The following article appeared in Journal of Hazardous Materials Advances Volume 9, February 2023, 100243 and may be found at: https://doi.org/10.1016/j.hazadv.2023.100243

Journal of Hazardous Materials Advances 9 (2023) 100243

Contents lists available at ScienceDirect

Journal of Hazardous Materials Advances

journal homepage: www.elsevier.com/locate/hazadv

Optimal GIS interpolation techniques and multivariate statistical approach to study the soil-trace metal(loid)s distribution patterns in the agricultural surface soil of Matehuala, Mexico

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ARTICLE INFO

Keywords:
Trace metal(loid)s
Soil contamination
Spatial distribution
GIS
Interpolation

ABSTRACT

Soils in the past mining areas are susceptible to trace metal(loid)s deposition and pose a health risk to humans. The purpose of this work is to evaluate the distribution patterns and contamination characteristics of trace metal(loid)s in the agricultural surface soils of past mining regions. The contaminated site is near an abandoned mining area, which is surrounded by land used for maize cultivation. The multivariate statistical approach and GIS interpolation techniques are often used in spatial distribution mapping to predict metal(loid)s concentrations for arsenic (As) and other trace metals (i.e., Al, Fe, Mn, and Sr) in areas that have not been sampled. The mean relative error (MRE) and root mean square error (RMSE) values were used to evaluate and correlate the efficiency of deterministic interpolation (Inverse Distance Weighting- IDW, Local Polynomial- LP, and Radial Basis Functions- RBF) as well as geostatistical interpolation methods (Ordinary Kriging- OK and Empirical Bayesian Kriging- EBK). The results revealed that all interpolation techniques predicted the mean concentration of trace metal(loid)s in soil with moderate accuracy. It was found that agricultural soils contained arsenic enrichment (up to 185 mg/kg), up to five times the background concentrations (35 mg/kg), and 8.5 times the Mexican guidelines (22 mg/kg). The statistical analysis with the cross-validation method revealed that IDW and LP consistently provided the most accurate predictions of trace metal(loid)s concentrations while OK, EBK, and RBF techniques are less accurate. Overall, these GIS interpolation techniques help in the prediction of trace metal(loid)s concentrations at unexplored sites and establish the requirement for the amelioration of agricultural surface soil.

1. Introduction

Due to the progressive growth of both agricultural and industrial sectors, soil quality has continued to deteriorate in the developing world (Soffianian et al., 2014; Othman et al., 2017; Akopyan et al., 2018). One of the main contributors to soil contamination in large mining areas nowadays is waste that contains heavy metal(loid)s. Once heavy metal(loid)s enters the soil, they can damage plants, impeding their development (Ruíz-Huerta et al., 2017; Chen et al., 2018). Moreover, heavy metal(loid)s in the soil can affect human health (Zhao et al., 2012; Li et al., 2014). Mining activities in the surrounding areas of Matehuala, Mexico stretch back over 200 years and are currently focused on the extraction of skarn deposits of heavy metals from a small hill range known as El Fraile that forms the western border of the Sierra Madre Oriental (Castro-Larragoitia et al., 1997; Razo et al., 2004; Martínez-Villegas et al., 2013). The deposition of demolition waste and blast furnace slag from a metal ore smelter that was operated around 60 years ago are among the other polluting activities in Matehuala city (Manz and Castro, 1997; Martínez-Villegas et al., 2013). Previous studies have revealed that the dissemination of tailings, mining wastes, sludges, and slags has affected children, wildlife, and crop production while contaminating soils, water, sediments as well as the ecological environment in Matehuala region that surrounds the mining district (Castro-Larragoitia et al., 1997; Razo et al., 2004; Chapa-Vargas et al., 2010; Martínez-Villegas et al., 2013). The majority of heavy metals in the environment are contaminated by anthropogenic activities, such as mineral extraction and smelting operations, manufacturing and industrialisation, as well as agricultural and domestic use of metal(loid)s and metal(loid)-containing compounds (Shallari et al., 1998; He et al., 2005). Even though heavy metals are naturally occurring elements throughout the earth's crust, they appear in the surface soil due to anthropogenic activities (Tchounwou et al., 2012; Briffa et al., 2020).

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https://doi.org/10.1016/j.hazadv.2023.100243
Received 30 November 2022; Received in revised form 13 January 2023; Accepted 18 January 2023

The soil classification method coupled with a soil map, and spatial interpolation are the two basic approaches that may be used to predict soil attributes in unsampled regions (Voltz and Webster, 1990; Goovaerts, 2011). All contour maps, including soil maps, represent the spatial distribution of soil properties as the conflict of polygons with fixed values (Goovaerts, 2011). Each mapping unit is assumed to be generally homogenous in this representation, and significant variations are expected to occur at the edges (Webster and Beckett, 1968). Geostatisticians have developed a number of methods to combine both mapping units and point measurement techniques in the estimation of soil attributes (Goovaerts, 2011; Mendes et al., 2020; Barrena-González et al., 2022). To locate areas with significant levels of contamination, and correlate the levels with potential sources, soil mapping has been carried out in many countries to investigate the spatial distribution of heavy toxic metal(loid)s in contaminated soils (Piedade et al., 2014). Such maps have been produced using various interpolation techniques with georeferenced soil sample points that are often organised in a grid (Long et al., 2020; Saha et al., 2022a).

The regional heavy metal(loid) distribution patterns and pollution assessments in soil are progressively integrating GIS-based techniques and multivariate statistical analysis. The majority of the studies have effectively differentiated between anthropogenic and natural sources for specific heavy metal(loid)s (Hou et al., 2017). The spatial distribution of heavy metal(loid)s has been estimated by using different types of statistical and geostatistical methods (Kumar et al., 2012; Saha et al., 2022a). The spatial distribution of soil parameters at the unsampled sites could not be determined using the traditional statistical approach. Therefore, geostatistics is an effective technique for analysing the spatial distribution of soil attributes and also for significantly reducing the variation of evaluation error and associated costs (Davis et al., 2009; Nickel et al., 2014; Bhunia et al., 2018; Fischer et al., 2021). GIS has become a valuable tool for monitoring environmental pollutants. To fill in any voids in the cognitive design model, interpolation of data can be utilised to forecast values in unsampled areas using adjacent observed values (Tobler, 1970; Fischer et al., 2021). Often a limited number of soil samples are obtained from study sites for chemical analysis due to time and resource limitations, resulting in poor datasets that potentially undermine experimental findings and conclusions (Liu et al., 2004). Through the multivariate statistical analysis, it was shown that the usage of sewage sludge, municipal wastages, suspended solids, and livestock manure was correlated with increasing levels of heavy metal(loid)s in soil (Cang et al., 2004; Wang et al., 2005; Dai et al., 2007; Huang et al., 2015). This study has employed appropriate GIS interpolation techniques, along with multivariate statistical analysis techniques, such as principal component analysis (PCA), cluster analysis (CA), and Pearson's correlation analysis for a realistic assessment of the extent of surface soil contamination.

The five common interpolation techniques for soil pollution studies and contamination mapping are (i) Inverse Distance Weighting (IDW), (ii) Local Polynomial (LP), (iii) Ordinary Kriging (OK), (iv) Empirical Bayesian Kriging (EBK), and (v) Radial Basis Functions (RBF). The IDW method involves a deterministic interpolation technique that generates an approximated area by taking into consideration the similarity of measured points and estimating the optimum weight required to limit the impact of locations at a specific distance (De Smith et al., 2007). The previous studies of trace metal(loid)s in the soil of Matehuala established that IDW was the optimal interpolation technique (Martínez-Villegas et al., 2018; Saha et al., 2022a; Saha et al., 2022b). The LP interpolation method only uses points in the predefined neighbourhood to match the specific polynomial order (Saha et al., 2022a). The Kriging method is based on a sequential interpolation technique that applies a semivariogram model to predict unknown values based on distance and variations in measured values (Paramasivam and Venkatramanan, 2019). According to the study of Zare-mehrjardi et al., (2010), cokriging and ordinary kriging were superior to the inverse distance weighting (IDW) technique for predicting the geographical distribution of soil characteristics. In another study, Robinson and Metternicht (2006) estimated the soil salinity, acidity, and organic matter using three distinct GIS interpolation techniques: kriging, IDW, and RBF. In iterative methods, RBF interpolation is a complex technique for creating high-precision interpolants of unstructured data, perhaps in elevated regions (Buhmann and Dyn, 1993). The precision of the interpolation depends on how accurately the boundaries and contaminated areas are defined; this also influences how accurately any pollutant is assessed (Xie et al., 2011). Various studies were conducted on the effectiveness of the preceding spatial interpolation techniques (Gotway et al., 1996; Panagopoulos et al., 2006; Shi et al., 2009). Moreover, the number of significant factors of the trace metal(loid) studies are also discussed in this work, such as: the traditional soil sampling approach, identifying, and correlating characteristics of trace metal(loid) concentrations as well as identification of sources.

