the lead content is less than the threshold limit and this portion is considered less risky. Although the highest value (0.91 mgL^{-1}) of lead detected from IcW1 and the lowest value (0.21 mgL^{-1}) detected from IcW5 from D2 and the concentration were decreased with distance, but all the concentration were higher than the recommended value 0.01 mgL^{-1} for drinking water [31]. Furthermore, D1 was found to be used in agricultural soil for irrigation of crops. Considering all the observed values of the physiochemical properties and lead content: higher than the standard level, it can be declared that both of the drainage canal D1 and D2 are risky for use in irrigation and drinking purpose.

The agricultural soils of the study area were irrigated with water from drainage channel D1 and another channel D2 has mixed with river water which is used for drinking and household by the nearby residents.

Highest lead content was found 4985 $mgKg^{-1}$ in S1 sampling site, closest to the industry and lowest content (78.48 $mgkg^{-1}$) in S9 soil sampling site. The uptake of lead in vegetables from soil, varies from plant to plant depending on the concentrations of metals in soils and irrigation water and excessive accumulation in agricultural soils not only result in soil contamination but also affect food quality and safety, which is quite evident in the level of Pb concentration (704.86 mgL^{-1}) in rice [32]. Considering the plant sample, the total lead content was found a following order: KS>GK>GR>PK>LS>DH>AS>PS. Elevated concentrations of Pb were observed in vegetable species that all the values were higher than the WHO permissible limits (0.3 $mgkg^{-1}$) for leafy vegetables, indicating lead might pose health risk from the consumption of these vegetable species [33].

Ecological risk assessment

In this study ecological risk assessment were described by using different pollution indices such as transfer Factor (TF), contamination factor (CF), geoaccumulation index (I_{geo}). TF is the measurement of plant capability to uptake metals from soil to plant tissue and was computed as the ratio of metal concentration in root and shoot of plants with the lead concentration in soils [34]. In this study, transfer factor was found to be in the order: DH <LS <KS <PK <GK <Gr <AS <PS (Table 1). Contamination factor (CF) was calculated from equation (1) which indicates the anthropogenic incorporation of elemental pollution and is widely used as a measure of overall contamination of soil in Asian countries [35]. Highest contamination factor was found 447.89 in S1 sampling site and all the sampling site observed as very strongly polluted as all the values were greater than 6 as suggested by Varol [36].

$$\mathrm{CF} = \frac{\mathrm{C_m}}{\mathrm{B_m}} \quad (1)$$

Where, C_m is the respective metal concentration in the soil and B_m is the respective metal concentration in the background soil. Muller proposed the geoaccumulation index (I_{geo}) which is widely used to assess the degree of soil contamination by trace metals which was calculated by using formula (2) in the current study [37, 38].

$$\mathrm{Igeo} = \log_2\left(\frac{C_n}{1.5B_n}\right) \quad (2)$$

C_n is the measured concentration of respective element, B_n is the background concentration of the respective element and 1.5 is the constant which is used to reduce the variation due to lithogenic effects on background element concentration [39]. According to Muller, the calculated values of geochemical index of sampling site S1 to S6 were more than 5 whereas S8 and S9 having values (2.71 and 2.23) between 2 and 3 as depicted in Table 1, so they can be marked as moderate to heavily polluted [37]. Lead accumulation in different species of the study decreased in the following order: grain plant < root vegetables <leaf vegetables [40].

Table 1. Ecological risk assessment of Pb for agricultural soil nearby lead acid battery industry

Plant sample	Transfer factor	Soil sample	Contamination factor (CF)	Degree of contamination	Geochemical index (Igeo)	Interpretation
KS	0.63	S1	447.89	Very strongly polluted	8.22	Extremely contaminated
GK	0.92	S2	255.78	Very strongly polluted	7.41	Extremely contaminated
PK	0.69	S3	240.81	Very strongly polluted	7.33	Extremely contaminated
LS	0.47	S4	186.70	Very strongly polluted	7.17	Extremely contaminated
DH	0.44	S5	150.41	Very strongly polluted	6.96	Extremely contaminated
GR	1.48	S6	14.18	Very strongly polluted	6.65	Extremely contaminated
AS	4.97	S7	215.90	Very strongly polluted	3.24	Heavily to extremely contaminated
PS	5.46	S8	9.84	Very strongly polluted	2.71	Moderate to heavily contaminated
-	-	S9	7.05	Very strongly polluted	2.23	Moderate to heavily contaminated

