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SPATIOTEMPORAL VARIATION OF SOIL HEAVY METAL CONTAMINATION IN URBAN INTERSECTION AREAS AND THE VICINITY OF A FORMER MILITARY MISSILE BASE IN RASEINIAI DISTRICT, LITHUANIA

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Abstract. The accumulation of heavy metals (HMs) in urban and protected area soils results from multiple anthropogenic emission sources, including vehicular traffic and historical land use. This study aimed at evaluating the levels and risk of HMs contamination in soils around Raseiniai District area, Lithuania. A total of 14 soil composite samples from urban and protected sites were collected at each site in the spring of 2011 and 2018. All investigated sites can be considered potential sources of hazardous pollution, including urban road intersections and the former Bedančiai military missile base, which now belongs to Dubysa regional park. Total concentrations of HMs (Cd, Pb, Cr, Ni, Cu, and Zn) were determined using an atomic flame absorption spectrophotometer. The assessment of HMs contamination in soils and potential ecological threat posed by each HM were conducted following the determination of their concentration levels and multiple contamination and environmental risk indices. Potential ecological risk index (*PERI*) values ranged from 15.2 to 55.4 in 2011 and from 8.6 to 19.8 in 2018 compared all tested sites, indicating a low ecological risk. Cd and Pb were the dominant contributors as to the total *PERI*, expressed as E_r, in both urban (53.9–69.9% and 22.3–13.2%, respectively) and protected areas (56.0–53.7% and 21.2–19.3%) throughout the entire monitoring period.

Keywords: soil heavy metals, spatial and temporal distribution, urban soil, military sites, contamination indices, potential ecological risk assessment.

1. Introduction

Over 20 million hectares of land worldwide and up to 2.5 million sites in Europe are contaminated with heavy metals (HMs), including Cd, Pb, Cr, Ni, Cu, and Zn, at concentrations that may exceed natural geochemical background levels or regulatory limits (Liu et al., 2018; Tóth et al., 2016; Barahouei et al., 2025). Such contamination poses a significant environmental concern due to the persistence, toxicity, and potential bioaccumulation of these elements in soils and ecosystems. Soil HM concentrations are driven by a complex interplay of natural and anthropogenic factors. Specifically, soil Cd, Cr, and Ni levels originate from the natural weathering of parent materials – primarily sedimentary rocks for Cd and ultramafic or mafic rocks for Cr and Ni. These geogenic inputs are supplemented by anthropogenic sources: vehicular emissions (Cd), industrial discharges from the alloy, pigment, and tannery sectors (Ni), and mining or dye manufacturing (Cr) (Khan et al., 2017; El-Naggar et al., 2025; Ao et al., 2022). Similarly, Cu accumulation stems from diverse human activities, including mining, dust deposition, wood treatment, metal scrap, and Cu-based fungicide use (Poggere et al., 2023). Regarding traffic-related pollutants, Pb is primarily introduced through historical leaded gasoline use and warfare activities, while tyre wear is a significant source of Zn due to usage of zinc oxide in tyre vulcanization process (Petrushka et al., 2024; Amin et al., 2025). Consequently, road traffic and industrial operations remain critical contributors to HM inputs in surface soils, particularly within urban and war-affected environments.

This study aimed to investigate the spatiotemporal distribution of HMs by focusing on two contrasting land-use types in the Raseiniai district, Lithuania: urban crossroads and a former military missile base. These

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areas were selected to represent intensive traffic activity and legacy military land use, respectively, enabling a comparative evaluation of diverse anthropogenic impacts on soil quality. Additionally, key factors contributing to HM pollution were identified.

2. Materials and methods

2.1. Study areas and sampling

In spring 2011 and 2018, a total of 14 composite topsoil samples were collected at depth of 0–20 cm from seven sampling locations using envelope principle and a stainless steel shovel. One composite soil sample was collected from five subsamples collected from the center and from four corners of a 1 m^2 grid. Two sampling sites (S1 and S2) were located near urban street intersections in Raseiniai city, while the remaining five sites (S3–S7) were situated near former military missile Bedančiai base within the Dubysa Regional Park (Figure 1 and Table 1).

