



# Ecological risk assessment and source apportionment of heavy metals contamination: an appraisal based on the Tellus soil survey

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**Abstract** It is imperative to comprehend the level and spatial distribution of soil pollution with heavy metals to find sustainable management approaches for affected soils. Selected heavy metals (Mn, Zn, Pb, Cu, Cr, Ni, As, Co, and Cd) and physiochemical parameters were appraised for 620 samples from industrial, agricultural and urban sites in Northern Ireland using the Tellus database. The findings of this study showed that among the analyzed heavy metals, Mn content was the highest and Cd content the lowest. Pearson's correlation analysis revealed that heavy metals were highly correlated with each other, signifying similar

sources for the heavy metals. Mixed factors (anthropogenic and lithogenic) were responsible for the contribution of heavy metals as revealed by multivariate statistical analysis. The results of contamination factor and enrichment factor analyses suggest that As, Cd, and Pb showed very high risk for pollution in the study area. The geoaccumulation index revealed that with the exception of Cd, all analyzed heavy metals showed severe accumulation in the soils. The potential and modified ecological risk indices inferred that Cd, As, and Pb represented ecological threats in the soils of Northern Ireland. The findings of this study will aid in forming approaches to decrease the risks associated with heavy metals in industrial, urban and agricultural soils, and help create guidelines to protect the environment from long-term accumulation of heavy metals.

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## Introduction

Soils are the main sink for accumulation of heavy metals in the environment through anthropogenic practices. Many heavy metals, unlike organic contaminants, do not sustain microbial or chemical

oxidation, so their overall accumulation in soils persists for a long time after their release (Wuana and Okieimen 2011). It is well known that heavy metals are hazardous to human health and the environment (Liu et al. 2009; Brevik et al. 2020). Elevated heavy metals in the soil are of great concern because of their tendency to bioaccumulate and their non-biodegradable properties, leading to long-term exposure for plants, microorganisms, and livestock, and their possible harm to both humans and animals via the food chain (Liu et al. 2009; Olgun and Atar 2011; Colaka et al. 2011; Rezapour et al. 2014). This disrupts the ecological soil balance, which in turn can affect field productivity (Cao et al. 2010), reduce food quality (safety and marketed) by means of phytotoxicity, reduce land usability for agricultural production which causes food insecurity, and pose problems with land tenure (Wuana and Okieimen 2011). Heavy metal exposure can occur through direct ingestion or interaction with polluted soil, by passing the metals up the food chain (soil–plant–human), and through the use of polluted water (Wuana and Okieimen 2011; Steffan et al. 2018; Elbehiry et al. 2020).

Heavy metals can be released into the environment from both natural and anthropogenic sources (Jung 2008; Trujillo-González et al. 2017). Emissions from rapidly expanding industrial sites, mining waste, the disposing of waste, paints, animal manures, sewage, pesticides, fertilizers, use of waste water, coal combustion residues, and the spillage of petrochemicals can contaminate soil with heavy metals (Wuana and Okieimen 2011; Steffan et al. 2018). Natural sources of heavy metal pollution include soil erosion, lithogenesis, weathering, and geological sources (Stafilov et al. 2010; Keshavarzi and Kumar 2019). However, it is also important to realize that many of the heavy metals are plant and human nutrients, required in small amounts for proper health and with narrow ranges from deficiency to toxicity (Brevik 2009; Steffan et al. 2018).

Risk assessment is an important analytical method that allows decision makers to handle polluted sites in a cost-effective way while protecting the health of the public and environment (Zhao and Kaluarachchi 2002). The spatial distribution, toxicity, and deficiency of soil metals are greatly affected by variations in parent rock, environmental qualities and pedological processes (Rezapour et al. 2014). Human exposure to toxic levels of heavy metals, especially arsenic (As),

chromium (Cr), copper (Cu), lead (Pb), and zinc (Zn), cause major health disruptions, including disrupting the nervous and digestive systems, damage to the circulatory system, various cancers, and several other unfavorable and adverse consequences (Lee et al. 2006; Arfsten et al. 2011; Peng et al. 2016; Brevik et al. 2020). In agricultural fields, major causes of heavy metal pollution are the application of excess fertilizers and pesticides, substandard manure and compost, manufacturing facilities and other anthropogenic practices (Brevik 2009; Keshavarzi and Kumar 2019). Further work is required on contamination indicators and environmental risk of heavy metals in soils (Kumar et al. 2016; Tepanosyan et al. 2017; Sihag et al. 2019; Kumar et al. 2019a, b; Heidari et al. 2019). In addition, a broad ranging approach to assessing heavy metals pollution has been applied by different researchers, including geoaccumulation index (Igeo), enrichment factor (EF), contamination factor (CF), and ecological risks index (RI) (Tian et al. 2017; Kumar et al. 2019a, b; Keshavarzi and Kumar 2019; Heidari et al. 2019; Aitta et al. 2019; Elbehiry et al. 2019).

The development of multivariate statistical and GIS-based approaches have revolutionized research on soil pollution and human health impacts over the last few decades (Brevik et al. 2016; Liu et al. 2016; Zuo et al. 2018). Multivariate approaches have been increasingly concerned with the source apportionment and distribution of heavy metals (Zhu et al. 2016; Jiang et al. 2017; Yegemova et al. 2018; Kumar et al. 2018; Barakat et al. 2019; Keshavarzi and Kumar 2019; Heidari et al. 2019). Heavy metal levels in the topsoil vary spatially based on landforms, parent material, geological features, topographical attributes, and soil physiochemical properties (pH, moisture regime, available phosphorus, organic matter content, etc.) (Rais et al. 2006; Wang et al. 2008; Zhu et al. 2016; Khaledian et al. 2017). Furthermore, studies to date have found different relationships between soil physiochemical properties and heavy metals. For example, some studies have shown positive correlations between pH and heavy metals (Kargar et al. 2015; Khaledian et al. 2017), others have shown negative correlations (Pereira et al. 2014; Perez-Esteban et al. 2014), and some of have shown no correlation at all (Cai et al. 2012; Zahra et al. 2014). Thus, an important issue is improved understanding of

the dynamic variation in these heavy metals and the factors that contribute to their levels in soils.

In the current study, we used the Tellus survey database. Tellus is a national program to gather geochemical and geophysical data across Ireland (Appleton et al. 2008). The geophysical characteristics of Northern Ireland were recently redefined by the Tellus airborne geophysical survey, which collected high-resolution electromagnetic data from the magnetic, radiometric, and active frequency domains (Lusty et al. 2009). The distribution of heavy metals in a portion of Northern Ireland was investigated using the Tellus soil geochemistry data in agricultural, industrial, and urban areas. It was hypothesized that the soils of the study location represented a threat to human and environmental health due to heavy metal accumulation from anthropogenic activities. This hypothesis was tested on 620 soil samples from the Tellus database. In order to evaluate different results, and to assimilate them into an inclusive dataset, multivariate statistical techniques were applied to find the plausible sources of heavy metals. Furthermore, contamination indices (CF, EF, PLI, Igeo, RI, and MRI) were also used on the data to determine the pollution level of HMs in the soils of Northern Ireland, because measuring the HMs concentrations alone does not provide a complete picture of the soils' pollution status. Therefore, to determine the relationships between the heavy metals and influencing factors, the present study aimed to (1) evaluate selected soil physiochemical properties (texture, pH, bulk density, phosphorus, As, Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, and Zn) and (2) assess and apply selected contamination indices (CF, EF, PLI, Igeo, RI, and MRI) and multivariate techniques (correlation, CA, and PCA) to Tellus data to determine the human health or ecological risk posed by various metals.

