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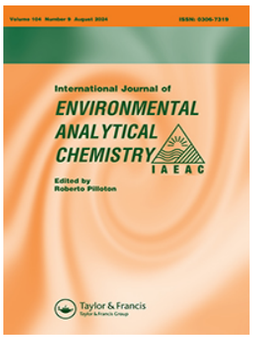

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Potentially toxic metals in road dust: An assessment of ecological and health risks at Woldia and Kobo towns in North Wollo, Ethiopia

Asamene Embiale Taye[a] and Bhagwan Singh Chandravanshi[b]

[a]Department of Chemistry, College of Natural and Computational Science, Woldia University, Woldia, Ethiopia; [b]Department of Chemistry, College of Natural and Computational Sciences, Addis Ababa University, Addis Ababa, Ethiopia

ABSTRACT

The accumulation of heavy metals (HMs) in the environment threatens the ecosystem and human well-being. This study investigated the amounts of Mn, Fe, Ni, Co, Cu, Zn, Cd, Hg, Pb, As, and Cr, as well as the associated health and ecological risks. Thirty samples of roadside dust were collected in the northern Ethiopian towns of Woldia and Kobo. The collected samples were digested using acid digestion method and the levels of heavy metals were analysed using inductively coupled plasma optical emission spectroscopy. The results showed that the average concentrations of HMs were 0.103 (As)-7.6 (Cd) and 0.083 (Cd)-3.80 (Fe) mg/kg in Woldia and Kobo, respectively. All the HM concentrations investigated in this study were lower than the average global concentration, except for Hg in both towns and Cd in Woldia town. Analysis of variance (ANOVA) for the heavy metal concentration showed a significant difference across each sampling site, while Ni and Hg did not show a significant difference. The hazard quotient (HQ) and hazard index (HI) values of all HMs for adults and children through the three exposure routes were lower than 1, which means that they had a low likelihood of noncarcinogenic risks. The pollution index, geoaccumulation index, ecological risk index, integrated pollution index and risk index parameters were used to estimate the ecological risk. The results demonstrated that the pollution level mainly varied between low and moderate levels, except in Woldia, which is highly polluted by Cd metal that further detailed research is highly recommended to find out the sources of this metal.

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Potential toxic metals; road dust; health risk; ecological risk; Ethiopia; ICP-OES

1. Introduction

Road dust pollution in large cities and small towns is one of the world's primary significant concerns. Road dust has a diverse chemical makeup, with the contents determined by the interactions of solid, liquid, and gaseous components. Hazardous chemicals such as HMs, polyaromatic hydrocarbons, and biological contaminants can also be transported. Heavy metals, among these pollutants, can be hazardous or damaging to the environment and living organisms even in low quantities [1–3]. The most important sources of HMs in road

CONTACT Asamene Embiale Taye asamene@wldu.edu.et

dust are biomass fuel burning, vehicle traffic, metallic equipment corrosion, water transport from surrounding soils, city building weathering, biological waste from domestic houses, road surface wear, road paint degradation, vehicle wear (tyres, brake linings, bodies, and others), and vehicle fluid and particulate emissions [2–6]. Heavy element-contaminated dust adversely affects the entire ecosystem due to its translocation, persistence, biomagnification, and bioaccumulation properties [3,5].

According to previous studies, certain HMs, such as Cu, Co, Mn, and Zn, are harmless for humans to consume in small quantities. However, several elements, such as Pb, As, Hg, and Cd, are hazardous even in very small amounts [2–7]. HMs bound in road dust can enter the human body by breathing, swallowing, and skin contact [8]. Once HMs enter the body, they can cause adverse effects on vital systems, such as the nervous, circulatory, excretory, reproductive, endocrine, and immune systems; cardiovascular and urogenital systems; and normal cellular metabolism. The adverse effects on the health of HMs are influenced by the toxicity, bioavailability, quantity, age of the exposed person, length of contact time and frequency with HMs, other factors, socioeconomic difficulties, and the individual's health status. For instance, the two most vulnerable groups are children and elderly individuals. This is due to immature or impaired immune systems and the unintentional ingestion of enormous amounts of dust through sucking their fingers or mouthing nonfood things [2–7,9]. Acute exposure to dust with high HM contamination or chronic exposure to dust with low HM contamination might have similar health impacts [3,5,10]. On the other hand, the accumulation of heavy metals in the environment prevents plants from undergoing photosynthesis, transpiration, or respiration [8].

Determining the HMs in road dust is important for characterising urban environmental quality, assessing pollution levels, and estimating the health impacts on inhabitants. Hence, many studies have been conducted around the globe regarding HM contamination in urban areas. The alarming increases in the number of old cars and three-wheel Bajajs, improper disposal of waste, and washing and maintaining vehicles at the roadside are the most common practices seen in most towns in Ethiopia. However, little research on this topic has been reported in Ethiopia [11–15]. As a result, there is a need for more studies on ecological and health risk assessments, as this study is the first attempt to investigate the level of HMs in road dust and assess the health risk for the inhabitants of Woldia and Kobo towns in North Wollo, Ethiopia. Additionally, the study quantified the environmental risk of heavy metals bound in road dust in the study towns [16,17].

2. Materials and methods

2.1. Sampling area description

The sampling area for this particular study was the North Wollo administrative zone, Ethiopia. This district has many towns, and most of its parts are unsuitable for agriculture since several parts of the zone are mountainous and have steep slopes. The altitude of the site varies from 600 to 4284 metres above sea level. It has an area of 12,172.50 km^2 and a population density of 123.25/km2. Among the towns in the district, Woldia is a large town with a population density of 130/km^2 and acts as the administrative centre for the community. The second sampling town for this study was Kobo, the second largest town in the zone, which acts as the administrative centre of the Raya Kobo district. Moreover,

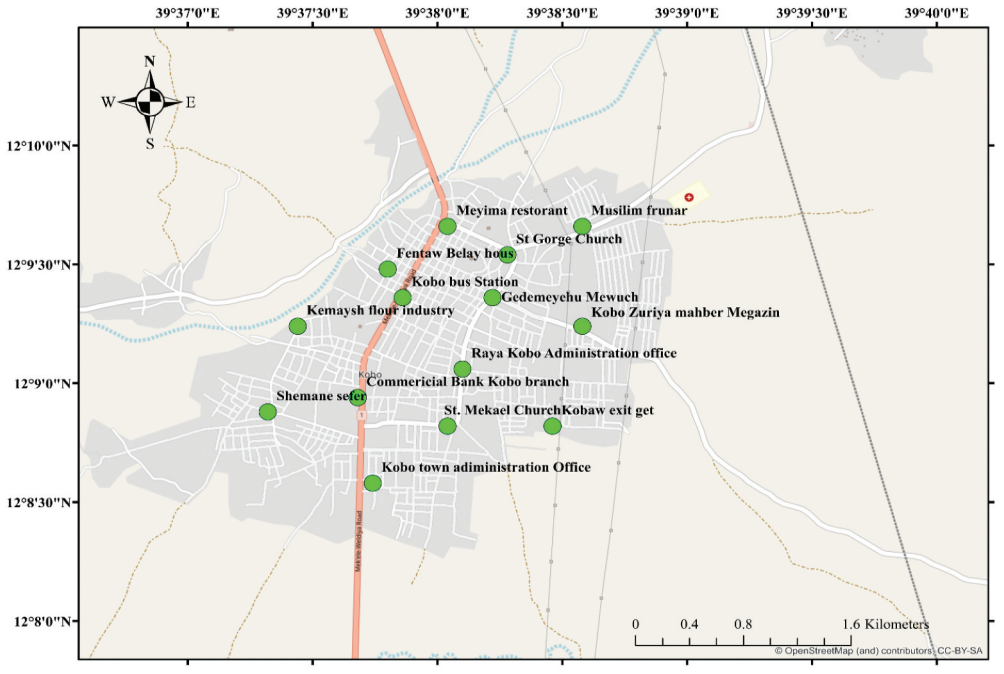

Figure 1. Sampling points in Kobo town.