The current study focuses on a contaminated environment where arsenic (As) is the predominant component in a mixture of inorganic contaminants, although other metals are not of much concern. The mobility of As is thought to be regulated in calcium-dominated conditions by calcium arsenates. However not being as strongly soluble as those found in other complex elements, these are known to remove significant quantities of arsenate from aqueous solutions (Rodríguez-Blanco et al., 2007; Martínez-Villegas et al., 2013; Mahlknecht et al., 2023). In natural environments, arsenic occurs in two forms, namely, arsenite, As(III), and arsenate, As(V), both forms being anions. The sorption of these two As forms on iron containing minerals have different characteristics (Deng et al., 2018; Vromman et al., 2018). As is mostly found as As(III) in reducing environments (Dixit and Hering, 2003). However, arsenite is much more toxic and soluble in water (Brusseau and Artiola, 2019; Saha et al., 2022c). The slower excretion rates for arsenite compared to arsenate and organic arsenic may be a factor in arsenite's higher level of toxicity (Kuivenhoven and Mason, 2019). In this study, the spatial distribution patterns of arsenic (As), aluminium (Al), iron (Fe), manganese (Mn), and strontium (Sr) in agricultural soil are evaluated using the geostatistical interpolation techniques, namely, OK and EBK, as well as deterministic interpolation techniques such as IDW, LP, and RBF. Following a different systematic approach, we investigated the effect of different types of soil (Calcisol and Gypsisol) on trace metal(loid)s concentration and try to associate concentration gradients with various contamination sources in semi-arid calcareous environments. The objectives of this study are as follows: (i) to estimate metal(loid)s concentration in agricultural surface soil around Cerrito Blanco, Matehuala, San Luis Potosi, Mexico, (ii) to evaluate the level of uncertainty surrounding a contaminated area using various interpolation techniques, and (iii) to investigate the relationship between the predicted accuracy and variance in soil trace metal(loid) components at the local level using various types of GIS interpolation techniques with multivariate statistical analysis.

2. Study area

The soil sampling sites were located in the town of Cerrito Blanco, around 6.8 km from the municipality of Matehuala in the northern part of the state of San Luis Potosi, Mexico. It covers a total land area of around 25.28 hectares and is located between 23°40′08″ N latitude and 100°34′44″ W longitude (Fig. 1). This area was chosen based on information obtained from previous studies on heavy metal distribution in agricultural soil (Chapa-Vargas et al., 2010; Martínez-Villegas et al., 2013; Ruíz-Huerta et al., 2017). Over the past 200 years, mining and metallurgical activities in Cerrito Blanco-Matehuala have resulted in a highly polluted area (Razo et al., 2004; Martínez-Villegas et al., 2018). The area has semi-arid weather, and the predominant vegetation is a microphallus shrub that supports modest cow grazing, mixed with maize croplands (Chapa-Vargas et al., 2010). In semi-arid conditions, this study

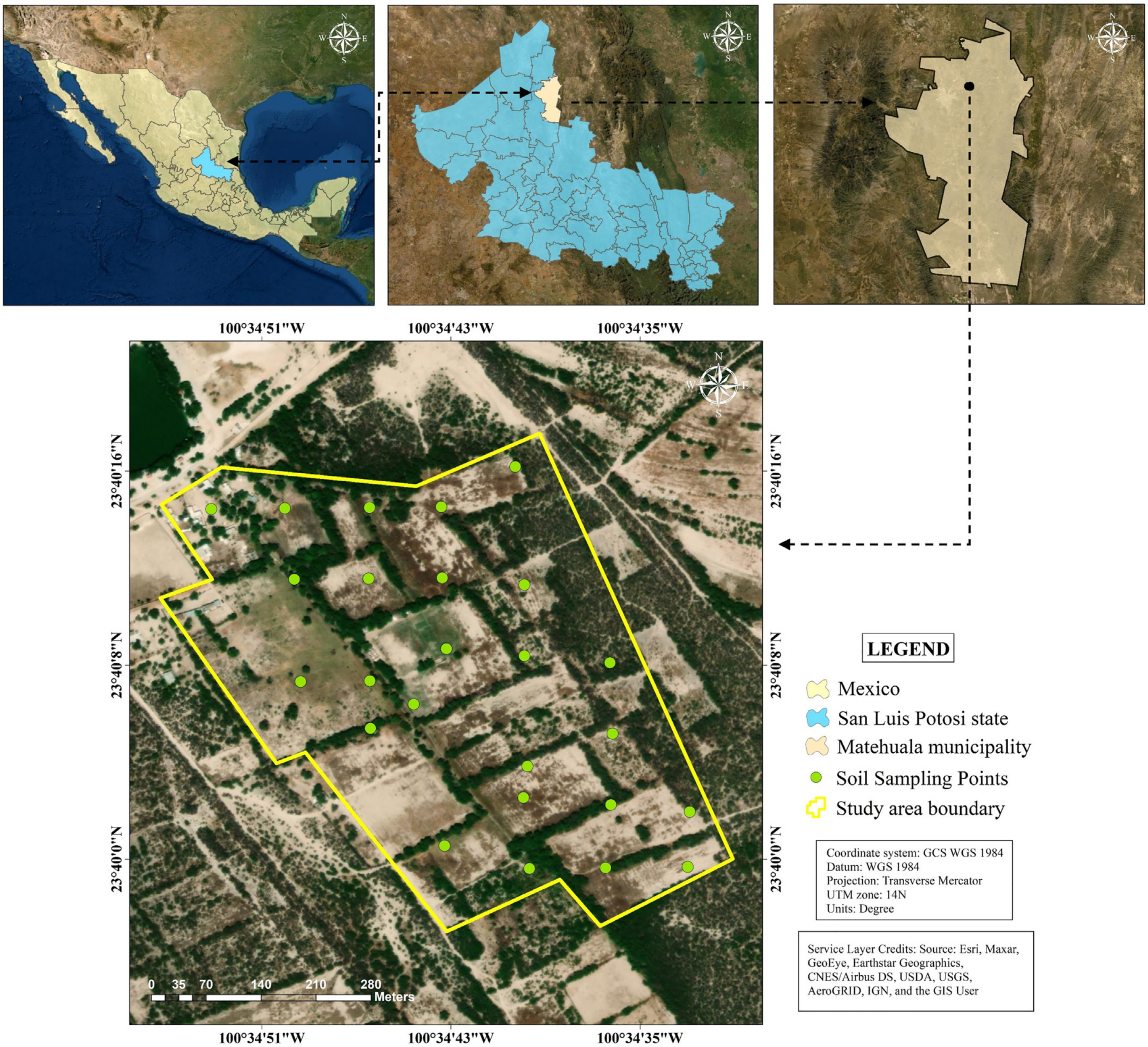

Fig. 1. Location of soil sampling points in the agricultural soil of Cerrito Blanco, Matehuala, San Luis Potosi, Mexico.

allowed the identification of the dispersion routes followed by arsenic and heavy metals to impact the environment. Calcisol ($CaCO_3$) and Gypsisol ($CaSO_4$) are the major soil variations in this region and there is one cultivating season due to low annual precipitation of 300 to 500 mm (INEGI, 2009). Therefore, surface water from the Matehuala municipality is utilised to irrigate crops as a supplement to rainwater (Ruíz-Huerta et al., 2017). Cultivating rain-fed maize in small plots maintained by subsistence farmers is a common practice in this study area, where contaminated water is frequently used to supplement the rainfall throughout the crop-growing season due to a lack of alternate supplies (Ruíz-Huerta et al., 2017). Other polluting activities in the area include the deposition of construction wastes, the extraction of heavy metal skarn deposits from El Fraile hill, and the slags from a metal ore smelter that operated in Matehuala until the 1960s (Castro-Larragoitia et al., 1997; Manz and Castro, 1997; Razo et al., 2004; Martínez-Villegas et al., 2013; Saha et al., 2022d). However, there is a lot of evidence that the soil in this study area is highly contaminated and it's a cause for concern.

3. Materials and methods

3.1. Soil sampling and analysis

For surface soil sampling, 25 soil samples were collected using an auger at the depth of 0–5 cm from the agricultural land using a systematic sampling approach based on a statistical method, according to the Mexican guidelines and recommendations (NMX-AA-132-SCFI-2001) (Secretaria de Economía, 2006). The soils were conserved in plastic bags and preserved at 4°C until soils were homogenised and crushed. The soil samples were air-dried at room temperature before being sieved to filter out the fraction <2 mm. Agricultural surface soil samples were digested using a slightly modified version of the ISO 11466:1995 procedure (Ruíz-Huerta et al., 2017). Inductively coupled plasma optical emission spectroscopy (ICP-OES) was used to estimate the metal(loid)s in digested soil samples (USEPA, 1994; Martínez-Villegas et al., 2018). An aqua regia solution (HNO_3:HCl, 3:1) was added to 1.0 gm of soil in a beaker. Total recoverable trace metal(loid)s in soils can be determined using this procedure (Ruíz-Huerta et al., 2017; Martínez-Villegas et al., 2018). Aqua regia digestion does not release residues that are not considered relevant for estimating elements of environmental importance's mobility and behavior (Niskavaara et al., 1997; Martínez-Villegas et al., 2018). The soil sample sites were georeferenced using a GPS device made by Garmin called an Etrex Personal Navigator.