## Materials and methods

### Tellus database

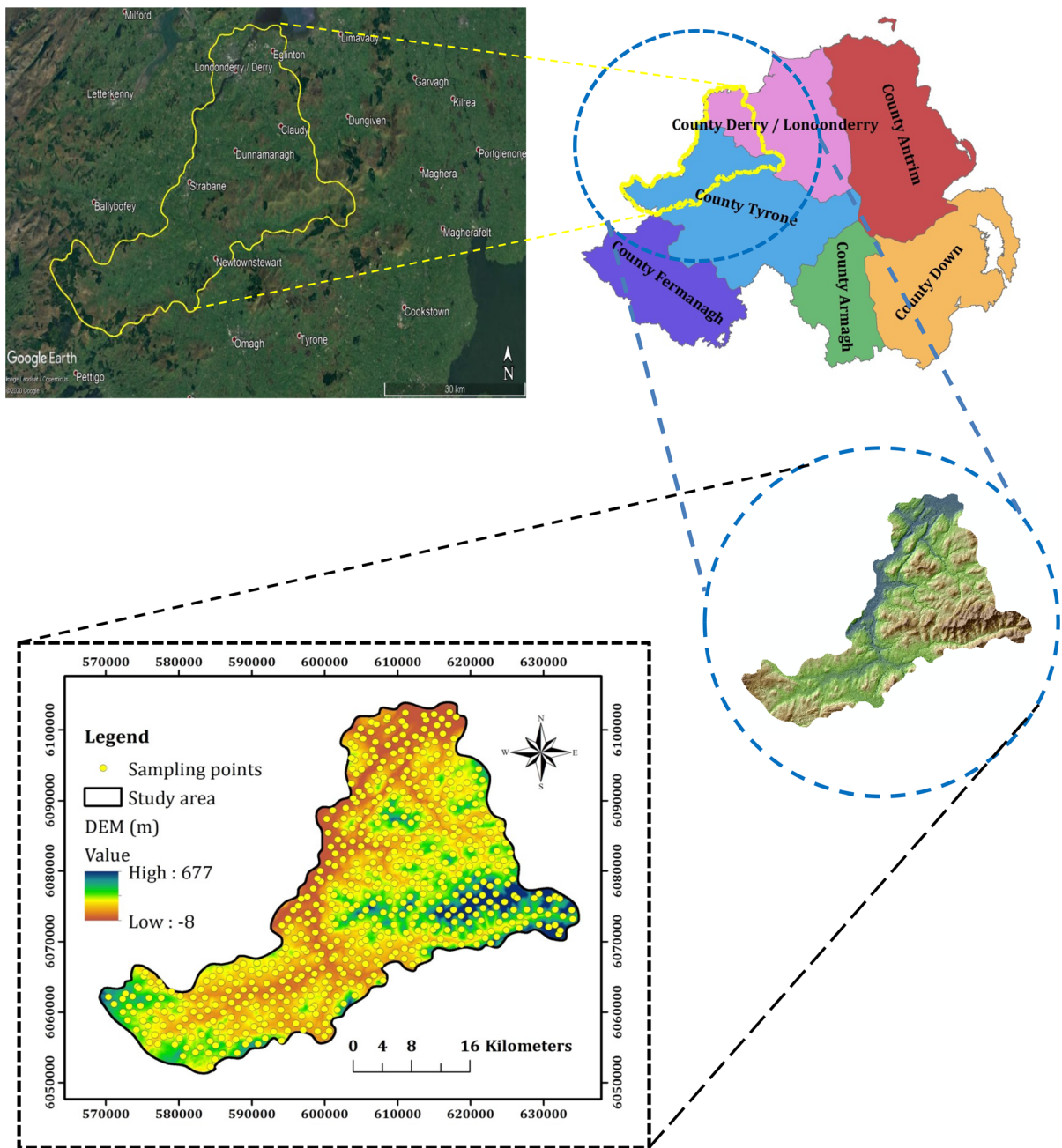
Tellus means “earth” in Latin. This has been used as an alternate name in mythology for Terra, the Roman Earth Mother goddess, and hence is the equivalent of the Greek goddess Gaia. The Tellus project was carried out by the Geological Survey of Northern Ireland and financed by the Department of

Communications, Climate Action and Environment. All Tellus data are made available online, free of charge. Tellus comprises two types of surveys: aerial geophysical data using a low-flying aircraft and field-based geochemical data surveys of stream sediment, stream water and soils (Young and Donald 2013; Gallagher et al. 2016). To date, Tellus surveying has been conducted in Northern Ireland and Ireland's border area, and 60% of the region has now been mapped with the Tellus geophysical survey. Tellus set up a product development process to create more oriented, user-centered data items, the need for which was defined through stakeholder feedback, independent reviews and government policy. Product development is being performed in five main areas: smart agriculture, climate action and education, mineral prospecting, and health and environment (Nice 2010).

### Description of the study area and data used

The study area follows along the boundary between Eglinton and Castlederg counties in the northwestern portion of Northern Ireland. It is situated between latitude 54° 38' N to 55° 4' N and longitude 7° 16' W to 7° 27' W with an average altitude of 148.68 m above mean sea level and an area of approximately 1270 km<sup>2</sup> (Fig. 1). A digital elevation model (US Geology Survey 2014) was downloaded from the Aster GDEM database and reintroduced to Universal Transverse Mercator (UTM) projection with a spatial resolution of 30 × 30 m to establish the topography of the study area. The slope gradient varies from 0.2 to 42 percent with an average of 9.4 percent. ArcGIS 10.5 (ESRI, USA) software was used to select a total of 620 soil samples (0 to 20 cm depth) extracted from the Tellus database in the defined study area based on the boundary location. The soil textures were classified as loam, clay loam, silt loam, and sandy loam. Previous research confirmed that increasing fertilizer inputs and imported feedstuffs led to high amounts of soil phosphorus in the study area (Foy et al. 2002). The main land covers in the study area include large areas of production agricultural land, pasture, and wetlands. Forested areas are scarce.

The climate in Ireland is strongly affected by its position on the eastern side of the Atlantic Ocean and on the western edge of Europe. This leads to a mild sea climate, which is characterized by winds from the south west (Creamer and O'Sullivan 2018). The mean



**Fig. 1** Digital elevation model (DEM), study area location and distribution of the soil samples

annual precipitation in the study area ranges from 959 to 1,320 mm, and mean annual temperature is about 8–9 °C (McElarney et al. 2015). The Köppen–Geiger climate classification is temperate oceanic (Cfb), which means the average temperature in the coldest month is above 0 °C, 1–3 months average above

10 °C, and there are no significant precipitation differences between seasons (Beck et al. 2018).

Earlier regional geochemical studies carried out as part of the Tellus project in the northern parts of Ireland (Young and Donald 2013; Gallagher et al. 2016) demonstrated the effects of widespread urbanization on the composition of topsoils, even outside of

major metropolitan regions (Creamer and O’Sullivan 2018), and frequent anomalies in topsoil have been found in small towns, villages, and communities across the country (Nice 2010). A significant proportion of Irish soils are marked by low internal drainage intensified by regionally high levels of precipitation. Because of their higher water content, poorly drained soils need longer time and energy to warm up in spring, which can delay crop/grass production (Creamer and O’Sullivan 2018). Around 49.5% (3.4 M ha) of Ireland’s total area is defined as marginal land that is subject to topography, relief, soil, and climate-related natural constraints. Such land is scattered across the country but mainly distributed along the west coast (Gardiner and Radford 1980; Creamer and O’Sullivan 2018). Excess soil moisture is considered to be a major limitation in terms of primary productivity in large parts of the Ireland region classified as significantly wet with impeded drainage (Collins and Cummins 1996) or low natural drainage status (Creamer and O’Sullivan 2018).

#### Analytical methods for selected heavy metals and laboratory analyses

Soil chemistry sample analysis techniques used to generate the Tellus database have been reported by Nice (2010) and Milne et al. (2013). They include soil loss-on-ignition at 450 °C to determine soil organic matter, determination of soil pH by CaCl<sub>2</sub> slurry, and multi-element total analyses by X-ray fluorescence spectrometry (XRFS) on pressed powder pellets for the heavy metals investigated in this study (Ingham and Vrebos 1994; Nice 2010; Milne et al. 2013). Topsoil physical properties, i.e., soil textural fractions (clay, silt, and sand) and bulk density, were extracted through the LUCAS database at the European scale (Panagos et al. 2012; Ballabio et al. 2016).

#### Collection of auxiliary information

The geology of the study area is characterized by metamorphic rocks and coastal deposits (Fig. 2). The major geological units are comprised of the Southern Highland Group, which is composed of pelitic and psammitic schist, phyllite, and marble. The major landforms of the region are plains followed by mid-slope ridges and small hills in plains. Landforms were

derived from the DEM using SAGA GIS software (Olaya and Conrad 2009) (Fig. 3).

#### Evaluation of soil contamination

Different contamination determinants and indices are used to assess the pollution levels and environmental hazards posed by heavy metals in soils, including the contamination factor (CF), enrichment factor (EF), geoaccumulation index (Igeo), potential ecological risk (RI), and modified potential ecological risk (MRI) indices (Tian et al. 2017; Kumar et al. 2018; Keshavarzi and Kumar 2019). Such pollution indices can demonstrate a qualitative threshold or requirement for each individual heavy metal’s ecological risk measurement.