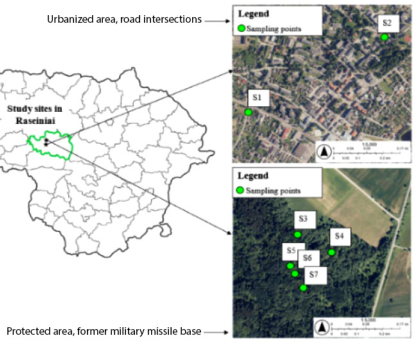

Figure 1. Study areas and soil sampling locations in Raseiniai District

Table 1. Locations and description of sampling sites in the area of Raseiniai District

Site	Coordinates	Description
S1	55° 22′ 51″ N, 23° 06′ 33″ E	Location in the vicinity of an urban road intersection within a residential area
S2	55° 23′ 4.0″ N, 23° 7′ 13.3″ E	Location in the vicinity of an urban road intersection within a public service area
S3	55° 27′ 31.9″ N, 23° 8′ 29.6″ E	Locations in the vicinity of a former military missile base
S4	55° 27′ 29.6″ N, 23° 8′ 37.4″ E	
S5	55° 27′ 27.9″ N, 23° 8′ 28.1″ E	
S6	55° 27′ 26.9″ N, 23° 8′ 29.1″ E	
S7	55° 27′ 25.0″ N, 23° 8′ 31.0″ E	

According to the Lithuanian Soil Classification and the national soil map, sampling sites S1 and S3–S4 were located on *Luvisols* (loam), site S2 on an *Albeluvisol* (sandy loam), while sites S5–S7 were situated in former military missile base areas classified as "other soils", representing anthropogenically transformed soil material. Data regarding soil types at specific locations were retrieved from Geoportal.lt website.

Following collection, samples were kept in hermetically sealed polyethylene bags until chemical analysis. The samples were air-dried under laboratory conditions and passed through a 2 mm sieve.

2.2. Analytical methods

The concentrations of HMs (in mg/L), including Cd, Cr, Pb, Cu, Ni, and Zn, were determined by flame atomic absorption spectrophotometry (Buck Scientific 210VGP) using specific lamps for each element. Heavy metal extraction from soil into solution was performed according to the standard method (International Organization for Standardization, 1998). Microwave-assisted acid digestion was performed using aqua regia at 180 °C (Tóth et al., 2016; Angelopoulou et al., 2017).

2.3. Contamination and ecological risk assessment methods

Assessment of HMs pollution in urban and protected soils of Raseiniai District was investigated by several indices, including geoaccumulation index (I_{geo}), contamination factor (C_f), degree of contamination (C_{deg}), as well as individual and overall potential ecological risk indices (E_r and *PERI*, respectively). The I_{geo} is commonly applied to assess metal accumulation in sediments, when their concentrations are above background or baseline concentration (Kusin et al., 2017). The 1.5 factor in Eq. (1) corrects for lithogenic-related background variability (Santos et al., 2020; Saher & Siddiqui, 2016). For the calculation of the I_{geo} and C_f, global geochemical background concentrations (C_b) were adopted from average shale values reported by Turekian and Wedepohl (1961). The *PERI* was calculated as the sum of all E_r values for HMs in soil, using previously determined C_f values and metal toxicity factors (T_r). The T_r values, which are Cr = 2, Ni = Cu = Pb = 5, Zn = 1, Cd = 30, were adopted from Yu et al. (2023). All Equations ((1)–(5)), used for the calculations of aforementioned indices, can be seen below:

$$I_{geo} = \log_2 \frac{C_{sample}}{1.5} \times C_b; \quad (1)$$

$$C_f = \frac{C_{sample}}{C_b}; \quad (2)$$

$$C_{\deg} = \sum C_i^n; \quad (3)$$

$$E_r = T_r \times C_f; \quad (4)$$

$$PERI = \sum E_r^i, \quad (5)$$

where C_{sample} – measured concentration of the test metal in soil (mg/kg), C_b – geochemical background concentration of the test metal (mg/kg). All categories and classes of indices shown below in Table 2.