3.2. Interpolation approaches for the spatial distribution of metals

A range of deterministic (i.e., generate maps from observed values) and geostatistical (i.e., tools make use of the statistical characteristics of the observed points) interpolation techniques were applied in the present study to derive the spatial distribution of trace metal(loid)s. This includes some widely used deterministic interpolation techniques, such as Inverse Distance Weighted (IDW), Local Polynomial (LP), Radial Basis Functions - Completely Regularized Spline (RBF-CRS), and geostatistical interpolation techniques, specifically Ordinary Kriging (OK) and Empirical Bayesian Kriging (EBK) (Arslan and Turan, 2015; Bhunia et al., 2018; Saha et al., 2022a). A brief overview comprising the technical underpinning of each technique used in this paper is presented below.

3.2.1. Deterministic interpolation techniques

3.2.1.1. Inverse distance weighted (IDW). One of the most widely used deterministic interpolation methods in soil research is the IDW which combines multivariate statistical analysis with GIS. In compliance with the inverse of distance to power, the weighing approach provided spatially adjacent points with higher weight than distant points, which was conceptually reasonable (Qi et al., 2020). For this work, IDW calculations were performed on adjacent observed points. It is implied that the known observed points regulate themselves independently of one another (Bhunia et al., 2018; Saha et al., 2022a).

$$Z = \sum_{i=1}^{n} \left(Z_i/d_i^p\right) / \sum_{i=1}^{n} \left(1/d_i^p\right)$$

Where Z denotes the approximate value at an interpolated point; Z_i denotes the computed values at point i; n denotes the total number of values obtained in interpolation; d_i denotes the distance between interpolated value Z and the computed value Z_i, and p denotes the weighting power.

3.2.1.2. Local polynomial (LP). The LP interpolation technique has been utilised in meteorological research for more than 50 years (Gilchrist and Cressman, 1954). This technique adjusts a unique polynomial equation for each region based on the maximum and minimum observed values, regions, observed neighbourhood types, and kernel types (Johnston et al., 2001; Antal et al., 2021). The purpose of polynomial interpolation is to identify a polynomial that can access a group of specified observation points. Overall, a global polynomial may cover the entire surface; however, it cannot perfectly match the surface when there is more natural variation (Liao et al., 2018). The LP method provides a number of advantageous characteristics, including efficiency and the ability to successfully detrend data in a variety of geostatistical models (Gribov and Krivoruchko, 2011).

$$Z_i = \left(1 - \frac{d_i}{R}\right)p$$

Where Z_i is the mean observed values made at the ith measurement point, d_i represents the difference between observed and predicted points, R denotes the neighbouring area carried into consideration, and p is the order of the polynomial function defined by the operator.

3.2.1.3. Radial basis functions (RBF). The RBF (also known as Spline) refers to a set of precise interpolation techniques that are based on artificial neural networks (ANN) (Johnston et al., 2001; Antal et al., 2021). The technique includes five distinct basis functions: thin-plate splines (TPS), spline with tension (ST), inverse multi-quadratic function (IMQ), completely regularized spline (CRS), and multi-quadratic function (MQ). RBF provides predictions about new values based on an operator-specified region, and each predicted value must carry through each measured value (Xie et al., 2011; Antal et al., 2021).

$$Z(x) = \sum_{i=1}^{m} a_i f_i(x) + \sum_{i=1}^{n} b_j \psi\left(d_j\right)$$

Where d_j represents the distance between each observed sample point and the estimated point x, and $\psi(d_j)$ represents the radial basis functions. The trend function $f_i(x)$ is regarded as a component of the basis for polynomials with degree m; n is the total number of known points considered in the interpolation.

In this study, we have assessed the completely regularized spline: radial basis functions (RBF-CRS). The following functional equations are used for this radial basis function case (Xie et al., 2011).

$$\psi(d) = \ln\left(\frac{cd}{2}\right)^2 + E_1(cd)^2 + \gamma$$

Where d represents the difference between the estimated and observed points, c represents the smoothing factor, E_1 represents the modified Bessel function, and γ denotes the Euler's constant. The methodological flowchart (Fig. 2) illustrates how soil samples are obtained as well as methods for identifying toxic metal(loid) and other trace metal(loid)s in the agricultural surface soil.

3.2.2. Geostatistical interpolation techniques

3.2.2.1. Ordinary Kriging (OK). Advanced geostatistical techniques like kriging are part of a different class of interpolation methods. Kriging interpolation approaches have been extensively used to characterise spatial differences in soil characteristics and have been regarded as one of the advanced geostatistical approaches that may produce an optimum and constructive estimation for an unsampled area (Hu et al., 2016). The basis of Ordinary Kriging (OK) is a statistical model that includes autocorrelation, or the statistical correlations between the observed points (Ghosh et al., 2020). However, geostatistical algorithms not only have the proficiency to generate a prediction surface but also offer some indication of the reliability or efficiency of the prediction (Oliver and Webster, 1990; Ghosh et al., 2020). OK emphasizes the function that is spatially associated. OK functions as a spatial interpolation predictor and is represented as the following weighted sum of the data:

$$Z(x) = \sum_{i=1}^{n} \lambda_i Z\left(x_i\right)$$

$Z(x)$ denotes the estimated value at point x, $Z(x_i)$ denotes the observed value at position x, λ_i indicates the weight applied to the residual of $Z(x_i)$, and n represents the number of sample data utilised at specific points within the neighbourhood.

3.2.2.2. Empirical Bayesian Kriging (EBK). Empirical Bayesian Kriging (EBK) optimizes the most challenging components using a sub-setting and simulation approach. EBK varies from traditional kriging methods in that it considers the error generated by predicting the semivariogram model (Krivoruchko, 2012). The predicted semivariogram is assumed by the EBK method to be the exact semivariogram for the interpolated area and a linear prediction with varying spatial dispersion (Ghosh et al., 2020). EBK is a combination of two geostatistical concepts: intrinsic random function kriging (IRFK) (Yaglom, 1955; Chiles and Delfiner, 1999) and linear mixed model (LMM, which is also known as simple kriging with the external trend in the geostatistical literature) (Banerjee et al., 2003; Diggle and Ribeiro, 2007; Schabenberger and Gotway, 2017). Since the same procedures are employed for IRFK and LMM model fitting, and integration of two distinct processes into a single computational model (Krivoruchko and Gribov, 2019; Gribov and Krivoruchko, 2020).