#### Contamination factor (CF)

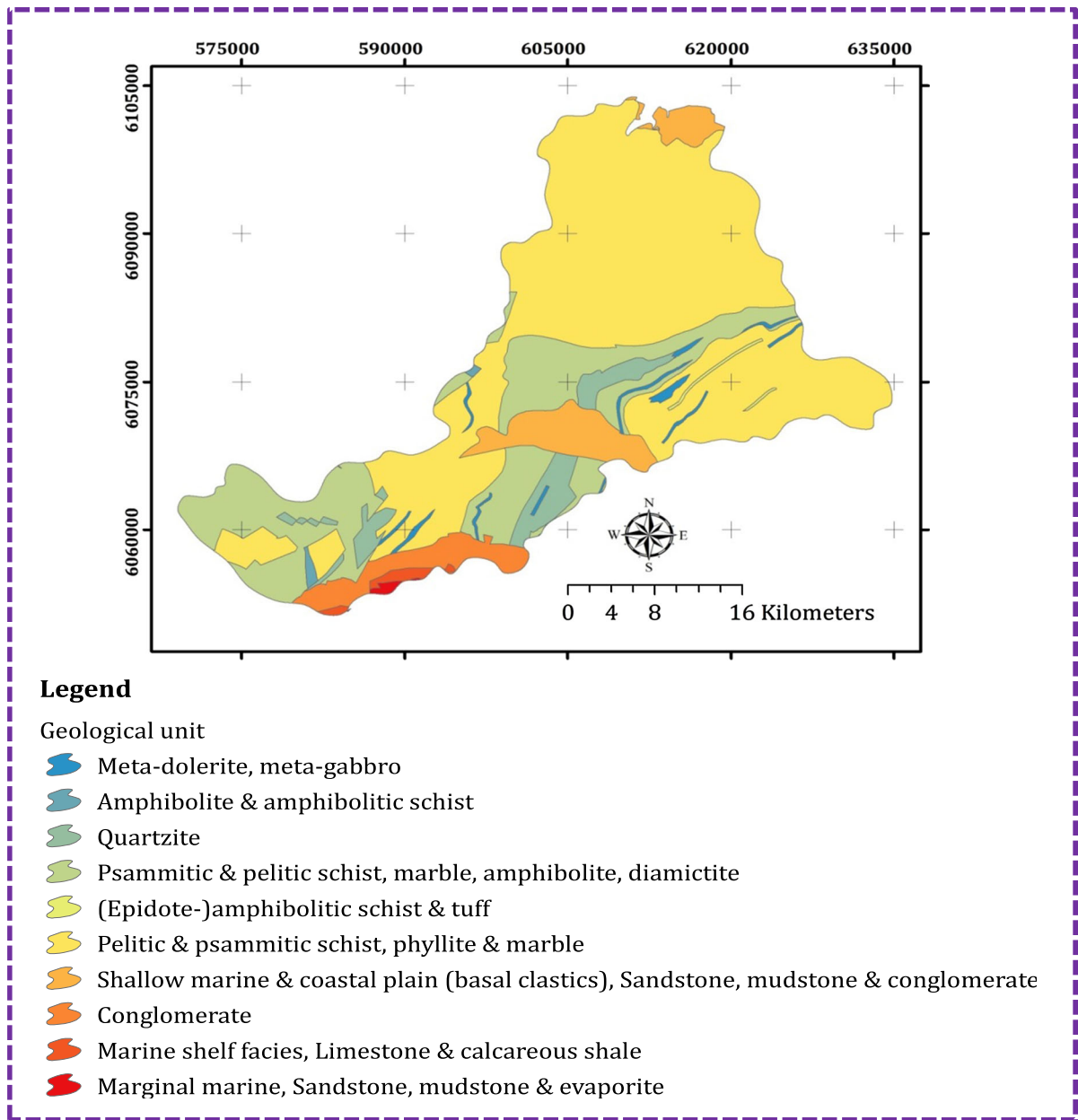
The contamination factor provides information on the anthropogenic contributions of heavy metals in soils (Ahmed et al. 2016). It was determined using Hakanson’s (1980) equation as follows:

$$CF = \frac{HM_s}{HM_b} \quad (1)$$

where  $HM_s$  and  $HM_b$  are the heavy metal’s concentrations at sampling locations and in the background environment, respectively. Values from Taylor and McLennan (1995) were used as heavy metal background values in this study. CF was interpreted using four classes: ( $CF > 6$ ; very high contamination), ( $3 < CF < 6$ ; high contamination), ( $1 < CF < 3$ ; moderate contamination) and ( $CF < 1$ ; low contamination) (Hakanson 1980; Keshavarzi and Kumar 2019).

#### Enrichment factor (EF)

The enrichment factor represents heavy metal enrichment as compared to the background heavy metals content (Sakram et al. 2015). Heavy metals that vary geochemically within the environment and are not capable of creating antagonism or compatibility issues with the measured heavy metals are employed as the heavy metal background (Chandrasekaran et al. 2015). Mn was used as the heavy metal background in this study and measured as:



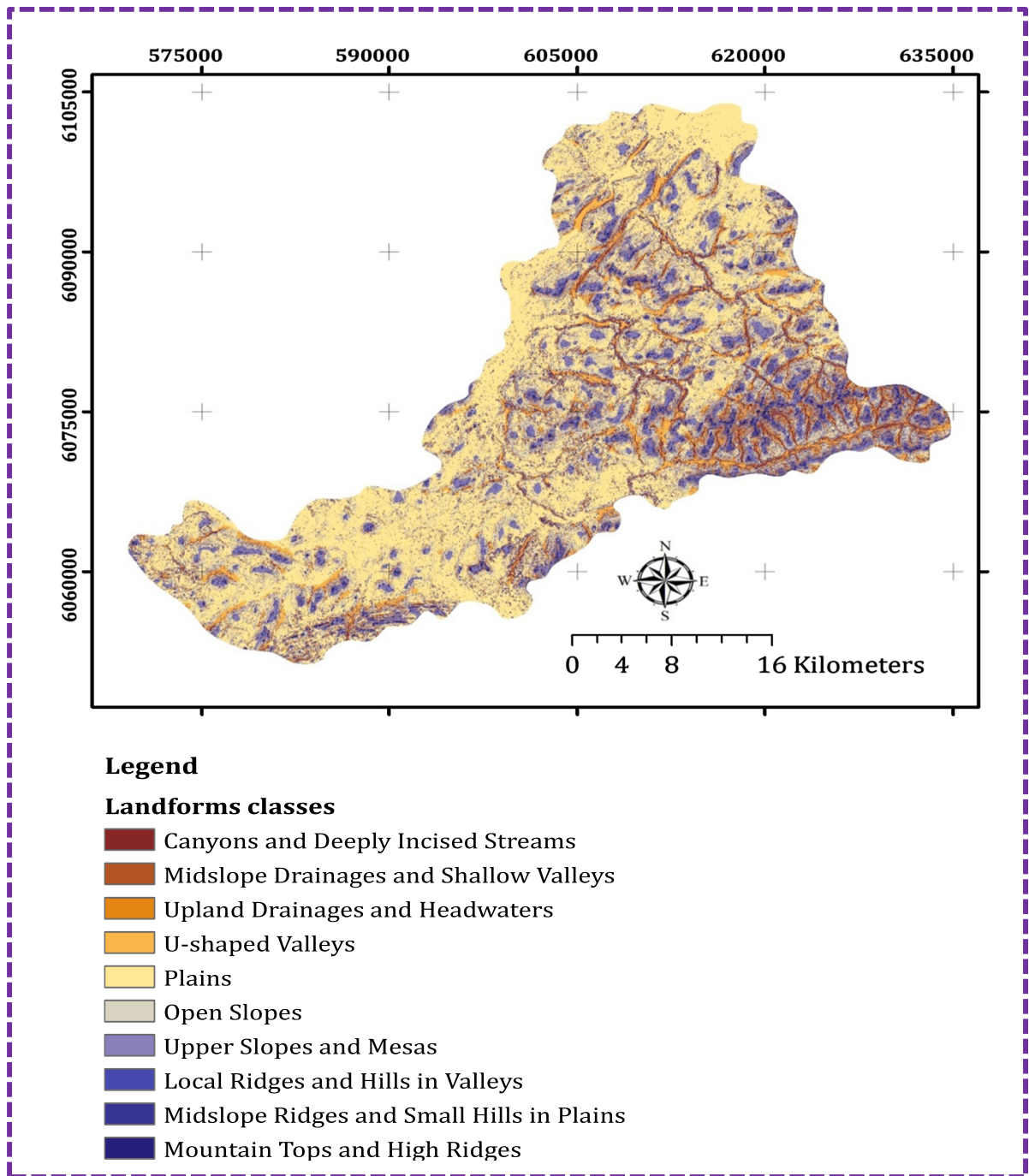
**Fig. 2** Geological units map of the study area (the data represent a seamless bedrock geological dataset encompassing Rep of Ireland and parts of Northern Ireland at the 1:500,000

scale. This dataset is part of the “Bedrock Geology 500 K Series” (Geological Survey of Ireland 2020))

$$EF = \frac{\frac{HM_s}{HM_b}}{\frac{Mn_s}{Mn_b}} \quad (2)$$

where HMs and HM<sub>b</sub> are the sample and background concentrations of the heavy metals, respectively, while Mn<sub>s</sub> and Mn<sub>b</sub> are the sample and background Mn concentrations, respectively.

The EF was interpreted using seven categories: (EF > 50; exceptionally high enrichment), (25 ≤



**Fig. 3** Landforms classes map of the study area (extracted from DEM, classification codes based on SAGA manual described by Olaya and Conrad (2009) contains Irish Public Sector Data

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EF < 50; very high enrichment), (10 ≤ EF < 25; high enrichment), (5 ≤ EF < 10; moderately high enrichment), (3 ≤ EF < 5; moderate enrichment),

(1 ≤ EF < 3; less enrichment) and (EF < 1; no enrichment) (Marrugo-Negrete et al. 2017; Keshavarzi and Kumar 2019).

