Research Article

Comprehensive Assessment of Heavy Metals in Soils

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ABSTRACT

Assessment of Heavy Metal (HM) pollution of soil could be index based or model driven. Each suffers from methodological limitations. Classification of regions with respect to different indices gave different results. The paper suggests an assumption-free method to get comprehensive index as geometric mean of chosen indices to reflect overall pollution status of HMs (I_{HM}) in soil as a Composite Index (CI), avoiding selection of weights and scaling.

The proposed aggregation is applicable for all types of indices including those having skewed distributions and can be applied even for data with small sample size. With reduced level of substitutability among component indices, the method facilitates formation of chain indices, satisfies time reversal test, and identifies the critical areas and contribution of the indices to I_{HM}. It produces no bias for polluted or non-polluted regions since effect of outliers is not much. It allows ranking and classification of regions in terms of I_{HM} values and assesses paths of improvement for each region over time. Possible to undertake statistical testing of $H_0: I_{HM\ i} = I_{HM\ j}$ for the *i*-th and *j*-th region at a particular time or $H_0: I_{HM(t)} = I_{HM(t+1)}$ for a particular region at two time periods. Since evaluation criteria/base period values vary across countries, meaningful comparison of countries can be made in terms of progress made on Year-to-Year basis. Based on theoretical advantages, the proposed aggregation by Geometric Mean (GM) is an improvement over the existing methods of finding overall pollution status of HMs (I_{HM}) in soils and is recommended.

Keywords: Heavy metals; Comprehensive assessment; Overall pollution status; Geometric mean; Chain indices; Time reversal test; Path of improvement

INTRODUCTION

Pollution of soil by Heavy Metals (HMs) from natural and anthropogenic sources like domestic/industrial waste water effluents, urban and agricultural runoff, fossil fuel combustion, atmospheric deposition, etc. have emerged as a major threat to human society. Major HMs affecting soils are Cadmium (Cd), Chromium (Cr), Copper (Cu), Lead (Pb), Mercury (Hg), etc. Toxicity of metals increase with concentration. Illustrative list of toxic effects of HMs are: Dysfunction of liver and renal problems by Pb, Cd, and Hg; cognitive functions may be affected by Pb, Hg which may introduce neurological changes; exposure to Cd can cause cancer, hypertension, even affect our reproductive system; Hg may result in fetal abnormalities, etc. [1-3]. Agricultural fields have shown significant contamination with HM. For example, predominant HM pollution of agricultural fields in India due to use of sewage water and industrial effluents were found [4,5]. Widespread HM pollution primarily due to rapid industrialization and urbanization has already affected public health, food safety and even the ecological environment.

Assessments of soil pollution due to HMs are either index based or model driven. While the former approaches involve use of a single pollution, extent of pollution, etc. the later approaches include enrichment factors, ecological hazard index, etc. [6], investigated pollution level due to concentrations of HMs using contaminant index (single factor), Nemerow index, spatial distributions of HMs, and assessed environmental health risk utilizing a United States Environmental Protection Agency recommended model [7]. Studies focusing on one specific HM cannot satisfy interests of landowners. Regulatory authorities and researchers who could be interested in approach of aggregation of several measures from individual sampling units to obtain sample information, based on which estimation of population parameters for further analysis and inferences including effectiveness of remedial action [8]. However, no agreed standard exists for evaluation of soil environmental quality [9].

While enrichment factors indicate (extent of increase of an element in sampling unit relative to average natural abundance; requiring selection of a background composition and also a

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reference element), Ecological risk index reflects the Potential Ecological Risk Index (PERI, or RI), and combine the single index of Ecological Risk Factor (ERI) and the Toxic Response factor of Individual metal (TRI).

Nemerow Pollution Index ($PI_{Nemerow}$) indicates soil quality in terms of the single Pollution Index (PI) and is computed as

$$PI_{Nemerow} = \sqrt{\frac{\frac{1}{n}\sum_{i=1}^{n} PI_i^2 + PI_{max}^2}{n}}$$

Where, PI_{max}: Maximum value of the single pollution index of all heavy metals

n: number of heavy metals.

Several indices are there for measuring effect of single factor, geo-accumulation, ecological risk and resulting human health risk, etc. Comprehensive assessment requires combining such indices on a methodologically sound approach to get a comprehensive index for analysis of overall pollution status of HMs. Clearly, comprehensive index is a Composite Index (CI) which is a real valued function f whose domain consists of set of individual indices, which could be unit-free or with different units, following different distributions and having different degrees of correlations.