Kobo and Woldia towns are the most fast-growing towns which might resulted more pollution as compared to other towns in the district, that is why this study focused on these towns. The annual rainfall and temperature in the region range from 525.8 to 919.6 mm and 11.4 to 24.7°C, respectively [18]. In addition, during the collection of samples, the geographic coordinates of each site were taken using handled GPS (model: Garmin GPS 73), then after the locations of each sampling points were drawn using ARIC GIS software 10.4.1. The geographical location of the sampling points for Kobo and Woldia towns are depicted in Figures 1 and 2 respectively.

2.2. Sample collection and laboratory analysis

A combination of purposive and simple random sampling techniques was employed for the sampling of dust samples. A total of 30 samples (16 from Woldia and 14 from Kobo) were collected to make up one bulk sample during the dry season (between December 2021 and February 2021). The selected months are due to high socio-economic movement and weather condition (high temperature and windy periods) which aggravate the formation and suspension of dust in air. The individual samples were taken at four different points along the roadside at distances of 3–5 m from each other. The sampling points are selected by considering various land uses, including education, places, street vendor activities, local market places, fuel plant, shops, the traffic density, topographic, student commuter routes, maintenance and servicing of cars, parks, residential and commercial. In addition, the rural community routes that allow inhabitants enter into the towns to were also considered during selecting the

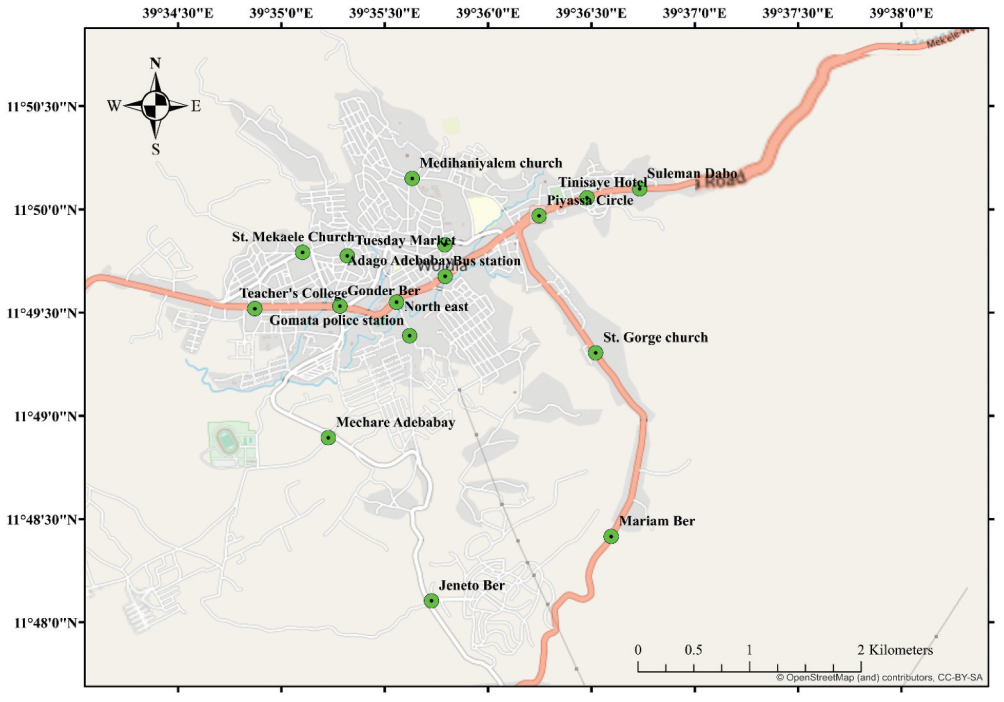

Figure 2. Sampling points in Woldia town.

sampling location. Moreover, Woldia town is located at the main highway that connects the Afar, Tigray and Amhara regional states, Similarly, Kobo town also found at the main high way that connected Tigray and Amhara regional states. where an enormous number of heavy vehicles are going daily, increasing the soil pollution by HMs. The sampling was carried out in three rounds (one sample each month) to get a representative sample. Moreover, during a single sample we have taken a sample at morning (6:00–8:00 Am), mid-day (12:00–2:00 pm) and afternoon (5:00–6:00 pm) to represent the temporary variations.

After the samples were collected in the laboratory, large pieces such as bricks, leaves, and paving stones were removed from the samples, and then, the samples were oven-dried and sieved through a 1 mm plastic sieve. After sieving, the sample was digested using an established method [19]. Thus, an acid digestion method has been employed, that 1.25 g of the dry weight dust was weighed and put into a beaker that contained 20 mL of aqua regia (HCl/HNO_3, 3:1), and then the mixture was heated on a hot plate until near dryness and it was cooled to ambient temperature followed by addition of 5.0 mL of hydrogen peroxide to complete the digestion, and then the resulting mixture was heated again to near dryness in a fume hood. The beaker was rinsed with 5 mL of deionised water. After letting the digested solution cool, it was transferred into a 100 mL standard volumetric flask and filled to the mark with deionised water. Finally, the average concentrations of Fe, Pb, Cu, Mn, Cr, Co, Ni, Zn, Hg, As, and Cd were measured using inductively coupled plasma optical emission spectroscopy (ICP–OES). The method accuracy was validated by performing a recovery test, while the method precision was determined

using the standard deviation calculation. All the individual measurements were carried out in triplicate.

2.3. Health risk assessment method

2.3.1. Exposure assessment

The US Environmental Protection Agency's [20] method was used to determine health risks arising from exposure to metallic elements in road dust. The three ways for road dust to enter the body are oral ingestion, inhalation, and skin contact. The exposure dose received through each of these routes over a lifetime can be estimated using the following equations: 1–4 [20–22]. Here, Ding is the daily dose taken via oral ingestion of road dust particles, Dinh is the daily dose taken via inhalation of resuspended dust particles, and Dder is the daily dose taken via skin absorption of dust-bound elements upon exposure to skin [2,3,7].

$$D_{ing} = Cx\frac{IngRxEFxED}{BWxAT}xCF \tag{1}$$

$$D_{inh} = Cx\frac{InhRxEFxED}{PEFxBWxAT} \tag{2}$$

$$D_{der} = Cx\frac{SLxSAxABSxEFxED}{BWxAT}xCF \tag{3}$$

$$LADD = \frac{CxEFxEDxCR}{ATxPEFxPEF} \tag{4}$$

where C is the average metal amount in the road dust samples (mg/kg); EF is the exposure frequency in days/year; BW is the average body weight of the exposed person in kg; IngR is the ingestion rate of the road dust in mg/day; ED is the exposure duration in years; AT is the averaging time in days; CF is the conversion factor in kg/mg; InhR is the inhalation rate of the road dust in m^3/day; SA is the surface area of the skin contact with the road dust in cm^2; AF is the skin adherence factor for the road dust in mg/cm^2day; LADD is the lifetime average daily dose in mg/kg day; PEF is the particle emission factor in mg/m^3; and CR is the contact frequency, which is equivalent to IngR for this study [23–26]. The exposure parameters used for the health risk calculations are shown in Table 1.

