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Elemental analysis of soils along the South African National Road (N3) — a combined approach including statistics, pollution indicators, and geographic information system (GIS)

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Abstract The South African National Road (N3) in the KwaZulu-Natal province is one of the major transportation routes from the Durban harbor. In this study, metal concentrations in *Bidens pilosa* L., which grows alongside the N3, and soil were determined using inductively coupled plasma — optical emission spectrometry to evaluate the impact of soil quality on the uptake. Furthermore, the distribution of Pb and Cd was mapped using the geographic information system (GIS) approach to identify the potential benefits of spatial data applications in soil studies. Plant concentrations of toxic metals, especially Pb, were high and were linked to high soil concentrations. The target hazard quotients indicated a low risk of adverse effects due to Cd exposure and increased risk due to As and Pb exposure. The carcinogenic risk was high for As and Cd exposure at all sites and Pb at 40% of the sites. Soil quality indicators (geoaccumulation indices and enrichment factors) showed soils to be moderate to heavily contaminated. Principal component analysis indicated different anthropogenic sources of contamination, including vehicular emissions and a combination of industrial, agricultural, and social impacts. Kriging interpolation depicted the spatial diffusion of Cd and Pb concentrations throughout the study area with different hot-spot areas of metal contamination for these two metals. The study demonstrated that the plants growing along national roads are not suitable for human consumption.

Introduction

While development in South Africa and other developing nations is increasing rapidly, the negative impact on the environment is growing exponentially. Heavy metal contamination in the environment is natural and anthropogenic and is a significant concern directly related to industrial development in environmental management (Zaidi et al., 2005). Natural land has been transformed for urban land use at an increasing rate and has resulted in chemical, physical, and biological changes and biodiversity changes (Nogaim et al., 2013). These changes result from an increase in the production of waste and

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effluent from a plethora of land uses and alterations in the natural landscape (Sakinatu & Ashraf, 2017). The soil along roads is a reservoir for pollutants and heavy metals that bioaccumulate and adversely affect biota, which result from vehicular emissions and improper waste disposal (Nogaim et al., 2013). South Africa, a developing nation with a high poverty index, has many motor cars, trucks, taxis, and buses of older models that emit high levels of environmental contaminants. These heavy metals are of great concern as they are introduced into the food chain via plants that absorb them from the soil (Steenland & Boffetta, 2000).

Trace metal analysis of edible vegetation and soil characteristics are excellent environmental indicators of the link between pollution and human impacts (Sahu & Basti, 2021). Pollution of metals, whether essential or non-essential, poses significant risks to living organisms as metals are non-biodegradable and remain in the soil over a long period even if point sources of pollution are removed, which results in higher uptake by living organisms (Mani & Kumar, 2014).

In isiZulu, *imifino* refers to a traditional vegetable. *Bidens pilosa* L. is a leafy green *imifino* that forms part of the staple diet to many people living in rural areas across South Africa. The herb is combined with maize meal to prepare dishes such as *isigwamba*; high poverty levels force communities to eat these inexpensive and readily available forms of nutrition. However, by consuming this herb that grows along roadsides, there is the danger of exposure to toxic chemicals emitted by vehicles. Therefore, this study aimed to assess the impact of vehicle pollution in the South African National Road (N3) between Durban and Hilton. It is one of the major transportation routes from the harbor. The elemental concentrations in the leaves of *Bidens pilosa* that grows along the N3 were investigated as a function of soil quality. Pollution indicators were applied to evaluate the degree of contamination on surface soils by anthropogenic sources, and these sources were examined using principal component analysis. Spatial distribution maps were produced for the two primary toxic heavy metals, Pb and Cd, to determine the potential of the geographic information system (GIS) approach for the identification of elevated concentrations along the N3.

Materials and methods

Description of sampling site and sampling

The study was conducted on the South African National Road (N3) from Durban to Cedara in KwaZulu-Natal. The study area was selected due to high automobile activity from daily commuters and leisure travelers and since it is the main logistic route for trucks from the Durban harbor to Johannesburg, which constitutes about 38% of the traffic. At peak times, traffic volumes exceed 2000 vehicles per hour. The N3 starts in the Central Business District (CBD) of Durban, heading west via a dual carriageway through Westville and Pinetown, through to the Toll Plaza situated at Marianhill, which leads to Cato Ridge, Camperdown, and Pietermaritzburg, after which, is a steep incline up Townhill towards Hilton and Howick, en route to Johannesburg.

Soil samples were obtained from twenty predetermined sampling locations along the N3 (Fig. 1) on the right-hand side of the road, where vegetation is grown as one heads west from Durban to Hilton. The sampling locations were mapped out via GIS to be a proportionate and statistically reduced variation of approximately 5 km apart while considering safe vehicle stopping areas. At each sampling location, three soil samples were collected systematically and directly behind the first sample. The first sampling point was 1 m away from the roadside; the second sampling point was 10 m away from the first point (11 m away from the roadside), and the third sampling point was 10 m away from the second point (21 m away from the roadside). Sixty soil samples were collected in total to compare concentrations relative to distance from the road. Sampling was conducted in April 2015 on a warm day (24 °C) with no rainfall or wind. Table S1 lists the sample locations with environmental factors for the area. Due to the horizon of the soil being the plow layer, which hosts plant roots that take up nutrients from the ground, soil samples were collected from a depth of 0–30 cm by using a hand auger (Loke et al., 2012).

Soil samples were manually homogenized using a plastic spoon after removing extraneous material such as leaves and rocks. The composited soil volume (500 mg) was reduced to 100 mg by coning
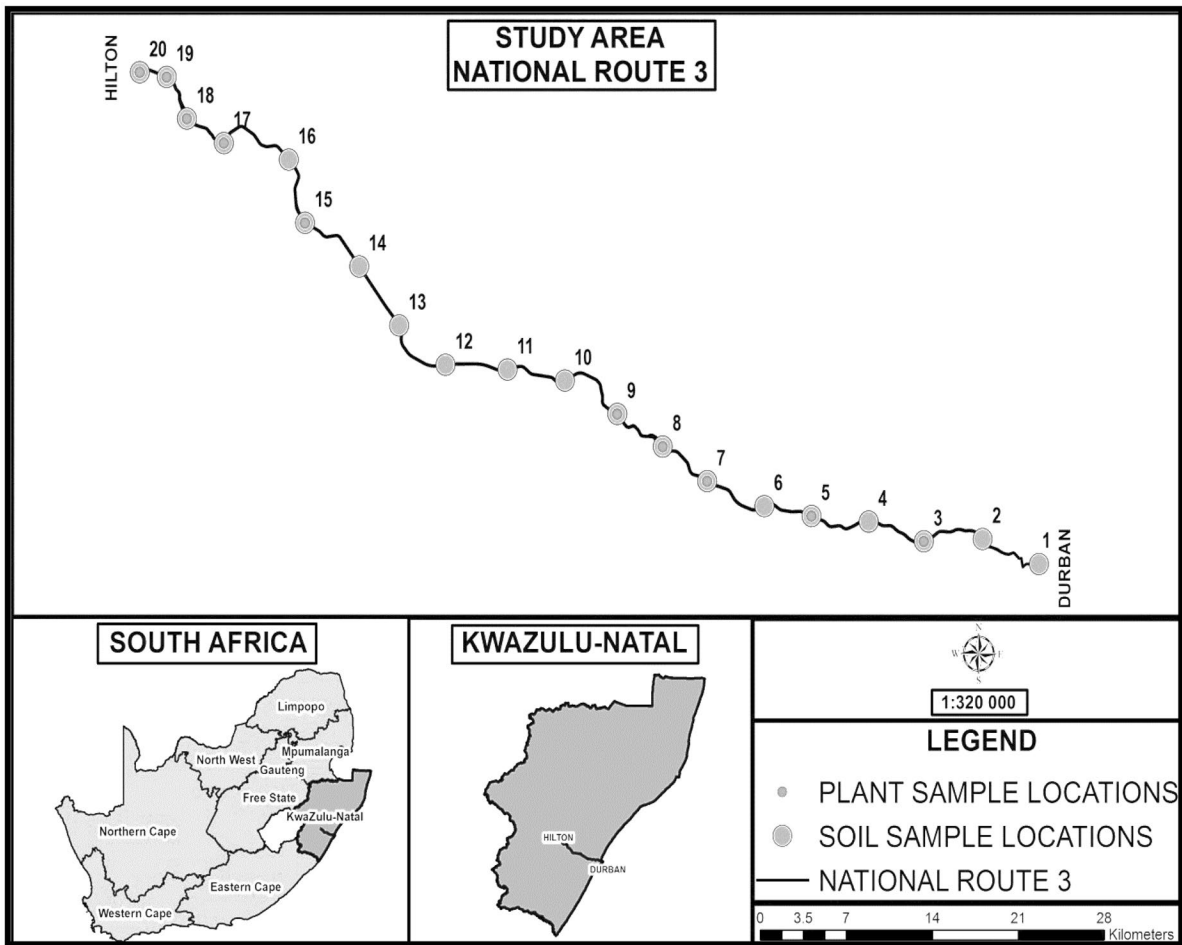
Fig. 1 Study area in relation to country and province