$$Z_i = y\left(s_i\right) + \varepsilon_i,\ i = \overline{1 \ldots \ldots K}$$

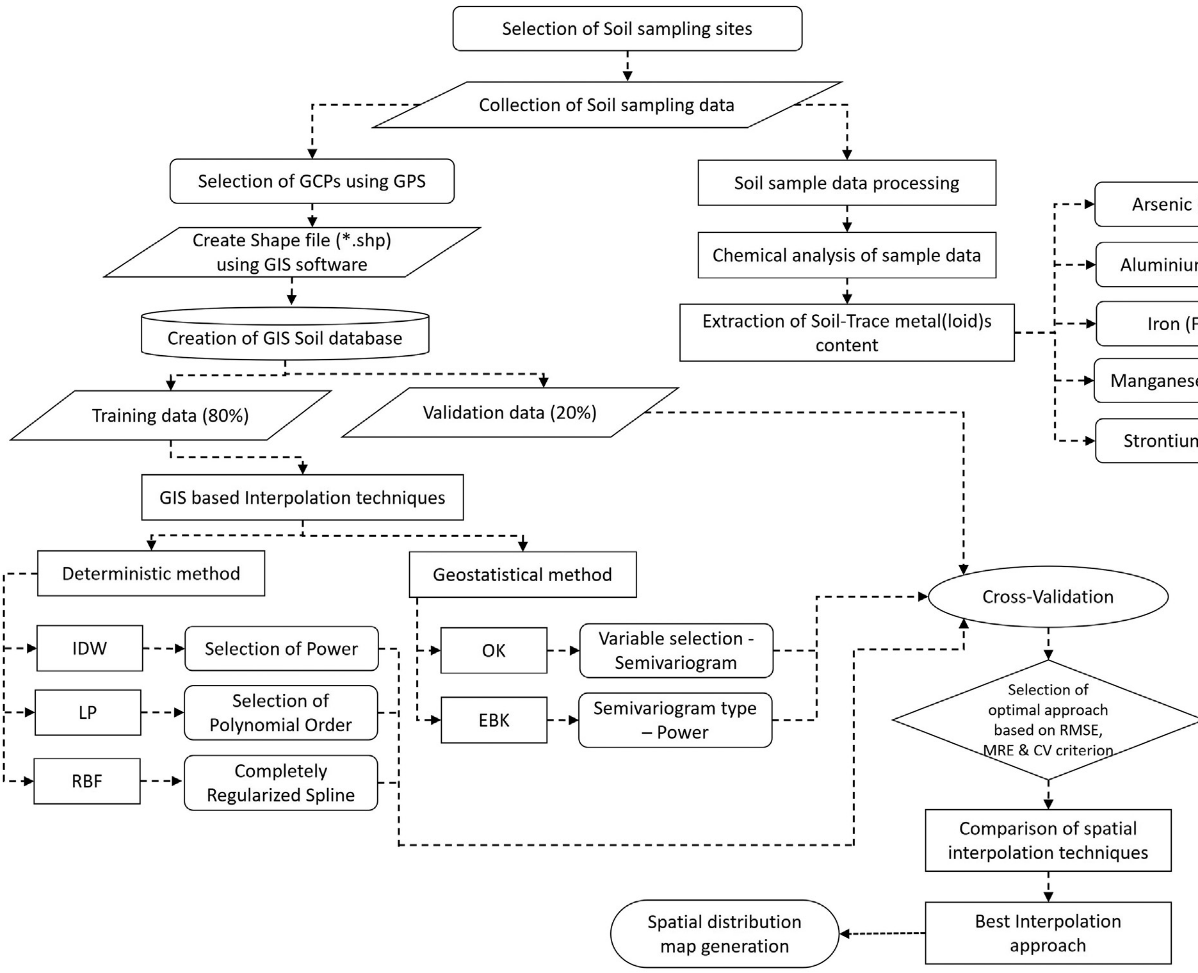

Fig. 2. Workflow for data acquisition and interpolation techniques.

Where Z_i represents the measurement made at the observed point s_i, $y(s)$ denotes the Gaussian process under the study location s, ε_i represents the measurement error, and K is the total number of measurements.

3.3. Cross-validation and accuracy assessments

The approaches that are frequently used to compare the interpolation techniques include cross-validation and validation using an independent data set. In this study, cross-validation was used because of the limited sample set. The distribution of error related to interpolation techniques was assessed by randomly parting observed sample points into two parts: i) for interpolation (80%) and ii) cross-validation (20%) (Ghosh et al., 2012; Hu et al., 2016; Saha et al., 2022a). The 20% of sample points that are set apart for cross-validation were compared to the corresponding predicted values, estimated using referred interpolation techniques. To evaluate the accuracy of predictions, the mean relative error (MRE), root mean square error (RMSE), and coefficient of variation (CV) derived from observed and predicted values at each soil sample point in the cross-validation portfolio were compared.

$$MRE = \frac{1}{n}\sum_{i=1}^{n}\left|\frac{Z^*(x_i) - Z(x_i)}{Z(x_i)}\right|$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}\left[Z^*(x_i) - Z(x_i)\right]^2}{n}}$$

$$CV = \frac{standard\ deviation\ of\ predicted\ values}{mean\ of\ predicted\ values}$$

Where $Z(x_i)$ denotes the observed value at the ith point, $Z^*(x_i)$ indicates the predicted value at the ith point, and n represents the number of observed sample data points.

3.4. Multivariate statistical analysis

Alternative methods for assessing the environmental conditions include multivariate statistical analysis (Ustaoğlu and Tepe, 2019; Kukrer et al., 2021). Elemental concentrations are highly variable in the natural environment. Therefore, statistical techniques must be used to interpret the big datasets for environmental information (Oprea, 2005). The data may be simplified, arranged, and classified using multivariate approaches to reveal meaningful patterns (Sheikhy Narany et al., 2014; Arslan and Turan, 2015). Multivariate statistical techniques are applied in environmental research to measure associations among more than two variables while taking their interactions into consideration (Oprea, 2005). The potential sources of the metal(loid)s and their distribution mechanisms were identified using a correlation matrix and principal component analysis (PCA). However, PCA was performed to reduce the dimension of the multivariate structure of the dataset while optimising the outcomes of subsequent analysis applied for assessing the environmental impact of metal(loid) contamination.

3.4.1. Principal component analysis (PCA)

Principal component analysis (PCA) is a widely applied technique for analysing a high-dimensional dataset that underpins the linear transformation of highly correlated data into uncorrelated variables (referred to as “principal components”) (Singh et al., 2004; Jolliffe and

Cadima, 2016; Goswami and Kalamdhad, 2022). The newly created uncorrelated components successively capture the variance in the dataset, i.e., first component captures most of the variance, and so on. A cumulative distribution plot of components is used to select the optimum number of components required for capturing the variance in the dataset. For example, in many highly correlated datasets, only the first few components could successfully capture more than 90-95% of the variance and thus can be used to replace a large dataset with a smaller set of components for any subsequent analysis. Thus, PCA facilitates the greater interpretability of the dataset with a reduced number of components while minimising the information loss. To assess the suitability of a dataset for a PCA analysis Kaiser-Meyer-Olkin (KMO) test is often applied. The KMO test is used to assess the suitability of the dataset for factor analysis (related analysis), in the present case, this approach is applied to normalised data using varimax rotation (Zhang et al., 2018; Keshavarzi and Kumar, 2020). Thus, before performing the PCA analysis, the feasibility of implementing this method on the dataset was assessed using the KMO measure of sample adequacy and Bartlett's test of sphericity (Akopyan et al., 2018). The following system of equations is used to mathematically express the linear transformation of multivariables as principal components:

$$Z_{ij} = x_{1i}y_{1j} + x_{2i}y_{2j} + x_{3i}y_{3j} + \cdots\cdots + x_{ik}y_{jk}$$

Where Z is the component value, x refers to the component loadings, y refers to the variable's measured value, j and k indicate the sample number and the total number of variables.

3.4.2. Cluster analysis

Cluster analysis is an unsupervised classification approach that facilitates pattern recognition and reveals the fundamental structure or inherent activity of a dataset. The approach utilises user-specified feature characteristics of the data in order to categorise the system's components into groups or clusters based on their proximity or similarity (Arslan and Turan, 2015; Alam et al., 2022). Data points that share similar characteristics/features were clustered together into one class during cluster analysis. Cluster analysis is a comprehensive approach that integrates a series of algorithms for identifying the optimum number of clusters and organising data across different clusters such that variance within the clusters is minimum while variance between the clusters is maximised. The K-mean clustering and Hierarchical clustering are among the widely applied clustering approaches (Shrestha and Kazama, 2007). The paper will be using hierarchical cluster analysis, which will also create a dendrogram plot illustrating the dataset in a layered structure. Furthermore, to prevent misclassification owing to significant variations in data dimensionality, cluster analysis, and PCA were performed on experimental data that had been normalised using z-scale transformation (Liu et al., 2003). The significance of variables with small variances usually grows, whereas the influence of variables with high variances tends to decrease (Singh et al., 2004).

4. Results and discussion

4.1. Descriptive statistics of metal(loid)s in soils

The concentrations of As, Al, Fe, Mn, and Sr in agricultural soils, together with soil permissible limits or reference values of metals at regional and international levels are shown in Table 1. The permissible or reference values of metal(loid)s in agricultural soil were As (22 mg/kg) (Martínez-Villegas et al., 2018), Al (10000 mg/kg) (US EPA Ecological Soil Screening Level for Aluminium, 2014), Fe (300 mg/kg) (Iyama et al., 2021), Mn (85 mg/kg) (Ashraf et al., 2021), Sr (200 mg/kg) (Essel, 2017). The mean concentrations of As, Al, Fe, Mn, and Sr in soil were 76.90 mg/kg, 241.85 mg/kg, 6933.74 mg/kg, 275.27 mg/kg, and 352.66 mg/kg, respectively. The concentration data of different trace metal(loid)s was shown using the box and whisker plots in Fig. 3. The mean concentrations of Al were within the soil's reference value. In relation to the permissible limit of metals in the soil, As, Fe, Mn, and Sr concentrations were 3.50, 23.11, 3.24, and 1.76 times higher, respectively. The metal(loid)s concentration showed decreasing trend (Fe > As > Mn > Sr > Al) in comparison with permissible or reference values, disclosing a concentration pattern in soil considerably impacted by industrial and past mining activities. The mean concentration of the most hazardous metal(loid) in this case, namely As was at an alarming level in mining areas, mining–smelting areas, and rapid urban areas in this study. As and Sr showed moderate to significant spatial variation with coefficients of variation (CV) of 50.05% and 36.56%, respectively. Although Sr present in the study location shows its natural origin and derives from the gypsum bed (Saha et al., 2022b). All the studied metal(loid)s in the soil samples had kurtosis values greater than zero, which show that their distributions are non-normal with heavy tail, representing values that are often far from the mean in extreme ranges (Cohen et al., 2020). Overall, the selected study area has higher concentrations of metal(loid)s than the majority of places studied, especially for As and Sr.