### Pollution load index (PLI)

The PLI was determined by considering the CF of each metal for a particular site; the equation used to calculate this was as follows:

$$PLI = (CF_1 \times CF_2 \times CF_3 \times CF_4 \times \dots \times CF_n)^{1/n} \quad (3)$$

where  $n$  corresponds to the number of metals. A PLI value less than 1 signifies unspoiled soil.

quality, while greater than 1 indicated deterioration of soil quality (Tomlinson et al. 1980).

### Geoaccumulation index (Igeo)

Igeo quantifies the level of heavy metal pollution. It was assessed as follows:

$$Igeo = \log_2 \frac{HM_s}{1.5 \times HMB} \quad (4)$$

where  $HM_s$  and  $HMB$  are the sample and background concentrations of the heavy metals, respectively. The constant 1.5 reflects changes in environmental concentrations of heavy metals (Wei and Yang 2010). Igeo was interpreted as: Igeo  $\geq 5$ ; extreme pollution, Igeo (4–5); severe pollution, Igeo (3–4); very high pollution, Igeo (2–3); high pollution, Igeo (1–2); strong pollution, Igeo (0–1); moderate pollution and Igeo  $\leq 0$ ; no pollution (Loska et al. 2004; Keshavarzi and Kumar 2019).

### Ecological risk assessment (RI and MRI)

To analyze the ecological risk posed by heavy metals in soils, the potential ecological risk index (RI) was calculated. It is determined as the multiplication of each heavy metal contamination factor and individual heavy metal toxicological response factor ( $Tr$ ), viz. Mn and Zn (1), Cu, Co, Ni and Cr (5), and As and Cr (2) (Kumar et al. 2018; Keshavarzi and Kumar 2019). It was calculated using the following equation:

$$RI = CF_n \times Tr \quad (5)$$

where  $CF_n$  and  $Tr$  are the individual heavy metal's contamination factor and toxicological response factor, respectively. The  $Tr$  can be supplemented with EF for measurement of potential ecological risk index, with a view to determining the ecological risks of anthropogenic and lithogenic additions of heavy metals to soils. The modified potential ecological risk

(MRI) was calculated based on EF (Kumar et al. 2018; Keshavarzi and Kumar 2019) using the following equation:

$$MRI = EF_n \times Tr \quad (6)$$

where  $EF_n$  and  $Tr$  are the individual heavy metals' enrichment factor and toxicological response factor, respectively. The risk assessment was interpreted as follows: > 320 (very high risk), 160–320 (high risk), 80–160 (considerable risk), 40–80 (moderate risk) and  $Er < 40$  (low risk) (Keshavarzi and Kumar 2019).

### Spatial distribution maps of RI and MRI

To illustrate the spatial distributions of ecological risk indices, kriging in ArcGIS 10.5 (ESRI, Redlands, CA, USA) software was performed as undertaken by various researchers (Liu et al. 2017; Masoud et al. 2016) to extrapolate their spatial distribution maps (Chen et al. 2016). The kriging method is one of the best linear unbiased techniques to provide reasonable spatial maps and gives stochastic uncertainty to the maps (Burrough and McDonnell 2015). To calculate the errors and investigate uncertainty assessment for the spatial maps, the cross-validation method (leave-one-out) was used and the prediction standard error for the kriged maps was measured and expressed as “standard error” maps (Grunwald et al. 2004).

### Statistical analysis

Descriptive statistics were calculated for all the analyzed parameters using PAST v.3.21 (Hammar et al. 2001). Pearson's correlation analysis was conducted to find the associations among soil physiochemical parameters and heavy metals using R-software v.3.5.1. Further cluster analysis (P, As, Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, and Zn) and principal component analysis (As, Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, and Zn) were performed on the dataset in order to find source apportionment of heavy metals by employing SPSS v.16 software (IBM, USA) and Minitab v.14. The varimax rotation method was used in the principal component analysis (PCA). The Kaiser–Meyer–Olkin (KMO) and Bartlett's test of sphericity was significant (KMO = 0.762 > 0.6,  $p = 0.000 < 0.005$ ) and demonstrates sample suitability. Cluster analysis was applied to the heavy metals



data employing Ward’s method (Kumar et al. 2018) and Euclidean distance as a measure of similarity.

## Results and discussion

### Descriptive statistics

The descriptive statistics for selected physiochemical parameters are shown in Table 1. The pH range was 2.86 to 7.29 with a mean value of 4.44. The acidic nature of the soils enhances the availability and movement of heavy metals in the soils (Tian et al. 2017). Among the textural properties, sand (28.5 to 93.1% with mean value 54.5%) is most abundant followed by silt (4.3 to 46.6% with mean value 29.6%) and clay (1.8 to 30.3% with mean value 15.7%). The high sand content is responsible for reducing the potentially toxic metals content of the soils owing to their low cation exchange capacity and leaching (Bhatti et al. 2018). The phosphorus content ranged from 138 mg/kg to 3220 mg/kg with an average value of 871.3 mg/kg. Heavy use of phosphorus fertilizers is associated with high phosphorus content in the soil and also acts as a sink for the immobilization of metals (McGowen et al. 2001). Bulk density averaged 1.09 g/cm<sup>3</sup> with a range of 0.96 to 1.23 g/cm<sup>3</sup>. The skewness and kurtosis of sand, silt, clay, pH, and bulk density were found to be less than one, signifying normal

distribution of data (Beaver et al. 2012). The kurtosis and skewness values of phosphorus were above one, demonstrating leptokurtic kurtosis and left-handed skewness (Beaver et al. 2012). The highest variance was observed for phosphorus followed by sand and silt, and greater variance of these parameters reflects higher degree of variation of each value from the average value for a particular parameter (Beaver et al. 2012). The coefficient of variation was also found to be higher for all the physiochemical parameters, indicating higher alteration of textural properties, pH, bulk density, and phosphorus.

The descriptive statistics for the heavy metals analyzed are given in Table 2. The mean values obtained from the analyzed metals showed the trend: Mn > Zn > Pb > Cu > Cr > Ni > As > Co > Cd. Variance analysis revealed that Mn content showed the greatest alteration among the studied sites followed by Zn, Pb, and As. Kurtosis and skewness values were greater than one for all metals, signifying leptokurtic kurtosis and right-handed skewness (Beaver et al. 2012). The high kurtosis indicated that all samples were gathered at moderately less values (Lu et al. 2010). The highest variance was observed for Mn followed by Zn and Pb, and greater variance of these heavy metals signifies a higher degree of variation of each concentration from the average concentration for Mn, Zn, and Pb than for other metals in the study (Beaver et al. 2012). All analyzed heavy metals

**Table 1** Descriptive statistics for selected physicochemical properties for the soils studied as reported in the Tellus database

Statistics	Sand (%)	Silt (%)	Clay (%)	pH (-)	Bulk Density (g/cm <sup>3</sup> )	Phosphorus (mg/kg)
Min	28.56	4.319	1.856	2.86	0.96	138
Max	93.16	46.60	30.34	7.29	1.23	3220
Mean	54.58	29.64	15.78	4.44	1.09	871.3
Std. error	0.61	0.39	0.23	0.03	0.002	14.12
Variance	232.82	96.60	34.17	0.69	0.003	123,660.4
Std.dev	15.25	9.82	5.84	0.83	0.05	351.65
Median	51.35	32.17	16.47	4.58	1.08	820
25 prcntil	41.33	22.11	11.17	3.765	1.04	626.2
75 prcntil	65.99	37.82	20.44	4.99	1.13	1060
Skewness	0.54	− 0.62	− 0.19	− 0.19	0.23	1.15
Kurtosis	− 0.73	− 0.62	− 0.82	− 0.41	− 0.81	3.68
Geom. mean	52.54	27.43	14.44	4.36	1.09	803.2
Coeff. var	27.96	33.16	37.04	18.6	5.16	40.36

**Table 2** Descriptive statistics for the heavy metals in soils of the study area as determined from the Tellus database. All units are in  $\mu\text{g/g}$  except Fe (% wt. eq.)