and quartering. Plant samples (*Bidens pilosa*) were collected from ten of the twenty soil sampling sites due to the plant's dispersed growing patterns. These samples were collected from roadside soils approximately 1 m away from the road. All samples (plant and soil) were placed in polyethylene bags and stored in cooler bags for transportation.

Sample preparation

Analytical reagent-grade chemicals were used and were supplied by Merck (Kenilworth, NJ, USA). All glassware was soaked in HNO_3 (3 M) then rinsed in double distilled water before use. Soil samples were air-dried, passed through a 2 mm mesh sieve to obtain the soil fraction, and crushed using a mortar and pestle to reduce the particle size to a powdered form.

Plant leaves (edible portion) were washed with double distilled water to remove extraneous matter, oven-dried at 40 °C, and then ground in a food processor (Russell Hobbs range) to obtain a powder. All samples (plant leaves and soil) were placed in polyethylene bottles and stored in a refrigerator at 4 °C until digestion within a week of collection.

Digestion and elemental analysis of samples

Digestions were performed using the CEM microwave accelerated reaction system (MARS 6, CEM Corporation, USA) according to the method as described by Moodley et al. (2012). All soil and plant (*Bidens pilosa*) samples were analyzed for the essential (Co, Cr, Cu, Fe, Mn, Ni, Se, and Zn) and toxic (As, Cd, Pb) elements by ICP-OES (PerkinElmer,

Optima 5300 Dual View, Billerica, Massachusetts, USA). These metals are considered the most interesting since they are likely contaminants from vehicle emissions (Bushell & Williamson, 1995). Details for digestion and elemental analysis appear in the Supplementary information. Method validation was performed using the CRM, White Clover (BCR-402) (Institute for Reference Materials and Measurement, European Commission, Joint Research Centre, Belgium), and Metals in Soil (D081-540) for soil samples (ERA, A Water Company, Milford, MA, USA). Certified reference materials were prepared and analyzed similarly to samples for method validation. All samples (plant material, soil, and CRMs) were analyzed in triplicate. The experimental and measured values for the CRMs are presented in Table S2 and show them within acceptable limits, thereby validating the method.

Pollution indicators

The degrees of metal pollution in seven grades ranging from uncontaminated to extremely contaminated are assessed using the geoaccumulation index (I_{geo}) according to the equation proposed by Muller (1969):

$$I_{geo} = \log_2\left[\frac{C_n}{1.5\,B_n}\right]$$

C_n is the concentration of the sample element, and B_n is the background/baseline concentration of the same element. The background concentrations for this study were used from a survey conducted by Herselman (2007) and Rudnick (2014). Factor 1.5 is incorporated into the equation to minimize variations in the background value due to lithologic (rock composition) variations (Stoffers et al., 1986).

An alternative method for determining levels of soil contamination is by use of the enrichment factor (EF) according to the following equation proposed by Muller (1969):

$$EF = \left(\frac{\frac{C_x}{C_{ref}}}{\frac{B_x}{B_{ref}}}\right)$$

C_x is the concentration of the soil element, C_{ref} is the concentration of the element in the earth's crust, B_x is the concentration of the reference element in the soil, and B_{ref} is the concentration of reference element in the earth's crust. The background concentrations for this study were used from a survey conducted by Herselman (2007) and Rudnick (2014). Due to Zn concentrations being known in South Africa, Zn is used as the reference element (Herselman et al., 2005; Mendiola et al., 2008).

Health risk assessment

The risk to human health as a result of consuming the plant (*Bidens pilosa*) growing along the N3 was evaluated by calculating the target hazard quotient (THQ) and carcinogenic risk (CR) for the elements in the plant. The THQ was calculated as per the USEPA Region III Risk-Based Concentration Table (USEPA, 2011). THQ provides the non-carcinogenic risk of exposure from the ratio of a determined level of a potentially hazardous metal to its reference dose considered toxic (Song et al., 2009).

$$THQ = [X] \times IR/(B_w \times RfD_0)$$

[X] is the metal concentration in the plant (mg/kg dry weight), IR is the ingestion rate of plant per person (0.033 kg per day) (Sharma et al., 2016), B_W is the average body weight of a South African adult (70 kg), and RfD_0 is the oral reference dose of metals that adults can be exposed to (mg kg^{-1} per day). The RfD_0 values are as follows: As (0.0003), Cd (0.001), Co (0.043), Cr (0.003), Cu (0.04), Fe (0.7), Mn (0.14), Ni (0.02), Pb (0.004), and Zn (0.3) (USEPA, 2011). A THQ < 1 indicates a low risk of adverse effects due to exposure to that element, while a THQ > 1 suggests possible health risks due to exposure to that element.

The CR estimates the likelihood of an individual developing cancer from exposure to a potential carcinogen over a lifetime (Kortei et al., 2020).

$$CR = ([X] \times IR /B_w) \times CPS_0$$

CPS_0 is the oral slope factor of the carcinogen in mg/kg BW per day, 1.5 for As, 0.0085 for Pb, and 6.3 for Cd. A value above 10^{-4} indicates a high probability of CR (Javed & Usmani, 2016).

Statistical analysis

All statistical analyses were done using International Business Machines Statistical Package for the Social

Sciences (IBM SPSS, Version 27, IBM Corporation, Cornell, New York). Principal component analysis (PCA) was used to reduce observed correlated variables to a smaller set of critical, independent variables. Cases used for statistics were based on points with no missing values for any variable used. The Pearson correlation matrix was explored prior to PCA. Correlations were considered to be significant at the 0.05 and 0.01 level (2-tailed). Sampling adequacy was confirmed using Kaiser-Meyer-Olkim (KMO = 0.53) suitability test and Bartlett's test of sphericity ($p < 0.0001$). Factor retention was considered if eigenvalues were > 1. The preferred rotation method was Varimax with Kaiser Normalization.

Geographic information system analysis

The sixty soil sampling points were interpolated; each sample point was analyzed in triplicate, and the mean was obtained and captured. Sample points were split into four subsets due to altitude and slope, which assisted in higher accuracy in interpolation and visually represented the information in smaller subsets. Subsets were broken up as follows: (1) Durban to Marianhill Toll (5–381 m above sea level), (2) Marianhill Toll to Cato Ridge (568–758 m above sea level), (3) Camperdown to Pietermaritzburg (758–625 m above sea level), and (4) Pietermaritzburg to Hilton (820–1061 m above sea level). A geostatistics method called Kriging was adopted to interpolate data due to the type of data sets and the required output. The Empirical Bayesian Kriging Method was used in this study (Krivoruchko & Gribov, 2019). Each subset was interpolated to a 200 m buffer of the N3 for Pb and Cd. Data were assessed for normality via the Shapiro–Wilk test. The experimental semi-variogram was fitted to the model to increase the accuracy in the Kriging process.