Table 2. Categories of contamination and ecotoxicological indices

Abbr.	Contamination/ecological risk intensity (class)	Ref.
I_{geo}	$I_{geo} \le 0$, uncontaminated (0) $0 < I_{geo} < 1$, uncontaminated to moderately contaminated (1) $1 < I_{geo} < 2$, moderately contaminated (2) $2 < I_{geo} < 3$, moderately to heavily contaminated (3) $3 < I_{geo} < 4$, heavily contaminated (4) $4 < I_{geo} < 5$, heavily to extremely contaminated (5) $5 < I_{geo} < 6$ extremely contaminated (6)	Santos et al. (2020), Skorbiłowicz et al. (2023)
C_f	$C_f < 1$, low contamination $1 \le C_f < 3$ moderate contamination $3 \le C_f < 6$ considerable contamination $C_f \ge 6$ very high contamination	Skorbiłowicz et al. (2023)
C_{deg}	$C_{deg} < 8$, low degree of contamination $8 < C_{deg} < 16$, moderate degree of contamination $16 < C_{deg} < 32$, considerable degree of contamination $C_{deg} > 32$, very high degree of contamination	Atiemo et al. (2011), Loska et al. (2004)
E_r	$E_r < 40$ low ecological risk $40 < E_r < 80$ moderate ecological risk $80 < E_r < 160$ considerable ecological risk $160 < E_r < 320$ high ecological risk $E_r > 320$ very serious ecological risk	Amin et al. (2025)
PERI	$PERI \le 150$, low ecological risk $150 < PERI \le 300$ moderate ecological risk $300 < PERI \le 600$ considerable ecological risk $PERI > 600$, very high ecological risk	Askari et al. (2020), Fatoba et al. (2016)

Notes: I_{geo} – geoaccumulation index; C_f – contamination factor; C_{deg} – degree of contamination; E_r – potential ecological risk factor; *PERI* – potential ecological risk index.

3. Results and discussion

Negative I_{geo} values for Cr, Ni, Cu, and Cd indicated the absence of contamination by these metals in soils from urban (S1–S2) and former military missile base (S3–S7) areas during 2011 and 2018, suggesting that these sites were not polluted by these HMs (Figure 2). Two sites (S1 and S7) presented moderate or heavy contamination with two HMs in 2011. S1 ranged from moderately to heavily contaminated with Zn (I_{geo} = 2.02) and from uncontaminated to moderately contaminated with Pb (I_{geo} = 0.62). This site belonged to third and first classes of the hazard category of this indicator according to Zn and Pb values, respectively. Meanwhile, S7 was moderately contaminated with Pb (I_{geo} = 1.07) and this site can be attributed to second class. Although the use of leaded gasoline in Lithuania ended in 2001, soils can retain Pb contamination for many years (Chen et al., 2010).

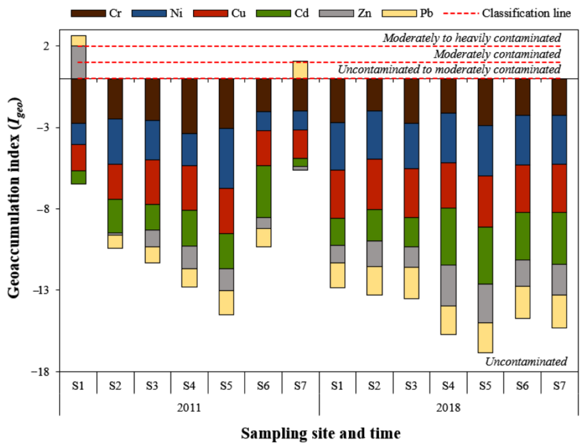

Figure 2. Spatiotemporal variations in I_{geo} of heavy metals (HMs) in urban (S1–S2) and former military missile base (S3–S7) soils during two monitoring years (2011 and 2018)

The presence of Pb in soil at the road intersection (S1) in 2011 could be attributed to the previous deposition of Pb in roadside environments resulting from the use of leaded gasoline. Earlier studies have likewise focused on the distribution of Pb in roadside soils, reflecting its widespread historical use as a gasoline additive (Akbar et al., 2006; Wheeler & Rolfe, 1979). Another study reported from moderate to strong Zn contamination in central and eastern parts of Lublin, Poland (Zgłobicki et al., 2025). Zinc on road surfaces primarily originates from tyre wear and safety fence corrosion, occurring mainly in the finest solid fractions of road runoff and drift (Blok, 2005).