Statistics	As	Cd	Cr	Co	Cu	Fe	Pb	Mn	Ni	Zn
Min	0.5	0.005	0.5	0.434	2.2	0.34	1.88	2.5	1.66	15.2
Max	217	2.69	140	45.9	143	10.7	545	4940	69.4	275
Mean	9.95	0.27	17.36	6.12	23.49	1.95	27.37	343.61	12.56	57.48
S.E	0.77	0.01	0.67	0.20	0.63	0.04	1.12	17.65	0.34	1.26
Variance	364.40	0.05	276.91	24.51	243.68	1.03	772.13	193,162.80	70.22	990.06
Std.dev	19.08	0.22	16.64	4.95	15.61	1.01	27.78	439.50	8.37	31.46
Median	5.8	0.22	14.8	5.19	22.3	1.93	21.1	229.5	11.5	50.7
25 prcntil	2.38	0.17	4.69	2.1875	9.69	1.12	15.725	102	5.58	35.9
75 prcntil	10.175	0.3	23.6	8.65	32.45	2.55	31.5	422.75	16.65	70.4
Skewness	6.76	5.30	2.39	1.95	1.34	1.50	11.43	4.98	1.34	2.24
Kurtosis	57.10	43.51	10.59	8.84	5.42	8.98	198.19	36.21	3.97	8.47
G.M	4.82	0.23	8.43	4.20	18.19	1.68	22.53	217.70	9.81	50.99
C.V	191.78	80.97	95.86	80.96	66.44	52.23	101.53	127.91	66.70	54.74
Irish soil limits <sup>a</sup>	1–50	0.1–1	5–250	1–25	2–100	–	2–80	20–3000	0.5–100	10–200
Swedish limits for soil <sup>b</sup>	–	0.4	120	30	100	–	80	–	–	350

<sup>a</sup>McGrath and Fleming (2006)

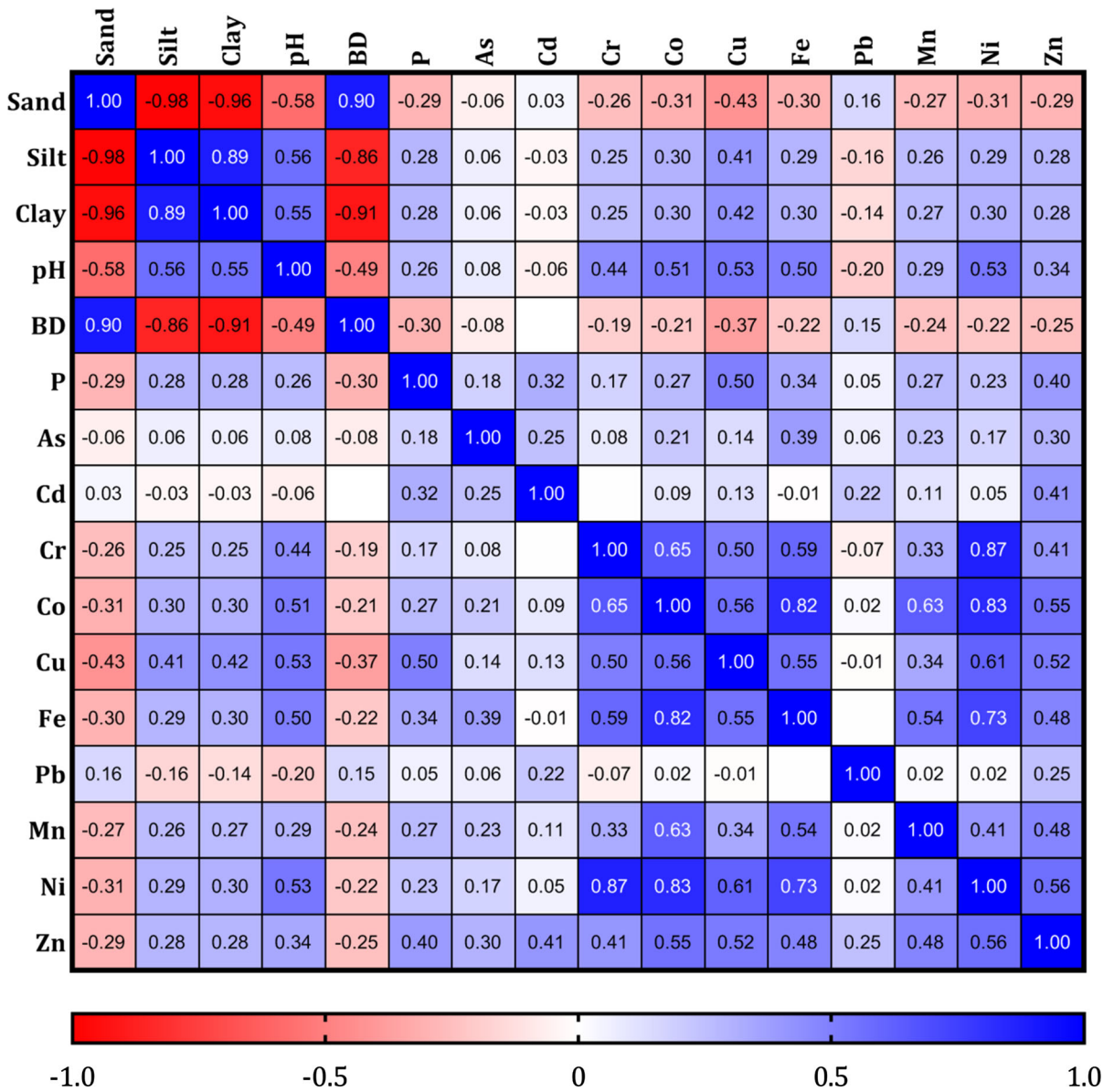
<sup>b</sup>Bhagure and Mirgane (2011); Swedish Environmental Protection Agency (2003)

showed a high coefficient of variation (52.2% to 191.7%), suggesting that human activities have had a large influence on them (Cai et al. 2015). The unevenness allied with the analyzed descriptive statistical data may be linked with activities that change land use and configuration like excavation, digging, weathering, and erosion that can alter the stability of soil conditions (Amuno 2013; Arenas-Lago et al. 2014). The coefficient of variation (CV) is mainly performed to elucidate the level of variability of heavy metals (Fu et al. 2010). Arsenic had the largest CV (191.7%) followed by Mn (127.9%) and Pb (101.5%), showing anthropogenic activities have a high impact on the content of these metals in the soil (Adimalla 2020). When the CV value is less than 10% it signifies low variability, whereas if greater than 90%, it signifies high variability (Fu et al. 2014). Guo et al. (2012) reported that moderately high CVs greater than 50% for heavy metals in soils were primarily impacted by anthropogenic activities, and in the present study all heavy metals showed CVs above 50%. After comparing heavy metals concentrations with Irish soil limits suggested by McGrath and Fleming (2006), it was estimated that 2.4% of the

samples exceeded their recommended maximum concentration for As, 1.1% exceeded their maximum recommended content for Cd, and 0.48%, 2.09%, 0.6% and 0.8% of samples surpassed their maximum recommended concentrations for Co, Pb, Mn, and Zn, respectively.

#### Correlation analysis

Pearson's correlation analysis was employed to investigate the correlations among the heavy metals (As, Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, and Zn) and physiochemical parameters (sand, silt, clay, pH, bulk density, and phosphorus) (Fig. 4). Cr, Co, Cu, Fe, Pb, Mn, Ni, and Zn were highly correlated with each other ( $p < 0.05$ ), signifying that human activities have likely played a large role in reaching their current concentrations (Wang et al. 2012). As and Cd (0.25) showed moderate correlation with other heavy metals and with the physiochemical properties except phosphorus. Ma et al. (2018) also reported correlations between Cr, Zn, Cu, and Ni and concluded that similar sources were responsible for those correlations. Phosphorus was positively correlated ( $p < 0.05$ ) with As



**Fig. 4** Pearson’s correlation analysis of heavy metals and physiochemical properties of soil. Blue color indicates positive correlation, while red color exhibits negative correlation at  $P < 0.05$ . Blank boxes represent no correlation among the parameters

(0.18), Cd (0.32), Cr (0.17), Co (0.27), Cu (0.50), Fe (0.34), Mn (0.27), Ni (0.23), and Zn (0.40), reflecting the contribution of agronomic practices like pesticide, fertilizer, and herbicide applications to the heavy metal content in these soils. The textural properties (0.55 to 0.58), pH, bulk density (0.49), and phosphorus (0.30) also showed high correlations ( $p < 0.05$ ) with each other. Sand and bulk density (0.90) were negatively correlated ( $p < 0.05$ ) with all heavy metals

except As, Cd, and Pb, showing that sandy texture has a negative effect on the accumulation of most heavy metals at this site, likely due to leaching of metals to lower soil layers which might expedite heavy metals uptake by plants (Rodriguez Martin et al. 2013). Silt and clay (0.89) were positively correlated ( $p < 0.05$ ) with all heavy metals except As (0.06), Cd (0.03), and Pb (0.14 to 0.16). Lead (0.20), As (0.15), Cd (0.22), and pH were also correlated with each other

( $p < 0.05$ ), signifying the impact of pH on heavy metals mobility and accumulation.