4.2. Spatial distribution of metal(loid)s

The spatial distributions of As, Al, Fe, Mn, and Sr in agricultural soil were estimated using various interpolation techniques and ArcGIS software, and the results are indicated in Fig. 4. All the metal(loid)s had a high concentration area (exception of Al metal), showing that anthropogenic activities in this area had a detrimental impact on the elemental concentrations. Fig. 4 indicates that greater than 90% of the study area had significant As concentrations (> permissible limit values), i.e., the small portion of the area of the northeast region was in the low concentration zone. The As contamination was observed over a gradient from a high of 90 mg/kg on the west and northwest side to a low of 8.03 mg/kg in the northeast; this could have been impacted by the use of As-contaminated groundwater used in irrigation. The mean Fe and Mn concentrations in agricultural soil are somewhat higher than the permissible limits compared with As and Sr, considering the fact that these two are prevalent in this area, as shown by the standard and permissible limit values. However, the ore smelting, slagging, and deposition of metal dust in soil from mining sites may be related to the enrichment of As concentration in soils in some specific areas. Similar spatial distribution patterns were observed for As, Fe, Mn, and Sr, with contamination dispersed along the study area. Sr in its elemental form occurs naturally in many compartments of the environment, including rocks, soil, water, and air. Sr compounds can move through the environment fairly easily because many of the compounds are water-soluble. Sr is always present in the air as dust, up to a certain level (Strontium (Sr) - Chemical Properties, Health and Environmental Effects, 2023). The spatial distribution patterns of each trace metal(loid) examined in agricultural soil may be connected to the consequences of anthropogenic activities, irrigation with contaminated waters, and their sources.

4.3. Comparison between deterministic and geostatistical interpolation techniques

To choose the optimal technique must be made comparisons based on the mean relative error (MRE), root mean square error (RMSE), and coefficient of variation (CV) in relation to several concepts. The prediction error will be depending on the size of the data, when it is near 0, it indicates that the prediction error is acceptable and ideal. The mean relative error is effective if the error is close to 0, which means that the root mean square error should be close to 1, and the prediction error can be evaluated to check if it is optimal (Park and Stefanski, 1998; Li et al., 2021a). So, the optimal technique is acceptable when the mean relative error is close to 0 and the root mean square error is close to 1; and the MRE and RMSE are near and the least (Li et al., 2021a; Saha et al., 2022a). The relative distribution of data points in a series around the

Table 1
Statistics of metal(loid)s concentrations in agricultural soils of Cerrito Blanco area.

	As	Al	Fe	Mn	Sr	pH
Mean (Measured)	76.90	241.85	6933.74	275.27	352.66	7.86
Standard Error	7.70	7.50	199.16	6.91	25.79	0.02
Median	72.18	252.17	7063.16	284.00	346.61	7.87
Standard Deviation	38.48	37.51	995.78	34.54	128.94	0.09
Sample Variance	1481.07	1406.70	991579.00	1192.86	16626.53	0.01
Kurtosis	1.41	6.17	6.19	4.09	9.41	6.12
Skewness	0.72	-1.93	-1.60	-1.84	2.49	-2.15
Range	177.34	193.22	5390.74	155.62	675.97	0.35
Minimum	8.03	107.33	3392.66	162.18	178.81	7.60
Maximum	185.37	300.55	8783.40	317.80	854.78	7.95
Sum	1922.48	6046.25	173343.53	6881.81	8816.54	N/A
Coefficient of variation (CV) (%)	50.05	15.51	14.36	12.55	36.56	N/A
Count	25	25	25	25	25	25
Confidence Level (95.0%)	15.89	15.48	411.04	14.26	53.23	0.05
Permissible/Reference values (mg/kg)	22	10000	300	85	200	N/A

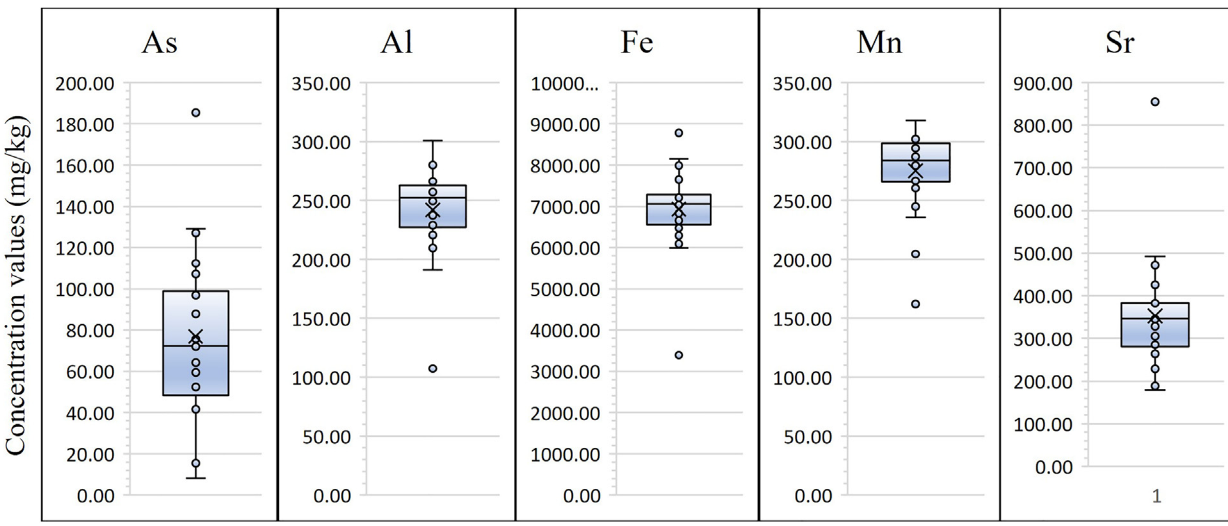

Fig. 3. Box and whisker plots of the different trace metal(loid)s concentrations.

mean is statistically measured by the coefficient of variation (CV). The CV demonstrates the degree of data variability in a soil sample in comparison with the value of the mean concentration of metal(loid)s. The higher degree of dispersion around the mean indicates an increase in the value of CV. The lower value of CV is preferred because the limited dispersion of data values is low relative to the mean. Therefore, in this study lower CV value has been considered.

The optimal interpolation approach for five selected metal(loid)s was identified by comparing deterministic and geostatistical interpolation techniques under various parameters in Table 2. According to the required criterion, the MRE value is closest to 0, the least values of RMSE and CV are given priority to compare the three deterministic interpolation techniques (IDW, LP, and RBF-CRS) and two geostatistical interpolation techniques (OK and EBK) for finding the optimal interpolation approach. In summary, the study area's five different soil- metal(loid) concentrations may be interpolated using the following techniques: IDW of power 2 for As, LP of polynomial order 3 for Al, IDW of power 3 for Fe, IDW of power 3 for Mn, and IDW of power 3 for Sr. According to the outcomes for the comparison of interpolation techniques, the IDW deterministic interpolation approach is the most suitable interpolation technique in this study.

The spatial distribution of trace metal(loid)s contamination map will often be more accurate when there are more collections of samples (Mueller et al., 2001). The collection of numerous samples is typically not practicable due to the expense of sample collection and processing (Xie et al., 2011). It can be observed in Fig. 4, approaches with lower CV values were better at mapping than those with higher CV values. Generally, the error estimations of interpolation techniques are minimised when the RMSE, MRE, and CV values are minimal for the trace metals, as shown in Table 2. The interpolation inaccuracy and the variations in interpolation techniques increased along with the higher values of RMSE, MRE, and CV. The interpolation techniques are used to decide how discrete data from a map are transformed into continuous data (Xie et al., 2011). The accuracy of the interpolation techniques depends on how effectively the interpolation approach represents the spatial variation and correlation patterns of soil properties. The study revealed that deterministic interpolation approaches are often simple to execute because models require fewer input parameters. In comparison, the geostatistical interpolation approach requires more effort to implement. But geostatistical approaches are usually acceptable as long as the following procedures are taken: statistical assessments, transformation of data, fitting of semivariance function, spatial structure analysis, etc (Xie et al., 2011; Liao et al., 2018; Saha et al., 2022a). However, the key concern for managing agriculture and the environment is having a comprehensive awareness of heavy metal distribution. The study shows that the IDW interpolation approach is optimal in comparison with any other geostatistical method for As, Fe, Mn, and Sr metal(loid)s based on the results of efficiency and error estimation of interpolation techniques. It may be concluded that IDW analysis uses a linear combination of values from the captured locations of events to provide a weighting estimate to the unknown locations by inversely calculating the distance between the event location to be estimated and the points captured to