Health risk assessment

The health risk assessment was evaluated by estimated daily intake of Pb, non-carcinogenic risk, carcinogenic risk. The estimated daily intakes (EDI) of toxic elements through vegetables in our study area were measured by using elements concentration in vegetables on fresh weight basis, daily consumption of vegetables and body weight and the EDIs were calculated by using following equation (3) [41].

$$\mathrm{EDI} = \frac{\mathrm{FIR} \times \mathrm{C}}{\mathrm{BW}} \quad (3)$$

Where, FIR is the vegetable consumption rate in fresh weight basis (g/person/day), C is the elements concentration in vegetables [mg/kg, fresh weight (fw)], BW is the body weight per person (60 kg in the current study) and the vegetables consumption rate for an adult person in the current study was 191 g on a fresh weight basis [42].

The non-carcinogenic health risk associated with the consumption of rice and vegetable species by the local inhabitants was assessed in this study based on the target hazard quotients (THQs) using the standard assumption for an integrate USEPA risk analysis. Higher THQ values mean a higher probability of experiencing long-term non-carcinogenic health effects. Equation (4) was used for the calculation of THQ [43, 44].

$$THQ = \frac{\mathrm{EFR} \times \mathrm{ED} \times \mathrm{EDI} \times \mathrm{CSFo}}{\mathrm{RFD} \times \mathrm{BW} \times \mathrm{AT}} \times 10^{-3} \quad (4)$$

Where EFR is the exposure frequency (350 days/year) and 365 days/year for cattle, ED is the exposure duration (73 years) equivalent to the average lifetime of human and 18 years for cattle; Oral reference, RfD dose 0.001 mg/kg/day for Pb, BW is the average body weight (adult, 60 kg and cattle, 244 kg) and AT is the average time for non-carcinogens (365 days/year 73 years and 365 days/year 18 year for cattle) [45, 46]. Whether the THQ is equal to or higher than 1, there is a potential health risk [47].

For carcinogens, risk was estimated as the incremental probability of an individual to develop cancer over a lifetime exposure to that potential carcinogen or excess individual lifetime cancer risk. Equation (5) was used for estimating the target carcinogenic risk [48]

$$TR = \frac{\mathrm{EFR} \times \mathrm{ED} \times \mathrm{EDI} \times \mathrm{FIR} \times \mathrm{CSFo}}{\mathrm{BW} \times \mathrm{AT}} \times 10^{-3} \quad (5)$$

The oral carcinogenic slope factor CSFo were 8.5×10^{-3} $(\mathrm{mg/kg/day})^{-1}$ for Pb from the Integrated Risk Information System database [46]. The EDIs of Pb was evaluated according to the concentration of Pb in the edible part of rice and vegetables and the respective consumption rate and are shown in table 2 [49]. Total daily intakes of Pb through the consumption of vegetables were higher than the maximum tolerable intake (MTDI) 0.21, indicated that these vegetables might pose risk to the consumers in the study area [50]. However, the EDI for animals from grazing on grasses was higher than that of from vegetables and which could be attributed to the higher risk for human when consuming milk or meat of those animal rate of vegetables. The EDI revealed that grass, rice and vegetables all are responsible to create health risk in the vicinity of agricultural field nearby lead acid battery industry in Bangladesh.

THQs due to consumption of grass, rice and vegetables are listed in Table 2. The THQ values for Pb in vegetable were less than 1 except PS and DH, which is considered safe for animal and human consumption. Conversely, THQ of Pb for Rice (1.11), and Spinach (1.12) were greater than 1.0 which can be a potential health risk for human health [47]. The consumption of Rice and Spinach grown on the polluted land were considered to be unsafe. Therefore, the consumers are at high risk due to the exposure of Pb from rice and vegetables which were associated with non-carcinogenic risk. It is recommended not to consume rice and vegetables growing in the study area as those can be exposed as a considerable risk.