Distributions of concentration of HMs vary for different locations, different sections of soil and are not normal for different sections of soil [10]. Large value of Coefficient of Variation (CV) of a HM shows uneven distribution of the HM concentration.

The paper describes limitations of existing indices and methods of obtaining comprehensive index as combination/aggregation of individual indices and suggests an assumption-free method to get comprehensive index reflecting overall pollution status of HMs in soil as a composite index, satisfying desired properties.

MATERIALS AND METHODS

Measures of pollution level of HMs

Single Factor assessment (SF): Concentration of HMs (mg/Kg) in soil are calculated and compared with respective standards. However, such standards vary across countries. Single-factor contaminant index P_i was considered [6] as

$$P_i = \frac{c_i}{s_i} \dots\dots\dots (1)$$

Where, C_i (mg/Kg) is the detected concentration of i-th HM and S_i (mg/Kg) is the evaluation criterion of i-th HM.

Nemerow pollution index P_N is given by

$$P_N = \sqrt{\frac{P_{i.max}^2 + P_{i.ave}^2}{2}} \dots\dots\dots (2)$$

Where, $P_{i.max}$ and $P_{i.ave}$ denote respectively the maximum and arithmetic mean of P_{is}. The index P_N reflects average pollution level of soil in terms of average of various pollutants and also helps to find more serious pollutants [11]. P_N has a one-to-one correspondence with Synthetic Pollution Index (SPI) computed as

$$SPI = \sqrt{\frac{P_{i\,Max} + P_{i\,Min}}{2}} \quad [12].$$

Here, each of P_i and P_N is unit free

Limitations

• Addition of P_i and P_j for $I \neq j$ is not admissible since evaluation

criterion of i-th HM and j-th HM are different, like average speed of two cars. For example, consider A=4/8=0.5 and B=3/12=0.25. Here, (A+B)/2=0.375 which is different from the pooled average of A and B=7/20=0.35.

• P_N is influenced by the maximum score. SPI considers two extreme values of data which could be unrealistic outliers and ignores other observations from the data. Thus, value of P_N or SPI depends heavily on heterogeneity of data. Better could be to find pooled average and pooled Standard Deviation (SD) based on all sample observations.

• Classification of soils based on increasing values of P_i under safe, slightly polluted, low polluted, moderately polluted and highly polluted needs to show efficiency of classification in terms of low value of within-group variance and high value of between-group variance. Same is true for classification on the basis of P_N values [13].

Geological Accumulation Assessment (GEO)

Muller [14], gave the formula for calculating Geo-accumulation index (I_{geo}) to assess pollution level for a single element by

$$I_{geo} = \log_2 \left[\frac{C_{Sample}}{1.5 \times C_{Background}} \right] \dots\dots\dots (3)$$

Where C_{Sample} and $C_{Background}$ denote respectively concentration of a HM in soil and the same in the background, taken from secondary data, which can change due to natural diagenesis. Background level concentration distinguishes concentration of the HM occurring by natural process and due to anthropogenic influence [15]. For each HM, separate geochemical baseline is determined for geologically different regions [16].

Considering I_{geoMax} (maximum I_{geo} value) and I_{geoAv} (average of I_{geo} value) of all HMs in a sample, [17] calculated arithmetic mean, SD, Geometric mean and median for I_{geo} and Nemerow index $\left(I_{IN} = \sqrt{\frac{P_{geoMax} + P_{geode}}{2}} \right)$ separately for eight different HMs without specifying most suitable averaging procedure.

Observations

I_{geo} is relative to the $C_{Background}$ and depends heavily on its value. For a given value of $C_{Background}$, $\frac{I_{geo(1)}}{I_{geo(2)}} \neq \frac{C_{Sample(1)}}{C_{Sample(2)}}$. For example, let $C_{Background}$ for Pb be 36.0 mg·kg^{-1}, $C_{Sample(1)}$=37 units and $C_{Sample(2)}$=38 units. Then, $\frac{C_{Sample(1)}}{C_{Sample(2)}} = \frac{37}{38} = 0.973684$; $I_{geo(1)} = \log_2(0.685185) = -0.5454$ and $I_{geo(2)} = \log_2(0.703704) = -0.507$.

Thus, $\frac{I_{geo(1)}}{I_{geo(2)}} = 1.07574$

Increase of I_{geo} per unit increase in C_{Sample} is different at different range of values. Let values of concentration of Pb in $C_{Sample}(3)$= 74 and $C_{Sample}(4)$= 75 respectively where $C_{Background}$ for Pb is 36.0 like the above example.