2.3.2. Risk characterization

Acute or chronic exposure to various HMs increases the risk of cancer and noncancer illnesses. The possible noncarcinogenic risks of the metals were estimated by Eqs. 5 and 6. The most popular parameters that can be used to quantify the possible noncarcinogenic risks of HMs are hazard quotient (HQ) and hazard index (HI) values. HI or HQ values less than or equal to unity (≤1) indicate no significant risk of noncarcinogenic health effects, while HI or HQ values > 1 indicate high possible noncarcinogenic adverse health effects [22].

On the other hand, the cancer risk from carcinogenic heavy metals was estimated based on the LCR (lifetime cancer risk) value. For carcinogenic elements, the LCR

Table 1. Exposure factors and parameter defaults [9,24,26].

Factor	Definition	Unit	Value	
			Children	adult
C	Concentration	mg/kg	This study	This study
IngR	Ingestion rate	mg/day	200	100
InhR	Inhalation rate	m^3/day	12.8	7.63
PEF	Particle emission factor	mg/m^3	$1.36x10^9$	$1.36x10^9$
SA	Exposed skin area	cm^2	5700	2800
SL	Skin adherence factor	mg/cm^2day	0.07	0.02
ABS	Dermal adsorption factor	Unitless	0.01	0.01
CF	Conversion factor	kg/mg	10^{-6}	10^{-6}
EF	Exposure frequency (site specific)	Days	350	350
ED	Exposure duration	Years	24	6
AT	Averaging time for noncancer risk	Days	365 x ED	365x ED
	Averaging time for cancer risk	Days	65 x 365	65 x 365
BW	Body weight (site specific)	kg	60.7	15

AT = EF × ED; the ED for adults for cancer risk is 65 years, which is the average life expectancy of an Ethiopian woman.

associated with exposure to that element is in the range of 10^{-6}–10^{-4} [21]. An LCR less than 10^{-6} is acceptable (unlikely to develop cancer), while an LCR greater than 10^{-4} is unacceptable (1 in 10,000 people who have developed cancer). However, an LCR in the range of 10^{-6}–10^{-4} is tolerable. The LCR for each carcinogenic metal in each exposure route was calculated using Eqs. 4 and 7. For the purpose of calculating the LCR, the reference dose (RfD) and slope factor (SF) for various exposure routes are provided in Table 2. The values for each parameter come from the USEPA and the Risk Assessment Information System of the US Department of Energy [7,27,28].

$$HQ = \frac{D}{RfD} \tag{5}$$

$$HI = \sum HQ_{ingestion,Inhalation,dermal} \tag{6}$$

$$LCR = LADDxSF \tag{7}$$

Table 2. The reference dose (mg kg^{-1} day^{-1}) and slope factor for each exposure route [7,9,27,28].

Metals	RfD_{inh}	RfD_{derm}	RfD_{ing}	SF_{ing}	SF_{der}	SF_{inh}
Fe	0.00022	0.07	0.84			
Cu	0.0402	0.012	0.04			
Mn	1.43E–05	0.00184	0.047			
Zn	0.3	0.06	0.3			
Pb	0.00352	0.000525	0.0035	0.085	0.0085	0.00008
Cr	2.86E–05	0.00005	0.003	42	0.0065	0.012
Cd	0.001	0.00001	0.001	6.3	6.3	0.0018
As	0.000301	0.000123	0.0003	0.151	1.5	0.0043
Ni	0.0206	0.0054	0.02	0.84	0.84	0.00024
Co	5.71E–06	0.016	0.02	9.8		
Hg	8.75E–05	0.000021	0.0003			

'RfD' is the reference dose; 'SF' is the slope factor; 'ing' is ingestion; 'inh' is inhalation; and 'derm' is dermal contact

2.4. Ecological risk assessment

2.4.1. Geoaccumulation index (I_{geo})

I_{geo} is used to evaluate the contamination level of elements. To compute it, equation 8 is used:

$$I_{geo} = log_2\left(C_n/_{1.5B_n}\right) \tag{8}$$

where C_n is the mean level of the metals in road dust (mg/kg), and B_n is the concentration of the metals (mg/kg) in the reference sample. To reduce the impact of potential variations in the background or control values, which may be related to lithogenic differences in the sediment, a factor of 1.5 is introduced. I_{geo} consists of seven classes, which are given as follows: $I_{geo} < 0$ means unpolluted environments; $I_{geo} > 5$ means an extremely polluted environment; $4 < I_{geo} < 5$ means strongly to very strongly polluted; $3 < I_{geo} < 4$ means strongly polluted; $2 < I_{geo} < 3$ means moderately to strongly polluted; $1 < I_{geo} < 2$ means moderately polluted; and $0 < I_{geo} < 1$ means unpolluted to moderately polluted [2,3,6,7].

2.4.2. Pollution index (PI)

As with the other indicators, the PI is used to determine the pollution level of a single metal, which is calculated using equation 9:

$$\mathrm{PI} = \mathrm{C_i}/\mathrm{C_{0i}} \tag{9}$$

where C_i is the concentration of a given i^{th} metal in the road dust sample (mg/kg) and C_{0i} is the level of HMs in the reference sample (mg/kg). Thus, according to the single metal pollution index criterion, the PI values for each HM are ranked as follows: low pollution level ($PI < 1$), moderate pollution level ($1 \leq PI \leq 3$) and high pollution level ($PI > 3$) [3,29]. Generally, the background values for each metal were quantified from samples collected in farmlands far from towns [30].

2.4.3. Integrated pollution index (IPI)

The IPIs are the arithmetic mean values of the PIs of the metals. Hence, based on the value of the IPI, the level of pollution was divided into three categories: low pollution ($IPI < 1$), moderate pollution ($1 < IPI \leq 2$) and high pollution ($IPI > 2$) [3].