Results and discussion

Analysis of the impact of soil concentrations on plant concentrations

The concentrations of essential elements (Co, Cr, Cu, Fe, Mn, Ni, and Zn) in plant leaves and soils from 10 sites along the South African National Road (N3) are presented in Table 1. Plant Co ranged from 0.3 (Westville) to 1.7 mg kg^{-1} (Hilton) with

concentrations at Drummond, Town Bush, Montrose, and Hilton exceeding the maximum permissible limit (MPL) (0.7 mg kg^{-1}) (Department of Water Affairs and Forestry, 2005). Soil Co was less than 18 mg kg^{-1}, which is less than its screening value for rehabilitation in all land uses (300 mg kg^{-1}) (Department of Environmental Affairs, 2012).

Plant Cr across all sites exceed the threshold value of 2.3 mg kg^{-1} (Codex Alimentarius, 2001), and soil Cr was above the MPL for South African soils (80 mg kg^{-1}), with Hilton (168.7 mg kg^{-1}) and Cedara (216.1 mg kg^{-1}) having extremely high concentrations (Water Research Commission, 1997). Accumulation of Cr within the soil is primarily due to industrial waste used in landfilling and untreated sewage exposure (Coetzee et al., 2020). Chromium is also used in corrosive preventative coatings in vehicle manufacture at approximately 10 g per vehicle. Herselman et al. (2005) found the baseline concentration of Cr (80–350 mg kg^{-1}) in South African soils to be higher than the world mean concentrations of other landscapes such as bedrock shale (90 mg kg^{-1}) and sandstone (35 mg kg^{-1}).

Plant Cu was above the MPL (30 mg kg^{-1}) for Cu in foods (Department of Health, 2004). Copper concentrations in soil ranged from 5 to 155 mg kg^{-1}. The MPL for Cu in South African soils was determined to range between 50 and 150 mg kg^{-1} (Herselman et al., 2005). Except for Cedara, soil concentrations were below this MPL. The world mean concentration for Cu in bedrock shale, the study area's predominant bedrock, is 50 mg kg^{-1} (De Vos et al., 2005).

Soils collected from certain sampling locations were also found to contain cadmium, chromium, copper, lead, and zinc at concentrations which exceed the maximum permissible concentrations of 2, 80, 6.6, 6.6, and 46 mg/kg, respectively, for soils used for agriculture in South Africa (WRC, 1997).

Although soil Fe was predominantly higher than plant Fe indicating exclusion, concentrations were above the MPL in vegetables (150 mg kg^{-1}) (WHO, 1996) (Table 1). Iron concentrations in the plant are consistent with previous studies on leafy vegetables in South Africa (Mahlangeni et al., 2016). Areas with high soil Fe have shale as the predominant bedrock within Dwyka and Pietermaritzburg geological formations. De Vos et al. (2005) predicted a 4.7 mg kg^{-1} world mean concentration of Fe in shale bedrock, which indicates Fe enriched soil content.

Table 1 Concentration (in mg kg^{-1}, Mean (SD), $n=3$) of the essential elements (Co, Cr, Cu, Fe, Mn, Ni, and Zn) in plants (dry mass) and soil from 10 sites along the South African National Road (N3)

Site	Co		Cr		Cu		Fe		Mn		Ni		Zn	
	Plant	Soil	Plant	Soil	Plant	Soil	Plant	Soil	Plant	Soil	Plant	Soil	Plant	Soil
1	0.3 (0.03)	3 (0.3)	11 (2)	35 (2)	46 (2)	26 (1.7)	504 (13)	5452 (158)	81 (2)	105 (15)	1.4 (0.2)	5 (1)	186 (4)	29 (8)
2	0.6 (0.1)	1 (0.1)	11 (0.7)	14 (5)	43 (4)	13 (1.1)	548 (12)	2418 (95)	154 (4)	110 (9)	1 (0.3)	1 (0.3)	62 (6)	5 (0.4)
3	1.6 (0.05)	1 (0.1)	14 (1.8)	2 (1)	69 (6)	85 (2.2)	1017 (26)	101 (8)	315 (7)	300 (23)	2.5 (0.1)	1 (0.1)	379 (6)	105 (2.2)
4	0.4 (0.18)	17 (2)	11 (1.7)	5 (2)	53 (3)	5 (0.5)	574 (23)	334 (22)	236 (6)	877 (25)	1.6 (0.2)	3 (0.3)	166 (4)	391 (4.6)
5	1.1 (0.48)	15 (2.2)	15 (1.7)	111 (10)	55 (3)	75 (2.4)	1030 (67)	26,599 (492)	350 (10)	966 (25)	2.7 (0.5)	16 (1.5)	153 (5)	133 (2.6)
6	0.5 (0.03)	17 (2.1)	10 (1)	124 (8)	48 (1)	106 (5)	781 (9)	30,475 (664)	210 (3)	1091 (33)	4.1 (0.6)	24 (1.9)	48 (4)	261 (3.3)
7	1.3 (0.08)	12 (1.6)	13 (1.8)	176 (10)	58 (4)	88 (2)	1104 (20)	31,172 (846)	247 (6)	495 (39)	3.4 (0.1)	24 (2.6)	315 (6)	282 (3.4)
8	1.2 (0.03)	10 (1.1)	9 (1.2)	118 (5)	58 (5)	32 (1.3)	718 (64)	25,477 (538)	187 (6)	503 (34)	1.9 (0.5)	14 (2)	179 (3)	51 (1.5)
9	1.7 (0.11)	17 (1.2)	9 (0.7)	169 (7)	65 (6)	95 (2.1)	644 (10)	47,798 (81)	750 (3)	597 (21)	9.3 (0.2)	21 (2)	200 (4)	99 (2)
10	0.8 (0.03)	16 (1.5)	9 (1.6)	216 (6)	48 (4)	155 (3.1)	510 (16)	46,421 (29)	153 (3)	1680 (19)	3.3 (1.2)	22 (1.4)	128 (6)	178 (2.2)

Sites: 1. Westville, 2. Marianhill, 3. Drummond, 4. Camperdown, 5. Scottsville, 6. Pietermaritzburg, 7. Town Bush, 8. Montrose, 9. Hilton, 10. Cedara

Manganese in plants across all sites was above the MPL for Mn in vegetation (6.6 mg kg^{-1}) (WHO, 1996) (Table 1). Plant Mn was comparable to *Laportea alatipes* from South African soils (260 mg kg^{-1}) (Mahlangeni et al., 2016). Three of the ten sites (Scottsville, Pietermaritzburg, and Cedara) exceeded the Mn screening value (740 mg kg^{-1}) for rehabilitation in all land uses (Department of Environmental Affairs, 2012). High Mn could be due to high trucking activities in the areas and naturally high Mn levels in the soil as Mn is commonly found within the earth's lithosphere.

All plant samples had Ni concentrations above the MPL of 0.18 mg kg^{-1} for Ni in vegetables (Department of Water Affairs and Forestry, 2005). Soil Ni across all sites was below the MPL in South African soils (50 mg kg^{-1}) (Water Research Commission, 1997).