The C_f indicates the degree of HM pollution in sediments by comparing measured metal concentrations with their pre-industrial background levels (Siddiqui & Saher, 2021). The highest C_f value for Zn (6.8), which denotes a very high contamination, was found at site S1, which receives a huge Zn discharge due to abrasion of heavy-duty vehicles (HDVs) tires (Figure 3a). The C_f value for Pb (2.3) at site S1 showed a moderate contamination. Second highest C_f value was found for Pb (3.15) at site S7, which denotes considerable contamination. The increase in Pb levels at site S7 can be linked to

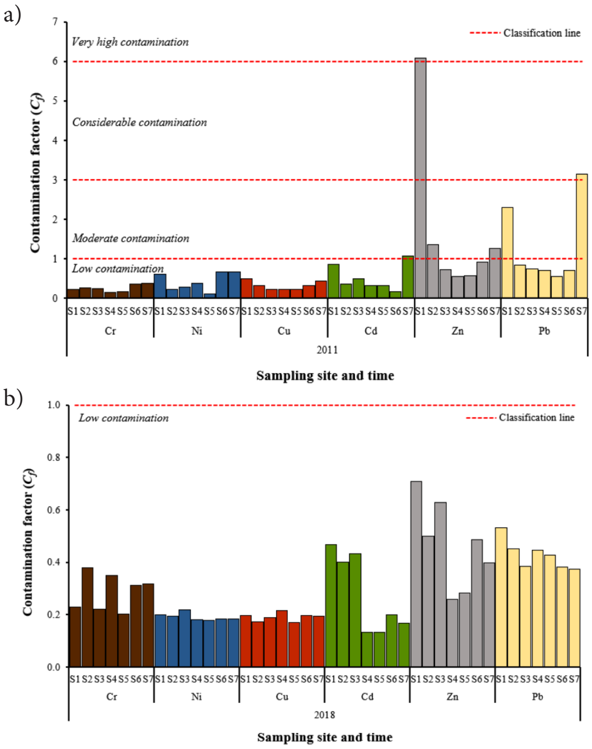

Figure 3. Spatiotemporal variations in C_f of HMs in S1–S2 and S3–S7 soils during two monitoring years: a – 2011, and b – 2018

historically intensive HDV traffic in missile base areas, as this sampling location was closest to the center of the former military missile base (approx. 66 m; Figure 4). Moderate contamination in 2011 was recorded for Pb at site S1 (2.3), Zn at sites S2 (1.36) and S7 (1.27), and Cd at site S7 (1.07). Cr, Ni, Cu and Cd (except site S7) at all sites had low contamination.

Meanwhile, in 2018, contamination by all HMs at all investigated sites was low (Figure 3b). A moderate increase in Cd concentrations near site S7 in 2011 could be associated with atmospheric deposition, which contributes to inputs of this metal into the environment (Shen et al., 2017).

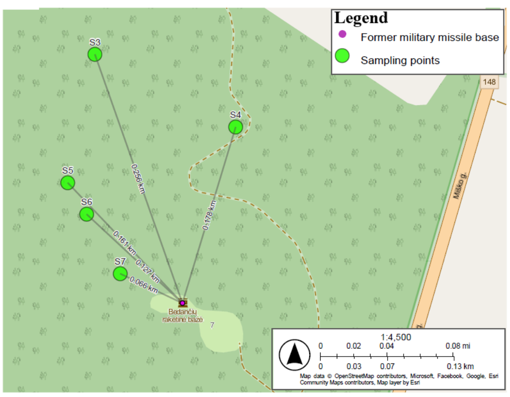

Figure 4. Spatial distribution of soil sampling sites (S3–S7) and straight-line distances to the former military missile base