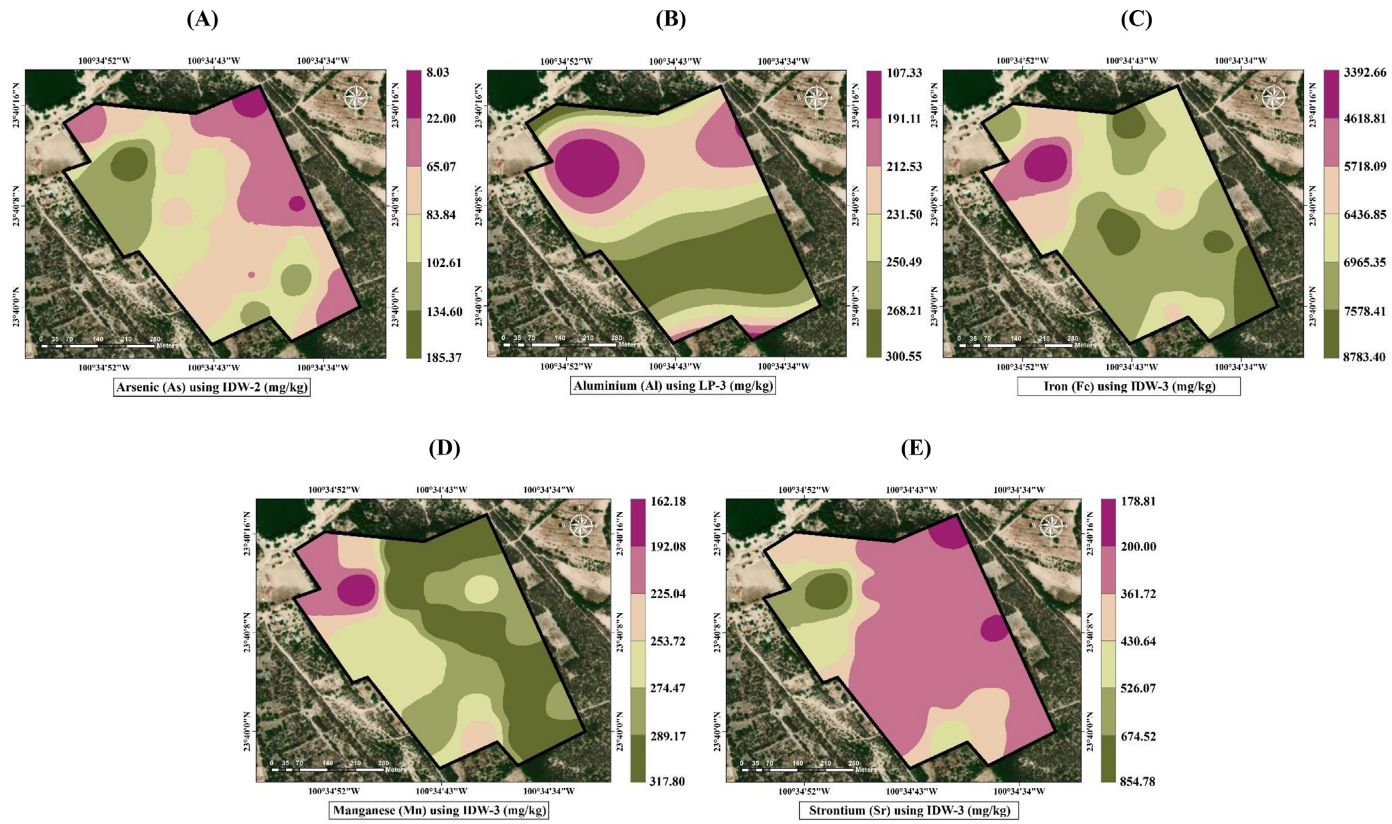

Fig. 4. Spatial distribution patterns of metal(loid)s of As, Al, Fe, Mn, and Sr content.

Table 2
Selection of optimal interpolation techniques for metal(loid)s concentrations in soil (optimal techniques are highlighted in bold).

Metal(loid)s	Predictive Errors	IDW1	IDW2	IDW3	LP1	LP2	LP3	OK	EBK	RBF-CRS
As	RMSE	24.05	**23.48**	23.45	22.15	25.73	20.41	22.23	26.32	28.31
	MRE (mg/kg)	0.35	**0.35**	0.36	0.27	0.31	0.26	0.31	0.38	0.29
	CV (%)	12.51	**13.04**	14.9	23.47	24.62	25.24	15.52	9.64	47.2
Al	RMSE	24.16	24.11	24.08	26.37	26.19	**21.11**	24.52	24.3	25.6
	MRE (mg/kg)	0.08	0.08	0.08	0.08	0.07	**0.07**	0.07	0.07	0.09
	CV (%)	8.01	8.46	8.74	6.86	6.02	**8.63**	7.91	5.98	8.43
Fe	RMSE	902.42	866.64	**840.79**	976.55	990.75	1081.39	857.53	949.63	1114.88
	MRE (mg/kg)	0.06	0.06	**0.06**	0.08	0.07	0.06	0.06	0.06	0.07
	CV (%)	4.95	4.23	**3.63**	5.3	3.89	6.42	3.94	4.66	8.94
Mn	RMSE	21.3	19.09	**17.39**	20.56	20.92	17.2	22.13	20.47	18.63
	MRE (mg/kg)	0.04	0.04	**0.04**	0.03	0.05	0.04	0.04	0.03	0.04
	CV (%)	3.7	3.26	**3.3**	4.35	3.78	4.29	3.99	3.64	2.9
Sr	RMSE	47.86	43.92	**40.71**	53.85	44.8	57.47	45.68	55.63	42.25
	MRE (mg/kg)	0.11	0.1	**0.09**	0.12	0.09	0.14	0.11	0.15	0.08
	CV (%)	12.62	11.57	**10.75**	14.03	17.12	17.86	12.36	9.43	12.08

obtain a weighting estimate (Ikechukwu et al., 2017). But for Al the LP interpolation technique is suitable, which may be the reason of limited exploration distance that might result in an empty region of the surface without any restrictions other than the sampling locations.

4.4. Pearson-correlation matrix of trace metal(loid)s

Pearson's correlation analysis was used to assess the relationship between various trace metal(loid)s in the soil. It is a detailed statistical tool that indicates the level of dependence between two variables (Belkhiri et al., 2010; Khound and Bhattacharyya, 2017). The computed correlation coefficients are shown in Fig. 5. The extraction of Fe as $FeCO_3$ solids might be the source of the weak correlation between As and Fe in surface soil (Lee et al., 2010; Khound and Bhattacharyya, 2017). One may also reach the conclusion that As might be released into irrigated water as a result of the oxidative dissolution of FeO(OH) or MnO(OH) as bacteria oxidise organic materials to produce energy (Ohno et al., 2005; Shamsudduha et al., 2008). Trace metal(loid)s typically have a significant positive correlation coefficient and may derive from the same or nearly adjacent sources.

It is observed that As and Sr (*r* 0.85), Al and Fe (*r* 0.78), Al and Mn (*r* 0.55), and Fe and Mn (*r* 0.55) all have a substantial positive correlation, indicating a consistent lithogenic source. The significant positive correlation between Fe and Mn in the soil shows that these two metals naturally originate through the decomposition of soils, rocks, and minerals (Khound and Bhattacharyya, 2017). Al, Fe, and Mn possess high positive correlations with each other, indicating the three metals' similar behaviour in this region, which is confirmed by the geographic dispersion. These results revealed that the parental materials and mineral components of the soil were responsible for these metals occurring natu-