The average concentration of Zn in the plants (181 mg kg^{-1}) exceeded the MPL for Zn in foods (40 mg kg^{-1}) (Department of Health, 2004). The MPL for Zn in South African soils was determined to range between 185 and 200 mg kg^{-1} (Herselman et al., 2005). Camperdown, Pietermaritzburg, and Town Bush had soil Zn concentrations above the upper limit. High bedrock concentrations of Zn in shale, the predominant bedrock in the high concentration areas, are 50–90 mg kg^{-1}, which indicates that Zn concentrations are much higher naturally in these

areas (De Vos et al., 2005). Zinc enhancement may also be connected to the traffic sector since ZnO, used in the rubber of motor vehicle tires, is a significant source of Zn (Hjortenkrans et al., 2006).

Plant As ranged from 0.4 (Hilton) to 2.9 mg kg^{-1} (Marianhill), with most sites exceeding the acceptable exposure limit for As in vegetable (1.0 mg kg^{-1}) (Department of Health, 2004) (Table 2). Target hazard quotients (THQs) ranged from 0.6 to 4.1, suggesting possible health risks from consumption of plants from all sites except Hilton due to As exposure, and the CRs at all sites were above the threshold of 1×10^{-4}, indicating long-term health risks. These areas are near the two major industrial hubs, Pinetown and Mkondeni, in an urban setting, pointing to a possible link between industrial effluent and As in plants.

Plant Cd was above the MPL of 0.2 mg kg^{-1} across all sites (Department of Health, 2004) (Table 2). Plants from Drummond (2.2 mg kg^{-1}) and Camperdown (2.3 mg kg^{-1}) had the highest concentrations, and Pietermaritzburg and Cedara had the lowest (0.5 mg kg^{-1}). THQs ranged from 0.2 to 1.0 indicating low risk due to Cd exposure from consumption, and the CRs at all sites were above 1×10^{-4}, indicating a high risk of developing cancer due to Cd exposure.

Plant Pb ranged from 8 (Marianhill) to 74 mg kg^{-1} (Pietermaritzburg). The MPL for Pb in vegetation is

Table 2 Concentration (in mg kg^{-1}, Mean (SD), $n = 3$) of the toxic metals (As, Cd, and Pb) in plants (dry mass) and soil from 10 sites along the South African National Road (N3) with the target hazard quotient (THQ) and carcinogenic risk (CR)

| | Concentration, mg kg^{-1} | | | | | | | | | | | |
| | As | | | | Cd | | | | Pb | | | |
Site	Plant	Soil	THQ	CR	Plant	Soil	THQ	CR	Plant	Soil	THQ	CR
1	2.2 (0.4)	0.01 (0.01)	3.5	1.56×10^{-3}	1 (0.13)	0.14 (0.01)	0.5	2.97×10^{-3}	20 (3)	181 (5)	2.4	8.01×10^{-5}
2	2.9 (0.3)	0.01 (0.01)	4.6	2.05×10^{-3}	1.5 (0.21)	0.14 (0.01)	0.7	4.46×10^{-3}	8 (1)	10 (0.4)	0.9	3.21×10^{-5}
3	1.8 (0.4)	2.47 (0.1)	2.8	1.27×10^{-3}	2.2 (0.11)	1.04 (0.01)	1.0	6.53×10^{-3}	15 (3)	3 (0.1)	1.8	6.01×10^{-5}
4	2 (0.5)	3.98 (0.2)	3.1	1.41×10^{-3}	2.3 (0.09)	0.87 (0.01)	1.1	6.83×10^{-3}	11 (2)	56 (0.2)	1.3	4.41×10^{-5}
5	2.6 (0.2)	0.28 (0.01)	4.1	1.84×10^{-3}	1 (0.02)	0.11 (0.01)	0.5	2.97×10^{-3}	15 (2)	133 (4)	1.8	6.01×10^{-5}
6	0.9 (0.1)	1.74 (0.1)	1.4	6.36×10^{-4}	0.5 (0.01)	0.15 (0.01)	0.2	1.49×10^{-3}	74 (4)	108 (6)	8.7	2.97×10^{-4}
7	1.9 (0.6)	5.48 (0.3)	3.0	1.34×10^{-3}	1.3 (0.12)	0.10 (0.01)	0.6	3.86×10^{-3}	32 (3)	318 (8)	3.8	1.28×10^{-4}
8	1 (0.02)	2.64 (0.2)	1.6	7.07×10^{-4}	0.7 (0.01)	0.26 (0.01)	0.3	2.08×10^{-3}	21 (2)	29 (1)	2.5	8.42×10^{-5}
9	0.4 (0.5)	9.17 (0.4)	0.6	2.83×10^{-4}	1.3 (0.01)	0.51 (0.01)	0.6	3.86×10^{-3}	31 (2)	126 (4)	3.7	1.24×10^{-4}
10	0.7 (0.01)	8.35 (0.3)	1.1	4.95×10^{-4}	0.5 (0.03)	3.05 (0.02)	0.2	1.49×10^{-3}	25 (3)	96 (3)	2.9	1.00×10^{-4}

Sites: 1. Westville, 2. Marianhill, 3. Drummond, 4. Camperdown, 5. Scottsville, 6. Pietermaritzburg, 7. Town Bush, 8. Montrose, 9. Hilton, 10. Cedara

set at 0.3 mg kg^{-1} (Department of Health, 2004); all plant samples exceeded this limit (Table 2). The plant accumulated high levels of Pb at Drummond with a bioaccumulation factor (BAF) of 5.2. The THQs ranged from 0.9 to 7.9, suggesting possible high risks due to Pb exposure if the plants are consumed. The CRs at Pietermaritzburg, Town Bush, Hilton, and Cedara exceeded the limit of 1×10^{-4}, which signifies the possibility of developing cancer.

Analysis of toxic metals (As, Cd, and Pb) in soil samples, three distances away from the main road

An analysis of soil samples collected from 20 sites across the N3 was conducted. Arsenic, Cd, and Pb were analyzed to determine the relationship between distance away from the main road (1 m, 11 m, and 21 m) and total soil concentrations. The MPL for As in South African soils is 2 mg kg^{-1} (Water Research Council, South Africa, 1997); five of the 20 sites (Drummond, Inchanga, Camperdown, Lynfield Park, and Hilton) had As concentrations above this limit at D1 (1 m away from the roadside) (Fig. 2). The results show average As concentrations from samples closest to the road (D1) to be 1.54 mg kg^{-1}, 11 m away from the road (D2) to be 1.85 mg kg^{-1}, and 21 m away from the road (D3) to be 2.25 mg kg^{-1}. It is clear

from the results that soil As concentrations increase with an increase in distance from the roadside.

Arsenic concentrations spiked at Sites 12 (Camperdown), 14 (Ashburton), 19 (Hilton), and 20 (Cedara). The high concentrations of As at Hilton and Cedara could be due to forestry, plantations, and agricultural practices close to the sample points (Fig. 2). Atafar et al. (2010) found fertilizers used in agricultural practices to cause dramatic spikes in As, Cd, and Pb concentrations in soils. Areas with As toxicity are to be rehabilitated to a concentration of 5.8 mg kg^{-1} in all land uses and 580 mg kg^{-1} to protect ecosystem health (Department of Environmental Affairs, 2012).

The MPL for Pb in South African soils is 56 mg kg^{-1} (Water Research Council, South Africa, 1997); only five of the 20 sites (Durban Harbour, Drummond, Inchanga, Cato-Ridge, and Montrose) had Pb concentrations below this limit 1 m away from the roadside (Fig. 3). Herselman (2007) found high natural total soil concentrations of Pb in South African soils, which would contribute to higher Pb concentrations in soil than other landscapes of the world.