Table 2. Health risk assessment of Pb for vegetables/food crops grown nearby lead battery industry

Plant sample	Estimated daily intakes (EDIs)	Non carcinogenic risk (THQ)	Carcinogenic risk (TR)
KS	0.34	0.25	1.48×10^{-2}
GK	0.50	0.37	2.19×10^{-2}
PK	0.44	0.32	1.92×10^{-2}
LS	0.85	0.62	3.69×10^{-2}
DH	1.53	1.11	6.65×10^{-2}
GS	58.08	0.66	$1.\ 60\times10^{-1}$
AS	1.30	0.94	5.65×10^{-1}
PS	1.55	1.12	6.72×10^{-2}

The target carcinogenic risk (TR) derived from the intake of Pb was calculated since it may promote both non-carcinogenic and carcinogenic effects depending on the exposure dose. The TR values of Pb for adults due to exposure to Pb from consumption of fish and vegetables are presented in Tables 2. In general, the excess cancer risk lower than 10^{-6} is considered to be negligible, cancer risk above 10^{-4} is considered unacceptable, and cancer risk lying between 10^{-6} and 10^{-4} is generally considered an acceptable range [46]. Considering the food-specific exposure, it was found that the TR values of Pb from consuming rice and vegetables were several times higher than the acceptable risk limit (10^{-4}), indicating that the inhabitants consuming these rice and vegetable species were exposed to Pb with a lifetime cancer risk. Based on the results of the present study, the potential health risk for the inhabitants due to Pb exposure through consumption of rice and vegetables should not be overlooked. It is thus suggested that constant monitoring of Pb in the milk and meat is needed in order to evaluate if any potential health risk to the consumers does exist through cattle.

"4. Conclusion"

Although the battery recycling industry has enormous economic impact on the socioeconomic development of Bangladesh but the environment risk is seemed to be alarming unless proper safety measure is not maintained. The battery recycling industry of the studied area even though have some safety measures like an effluent treatment plant (ETP) and an air treatment plant (ATP), but they were not using it every time being costly to run. The lead plates were pulled out from inside the plastic casings and left on the floor. Then the casings were washed in a concrete tank in which the water had turned black with lead dust and acid. The acid was then poured into the drain from where it washed down to the cannel that ultimately connects with River Bhairav half a km and Beel Dakatia 2 km away. The study was carried out to estimate the lead contamination associated with ecological and health risk in agricultural soil, water and plants in the vicinity of the lead acid battery recycling industry. High Pb concentration found in the soil, plant and water samples collected within 2 km radius which indicates very strongly polluted according on the basis of contamination factor. The geochemical indices suggest that up to S6 sampling site, the contamination is extreme; indicating severe risk of lead contamination for the ecosystem of the study area. Based on the results of the present study, lead concentration in rice, grass and other vegetables found to have high Pb concentration compared to permissible limit and carcinogenic risk indices were several times higher than the acceptable risk limit, demonstrating that a lifetime potential cancer risk can be take place from consuming these rice and vegetable species. The amount of Pb might contribute to a major threat of Pb entrance in the food chain along with potential ecological and health risk. As higher lead content has found in rice and vegetables compared to the permissible limit in agricultural soil so it is highly recommended to the farmers in the nearby area, not to grow edible crops in the contaminated sites and irrigated with the waste water. Particularly, the excessive Pb concentration found in grass with potential carcinogenic risk, indicating the threat of Pb poisoning to the people who would consume the meat or milk of the livestock grazing the grasses grown when the agricultural fields remain bare. It is thus suggested that, constant monitoring of Pb in the milk and meat is needed in order to evaluate if any potential health risk to the consumers does exist through cattle grazing grass from that area. Furthermore, a long-term risk assessment needs to be carried out on the leach ability and migration potential of Lead (Pb) at the contaminated sites. The residence need to be awaked about the lead pollution happening due to the negligence of the industry authority and also possible lead poisoning from intake of rice and vegetables grown nearby agricultural land around 2 km away from the industry. Moreover, different remediation measures should be taken promptly to remove/reduce excising metal (Pb) contamination.

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