2.4.4. Ecological risk index (ERI) and risk index (RI)

The ecological risk for particular metals can be quantified by the ERI, whereas the RI quantifies the ecological risk of a group of metals. The ERI is computed using formulas 10 and 11:

$$ERI = TrxPI \tag{10}$$

$$RI = \sum_{1}^{n} ERI \tag{11}$$

where Tr indicates a toxic-response factor, and each element has its own Tr values, which are given as a standard (i.e. Mn = 10, Ni = 5, Cu = 5, Zn = 1, Cd = 30, Hg = 40, Pb = 5, As = 10, and Cr = 5) [4,8,31,32]. The levels of pollution based on the ERI are divided into

five categories: low pollution (ERI <40), moderate pollution (40 ≤ ERI < 80), high pollution (80 ≤ ERI < 160), severe pollution (160 ≤ ERI < 320) and severe pollution (ERI ≥320), whereas the RI is divided into four categories: very high pollution level (RI ≥ 600), high pollution level (300 ≤ RI < 600), moderate pollution level (150 ≤ RI < 300) and low pollution level (RI < 150) [17,29].

2.4.5. Statistical analysis

All the statistical analyses were performed using SPSS software (version 21), Origin software (version 2018), and Microsoft Office Excel 2016. Correlation analysis and ANOVA were performed to explore the possible relationships between the studied element concentrations and sampling sites. Unless indicated in the study, all the statistical analyses were performed at a confidence level of 95%.

3. Results and discussion

3.1. Heavy metal concentrations in road dust

The HM arithmetic means and maximum and minimum values for road dust in Woldia and Kobo towns are presented in Table 3. The concentrations of Mn, Fe, Ni, Co, Cu, Zn, Cd, Hg, Pb, As, and Cr varied between 2.01 and 2.77, 3.14 and 5.08, 1.82 and 2.87, 2.11 and 2.82, 0.8 and 0.88, 1.44 and 1.57, 0.083 and 7.60, 0.12 and 0.18, 0.57 and 0.66, 0.10 and 0.15, and 1.41 and 2.05, respectively. The total mean heavy metal concentrations ranged from 0.112 (As) to 3.98 (Fe) mg/kg, with Fe > Cd > Ni > Co > Mn > Cr > Zn > Cu > Pb > Hg > As in decreasing order. The variation in the concentration might be due to differences in activities, weather conditions, and sources (anthropogenic or natural).

All the HM concentrations investigated in this study were lower than the average global concentration, except for Hg in all towns and Cd in Woldia town [30]. This incremental increase in Cd and Hg may be due to the emission of Cd and Hg from the discharge of motor oils, grease, fuel burning, pigments, and Ni-Cd batteries in the towns [33]. The previous study in farm land of the same region showed high level of Cd, which might be volcanic activity, the gradual process of erosion and abrasion of rocks are also the main sources for the area [34]. Moreover, the levels of HMs across each sampled town

Table 3. HM concentrations in road dust from Woldia and Kobo towns.

Heavy metals											
Sampling place		Mn	Fe	Ni	Co	Cu	Zn	Cd	Hg	Pb	As	Cr
Woldia	Maximum	2.510	3.170	2.820	2.130	0.880	1.570	7.640	0.130	0.660	0.110	1.480
	Minimum	2.440	3.140	2.750	2.110	0.850	1.550	7.530	0.120	0.630	0.100	1.440
	Mean	2.477	3.157	2.797	2.117	0.863	1.563	7.603	0.127	0.643	0.103	1.463
	SD	0.035	0.015	0.040	0.012	0.015	0.012	0.064	0.006	0.015	0.006	0.021
Kobo	Maximum	2.080	3.870	2.870	2.380	0.810	1.240	0.090	0.130	0.590	0.120	2.050
	Minimum	2.010	3.760	2.800	2.350	0.800	1.230	0.080	0.120	0.570	0.100	1.990
	Mean	2.050	3.800	2.833	2.367	0.807	1.233	0.083	0.127	0.577	0.110	2.017
	SD	0.036	0.061	0.035	0.015	0.006	0.006	0.006	0.006	0.011	0.010	0.031
Reference	Maximum	2.770	5.010	1.840	2.820	0.870	1.150	0.120	0.180	0.620	0.150	1.410
	Minimum	2.700	4.970	1.820	2.790	0.860	1.140	0.110	0.170	0.610	0.130	1.410
	Mean	2.743	4.990	1.830	2.803	0.863	1.147	0.113	0.173	0.613	0.140	1.410
	SD	0.0378	0.020	0.010	0.015	0.006	0.006	0.006	0.006	0.006	0.010	0.0001

SD means standard deviation and RF reference values

were also compared with the reference values, and the results showed that the Zn, Cr and Ni concentrations at all sites and the Pb and Cd concentrations in Woldia were above the reference values. The results of this study are similar to those of a study conducted in Mexico City [9]. However, the level of Cd in similar other studies showed lower than the present study [35,36]. The variation in type, frequency and source of Cd might be resulted such variation. The level of HMs other than Cd studied in this study was also compared with previous similar studies, and the results showed that the majority of HMs in the present study were found to be lower [35,37,38].

The average values of the HMs in each sampling site were compared to the FAO/WHO standards, which showed that all metals except Cd in Woldia were below the standards. Woldia is the district's largest town, where many activities that may cause heavy metal sources are more common and tremendous than those in other towns. The activities include parking and maintaining different tracks and automobiles, generating a large amount of municipal waste along with improper disposal, the availability of many metal-wood workshops and garages throughout the town, and the use of biomass fuels for cooking activities. The availability of many pollution sources might be the reason for the very high cadmium level in Woldia town compared to other sampling sites. More importantly, there are a variety of materials, such as Cd-containing alloys, metal plating, pigments in yellow or brown paint (for colouring plastics, glass, and enamels), and Cd-battery-powered cars along the town's roadside. Therefore, anthropogenic activities might be the primary sources of Cd contamination in the environment. Hence, authorities and stakeholders in towns must monitor the issue because background concentrations are higher [33,36,39,40].

An ANOVA test for the heavy metal concentration showed a significant difference across each sampling site. However, a simple ANOVA test did not show where the significant change occurred. Therefore, to determine where the change can occur, a post hock test was performed, and the results revealed that all metals significantly differed across each sampling site, except for the reference values of Cr and Cu, which did not significantly differ from those in Woldia. Ni and Hg levels did not differ significantly among Woldia and Kobo. The percentage contributions of metals across the sampling towns were as follows: Mn (Woldia > Kobo), Fe (Kobo > Woldia), Ni (Kobo > Woldia), Co (Kobo > Woldia), Cu (Woldia > Kobo), Zn (Woldia > Kobo), Cd (Woldia > Kobo), Hg (Woldia > Kobo), Pb (Woldia > Kobo), As (Kobo > Woldia), and Cr (Kobo >Woldia). The order of the sampling towns may have changed due to variations in the sources, such as the surrounding soil, road paving components (e.g. asphalt and concrete), transport vehicles (originating from wheels, vehicle bodies, brake systems, and exhaust emissions), pollutants from the combustion of fossil fuels, and atmospheric particle sedimentation [16,38].

3.2. Human health risk assessments

3.2.1. Noncancer risk assessment

The health risk of HM pollutants to exposed persons was evaluated using the US EPA health risk model. The calculated exposed doses of the 11 HMs for three exposure routes (hand-to-mouth intake (Ding), respiration intake (D_{inh}), and dermal intake (D_{der})) for children and adults in Woldia and Kobo towns are indicated in Table 4. Despite the Dder values for the adults all exceeding those for the children, the Ding and D_{inh} values

Table 4. The exposure amount in three exposure routes for adults and children at different sampling areas.