The C_{deg} indicates integrated pollution degree of sediments (Suresh et al., 2012). According to the C_{deg}, among all sites, only site S1 showed a moderate level of pollution (10.6) in 2011 (Figure 5). According to the traffic intensity map of Lithuanian roads (*ArcGIS.com*), at the intersection where soil samples from site S1 were collected, the total daily traffic volume on road No. 3512 reaches 1,291 vehicles per day. This traffic volume falls into the moderate traffic intensity category and may result in increased HMs emissions. In 2018, all sites exhibited a low contamination level, with C_{deg} values below 8. The decrease in HMs contamination at site S1 over time could largely be attributed to reductions in Zn and Pb concentrations, which decreased by factors of 8.6 and 4.3, respectively, when comparing their concentrations in roadside soil at the road intersection in 2011 and 2018. Since HDV traffic has a strong influence on roadside HM contamination, especially Zn and Pb pollution through tire wear, brake abrasion, and road dust resuspension, it is likely that the observed decrease in 2018 may be partly attributed to the local fees introduced in 2015 for HDVs on local roads in the Raseiniai District (Raseiniai District Municipality Council, 2015). According to available traffic data (*map.eismoinfo.lt*), the average annual daily traffic intensity (AADTI) of HDVs on road No. 3512, which intersects the studied junction where the composite sample from site S1 was collected, decreased by nearly twofold when comparing 2015 and 2021. Compared with the overall AADTI, the proportion of HDVs, including trucks and articulated vehicles, declined from 34% in 2015 to 17.5% in 2021. This temporal shift in traffic composition provides important context for the observed changes in Zn and Pb contamination levels in roadside soils, suggesting a reduced contribution of traffic-related HM inputs over time. The temporal alignment between policy implementation and observed contamination decline suggests that traffic management measures were an important contributing factor.

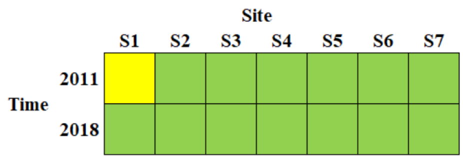

Figure 5. A heatmap illustrating the spatiotemporal variation of the contamination degree index (C_{deg}) of HMs. The yellow color denotes a moderate degree of contamination, while the green color indicates a low degree of contamination

Based on the results of the potential ecological risk index (E_r) for individual HM, all HMs exhibited a low potential ecological risk at all sampling sites during both study periods due to E_r < 40 (Table 3). Among all HMs, Cd exhibited the highest ecological risk in both urban (S1–S2) and former military missile base (S3–S7) soils across all sites and study years. In 2011,

Table 3. Potential ecological risk factors (E_r) for single HM of soil and total potential ecological risk index (*PERI*) in 2011 and 2018

Time	2011							2018						
Sampling site	E_r						PERI	E_r						PERI
	Cr	Ni	Cu	Cd	Zn	Pb		Cr	Ni	Cu	Cd	Zn	Pb	
S1	0.44	3.01	2.44	26	6.08	11.5	49.5	0.46	1.00	0.98	14	0.71	2.66	19.8
S2	0.53	1.10	1.67	11	1.36	4.3	19.9	0.76	0.97	0.86	12	0.50	2.26	17.3
S3	0.51	1.40	1.11	15	0.74	3.8	22.5	0.44	1.10	0.95	13	0.63	1.93	18.0
S4	0.29	1.91	1.11	10	0.56	3.5	17.4	0.70	0.90	1.08	4	0.26	2.23	9.2
S5	0.36	0.59	1.11	10	0.58	2.8	15.4	0.41	0.89	0.84	4	0.28	2.14	8.6
S6	0.73	3.38	1.67	5	0.92	3.5	15.2	0.62	0.92	0.98	6	0.49	1.91	10.9
S7	0.76	3.38	2.22	32	1.27	15.8	55.4	0.64	0.91	0.97	5	0.40	1.88	9.8

sites S1–S2 showed the sequence Cd > Pb > Zn > Ni/Cu > Cr, whereas sites S3–S7 followed Cd > Pb > Ni > Cu > Zn > Cr. In 2018, the sequence shifted to Cd > Pb > Ni > Cu > Cr/Zn in urban soils and to Cd > Pb > Cu > Ni > Cr > Zn in former military missile base soils. Calculated PERI values ranged from 55.4 (site S7) to 15.2 (site S6) in 2011 and from 19.8 (site S1) to 8.6 (site S5) in 2018. Higher PERI values were observed at site S1 due to highest Zn and Pb contents (Figure 6).

Contribution from each metal to total *PERI* is presented in Figure 7. It can be seen that Cd constituted the highest proportion of E_r for individual element to total *PERI* during both monitoring years, while contributions of Pb was second most important at all locations. Cd and Pb posed the greatest ecological risk due to their high toxicity coefficients, particularly in the case of Cd, despite its concentrations being much lower than those of the other HMs. Similar results were reported in another study assessing the potential ecological risk of HM contamination in estuarine surface water sediments, where both metals also exhibited the highest ecological risk due to their high toxicity coefficients (Lei et al., 2016).