The results show average Pb concentrations from samples closest to the road (D1) to be 118.93 mg kg^{-1}, 11 m away from the road (D2) to be 99.40 mg kg^{-1}, and 21 m away from the road (D3) to be 71.83 mg kg^{-1} (Fig. 3). Typically, soil Pb concentrations decrease
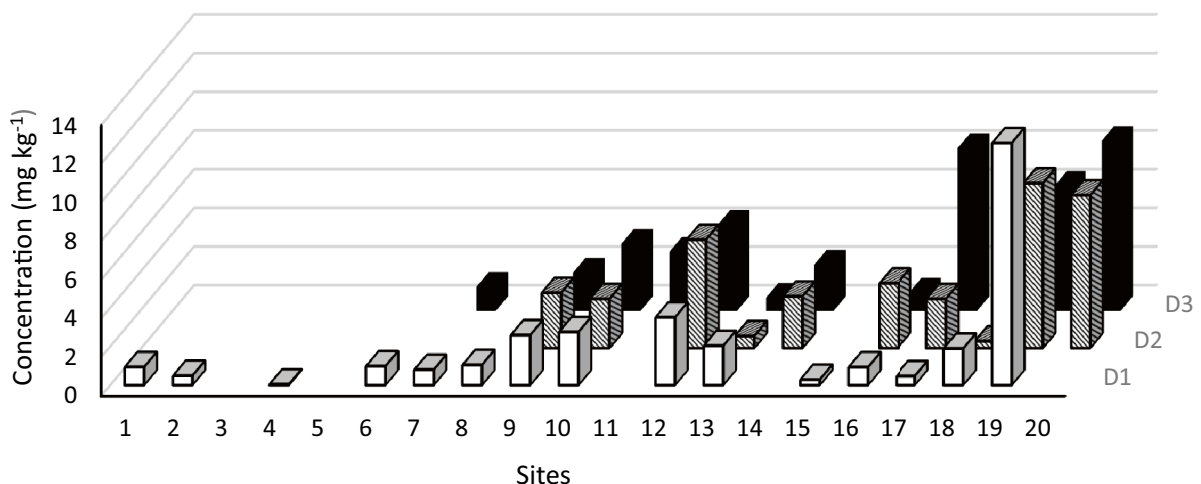

Fig. 2 Concentration of arsenic (As) in soil samples collected along the South African National Road (N3) with distance away from the roadside (D1, 1 m away from the roadside; D2, 10 m away from D1; and D3, 10 m away from D2). Sites: 1. Durban Harbour, 2. Durban CBD, 3. Westville, 4. Pinetown, 5. Marianhill, 6. Marianhill Toll, 7. Hillcrest, 8. Shongweni, 9. Drummond, 10. Inchanga, 11. Cato-Ridge, 12. Camperdown, 13. Lynfield Park, 14. Ashburton, 15. Scottsville, 16. Pietermaritzburg, 17. Town Bush, 18. Montrose, 19. Hilton, 20. Cedara
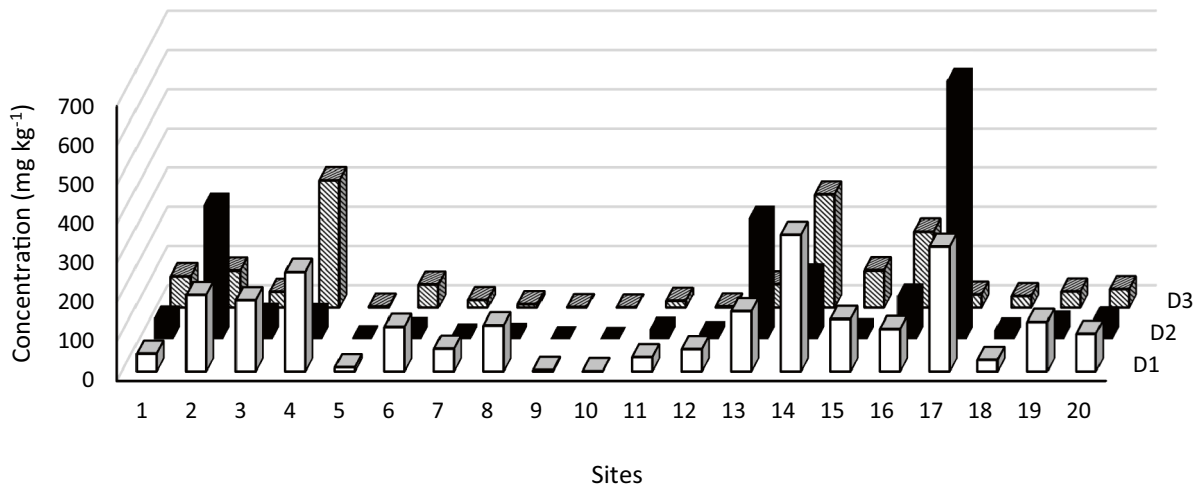
Fig. 3 Concentration of lead (Pb) in soil samples collected along the South African National Road (N3) with distance away from the roadside (D1, 1 m away from the roadside; D2, 10 m away from D1; and D3, 10 m away from D2). Sites: 1. Durban Harbour, 2. Durban CBD, 3. Westville, 4. Pinetown, 5. Marianhill, 6. Marianhill Toll, 7. Hillcrest, 8. Shongweni, 9. Drummond, 10. Inchanga, 11. Cato-Ridge, 12. Camperdown, 13. Lynfield Park, 14. Ashburton, 15. Scottsville, 16. Pietermaritzburg, 17. Town Bush, 18. Montrose, 19. Hilton, 20. Cedara

with an increase in distance from the roadside. High soil Pb concentrations close to the roadside are evident in the Ashburton and Mkondeni areas (Site 14) due to increased industrial activities and truck depots. The emerging rural and smallholding land uses in the area could be at risk as subsistence crops grown in contaminated soils could absorb high Pb levels, resulting in toxic health effects. Lead concentrations spike 11 m away from the roadside at Site 2 (Durban CBD), 13 (Lynfield Park), and 17 (Town Bush). Samples from the Durban CBD were collected closer to the Spaghetti Junction, a high traffic zone with many on- and off-ramps and start-stop conditions resulting in higher vehicular emissions. High Pb concentrations at Lynfield Park are possibly due to fertilizers' leaching from agricultural activities in the vicinity, a rock quarrying mine, and industrial activities very close to the N3. High Pb concentrations at Town Bush or the foot of Town Hill could be due to engine strain on vehicles and trucks to power up a steep hill, excessive braking downhill, and stormwater run-off of herbicides, pesticides, and fertilizers from plantations in the vicinity, which are carried further away from the roadside.

Lead concentrations spike 21 m away from the roadside at Sites 4 (Pinetown) and 16 (Pietermaritzburg) (Fig. 3). Pinetown is a high industrial activity zone with high trucking activities. Pietermaritzburg,

the capital and second-largest city in KwaZulu-Natal, is a regionally important industrial hub and central economic hub of the uMgungundlovu District Municipality. Areas with Pb toxicity are to be rehabilitated to a concentration of 20 mg kg^{-1} in all land uses and 100 mg kg^{-1} to protect ecosystem health (Department of Environmental Affairs, 2012). The findings indicate high Pb concentrations along the N3.

The MPL for Cd in South African soils is 2 mg kg^{-1} (Water Research Council, South Africa, 1997). Except for Sites 6 (Marianhill Toll, 2.8 mg kg^{-1}) and 20 (Cedara, 3.0 mg kg^{-1}), all soil samples were below this limit (Fig. 4). High soil Cd at Marianhill Toll Plaza could be due to traffic density and the impacts thereof. High soil Cd at Cedara could be due to agricultural practices in the area and proximity to a freeway interchange with a moderate increase in traffic (not too high as the region is not densely populated). Areas with Cd toxicity are to be rehabilitated to a concentration of 7.5 mg kg^{-1} in all land uses and 37 mg kg^{-1} to protect ecosystem health (Department of Environmental Affairs, 2012).