Adults						
	Woldia			Kobo		
Type of metal	D_{inh}	D_{der}	D_{ing}	D_{inh}	D_{der}	D_{ing}
Fe	7.33E–10	1.99E–07	4.99E–06	8.83E–10	2.40E–07	6.00E–06
Cu	2.00E–10	5.44E–08	1.36E–06	1.87E–10	5.09E–08	1.27E–06
Mn	5.75E–10	1.56E–07	3.91E–06	4.76E–10	1.29E–07	3.24E–06
Zn	3.63E–10	9.85E–08	2.47E–06	2.86E–10	7.77E–08	1.95E–06
Pb	1.49E–10	4.05E–08	1.02E–06	1.34E–10	3.64E–08	9.12E–07
Cr	3.40E–10	9.22E–08	2.31E–06	4.69E–10	1.27E–07	3.19E–06
Cd	1.77E–09	4.79E–07	1.20E–05	1.93E–11	5.23E–09	1.31E–07
As	2.39E–11	6.49E–09	1.63E–07	2.56E–11	6.93E–09	1.74E–07
Ni	6.50E–10	1.76E–07	4.42E–06	6.58E–10	1.79E–07	4.48E–06
Co	4.92E–10	1.33E–07	3.34E–06	5.50E–10	1.49E–07	3.74E–06
Hg	2.93E–11	7.94E–09	1.99E–07	2.95E–11	8.03E–09	2.01E–07
Children						
Fe	1.13E–09	1.13E–07	4.04E–05	1.36E–09	1.36E–07	4.86E–05
Cu	3.08E–10	3.09E–08	1.10E–05	2.88E–10	2.89E–08	1.03E–05
Mn	8.85E–10	8.87E–08	3.17E–05	7.32E–10	7.34E–08	2.62E–05
Zn	5.58E–10	5.60E–08	2.00E–05	4.40E–10	4.41E–08	1.58E–05
Pb	2.30E–10	2.30E–08	8.22E–06	2.06E–10	2.07E–08	7.38E–06
Cr	5.23E–10	5.24E–08	1.87E–05	7.21E–10	7.22E–08	2.58E–05
Cd	2.72E–09	2.72E–07	9.72E–05	2.97E–11	2.97E–09	1.06E–06
As	3.68E–11	3.69E–09	1.32E–06	3.93E–11	3.94E–09	1.41E–06
Ni	9.99E–10	1.00E–07	3.58E–05	1.01E–09	1.01E–07	3.62E–05
Co	7.56E–10	7.58E–08	2.71E–05	8.46E–10	8.47E–08	3.03E–05
Hg	4.50E–11	4.51E–09	1.61E–06	4.54E–11	4.55E–09	1.62E–06

D_{in} means inhalation dose, D_{der} means dermal dose and D_{ing} means ingestion dose

of Fe, Cu, Mn, Zn, Pb, Cr, Cd, As, Ni, Co, and Hg for the children > those for the adults. Furthermore, the results revealed that among the three exposure methods, the Ding value for children was much greater than the D_{inh} and D_{der} values. The findings of the present work were in line with the work of [41]. The similarity in results may be because children can easily come into contact with urban dust through their hands and mouths.

The overall intake doses by adults regardless of the HMs at Woldia and Kobo through inhalation, dermal contact, and ingestion exposure routes were 4.59E–09, 1.26E–06, 3.12E–05 and 2.84E–09, 7.69E–07, 1.93E–05 mg/kg, respectively. Similar calculations were performed for children, and the values were 8.19E–09, 8.2E–07, and 2.94E–04 and 5.72E–09, 5.73E–07, and 2.05E–04 mg/kg for Woldia and Kobo through inhalation, dermal contact, and ingestion exposure routes, respectively. Furthermore, the overall intake dose, regardless of both exposure route and HM concentration, was also calculated, and the results revealed that the intake levels of adults and children living in Woldia and Kobo were 3.25E–05 and 2.01E–05 and 1.61E–05 and 1.77E–06 mg/kg, respectively. From this result, one can conclude that the inhabitants of Woldia town take in greater amounts of pollutants than residents of Kobo town.

The HQs calculated for the noncarcinogenic risks of Fe, Cu, Mn, Zn, Pb, Cr, Cd, As, Ni, Co, and Hg for adults and children through the three exposure routes were lower than 1. The details of the results for adults and children are given in Table 5. The highest HQs at Kobo and Woldia were Cr (through ingestion) and Cd (through dermal contact), respectively.

The HQ values for inhalation, dermal contact and ingestion were Cd < Fe < Cr < Mn < Co, Zn < Fe < Cu < Co < Ni < As < Pb < Mn < Hg < Cr < Cd and Fe < Zn < Cu < Mn < Co < Pb

Table 5. HQ and HI of each heavy metal for adults and children living in different functional areas (the threshold value of HQ and HI for each metal is 1).

Adults												
Site	HQ	Fe	Cu	Mn	Zn	Pb	Cr	Cd	As	Ni	Co	Hg
Kobo	HQ_{inh}	4.01E−06	4.66E−09	3.33E−05	9.55E−10	3.81E−08	1.64E−05	1.93E−08	8.49E−08	3.19E−08	9.63E−05	3.37E−07
	HQ_{der}	3.42E−06	4.24E−06	7.02E−05	1.30E−06	6.93E−05	0.002543	0.000523	5.64E−05	3.31E−05	9.32E−06	0.000382
	HQ_{ing}	7.15E−06	3.19E−05	6.89E−05	6.49E−06	0.00026	0.001062	0.000131	0.000579	0.000224	0.000187	0.000671
Woldia	HQ_{inh}	3.33E−06	4.99E−09	4.02E−05	1.21E−09	4.24E−08	1.19E−05	1.77E−06	7.95E−08	3.15E−08	8.61E−05	3.35E−07
	HQ_{der}	2.84E−06	4.53E−06	8.49E−05	1.64E−06	7.72E−05	0.0018	0.047923	5.28E−05	3.26E−05	8.34E−06	0.000378
	HQ_{ing}	5.94E−06	3.41E−05	8.33E−05	8.23E−06	0.00029	0.0008	0.012011	0.000542	0.000221	0.000167	0.000663
HI	Kobo	1.46E−05	3.61E−05	0.000172	7.79E−06	0.00033	0.0036	0.000654	0.000636	0.000257	0.000293	0.001054
	Woldia	1.21E−05	3.86E−05	0.000208	9.87E−06	0.00037	0.00263	0.059936	0.000595	0.000254	0.000262	0.001042
Children												
Kobo	HQ_{inh}	6.17E−06	7.17E−09	5.12E−05	1.47E−09	5.86E−08	2.52E−05	2.97E−08	1.31E−07	4.91E−08	0.000148	5.19E−07
	HQ_{der}	1.94E−06	2.41E−06	3.99E−05	7.36E−07	3.93E−05	0.001444	0.000297	3.20E−05	1.88E−05	5.30E−06	0.000216
	HQ_{ing}	5.78E−05	0.000258	0.000558	5.25E−05	0.002108	0.008596	0.001061	0.004688	0.001811	0.001513	0.005412
Woldia	HQ_{inh}	5.12E−06	7.67E−09	6.19E−05	1.86E−09	6.53E−08	1.83E−05	2.72E−06	1.22E−07	4.85E−08	0.000132	5.14E−07
	HQ_{der}	1.61E−06	2.57E−06	4.82E−05	9.33E−07	4.38E−05	0.001047	0.027218	3.00E−05	1.85E−05	4.74E−06	0.000215
	HQ_{ing}	4.80E−05	0.000276	0.000674	6.66E−05	0.002349	0.006235	0.097207	0.00439	0.001788	0.001353	0.00537
HI	Kobo	6.59E−05	0.00026	0.000649	5.33E−05	0.002147	0.010065	0.001358	0.00472	0.00183	0.001667	0.005629
	Woldia	5.48E−05	0.000278	0.000784	6.75E−05	0.002393	0.007301	0.124428	0.00442	0.001807	0.001491	0.005585