Here, $\frac{I_{geo(3)}}{I_{geo(4)}} = 0.959274 \neq \frac{I_{geo(1)}}{I_{geo(2)}}$

I_{geo} could be expressed as $I_{geo} = \log_2 \left[\frac{P_i}{1.5} \right]$ when geochemical background value and evaluation criterion of i-th HM are same. Thus, I_{geo} and P_i are likely to be highly correlated.

C_{Sample} could be better calculated as pooled mean.

$C_{Background}$ may not be available for areas considered as free from any HM pollutants where the geochemical baseline map is under progress. Non-availability of $C_{Background}$ at Brahmaputra valley, North East India was found [18].

Classification based in I_{geo} may not be beyond questions.

Ecological Risk (ER)

ER reflects pollution level due to accumulation of HMs in soil by the interaction effects of several HMs and environmental associations with HMs. Potential ecological Risk Index (RI) integrates ER value of each HM as follows:

$$CF = \frac{C_{Sample}}{C_{Crust}} \quad .... \ (4)$$

$$ER = T_r \times CF \quad .... \ (5)$$

$$RI = \sum E_r \quad .... \ (6)$$

Where, T_r denotes the toxicity coefficient and CF denotes the pollution factor.

Multiplicative aggregation was favored by [19], to obtain Pollution Load Index (PLI) for a station by

$$PLI = \sqrt[n]{\prod CF_i} \quad .... \ (7)$$

Where, $PLI > 1 \Rightarrow Pollution$

For the i-th HM in soil, [20] suggested computation of potential ecological Risk Index (RI) by

$$RI = \sum_{i=1}^{m} E_r^i = \sum_{i=1}^{m} T_r^i \times C_f^i = \sum_{i=1}^{m} T_r^i \times \frac{C^i}{C_n^i} \quad .... \ (8)$$

Where,

C^i: Total amount of i-th HM,

C_n^i: Corresponding background value of the i-th HM

T_r^i: Coefficient of toxicity of i-th HM, taken as 5,1,5,30, and 5, respectively for Cu, Zn, Pb, Cd, and Ni [21].

E_r^i : Single factor ecological risk index of i-th HM and

Higher value of E_r^i indicates higher risk degree of RI.

Classification of regions with respect to different indices gave different results. Unpolluted regions (with $PLI<1$) had heavy contaminations in terms of I_{geo} values [22].

Human Health Risk assessment (HHR)

HHR is a tool to assess and manage adverse health effects on humans beings exposed to HMs in polluted environmental media through ingestion, dermal absorption, and breathing inhalation. It also determines the measures required to comply with the relevant health and safety legislation and, thus, reduce level of occupational injuries and ill-health Potential Non-Carcinogenic health Risk (PNCR) and Potential Carcinogenic health Risk (PCR) for more than one HM can be estimated by sum of HQ for each metal and expressed as unit-free Hazard Index (HI) [23].

$HI>1 \Rightarrow$ existence of PNCR. Total PNCR$>1 \times 10^4$, implies high carcinogenic risk to human beings. If total PNCR$<1 \times 10^{-6}$, PCR due to exposure to HM could be negligible.

Comprehensive assessment

Analytic Hierarchy Process (AHP) was used by [10], to combine four indices (*SF*, *GEO*, *PER* and *HHR*) involving subjective assessment of relative importance of the indices using Saaty's scale consisting of 9-point items (1-9) followed by selection of weights and comprehensive assessment scores (*T*) by weighted sum. Here, higher value of *T* implies lower pollution level. Major disadvantages of AHP are: Determination of importance through subjective approach using pairwise comparisons; Saaty scale

depends on the trial perceptions and is not meaningful [24]; AHP is not a legitimate technique [25]; use of AHP along with others Multi Criteria Decision Making (MCDM) methods was suggested [26]. But, results of Data Envelopment Analysis (DEA) depend significantly on selection of inputs and outputs and number of inputs and output variables [27].

Principal Component Analysis (PCA) was used [12], to determine the sources of soil HMs, without verification of normal distribution of variables under investigations, which is an important assumption of PCA. Non satisfaction of assumptions of techniques used may distort the results. PCA considers the low variance components in the data as noise and thus, those components are thrown away, even if those components play major roles in decision making.

PCA is sensitive to scaling, presence of outliers and may not be suitable for small samples. PCA weights are not constant over time and space and thus, comparisons are difficult. Sava [28], observed PCA weights for data accumulated for 11-years and averaging over each factor for year-wise data were different. Use of PCA was not favoured by Environmental Sustainability Index [29].