Pollution indicators

A geoaccumulation index (I_{geo}) and enrichment factor (EF) were calculated for each of the trace metals
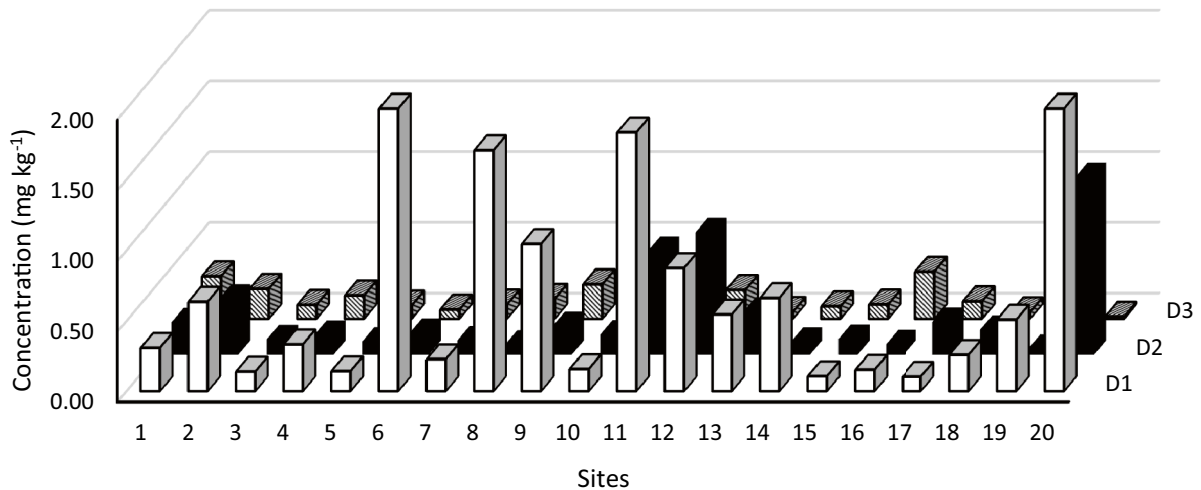
Fig. 4 Concentration of cadmium (Cd) in soil samples collected along the South African National Road (N3) with distance away from roadside (D1, 1 m away from the roadside; D2, 10 m away from D1; and D3, 10 m away from D2). Sites: 1. Durban Harbour, 2. Durban CBD, 3. Westville, 4. Pinetown, 5. Marianhill, 6. Marianhill Toll, 7. Hillcrest, 8. Shongweni, 9. Drummond, 10. Inchanga, 11. Cato-Ridge, 12. Camperdown, 13. Lynfield Park, 14. Ashburton, 15. Scottsville, 16. Pietermaritzburg, 17. Town Bush, 18. Montrose, 19. Hilton, 20. Cedara

(As, Cd, Co, Cr, Cu, Ni, Pb, and Zn) to evaluate for soil contamination (Table 3). The results showed soils to be uncontaminated by As, Co, and Ni. The I_{geo} values and EFs for Cr indicate soil to be uncontaminated with minimal enrichment. The I_{geo} values for Cu indicate soil to be uncontaminated to moderately contaminated, with Sites 2 (Durban CBD) and 6 (Marianhill Toll) being heavily contaminated. For Cu, EFs indicate moderate (Sites 8, 9, 13, 14, 16, 17, 19, and 20) and significant (Sites 2 (Durban CBD) and 6 (Marianhill Toll)) enrichment. For Zn, EFs indicate moderate (Sites 4, 8, 13, 14, 15, 16, 17, and 20) and significant (Sites 2 (Durban CBD) and 12 (Camperdown)) enrichment while I_{geo} values indicate moderate to heavy contamination (Sites 2 (Durban) and 17 (Town Bush)).

I_{geo} values for Pb indicate soil at Sites 14 (Ashburton) and 17 (Town Bush) to be moderate to heavily contaminated. The EFs show Sites 3, 15, 16, 19, and 20 to be moderately enriched and Sites 2 (Durban CBD), 14 (Ashburton), and 17 (Town Bush) to be significantly enhanced. I_{geo} values for Cd indicate Sites 6, 8, 11, 12, and 20 to be moderate to heavily contaminated, and EFs show Sites 2, 8, 9, 12, and 13 to be moderately enriched and Sites 6 (Marianhill Toll), 11 (Cato-Ridge), and 20 (Cedara) to be significantly enriched.

Statistical analysis

Pearson's correlations were used to find relationships between metals in the soil. The results showed the significant correlations between metals in the soil. Some of these were between Mn, Ca, Cu, and Cd ($r > 0.7$, $p < 0.01$), and Fe, Cr, and Ni ($r > 0.8$, $p < 0.01$) suggesting strong associations between these metals in the soil and possible common sources (Table 4). Principal component analysis (PCA) was used to reduce observed correlated variables to a smaller set of critical, independent variables for easier identification of anthropogenic or lithogenic sources. In this study, three principal components were extracted. Component 1, with higher loadings of Ca, Cd, Cu, and Mn explained 28.1% of the total variation (79.8%). Component 2, with high loadings of Cr, Fe, and Ni explained 27.7% of the total variation, while component 3, with high loadings of Co, Mg, Pb, and Zn explained 23.9% of the total variation (Fig. 5).

High Cd levels in the environment are usually attributed to fertilizers in agriculture, and exposure to untreated sewerage near wastewater treatment works and unmaintained sewerage pipelines (Gulten, 2011). Anthropogenic sources of diffuse copper contamination include manure, sewage sludge, and local industrial contamination (Panagos et al., 2018). The

Table 3 Geoaccumulation index (I_{geo}) and enrichment factors (EF) for each element from the twenty sites along the South African National Road (N3)

Sites	As EF	As I_{geo}	Cd EF	Cd I_{geo}	Co EF	Co I_{geo}	Cr EF	Cr I_{geo}	Cu EF	Cu I_{geo}	Ni EF	Ni I_{geo}	Pb EF	Pb I_{geo}	Zn EF	Zn I_{geo}
1	0.1	−3.0	1.9	0.9	0.2	−2.3	1.1	0.1	1.6	0.6	0.2	−2.6	1.9	0.9	1.9	0.9
2	0.1	−3.9	2.8	1.5	0.4	−1.4	1.6	0.6	*11.5*	*3.5*	0.4	−1.2	*6.5*	*2.7*	*5.4*	*2.4*
3	0.0	ND	0.8	−0.4	0.1	−3.3	0.3	−1.6	0.6	−0.8	0.1	−3.5	2.9	1.5	0.4	−1.2
4	0.0	−7.7	1.4	0.5	0.7	−0.6	0.7	−0.6	*6.6*	*2.7*	0.3	−1.9	*6.6*	*2.7*	3.1	1.6
5	0.0	ND	0.7	−0.5	0.0	−4.6	0.1	−2.9	0.3	−1.8	0.0	−5.5	0.2	−2.6	0.1	−3.7
6	0.1	−2.9	*6.3*	*2.6*	0.5	−1.1	1.1	0.1	*14.5*	*3.8*	0.3	−2.0	2.0	1.0	0.2	−2.2
7	0.1	−2.8	1.0	−0.1	0.1	−3.6	0.4	−1.4	0.3	−1.7	0.1	−4.0	0.9	−0.2	0.4	−1.2
8	0.1	−2.8	4.3	*2.1*	0.2	−2.6	0.5	−1.1	2.2	1.1	0.1	−3.0	1.4	0.5	2.1	1.1
9	0.3	−1.5	3.4	1.7	0.0	−4.9	0.0	−6.0	2.0	0.9	0.0	−5.8	0.1	−4.2	1.6	0.6
10	0.4	−1.3	1.0	0.0	0.0	−8.3	0.0	−6.0	1.5	0.6	0.0	−7.4	0.0	−8.7	1.5	0.6
11	0.4	−1.2	*6.2*	*2.6*	0.4	−1.4	0.1	−4.3	1.1	0.1	0.0	−4.8	0.8	−0.3	1.6	0.6
12	0.6	−0.7	4.4	*2.1*	0.6	−0.7	0.0	−4.4	0.1	−3.3	0.1	−4.3	0.8	−0.4	*5.8*	*2.5*
13	0.2	−2.6	2.0	1.0	0.5	−1.1	0.7	−0.4	2.0	1.0	0.2	−2.2	*5.4*	*2.4*	2.5	1.3
14	0.4	−1.5	1.9	0.9	0.4	−1.3	0.7	−0.4	2.7	1.4	0.4	−1.5	*8.3*	*3.0*	2.4	1.3
15	0.0	−4.7	0.7	−0.5	0.6	−0.8	1.0	0.0	1.7	0.8	0.3	−1.9	2.6	1.4	2.0	1.0
16	0.2	−2.0	1.3	0.3	0.6	−0.7	1.2	0.2	2.4	1.3	0.4	−1.3	4.3	*2.1*	3.9	1.9
17	0.5	−0.9	1.0	0.0	0.4	−1.2	1.7	0.7	2.0	1.0	0.4	−1.3	*10.5*	*3.4*	4.2	*2.1*
18	0.4	−1.4	1.1	0.1	0.4	−1.4	1.1	0.1	0.7	−0.5	0.2	−2.1	0.8	−0.3	0.8	−0.4
19	1.3	0.3	1.2	0.3	0.6	−0.7	1.6	0.6	2.2	1.1	0.4	−1.5	2.2	1.1	1.5	0.5
20	1.2	0.2	*9.7*	*3.3*	0.6	−0.8	2.0	1.0	3.5	1.8	0.4	−1.4	2.0	1.0	2.7	1.4