< As < Hg < Cr < Cd, respectively. From the patterns, one can conclude that Cd poses a relatively high noncarcinogenic risk compared to other HMs. A comparison of the three exposure routes for children and adults revealed that the HQ values for children were greater than those for adults. In addition, the HQder values of skin exposure for adults were greater than those for children (Table 5).

In addition to HQ values, HI values are also used for estimating the hazard risk of single metal intake in three exposure routes and for determining the synergetic effect of different metals on the exposed person. The findings revealed that the HI values for adult residents at Kobo and Woldia ranged from 7.79E–06 (Zn) to 3.62E–03 (Cr) and from 8.97E–06 (Zn) to 5.99E–03 (Cd), respectively. However, for children, the HI values ranged from 5.33E–05 (Zn) to 1.00E–02 (Cr) and from 5.48E–05 (Fe) to 1.24E–13 (Cd). Furthermore, the HI values for inhabitants of Kobo and Woldia, regardless of the type of HM, were 7.08E–03 and 6.54E–02, respectively. Similarly, 2.84E–02 and 1.49E–01 were found for children in Kobo and Woldia, respectively.

Generally, the overall hazard risk of the investigated HMs, regardless of the sampling town, was also calculated using HI. The findings confirmed that the HI values for the investigated HMs across inhalation, dermal contact, and oral ingestion methods were less than one, implying that adult and child residents were less likely to experience noncariogenic risks. However, children had a greater HI for any of the three exposure routes than did adults. The findings also revealed that the HI values for inhalation < dermal contact < oral ingestion for adults and children. Among the HMs analysed, Cd contributed a significant portion to the total amount of HI for adults and children.

3.2.2. Cancer risk assessment

The lifetime cancer risks of Pb, Cr, Cd, As, Ni and Co for adults and children through inhalation, ingestion, and dermal contact exposure routes were calculated and are given in Table 6. The results showed that the risk of metals through inhalation, dermal contact and ingestion ranged from 4.01E–15 (Pb at Kobo) to 2.10E–12 (Cr at Kobo), 4.26E–12 (Pb at Kobo) to 4.16E–09 (Cd at Woldia) and 1.35E–12 (As at Woldia) to 1.35E–092 (Cr at Kobo), respectively. Similarly, the cancer risk for adults through inhalation, skin contact and oral

Table 6. Lifetime cancer risk values for carcinogenic metals via ingestion, dermal contact and inhalation exposure routes.

Children							
	Exposure route	Pb	Cr	Cd	As	Ni	Co
Woldia	Ing	4.74E–12	5.33E–09	4.16E–09	1.35E–12	2.04E–10	1.80E–09
	Der	4.74E–13	8.25E–13	4.16E–09	1.34E–11	2.04E–10	
	Inh	4.46E–15	1.52E–12	1.19E–12	3.84E–14	5.83E–14	
Kobo	Ing	4.26E–12	7.35E–09	4.54E–11	1.44E–12	2.07E–10	2.01E–09
	Der	4.26E–13	1.14E–12	4.54E–11	1.43E–11	2.07E–10	
	Inh	4.01E–15	2.10E–12	1.3E–14	4.10E–14	5.90E–14	
Adult							
Woldia	Ing	4.69E–12	5.27E–09	4.11E–09	1.33E–12	2.02E–10	1.78E–09
	Der	4.69E–13	8.16E–13	4.11E–09	1.33E–11	2.02E–10	
	Inh	4.41E–15	1.51E–12	1.17E–12	3.80E–14	5.76E–14	
Kobo	Ing	4.21E–12	7.27E–09	4.49E–11	1.42E–12	2.04E–10	1.99E–09
	Der	4.21E–13	1.12E–12	4.49E–11	1.42E–11	2.04E–10	
	Inh	3.96E–15	2.08E–12	1.28E–14	4.06E–14	5.83E–14	

ingestion exposure routes ranged from 3.96E–15 (Pb at Kobo) to 2.08E–12 (Cr at Woldia), 4.21E–13 (Pb at Kobo) to 4.11E–09 (Cd at Woldia) and 1.33E–12 (As at Woldia) to 7.27E–09 (Cr at Kobo), respectively. Generally, all risk values for both children and adults through any of the three exposure routes were lower than the USEPA recommended value range (10E–6 to 10E–4) and the International Commission on Radiation Protection (ICRP) (5.0E–5). Thus, one can conclude that the cancer risk due to these metals is relatively low. In addition, although the computed LCR for each exposure route was less than the US EPA's recommended threshold, the overall trend demonstrated that adults are at lower risk than are children.

The sum of all cancer risk values for Pb, Cr, Ni, Cd, As and Co in all exposure routes for children and adults was also 3.81E–08 and 3.76E–08, respectively, which are also below the recommended values of the US EPA. We also calculated the percent contribution of each metal to the total cancer risk. The findings showed that Cr, Cd and Co accounted for 56%, 23% and 18% of the total for children and adults, respectively, and are the main carcinogenic elements.

3.3. Ecological risk assessment

Different parameters, including geoaccumulation, the pollution index, the ecological risk index, the integrated pollution index, and the risk index, were used to estimate the pollution levels of HMs in the study towns, and the findings are provided in Table 7. The average Igeo values for the heavy metals in Woldia and Kobo towns ranged from −1.245 (Fe) to 5.526 (Cd) and −1.168 (Fe) to 0.046 (Ni), respectively.

Road dust pollution was relatively low in all the sampled towns except Woldia (which is extremely polluted by Cd and moderately polluted by Ni) and Kobo (which is moderately polluted by Ni). The sum effect of all measured heavy metals on the pollution level of the towns was determined using IPE and RI parameters. Thus, the pollution levels of roadside dust from the IPE in the towns of Kobo and Woldia indicated that the towns were at low pollution and extremely polluted levels, respectively. However, as shown in Table 7, the values of IPE in Kobo town are close to one, which reveals that the pollution level for this

Table 7. Ecological risk assessment of different elements in different towns using different pollution index parameters.