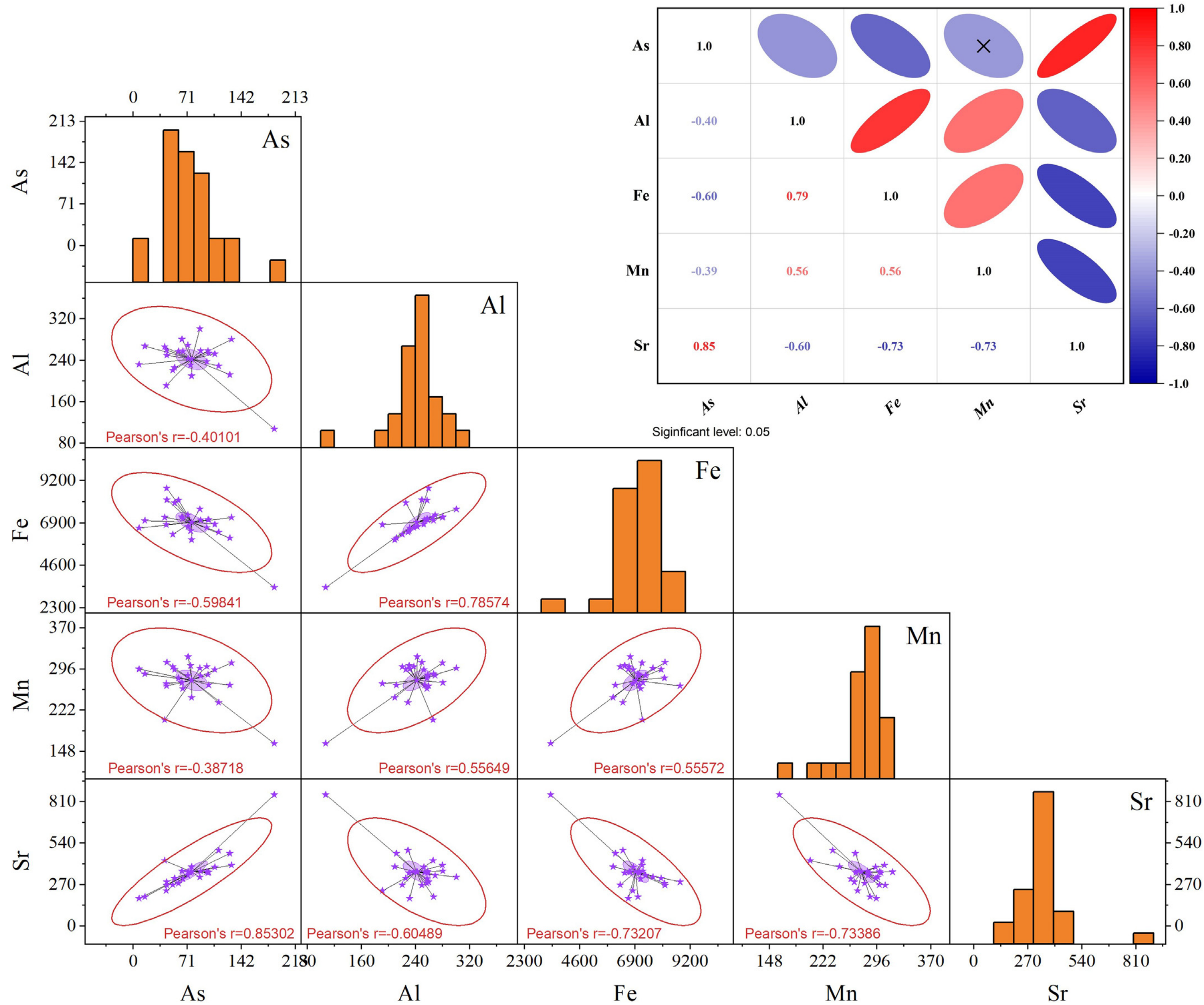

Fig. 5. Scatter matrix analysis to determine relationships between various trace metal(loid)s.

rally in the soil. The main natural sources were the parent rock materials since these trace metals are mostly found in the Earth's crust (Taylor and McLennan, 1995; Keshavarzi and Kumar, 2020). As and Sr had a negative correlation with other trace metals as opposed to metal(loid)s synergistic effects and indicating the different sources. In general, As in the soil could derive from As-contaminated irrigational water, pesticides, industrial emissions, etc., (Chung et al., 2014) and Sr in the soil is sourced from the lithosphere composition of the crustal, groundwater, and mining activities (Li et al., 2021b; Wang et al., 2021). In this study, the source of As is irrigational contaminated water from metal arsenate dissolution upstream, while Sr derives from natural gypsum beds.

4.5. Principal component analysis for trace metal(loid)s in soil

To evaluate the possible source of arsenic contamination and other trace metals in the regions, we have furthermore used principal component analysis (PCA) to cluster elements that have related patterns of distribution. Five trace metal(loid)s were chosen for each to compute the extensive concentration levels in accordance with the principal component score. Since it has been confirmed that principal component analysis (PCA) with VARIMAX normalised rotation is an effective method for identifying the source of hazardous pollutants; it was utilised to ascertain the distribution of trace metal(loid)s in the soils by several researchers (Hu et al., 2013; Arslan and Turan, 2015; Keshavarzi and Kumar, 2020). The results of the Keiser-Meyer-Olkin (KMO) measure of sampling adequacy and the Bartlett's test of sphericity were 0.64 and 97.04 ($p<0.001$), respectively, showing that the dimension reduction was reasonable. The fraction of typical variation generated by inherent variables is measured by the KMO, which is an indicator of sampling adequacy. Principal component analysis may be effective when the KMO value is close to 1 (Khound and Bhattacharyya, 2017). The results of Bartlett's test of sphericity determine either the variables are not connected, or the correlation matrix is an identical matrix (Shrestha and Kazama, 2007; Khound and Bhattacharyya, 2017).

Two main components were identified by PCA, which accounted for 84.62% of the data variability. The first principal component (PC1) and the second principal component (PC2) represented 70.10% and 14.52% of the total variance, respectively (Fig. 6). Eigenvalues are a reliable selection criterion for component factors, according to the Kaiser Criterion (Kaiser, 1960). It is appropriate to consider an eigenvalue as a factor if it is more than one. But according to the variance extraction criteria, it should be more than 0.7; if the variance is lower than 0.7,

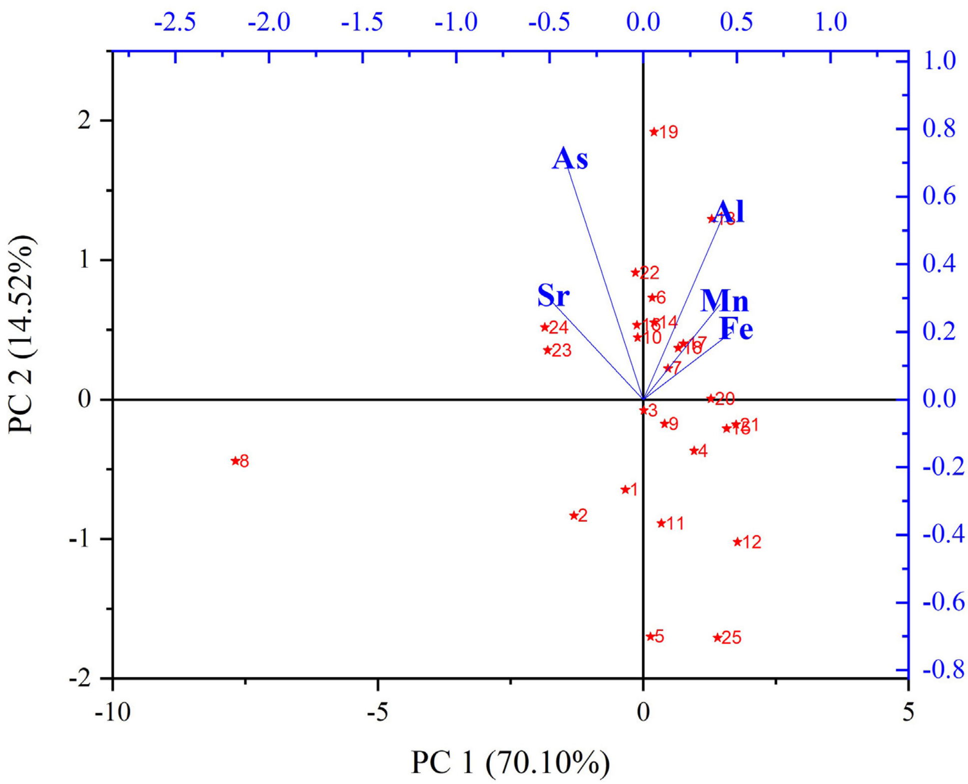

Fig. 6. PCA component weights plot.

Table 3
PCA results of trace metal(loid)s (components greater than 0.32 are shown in bold).