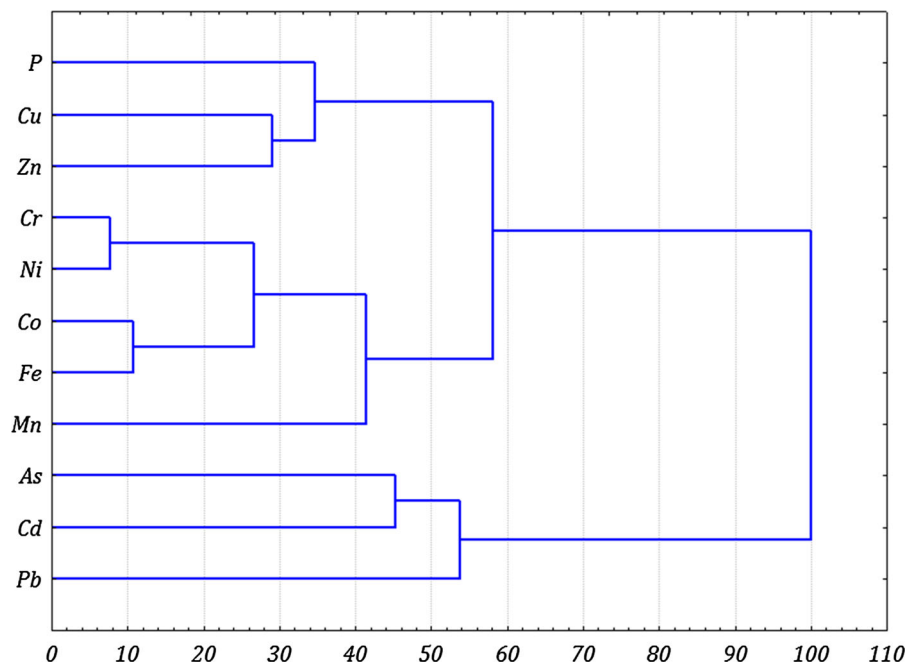
### Multivariate analysis

Cluster analysis (CA) was applied to the heavy metals data (Fig. 5) to investigate which metals likely had the same sources. Ward's method is a criterion used in hierarchical CA. Ward (1963) suggested this method for common agglomerative hierarchical clustering practice, where the criterion for selecting the pair of clusters to amalgamate at every step is dependent on the ideal value of an independent function. Various researchers such as Moghtaderi et al. (2020), while working on soil heavy metal contamination in Southwest Iran, and Kumar et al. (2020) on heavy metal contamination in India, applied Ward's method for CA. Moreover, it is the most commonly employed method for environmental sample analysis (Strauss and von Maltitz 2017).

CA results showed two main groups: cluster I (As, Cd, and Pb) and cluster II (Cr, Ni, Co, Mn, Fe, P, Cu, and Zn). Furthermore, cluster II was subgrouped into Cr and Ni and Co and Fe, with these two subgroups forming a higher level subgroup that also included Mn,

along with another subgroup consisting of Cu, Zn, and P.

PCA was conducted to investigate relationships between the metals (Table 3 and Fig. 6). The eigenvalues were greater than one for the first two components and accounted for 61.2% of the total variance. PC1 accounted for 45.9% of the variation and had loadings on Co, Cu, Fe, Mn, Cr, Ni, and Zn. Arsenic, Pb, and Cd loadings were grouped into PC2 and accounted for 15.2% of variance. After rotation of the component matrix, PC1 accounted for 44% of the variance and had same loadings as PC1 from the component matrix, while PC2 accounted for 17% of the variation and also had the same loadings as PC2 from the component matrix. Gasoline additives and release by dust are responsible for most of the Pb content in soil and the broader environment and is a key source of contamination in urban areas (Chen et al. 2016). The addition of Cd is due to impurities in detergents, petroleum goods, and fertilizer (Kumar et al. 2019a). Sewage sludge and fly ash are the main sources associated with Cr in the soil (Dogra et al. 2019). Zinc sulfides, zinc chloride, and zinc acetate are applied as wood preservatives and during the dyeing of fabrics and account for much of the Zn in soil (Kumar et al. 2019b). Nickel is contributed to the

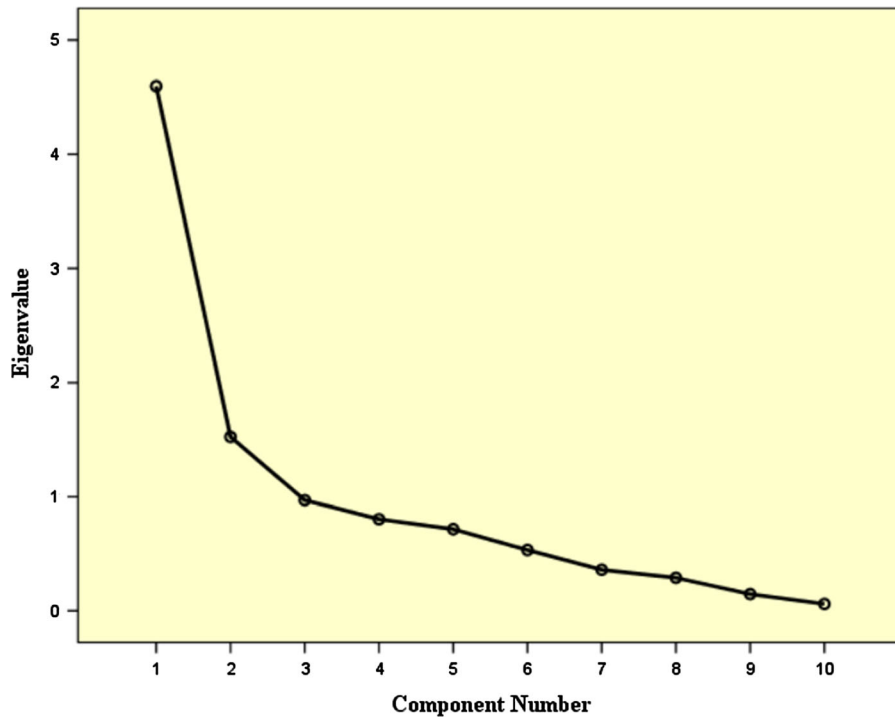


**Fig. 5** Cluster analysis of heavy metals in soils from Northern Ireland

**Table 3** Principal component analysis of heavy metals in the studied soils as indicated in the Tellus database

Components	Initial eigenvalues			Extraction sums of squared loadings			Rotation sums of squared loadings		
	Total	Var %	Cumulative %	Total	Var %	Cumulative %	Total	Var %	Cumulative %
1	4.59	45.96	45.96	4.59	45.96	45.96	4.41	44.09	44.09
2	1.52	15.24	61.20	1.52	15.24	61.20	1.71	17.11	61.20
3	0.96	9.69	70.90						
4	0.80	8.03	78.93						
	Component matrix			Rotated component matrix					
	PC1	PC2		PC1	PC2				
As	0.349	<b>0.443</b>	As	0.229	<b>0.516</b>				
Cd	0.188	<b>0.762</b>	Cd	0.006	<b>0.785</b>				
Cr	<b>0.774</b>	– 0.326	Cr	<b>0.830</b>	– 0.125				
Co	<b>0.907</b>	– 0.117	Co	<b>0.908</b>	0.110				
Cu	<b>0.720</b>	– 0.062	Cu	<b>0.713</b>	0.118				
Fe	<b>0.858</b>	– 0.124	Fe	<b>0.862</b>	0.092				
Pb	0.064	<b>0.606</b>	Pb	0.088	<b>0.603</b>				
Mn	<b>0.651</b>	0.084	Mn	<b>0.610</b>	0.242				
Ni	<b>0.900</b>	0.217	Ni	<b>0.925</b>	0.012				
Zn	<b>0.725</b>	0.431	Zn	<b>0.596</b>	0.591				

Bold values represent significant loadings



**Fig. 6** PCA scree plot showing first two components with eigenvalues greater than one

atmosphere by power plants where it becomes attached to dust particles that are washed into the soil by precipitation. Use of inorganic fertilizers adds Cu to the soil, while the steel and iron industries and dust from iron mines are responsible for most of the anthropogenic Fe in soil (Kumar et al. 2019b). Zhu et al. (2017) also observed that Cu and Zn were contributed by anthropogenic activities. Tepanosyan et al. (2018) reported loadings of Mn, Cu, and Zn on PC1 and concluded that human inputs contributed to the soil content of these metals.

#### Appraisal of pollution by CF, EF, PLI, and Igeo

The CF was computed for each heavy metal studied (Table 4). Results showed that 17.9% of sampling sites showed low, 22.9% moderate, 30.8% high, and 28.3% very high contamination by As. For Cd 5%, 68.3%, 20.9%, and 5.6% of sites showed low, moderate, high, and very high contaminations, respectively. In the cases of Zn, Cr, Co, and Cu, 75.1%, 90.3%, 81.7%, and 57.2% of sampling sites had low contamination, respectively, while 24.8%, 9.6%, 18.2%, and 42.7% of sites showed moderate contamination, respectively. The results for Mn and Pb indicated that 82.4% and 43.8% sites showed low contamination, 15.9% and 51.1% moderate contamination, 0.96% and 4.5% high contamination, and 0.64% and 0.48% of sampling sites showed very high contamination, respectively. In the case of Ni, 82.5% and 17.4% of sampling sites showed low contamination and moderate contamination, while no sites showed higher Ni contamination values. Wang et al. (2020) reported CF values for different heavy metals such as Pb (1.54), Cd (1.13), Cu (1.39), As (1.14), Zn (1.21), and Cr (1.43) in the soils of Jiangsu Province and reported moderate contamination of heavy metals which is in confirmation with our findings.