Italicized numbers represent high enrichment or contamination

Sites: 1. Durban Harbour, 2. Durban CBD, 3. Westville, 4. Pinetown, 5. Marianhill, 6. Marianhill Toll, 7. Hillcrest, 8. Shongweni, 9. Drummond, 10. Inchanga, 11. Cato-Ridge, 12. Camperdown, 13. Lynfield Park, 14. Ashburton, 15. Scottsville, 16. Pietermaritzburg, 17. Town Bush, 18. Montrose, 19. Hilton, 20. Cedara

primary anthropogenic sources of environmental Mn include municipal wastewater discharges, sewage sludge, mining and mineral processing, and emissions from alloys, steel, and Fe production (Department of Environmental Affairs, 2010). Therefore, fertilizer runoff from agricultural applications and wastewater discharges including sewage sludge in the region appear to be possible sources for trace metals in the environment due to component 1.

Table 4 Pearson's correlation matrix showing significant relationships between metals in the soil

	Fe	Mg	Mn	Ca	Co	Cr	Ni	Zn	Cu	Pb
Mg	0.5[*]									
Mn	0.2	0.6[*]								
Ca	0.2	0.8[**]	0.8[**]							
Co	0.6[**]	0.8[**]	0.4	0.5[*]						
Cr	0.9[**]	0.6[**]	0.3	0.3	0.6[**]					
Ni	0.8[**]	0.8[**]	0.4	0.5[*]	0.7[**]	0.9[**]				
Zn	0.1	0.5[*]	0.0	0.3	0.5[*]	0.3	0.4			
Cu	0.1	0.4	0.8[**]	0.6[**]	0.3	0.4	0.4	0.1		
Pb	0.3	0.5[*]	0.2	0.5[*]	0.4	0.4	0.7[**]	0.4	0.3	
Cd	0.2	0.2	0.7[**]	0.4	0.2	0.2	0.1	0.0	0.4	−0.1

[*]Correlation is significant at the 0.05 level (2-tailed)

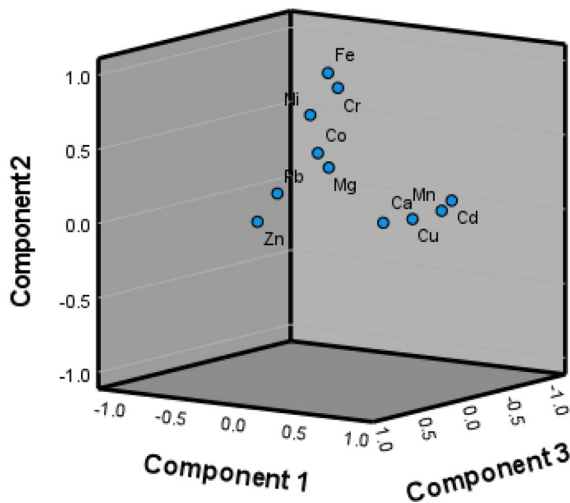[**]Correlation is significant at the 0.01 level (2-tailed)
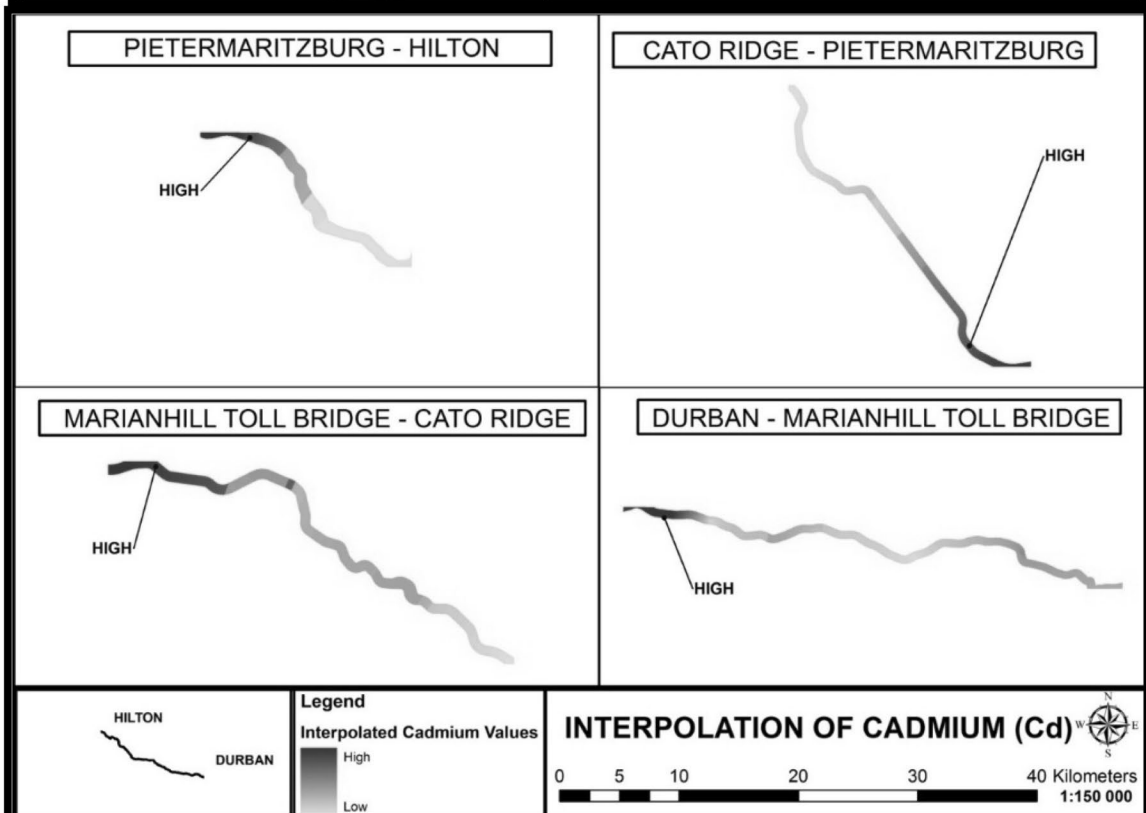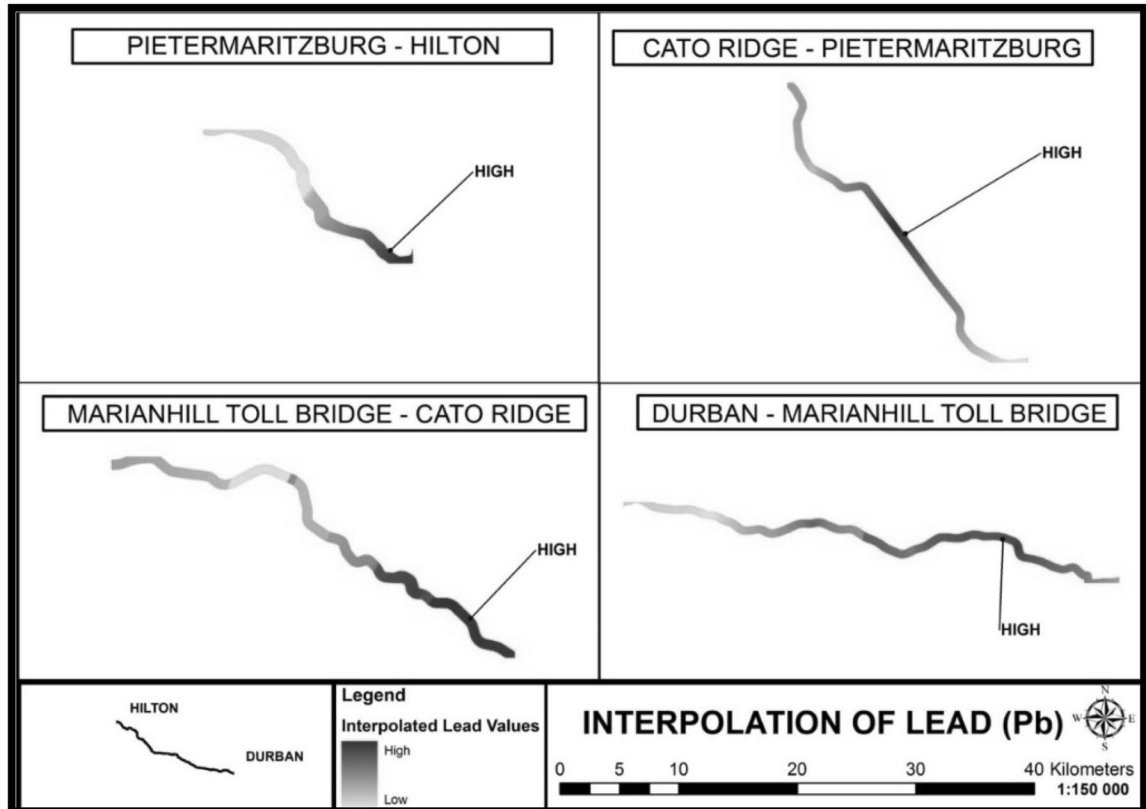
Fig. 5 Component plot in rotated space