Metal(loid)s	Principal components PC1	PC2	Communalities
As	-0.416	**0.704**	0.606
Al	**0.427**	**0.545**	0.640
Fe	**0.471**	0.199	0.779
Mn	**0.411**	0.282	0.593
Sr	-0.503	**0.328**	0.887
Eigen value	3.205	1.026	
% of variance	70.10	14.52	
Cumulative %	70.10	84.62	

then it should not be considered as a factor (Factor Analysis - Statistics Solutions, 2021). In the assessment of PCA patterns, component values more than 0.71 are generally considered good, while those below 0.32 are considered inadequate (Nowak, 1998; Hu et al., 2013). The loadings of each trace metal(loid)s in principal components and communities are shown in Table 3, and the PCA component weights plot is shown in Fig. 6. The interpretation of the difference between PC1 and PC2 should not be based on absolute values only, but rather on the interaction of all PCA components. Based on the directions of the corresponding arrows on the weighting plot (Fig. 6), PC 1 discriminates observations or soil samples based on heavy metal concentration with high contents listed on the right half and low ones listed on the left. PC1 was influenced by Al, Fe, and Mn, contributing 70.10% of the total variance and it might be affected by both lithogenic and anthropogenic influences. Fertilisers and agrochemicals might be the minor contributors to trace metal(loid)s contamination in agricultural soils. In addition to the anthropogenic factors of trace metal(loid)s to fertilisers, limestone and compost were already present in the surface soil in lower concentrations. However, it has been observed that Fe and Mn are present in the parent components of soils. Since the agricultural activities in the region used rigorous tillage for many years, long-term fertiliser usage may be a significant contributor to the formation of trace contaminants in the surface soil of the study area (Castro-Larragoitia et al., 1997; Razo et al., 2004). The component loading of Al (0.427) was not particularly in PC1; it was partially reflected in PC2 with a component loading of 0.545 and indicating a quasi-independent behaviour within this group. PC2 was dominated by As, Al, and Sr, contributing to 14.52% of the total variance with As having the highest loading of 0.704. It indicated mining and metallurgical factors, specifically for the deposition of construction wastes, metal ore smelter, and past mining activities. The significant source of As contamination in industrialised and water-scarce areas of Cerrito Blanco, Metehuala is irrigational groundwater. It can be a significant source in this study because all irrigational water for agricultural land came from the Matehuala-Cerrito Blanco hydraulic complex. During the single growing season, the hydraulic complex is used to supplement rainwater for irrigation (Ruíz-Huerta et al., 2017). The surface and groundwater in the area have been contaminated due to previous mining activities (Martínez-Villegas et al., 2018). Moreover, it is assumed that previous mining operations contributed to the deposition of harmful metal(loid)s in the soil in this study region.

4.6. Cluster analysis of trace metal(loid)s

The relative homogeneous groups of trace metal(loid)s were identified in this study using a hierarchical cluster analysis method. The group average cluster method of linkage with correlation distance type was used to interpret the datasets produced for this study as a measurement of similarity. As and Sr formed a cluster and had a strong correlation with each other, as shown in Fig. 7. The significant correlations were also found between Al and Fe, suggesting similar distribution patterns. But Al and Fe cluster groups were also associated with Mn at a later stage. However, it can be assumed that Mn was not found in interactions with the other trace metal(loid)s, indicating that it was separated from other metal(loid)s in the soils. The dendrogram of cluster analysis suggests that there is a substantial variation between these trace metal(loid)s.

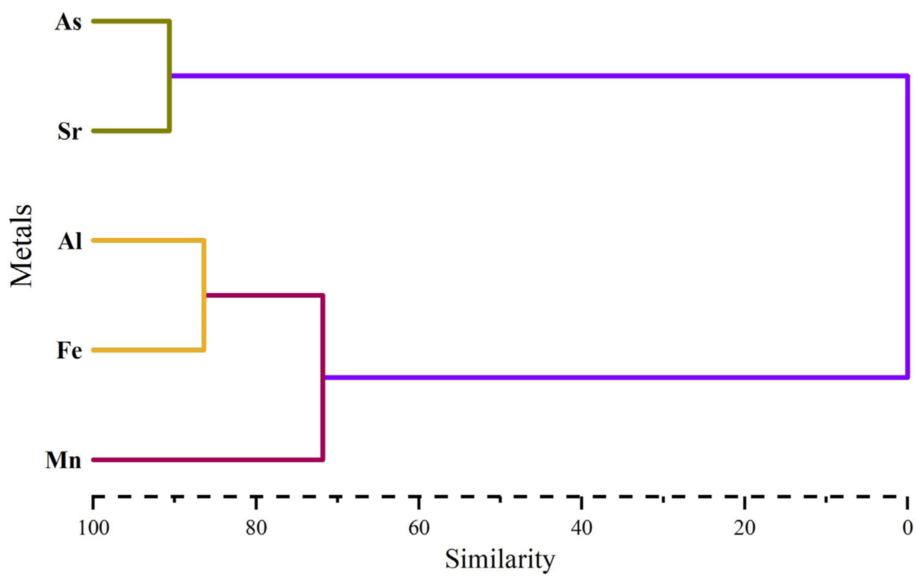

Fig. 7. Hierarchical dendrogram of trace metal(loid)s by cluster analysis.

Based on this study, it is clear that the GIS is an effective technique for data sampling, data processing, and producing graphical output for spatial analysis. The results of the spatial distribution mapping, principal component analysis, Pearson-correlation matrix, and hierarchical cluster analyses are determined on the overall distribution characteristics of the trace metal(loid)s. PCA indicates the quantity and form of contamination that could occur as a result of the variation in the dataset rather than specifically identifying and quantifying contaminant sources (Goswami and Kalamdhad, 2022). Therefore, this study evaluates or verifies the suggestions of the sources by examining a map of soil and satellite images nearby the Cerrito Blanco area, Mexico. Overall, the study indicates that the two groups might be used to classify the trace metal(loid)s in the surface soils. The first group comprised Al, Fe, and Mn which collectively characterise the most significant component. The second group consists of As and Sr; whereas As is predominantly influenced by anthropogenic activity and Sr is mostly originated from natural sources. The results of this study revealed considerable As contamination, which may be due to past mining and metallurgical activities, contaminated irrigational water for farming, and ore smelting. But there is still concern about the toxicity of the agricultural surface soil with negligible concentrations of the other trace metals. To address the concerns of worldwide trace metal(loid)s-soil contamination that occurs within the limitations of current legislation, a technically feasible, ecologically sustainable, and economically acceptable method is needed. It is necessary to conduct an in-depth study on anthropogenic and natural sources of pollution and put into effect a variety of protective methods for soil pollution sources.

5. Conclusions

Agricultural surface soil in the Cerrito Blanco area, Matehuala, Mexico has been studied to estimate the concentration level and spatial distribution pattern of As, Al, Fe, Mn, and Sr. The comparative abundance of various interpolation approaches has led to the use of several algorithms, and studies are being conducted to determine the optimal approach for defining the spatial distribution of trace metal(loid)s. The effectiveness and error predictions of interpolation techniques are used to assess the optimal approaches. The results of this study have revealed that different interpolation techniques have distinctive aspects and applicability contexts. The study indicates that IDW and LP are more suitable interpolation approaches than other geostatistical techniques. IDW is the most effective interpolation approach for the conditions of limited spatial scale, the average number of sampling data, and the systemic approach of data sampling with high spatial autocorrelation. This study also revealed that the assessment of soil sampling data using multivariate statistical approaches, such as principal component analysis and cluster analysis, provides some relevant evidence that is not immediately accessible. The concentrations of all selected metal(loid)s are higher than their permissible limit or reference values in the studied soil, except for Al metal. Fe and Mn concentrations might be influenced by both anthropogenic and natural activities while Sr mostly originated from natural sources, and As came from past mining and metallurgical activities as well as from the As-contaminated groundwater use. The dispersion of industrial operations was spatially coherent with the accumulation of metal(loid)s in agricultural soil. The growth of plants is positively influenced by a few metal components in agricultural soils. But once their levels in agricultural topsoil reach certain limits, they exert a negative influence. In this study area, As, Fe, Mn, and Sr concentrations in agricultural soil exceeded the permissible or reference values at regional and international levels. It indicates that if the concentrations of these metal(loid)s continue to rise, the normal development of plants will be adversely affected. Therefore, necessary action must be taken to stop further enrichment of these metal(loid)s. While the initial risk assessment required limited resources, future work would require more soil sampling, contamination monitoring, and risk further analysis for the intermediate-priority regions.

Funding

This study was financially supported by the Engineering and Physical Sciences Research Council (EPSRC-IAA), UKRI, and Heriot-Watt University, Edinburgh internal research grant (under award number D19R12025) through the Project 748502: Spreading The Chemical Free Arsenic Removal Technology To Various Communities In India, Bangladesh And Mexico.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Arnab Saha: Conceptualization, Methodology, Software, Validation, Formal analysis, Resources, Writing – original draft, Visualization.

Bhaskar Sen Gupta: Conceptualization, Formal analysis, Investigation, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Sandhya Patidar:** Methodology, Validation, Data curation, Writing – review & editing, Supervision. **Nadia Martínez-Villegas:** Formal analysis, Investigation, Resources, Data curation, Writing – review & editing.

Data Availability

Data will be made available on request.

Acknowledgments

Thanks to the Institute of Infrastructure and Environment, EGIS, Heriot-Watt University, Edinburgh. The authors are thankful to The School of Energy, Geoscience, Infrastructure and Society (EGIS), Heriot-Watt University, Edinburgh for providing a student bursary to the first author for doctoral research through the James Watt Scholarship. The author would also thanks to IPICyT, San Luis Potosi, Mexico for providing feedback and support.

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