The EF was also determined for each heavy metal (Table 4). The EF results for As indicated that 7.4%, 25%, 51.7%, 11.1%, and 4.6% of the sampling sites indicated less enrichment, moderate enrichment, moderately high enrichment, high enrichment, and very high enrichment, respectively. For Cr, 91.2%, 8.06%, and 0.64% of sampling sites showed less enrichment, moderate enrichment and moderately high enrichment, respectively. The results for Cd indicated that 14.03% sites showed less enrichment, 33.8% moderate enrichment, 36.2% moderately high

enrichment, 11.6% very high enrichment and 4.1% exceptionally high enrichment. The results for Co and Cu showed that 92.5% and 54.5% of sampling sites had low pollution, while 7.4% and 40.8% sites showed modest enrichment, respectively. The EF results for Pb and Zn indicated that 42.2% and 56.6% sites showed less enrichment, 32.1% and 37.2% sites showed modest enrichment, 20.1% and 3.7% significant enrichment, 4.35% and 0.32% very high enrichment, and 1.12% and 0.16% life-threatening enrichment, respectively. For Ni 83.5% and 16.2% sampling sites showed less and modest enrichment. Pan et al. (2016), working on soils in Shanxi Province, found EF values of 1.52 (As), 2.86 (Cd), 1.34 (Cr), 1.50 (Cu), 0.96 (Pb), 1.39 (Ni), and 1.21 (Zn), and indicated that human inputs were responsible for enrichment of heavy metals in the area. Our results were also in corroboration with Cai et al. (2019). They reported EF values for Cu (1), Pb (1.54), Zn (1.33), Ni (0.98), Cd (1.55), As (0.68), and Cr (0.71) in agricultural soils of Guangdong and concluded that heavy metals showed less enrichment in the area.

The pollution load index (PLI) was calculated to determine the collective influence of As, Cd, Cr, Co, Cu, Pb, Mn, Ni, and Zn in the soils of the study area. Results showed that 42.4% of samples indicated that soil quality was being deteriorated by heavy metals, while 57.5% samples indicated that soil quality was not impacted by heavy metals. The range of PLI found in the present study was 0.057 to 10.57 with an average value of 1.06. Our PLI results are in corroboration with Charzyński et al. (2017). They reported PLI values of 0.1 to 2.8 in their studies of soils in northwest Poland. Xiao et al. (2019) reported that 57.6% of soil samples in their study area in Shaanxi, China, exhibited moderate pollution, while 43.4% samples showed high pollution of heavy metals (Zn, As, Cd, Cu, and Pb) based on PLI.

The Igeo index was determined for the studied metals (Table 4). Results for Cu, Pb, Zn, Mn, and Ni were above 5, representing extreme pollution for all sampling sites. As and Co Igeo results indicated that 31.4% and 0.32% of sampling sites showed no pollution, 25.9% and 12.2% moderate pollution, 17.2% and 7.09% strong pollution, 8.54% and 3.87% high pollution, 4.5% and 3.06% very high pollution, and 12.2% and 73.3% severe pollution. The Igeo results for Cd indicated no pollution for all sampling sites. Mehr et al. (2017) reported Igeo values for Cd

**Table 4** CF, EF, and Igeo indices for the studied heavy metals

Contamination factor (CF)									
	As	Cd	Cr	Co	Cu	Pb	Mn	Ni	Zn
Min	0.33	0.05	0.01	0.04	0.09	0.09	0	0.08	0.21
Max	144.67	27.45	4	4.59	5.72	27.25	9.15	3.47	3.87
Mean	6.64	2.77	0.49	0.61	0.94	1.37	0.64	0.63	0.81
Enrichment factor (EF)									
	As	Cd	Cr	Co	Cu	Pb	Ni	Zn	
Min	0.22	0.15	0.0040	0.16	0.07	0.14	0.06	0.12	
Max	854.39	308.57	9.95	11.71	42.68	69.32	22.25	82.14	
Mean	16.67	10.55	0.95	1.17	2.25	5.37	1.43	2.37	
Geoaccumulation index (Igeo)									
	As	Cd	Cr	Co	Cu	Pb	Mn	Ni	Zn
Min	0.15	0.0001	3.51	0.87	11.04	7.55	270.93	6.66	216.58
Max	65.32	0.05	983.36	92.12	717.45	2187.49	535,351.70	278.55	3918.41
Mean	3.00	0.01	121.94	12.27	117.87	109.84	37,237.76	50.43	818.96

(0.32), As ( 0.8), Co ( 0.99), Cr ( 1.45), Cu (0.24), Ni ( 1.05), Pb (1.64), and Zn (1.33) and reported no pollution to strong pollution of heavy metals in the area which are consistent with our results. Xiao et al. (2019) also reported Igeo values for Cd (1.78), Zn (0.75) and As (0.39), signifying accumulation of these metals in the soils of Shaanxi.

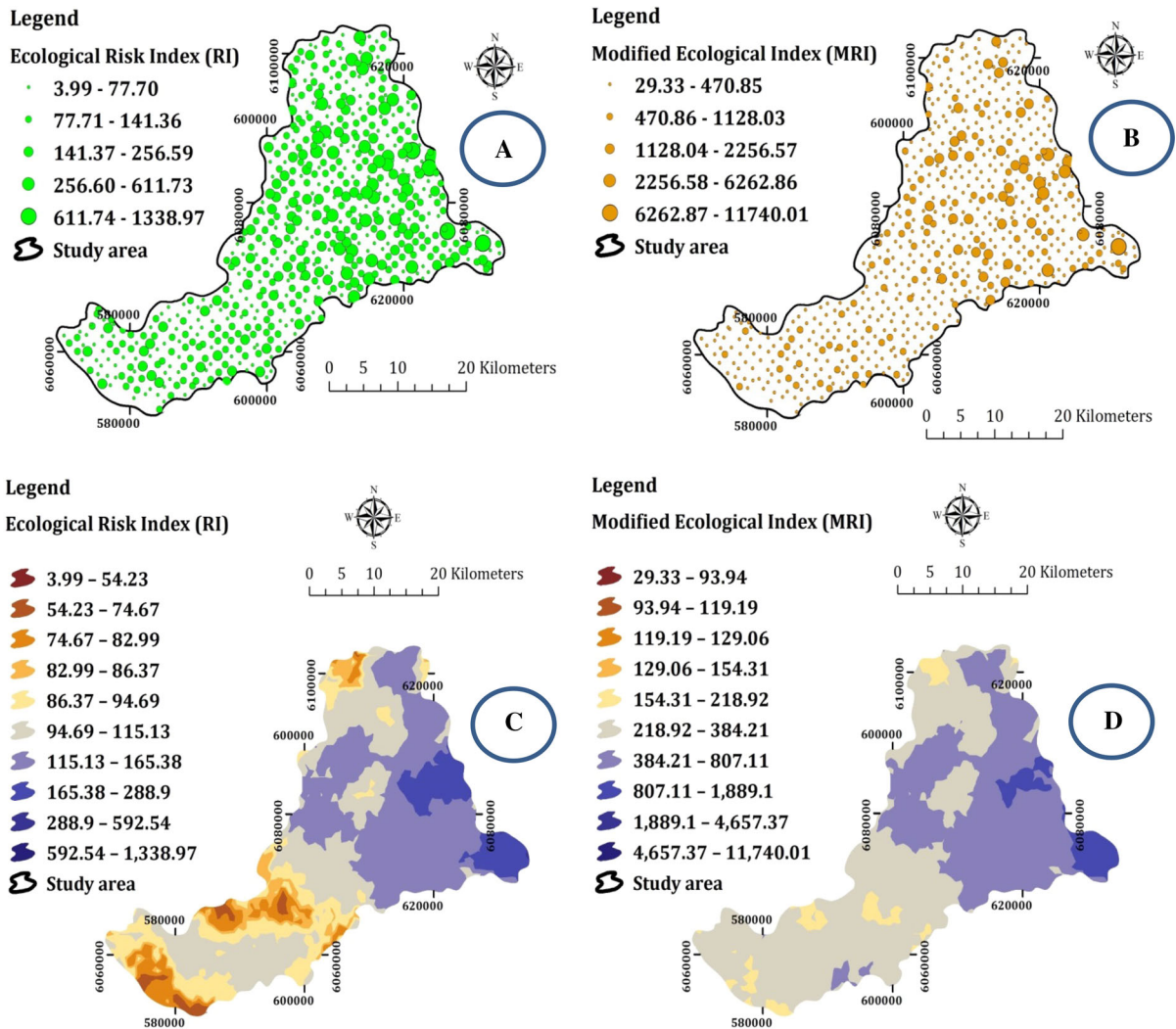
**Ecological risk assessment (RI and MRI)**

The potential ecological risk (RI) was determined for the studied metals (Table 5). The results of potential ecological risk (RI) for Cd showed that 11.9% of sites

reported low risk, 53.7% moderate risk, 27.7% considerable risk, 5.4% high risk, and 1.12% of sites showed very high risk, respectively. RI results for As showed that 94.5% of sites showed low risk, 3.8% moderate risk and 0.8% high risk and very high risk, respectively. RI values for Co, Cr, Mn, Ni, and Zn were below 40, indicating low risk from these metals. RI values for Pb showed that 99.6% of sites showed low risk. However, the average RI value was less than 150, signifying low ecological risk. Islam et al. (2017) reported RI values as follows Cr (2.8), Ni (2.9), Cu (14), As (51), Cd (130), and Pb (11) in their study on agricultural soils of Bangladesh and concluded that the