	Kobo			Woldia		
Metals	I_{geo}	PI	ERI	I_{geo}	PI	ERI
Mn	−1.004	0.748	7.482	−0.731	0.904	9.04
Fe	−1.168	0.667	-	−1.245	0.633	-
Ni	0.046	1.548	7.74	0.027	1.528	7.642
Co	−0.827	0.845	-	−0.988	0.756	-
Cu	−0.677	0.938	4.692	−0.58	1.003	5.017
Zn	−0.484	1.072	1.072	−0.142	1.359	1.359
Cd	−0.991	0.755	22.636	5.526	69.118	2073
Hg	−1.006	0.747	29.88	−1.017	0.741	29.647
Pb	−0.665	0.946	4.73	−0.509	1.054	5.27
As	−0.933	0.786	7.857	−1.028	0.736	7.357
Cr	−0.068	1.43	7.152	−0.531	1.038	5.188
IPE	-	0.953	-	-	7.17	-
RI	-	-	93.24	-	-	2144

I_{geo} is the geoaccumulation index, PI is the pollution index, ERI is the ecological risk index, IPE is the integrated pollution index and RI is the risk index.

town is close to moderate. Furthermore, the effects of individual metals on pollution levels were evaluated using IP and ERI parameters. The ERIs and IRs for both towns revealed that Cd, Hg, Cr, Ni, Zn and Pb were the most abundant contaminants. Thus, the ERIs for HMs measured in the study site at the Kobo and Woldia sites were classified as indicating low pollution, except for Woldia (which is extremely polluted by Cd). The IP calculations revealed that all towns were classified as having low pollution levels, except Kobo (moderately polluted by Ni, Zn and Cr) and Woldia (moderately polluted by Cr, Ni, Zn and Pb; highly polluted by Cd). The lower level of PI revealed that no significant contribution of from anthropogenic sources [38].

Generally, almost all parameters used for ecological risk analysis showed that the pollution level of Woldia town fluctuated from moderate to extreme levels and that of Kobo fluctuated between low and moderate pollution levels. The moderate and extreme pollution levels that occurred in both towns might be because activities included improper disposal of large domestic waste at the roadside, availability of so many garages along the streets, parking service of a large number of large trucks, relatively high population, and three-wheel Bajajs. Hence, maintaining good personal hygiene, reducing the likelihood of hand-to-mouth behaviour, wearing a face mask, and using proper skin protection measures when walking along the roadside, especially for children who have rapid bone growth and are at greater risk of exposure, are the most recommended techniques for reducing contamination [8]. Moreover, the dynamics of HMs concentration along different seasons and sampling locations were not performed, which needs further study to prevent further increments of HMs, especially Cd. The detailed study is also necessary for giving advice for policymakers and stakeholders.

3.4. Method validation

Validation of the method was carried out using the calibration method and employing matrix spike recovery tests, and the results are presented in Table 8. Thus, a series of standard solutions of 0.01, 0.056, 0.56, 0.84, 1.12 and 1.4 ppm (for Pb, As, Cd and Cr); 0.01, 0.056, 0.128, 0.192, 0.64, 1.28, 1.92 and 2.56 (for Ni, Co and Hg); 0.2, 0.3, 0.4, 0.5, 1, 2, 2.5, and 3 (for Cu, Zn, and Mn); and 0.4, 0.6, 1, 2, 4, and 5 for Fe were prepared for ICP–OES. The results of the calibration curve equation and r correlation coefficients for Pb, As, Zn, Cd, Cu, Ni, Co, Hg, Fe, Mn and Cr were found to be $y = 900.34\times + 4.5374$ with $R^2 = 0.9963$, y =

Table 8. Recovery test for the examined heavy metals.

Type of HMs	Amount before spiked (mg/kg)	Amount spiked (mg/kg)	Amount after spiked (mg/kg)	% Recovery
Mn	2.050	1	3.10	105
Fe	3.800	2	5.87	104
Ni	2.833	1	3.78	94.7
Co	2.367	1	3.41	104
Cu	0.807	1	1.87	106
Zn	1.233	1	2.13	90
Cd	0.083	1	1.12	103
Hg	0.127	1	1.17	104
Pb	0.577	1	1.58	100
As	0.110	1	1.21	110
Cr	2.017	1	3.05	103

3493.8× − 93.737 with $R^2 = 0.9972$, y = 33125× + 102.25 with $R^2 = 0.9999$, y = 39443× − 133.6 with $R^2 = 1$, y = 5234.6× − 65.04 with $R^2 = 0.9967$, y = 9354.2× − 192.78 with $R^2 = 0.9957$, y = 2440.8× + 25.11 with $R^2 = 0.9998$, y = 10578× − 65.881 with $R^2 = 0.9999$, y = 37575× + 147.95 with $R^2 = 1$ and y = 6488.8× − 81.911 with $R^2 = 0.9976$, respectively. Thus, the correlation coefficients for all HMs found between R2 = 0.9957 and R2 = 1 are within the recommended acceptable range.

3.4.1. Recovery test

The method's accuracy was also checked using a standard spiked method, and the findings indicated that the percentage recovery values of the analysed HMs were between 90 (Zn) and 110% (As), which is within the recommended acceptable range.

3.5. Pearson correlation

Examining correlations provides clues on the sources of pollution, distribution, and similarities of HM behaviours [42]. The Pearson correlations for the analysed metals for the selected towns in North Wollo were analysed ($p < 0.01$ and $p < 0.05$), and the findings are presented in Table 9. Mn in combination with Cu, Zn, Cd, and Pb; Fe in combination with Co and Cu; As in combination with Ni; Cr in combination with Co; Cu in combination with Cd and Pb; Zn in combination with Cu and Pb; and Cd in combination with Pb had a significant positive correlation with $r > 0.75$, which indicated that they might have similar or common sources. On the other hand, a significant negative correlation ($r > -0.75$) was detected between Fe and Zn, Cd and Pb; between Mn and Fe, Co and Cr; between Co and Cu, Zn, Cd and Pb; and between Cr and Cu, Cd, Pb and Zn, indicating that these metals come from different sources.

3.6. Hierarchical cluster analysis (HCA) and principal component analysis (PCA)

The two most common techniques used to pinpoint likely HMs sources are principal component analysis and hierarchical clustering [43]. Thus, HCA was carried out for HMs using Euclidean distance as a measure of similarity, and the results are shown in Figure 3. Accordingly, five clusters were found at the Euclidean distance 2 including cluster 1 (contains Mn, Co and Ni), cluster 2 (contains Zn, Cr), cluster 3 (contains Fe only), cluster

Table 9. Pearson's correlation matrix between HM concentrations in road dust samples from Woldia and Kobo towns.