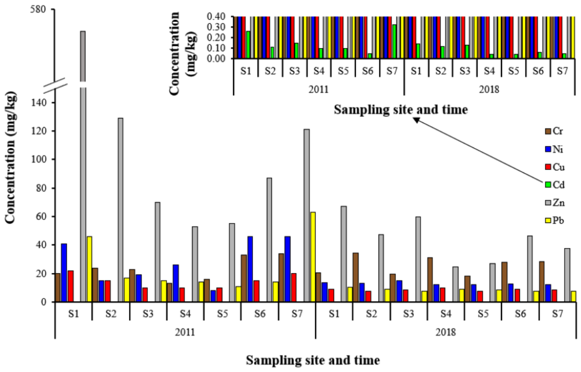

Figure 6. Average concentrations of HMs in the sampling sites during monitoring years of 2011 and 2018

a) b)

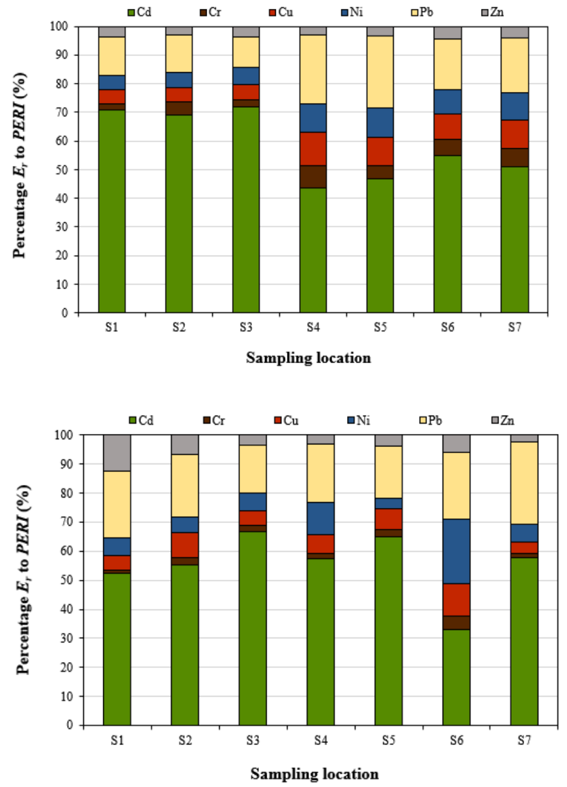

Figure 7. E_r of individual HM percentage to total *PERI* in S1–S2 and S3–S7 soils during two monitoring years: a – 2011; b – 2018

4. Conclusions

1. This study evaluated the spatial and temporal variation of heavy metal (Cr, Ni, Cu, Cd, Zn, and Pb) contamination in soils collected from urban (S1–S2) and former military missile base (S3–S7) areas during 2011 and 2018 using multiple contamination and ecological risk indices. Concentrations of all HMs were lower in 2018 than in 2011, indicating a general decline in soil contamination over time.
2. In 2011, the highest concentrations of Zn and Pb were recorded at sites S1 (urban intersection) and

S7 (near the former military base). By 2018, these concentrations had significantly decreased.

3. Based on Geoaccumulation Index (I_{geo}) and Contamination Factor (C_{deg}) assessments, most sites were classified as uncontaminated or low contaminated during both study years. A "moderate contamination" level (C_{deg}) was only identified at site S1 in 2011. Higher contamination observed at site S1 can be attributed to its location at a major road intersection in the central part of Raseiniai city, where intensive traffic activity likely contributes to increased HM inputs.
4. Site S7, which is situated closest to the former military missile base (approximately 66 m from the base center), showed higher values of certain indices in 2011 but exhibited a clear reduction by 2018, suggesting gradual natural attenuation or reduced anthropogenic influence over time.
5. The potential ecological risk index remained within the low-risk category for all sites and years, though Cd provided the highest individual contribution to the total risk due to the highest toxicity factor (30) among all studied metals.
6. The findings confirm a positive temporal trend in soil quality improvement. However, continuous monitoring, particularly at anthropogenically impacted sites, remains essential to ensure long-term environmental safety.

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