**Table 5** Ecological risk appraisal of metalloids in soils of the study area

	Potential ecological risk of each metal (Er)									RI
	As	Cd	Cr	Co	Cu	Pb	Mn	Ni	Zn	
Min	0.67	1.53	0.03	0.22	0.44	0.47	0.00	0.42	0.21	3.99
Max	289.33	823.47	8.00	22.95	28.60	136.25	9.15	17.35	3.87	1338.97
Mean	13.27	83.10	0.99	3.06	4.70	6.84	0.64	3.14	0.81	116.55
	Modified potential ecological risk of each metal (mEr)									MRI
	As	Cd	Cr	Co	Cu	Pb	Ni	Zn		
Min	0.44	4.36	0.008	0.81	0.37	0.70	0.28	0.12		7.11
Max	1708.7	9257.1	19.89	58.53	213.40	346.5	111.2	82.14		11,797.7
Mean	33.34	316.4	1.90	5.85	11.26	26.86	7.16	2.36		405.23



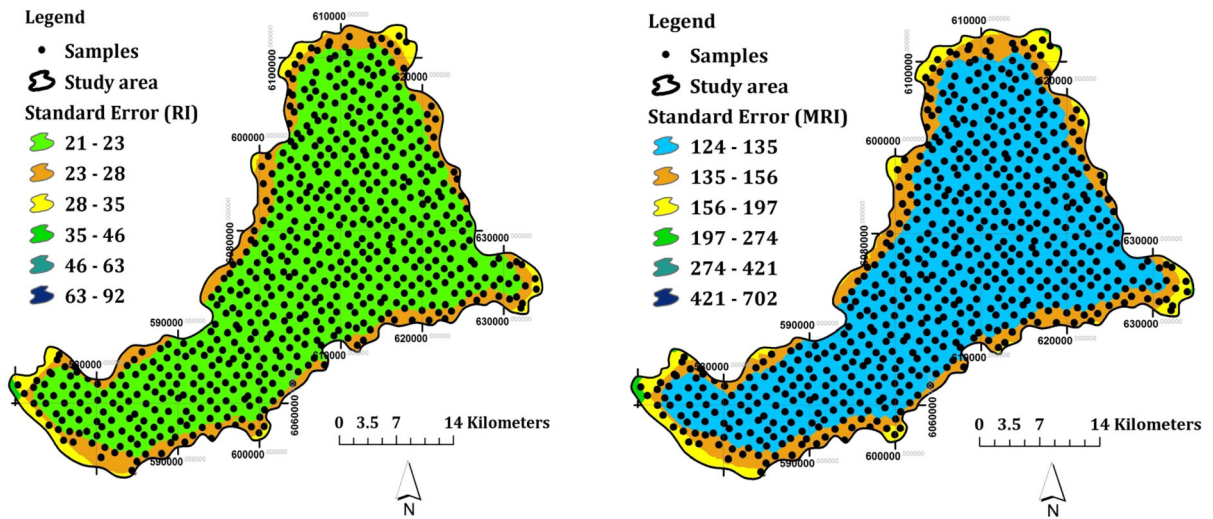
**Fig. 7** Maps showing the spatial distribution of RI and MRI in the study area. **a** and **b** show point data, **c** and **d** show kriged distributions

use of fertilizers and pesticides has great impact on soil heavy metals concentrations. In this study, RI values showed that Cd is the prime contaminant in the area, which agrees with Islam et al. (2017), indicating that agricultural management is a potential source of metals accumulation in the Northern Ireland soils. Zhao et al. (2020) reported RI values for Cd (85.51), Cu (6.67), Zn (0.81), Pb (4.94), Ni (3.88), and Cr (1.18) in soils of southeastern China and concluded that Cd was the prime contaminant in the soils of their study area; this agrees with the results of this study.

The modified potential ecological risk (MRI) was assessed for the studied metals (Table 5). MRI values

for As, Cd, and Pb indicated that 84.1%, 5.8%, and 81.2% of study sites showed low risk, 11.1%, 20.3%, and 10.3% moderate risk, 2.2%, 23.8%, and 6.7% considerable risk, 1.1%, 23.3%, and 1.2% sites high risk, and 1.2%, 26.6%, and 0.32% very high risk, respectively. MRI values for Cr, Ni, Zn, and Co were less than 40, demonstrating low risk. The MRI values for Cu showed that 99% of the sites were low risk. The spatial maps and prediction standard errors for spatial distributions of ecological risk indices are given in Figs. 7 and 8. Pandit et al. (2020) reported MRI values of Cr (2.5), Ni (1.6), Cu (8.8), Zn (8.4), Pb (5.7), and Co (8.8) while working on soils of the Siwalik region in India; from these values, they concluded that





**Fig. 8** Prediction standard errors for spatial distributions of ecological risk indices

anthropogenic activities contributed to the heavy metals concentrations in their soils. The prediction standard errors for the spatial distributions of ecological risk indices in most parts of the study region ranged between 21–23 and 124–135 for RI and MRI, respectively (Fig. 8). Grunwald et al. (2004) discussed different sources of errors on spatial maps and mentioned that the total number of samples, distances, and short-range spatial variability could either reduce or increase uncertainty and standard errors. Kriging is preferred for local estimation but can have higher errors along the boundary of a study area (Fig. 8). Least-square interpolation algorithms such as kriging tend to smooth out local details regarding the spatial variation of the mapped attribute, with small values typically overestimated and large values underestimated (Grunwald et al. 2004). The results of CF, EF, RI, and MRI all indicated that Cd and As are the key contaminants in the study area, while the Igeo index suggested that Mn, Cu, Cr, Pb and Zn are the main contaminants in the area. Attention should be paid to reduce pollution by these heavy metals, and remediation options may need to be considered.

**Other considerations and future needs**

The Tellus XRFs data provide total elemental analysis; however, the risks posed to human and environmental health depend on factors other than the total concentration of a given heavy metal (Morgan 2013;

Brevik et al. 2020). The various pollution indices utilized in this study are one way to provide better information about health risks than just total elemental concentration (Antoniadis et al. 2017; Aitta et al. 2019). Soil chemical properties including organic matter content, pH, the percentage and mineralogy of clays, antagonism/competition/interactions with other ionic species in the soil, the presence of minerals that may sorb heavy metals, and the species (oxidation state) of various heavy metals present, among others, also make a difference in health risks (Brevik 2009; Morgan 2013; Khaledian et al. 2017; Vardhana et al. 2019; El-Ramady et al. 2020). Therefore, there is a need for future studies that investigate these aspects of heavy metals in the soil and their influence on human and environmental health in this region.

**Conclusions**

This study appraised the presence of heavy metals and investigated relationships between the metals and selected physiochemical soil properties in the urban, industrial, and agricultural soils of a portion of Northern Ireland. Among the appraised metals, Mn had the highest maximum values and Cd had the lowest. Pearson’s correlation analysis demonstrated strong correlations between heavy metals and physiochemical properties, indicating that all or most of the heavy metals had the same origin and the impact of

physiochemical properties on the availability and mobility of heavy metals. Multivariate analysis indicated that anthropogenic and lithogenic factors have influenced heavy metals contents in the study area. Furthermore, CF, EF, and Igeo revealed that heavy metals showed modest to extreme enrichment in the soils. The results of ecological risks analyses showed that Cd is the primary heavy metal accountable for ecological threats in the soils of this portion of Northern Ireland. In impending studies, other probable heavy metals sources and sinks (leakage, wind erosion, and evaporation) might be measured to precisely compute a whole mass balance to advance the formation of heavy metals inventories.

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**Author contributions** A.K. and G.E. were involved in the conceptualization; A.K. and G.E. were involved in the data curation; A.K., V.K., and G.E. were involved in the formal analysis; A.K., V.K., G.E., and E.C.B. were involved in the investigation; A.K., V.K., G.E., and E.C.B. were involved in the methodology; A.K., V.K., G.E. contributed to software; G.E. was involved in the supervision; A.K., V.K., G.E., and E.C.B. were involved in the validation; A.K., V.K., G.E., and E.C.B. were involved in the writing and editing.

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**Availability of data and materials** The LUCAS 2009 topsoil database has been available for download since September 2013. Tellus is a project of the Geological Survey Ireland and is funded by the Department of Communications, Climate Action and Environment. All data from Tellus are made available free of charge online. The data that support the findings of this study are openly available at <https://www.gsi.ie/en-ie/programmes-and-projects/tellus/Pages/default.aspx>.

**Code availability** There is no custom code for this research. All software used in this paper are mentioned in the Materials and Methods.

#### Compliance with ethical standards

**Conflict of interest** The authors declare no conflicts of interest.

**Consent to participate** No human research subjects were used in this study.

**Consent to publish** No human research subjects were used in this study.

**Ethical approval** No human or animal research subjects were used in this study.

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