Metal	Mn	Fe	Ni	Co	Cu	Zn	Cd	Hg	Pb	As	Cr
Mn	1										
Fe	−.995**	1									
Ni	−.431	.494	1								
Co	−.980**	.982**	.541	1							
Cu	.964**	−.937**	−.287	−.950**	1						
Zn	.995**	−.994**	−.473	−.993**	.958**	1					
Cd	.992**	−.994**	−.504	−.996**	.951**	.999**	1				
Hg	−.022	.062	.098	−.047	.118	−.007	−.003	1			
Pb	.968**	−.956**	−.449	−.945**	.954**	.954**	.951**	−.101	1		
As	−.419	.440	.809	.487	−.374	−.424	−.445	.158	−.573	1	
Cr	−.978**	.985**	.562	.998**	−.933**	−.993**	−.997**	.000	−.939**	.483	1

** *Correlation is significant at the 0.01 level (2-tailed).*

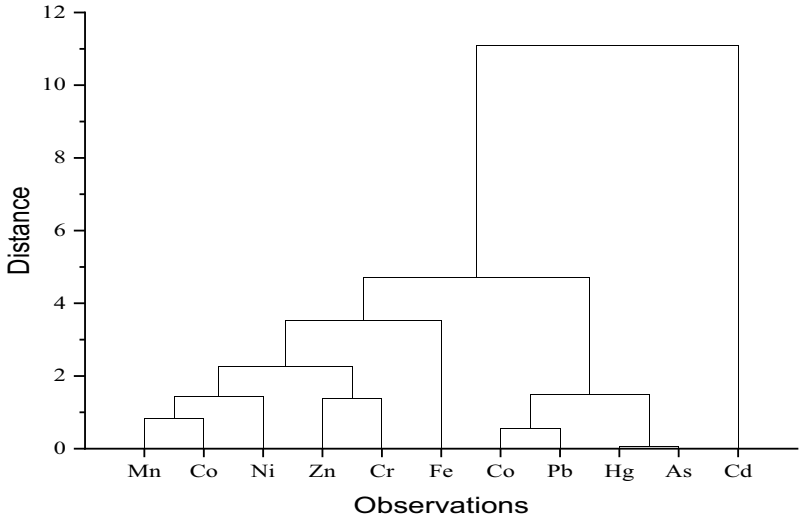

Figure 3. Dendogram obtained from HCA of HMs contents in road dust samples.

4 (contains Co, Pb, Hg and As) and cluster 5 (contains Cd only). Hence, the HMs grouped under the same cluster might have similar or common sources (vehicular, traffic movement, car bodies, Cd-Ni batteries, pigments and tyre wear) [37]. However, Fe and Cd cluster were found in separate cluster that showed they are possible originated from different sources [43].

Moreover, PCA analysis was also performed to investigate the probable sources of HMs under study. 99.99% of the total variance was explained by the first two principal components (PCs). Thus, 56.23% and 43.75% of the total variance was explained by PC1 and PC2, respectively. PC1 characterised by high loading of Cr, Co, Mn, and Ni, while PC2 is by Cd only. The details of the result are depicted in Figure 4. As shown in Figure 4, the obtained result from PCA analysis is quite similar to HCA analysis. Cr, Co, Mn and Ni; As and Hg; Pb, Co and Zn were showed a good correlation, which possibly have similar sources. The Cd metal have loosely correlated with the rest of metals under investigated that might have its own possible sources.

4. Conclusion and recommendations

The average values of the HMs in roadside dust samples from selected towns in North Wollo, Ethiopia, were assessed, and the findings showed that except for Cd in Woldia town, all the HM concentrations were below the limits of the FAO/WHO guidelines. Overall, the order of the mean HMs was Fe > Ni > Mn > Co > Cd > Cr > Zn > Cu > Pb > Hg > As. The spatial distribution of pollution levels in selected towns was assessed using Igeo, IP, IPE, ERI, and RI parameters, in which the distribution level of HMs varied mainly between low and moderate pollution levels, except in Woldia, where Cd was highly polluting.

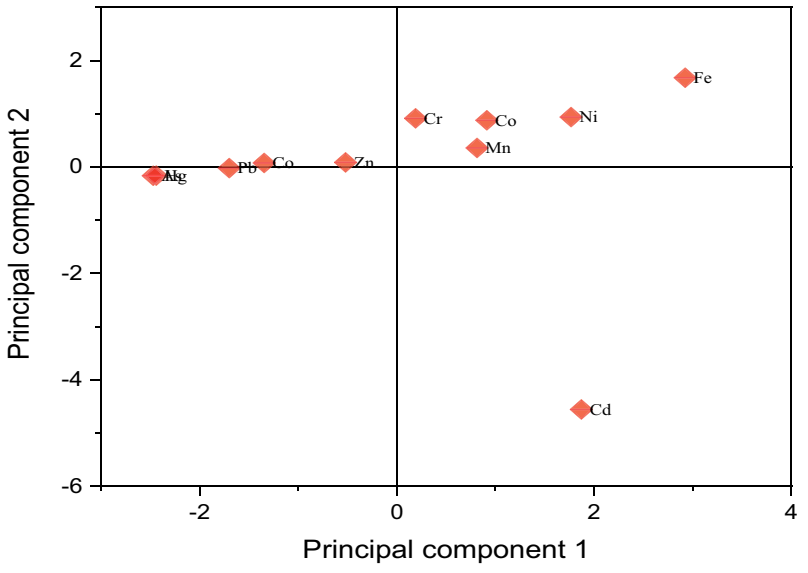

Figure 4. Score plot for PCA analysis.

The health risks associated with oral ingestion, dermal/skin contact and inhalation were assessed using a methodology defined by the USEPA for adults and children. The findings revealed that adults are less exposed than children.

Among the three routes of exposure, hand-mouth intake or oral ingestion was the primary exposure route for both children and adults, followed by skin exposure and then respiratory/inhalation exposure. The HQ values for each exposure route (HQ_{ing}, HQ_{der} and HQ_{inh}) and HI values for each exposure route (HI_{ing}, HI_{der} and HI_{inh}) for children and adults are less than one, showing that there is a low likelihood of noncarcinogenic hazards. The lifetime carcinogenic risk (LCR) for children and adults was also calculated, and the results showed that the total carcinogenic risk of the six metals was 3.81E–08 and 3.76E–08, respectively, which revealed that the probability of developing carcinogenic risk was low. The individual metal contributions to the LCR were calculated, and Cr, Cd and Co accounted for 56%, 23% and 18%, respectively. The Pearson correlation test revealed that Mn, Cu, Zn, Cd and Pb; Fe, Co and Cu; Ni and As; and Co and Cr have similar or common sources.

The analysis of this work needs to include seasonal and special variation analysis within the town, which may underestimate the pollution status of street dust in the towns. Moreover, the number of vehicles, especially three-wheel Bajajs, is rapidly increasing in addition to the rapid growth rate of the town. Thus, a continuous measurement analysis for HMs and other toxic pollutants is necessary to understand the dynamics of HMs pollution, which might helpful to prevent further increments of contaminants and take remedial action for particular pollutants. Moreover, as the growth rate of towns increases, municipal waste increases, and the expansion of industries with high industrial waste and poor disposal mechanisms might aggravate environmental pollution hazards. Thus, selecting

proper municipal waste disposal mechanisms, develop and implement proper rules, garage locations are highly recommended to prevent future pollution hazards in both towns.

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Author contribution

Asamene Embiale Taye designed the study, executed the study, analysed the data and wrote the manuscript. Bhagwan Singh Chandravanshi contributed to the manuscript preparation and proof-reading. All authors approved the final version of the manuscript.

Data availability statement

The data used to support the findings of this study are included in this study.

Ethical responsibilities of authors

All authors have read, understood, and complied as applicable with the statement on 'Ethical responsibilities of Authors', as found in the Instructions for Authors.

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