the proper functioning of electric cars. Other anthropogenic sources of environmental Co include fast-drying paints and varnishes and burning of fossil fuels (Agency for Toxic Substances & Disease Registry, 2004). Magnesium is preferred to Al for automotive use; when alloyed, Mg has the highest strength-to-weight ratio of all the structural materials (Suman, 2010). Therefore, vehicular emissions and possibly paint appear to be common sources of metals in the environment due to component 3.

Geographic information system analysis

Lead and Cd analyzed for the distance away from the roadside form part of the materials and manufacturing processes of vehicles and contribute to total tailpipe emissions. GIS analysis was therefore conducted on these two toxic trace metals to determine their spatial distributions in the roadside verges.

Although a high natural Pb presence has been determined in South African soils (Herselman, 2007), the trend evident from the interpolated distribution maps for Pb indicates high concentrations in densely populated areas and the immediately surrounding areas (Fig. 6). The high Pb levels in soil demonstrate the link to anthropogenic sources, as was established in the elemental analysis and confirmed by this study's statistical analysis. When Pb concentrations are overlaid onto a geological map, there is an apparent correlation between concentrations in the soil and parent rock lithology (Rezapour et al., 2014). However, further analysis of the composition of parent rock is required to accurately predict a correlation between the findings and the natural content of Pb within the soil, which is related to the trace metal composition of the parent rock.

The interpolated distribution maps demonstrate the link to anthropogenic sources, as was determined in the elemental analysis, with high soil Cd concentrations in areas of high traffic density and sustained agricultural practices (Fig. 6). Superimposition of the maps for Cd and Pb indicates no correlation between these two metals in the soil. The non-superimposition implies different sources of contamination and other deposition characteristics to the natural environment. This GIS finding confirms the observation

Chromium is released into the atmosphere by by-products of fossil fuel combustion and waste incineration, and various industrial processes produce effluents and solid wastes such as chrome-plating, leather-tanning, and dye-manufacturing industries (Coetzee et al., 2020; Tian et al., 2012). Anthropogenic sources of Fe include fossil fuel and biofuel combustion, waste combustion, and steel manufacturing (Yoshida et al., 2018). Extensive distribution of Ni in the environment is primarily due to anthropogenic activities such as the burning of fossil fuels in industrial processes (Department of Environmental Affairs, 2010). Therefore, industrial activities and the burning of fossil fuels in the region appear to be common sources of trace metals in the environment due to component 2.

Lead in the environment results from industrial emissions, vehicle exhaust emissions, and paint, including road-marking paint containing Pb (Bigdeli & Seilsepour, 2008). The breakdown of tires from vehicles introduces Pb into the environment, and Pb is also used in other vehicle manufacturing processes (Giannouli et al., 2007). Elevated Zn concentrations in the environment are run-off from galvanized steel and surfaces painted with zinc-containing paints. At the same time, most of the Zn enhancement may be connected to the traffic sector (Szwalec et al., 2020). The use of ZnO in the rubber of tires is also a significant Zn source in the environment (Hjortenkrans et al., 2006). Cobalt is used in car batteries and is critical to

INTERPOLATION OF LEAD (Pb)

INTERPOLATION OF CADMIUM (Cd)

by statistical analysis. The GIS analysis shows Pb and Cd concentrations to be directly impacted by the area's topography and geopedological processes.

Conclusions

The study results showed plant concentrations of the studied essential and toxic metals to be above acceptable limits. Soil As and Pb were high, with soil Pb exceeding threshold values across all sites. The plant showed potential for phytoremediation of soils as it accumulated Cd and Pb in the leaves. Target hazard quotients indicated adverse effects to health due to As and Pb exposure and carcinogenic risk due to the toxic metals studies, especially As and Cd. Pollution indicators revealed moderate to heavy contamination and significant enrichment in some areas for As, Cu, Pb, and Zn with a higher frequency for Pb enrichment. Statistical analyses indicated common sources for different metals with anthropogenic sources including vehicular emissions, agricultural practices, commercial and industrial activities, and contaminated groundwater. The Kriging interpolation study showed pollution by Cd and Pb to dominate different regions of the N3, as was observed by principal component analysis. Interpolation suggested natural factors such as slope for stormwater run-off and anthropogenic factors, including effluents from industrial areas, agricultural and forestry practices, high traffic density, and vehicular emissions due to engine strain primarily contribute to high Cd and Pb concentrations along the N3. The road transport sector is a crucial source of heavy metal contamination as it is the preferred method of transport by most industries. Therefore, this study provides insight into the impacts of vehicle pollution and commercial and industrial activities in the surrounding environment. The government should be required to rehabilitate contaminated land to acceptable trace metal concentrations, possibly via hyper-accumulator plants such as *Bidens pilosa* and other natural and technically engineered methods. Interventions can be through policy on social and environmental responsibility and may reduce the negative impacts of metal pollution.

Availability of data and materials   All data generated or analyzed during this study are included in this published article.

Declarations

Ethics approval and consent to participate   Not applicable.

Consent for publication   Not applicable.

Competing interests   The authors declare that they have no competing interests.

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