Cluster analysis for identification of 10 HM accumulation patterns in 47 soil samples was undertaken [30], and three clusters were found, each with a characteristic composition groups. While Cluster-1 showed high concentrations of Zn and Pb, Cluster-2 had high Cu concentration and major concentrations at Cluster-3 were Cr and Cu implying overlapping of Cluster-2 and Cluster-3. Hierarchical Cluster Analysis (HCA) on normalized datasets was used [31], to avoid problems due to scaling of variables and interconnections among the contaminants. However, non-inferential Cluster analysis assumes that the variables are independent. If variables are not independent, the assumption gets violated requiring further necessary action to deal with the situation.

Comprehensive assessment by weighted sum approach falls on the class of linear model (without testing linearity) with full compensability where poor effect of some pollutants can be well compensated by high effects of other pollutants. Different methods of selection of weights satisfying $W_i>0$ and $\Sigma W_i=1$ give different results and no weighting system is above criticism [32]. The multiplicative aggregation with non-linearity properties by $P_i = \Pi_j I_{ij}^{wj}$ has been proposed [33], which prevents rank reversal phenomenon and practically no substitution problems [34].

Proposed method

Ignoring the stage of selection of indicators, following method by [35] is proposed for Comprehensive Assessment of HMs in Soils as a Composite Index (CI) which avoids selection of weights and scaling of raw data. The method requires ensuring that each indicator/index is positively related to CI.

Set up and methodology

Take pooled average of the sample observations. For a region, consider the vector $X = (X_{1c}, X_{2c}, \ldots \ldots, X_{nc})^T$ showing performance of n-indices in the current period and vector $T = (X_{10}, X_{20}, \ldots \ldots, X_{n0})^T$ showing evaluation criteria/base period values. Instead of arithmetic mean of ratios, overall pollution status of HMs (I_{HM}) as CI for the t-th time period may be defined as the Geometric mean i.e.

$$I_{HM\,c0} = \sqrt[n]{\frac{X_{1c}.X_{2c}........X_{nc}}{X_{10}.X_{20}........X_{n0}}} \quad .... (9)$$

Or equivalently by,

$$I_{HM\,c0} = \frac{X_{1c}.X_{2c}........X_{nc}}{X_{10}.X_{20}........X_{n0}} \quad .... (10)$$

Multiplicative aggregation given in (9) and (10) can be applied for all types of indices including those in percentages or having skewed distribution and can be applied even for data with small sample size. In line with general convention of index value=100 in the base year, one can take $I_{HMc0} \times 100$ to reflect changes in percentage.

Properties

For a given region, each of (9) and (10) depicts overall improvement/decline of soil pollution by HMs in the current year with respect to the target values or base year or previous year by a unit free continuous function satisfying the following desired properties.

- Independent of order of the chosen indices and independent of change of scale

- Reduces level of substitutability among the component indices.

- Facilitates formation of chain indices like $I_{HM20} = I_{HM21} \times I_{HM10}$

- Satisfies time reversal test $I_{HMc0} \times I_{HM0c} = 1$

- Facilitates drawing of I_{HMc0} graph over time showing path of improvement/decline for a region across time for comparison of regions with respect to such paths. Replacing the base period vector by the vector for the previous year gives improvement in I_{HM} on year-to-year basis.

- Produces no bias for polluted or non-polluted regions since I_{HMc0} is least affected by outliers.

- Distribution of GM approaches the lognormal form and thus, computation of mean and SD are admissible [36].

Taking log on both sides of equation (9),

$$\log I_{HMc0} = \frac{1}{n}\sum_{i=1}^{n} \log[\frac{x_{ic}}{x_{i0}}] \quad .... (11)$$

Thus,
$$\log SI_{HM} = \left[\frac{1}{n}\sum_{i=1}^{n}(\log\frac{x_{ic}}{x_{i0}} - \log I_{HMc0})^2\right]^{\frac{1}{2}} \quad .... (12)$$

Where, SI_{HM} denotes Geometric Standard Deviation (GSD)

Since, log (GSD of X_1, X_2, ..., X_n)=usual SD of $\log X_1$, $\log X_2$, ..., $\log Xn$.

The ratio $\frac{\log SI_{HM}}{\log I_{HMc0}}$ is analogous to Coefficient of Variation (CV) and indicates consistency of the data.

Help to estimate population parameters. For large data, sample GM is taken as population GM and standard error of GM is estimated by $\frac{GM \times GSD}{\sqrt{n-1}}$.

Thus, statistical tests of hypotheses regarding equality of GM's can be performed across time and space using t-tests on the logarithms of the observations.

Possible to construct separate index for each subset of indices.

Relative importance and contribution of each index can be easily found.

Critical indices are those for which $\frac{X_{it}}{X_{i0}} < 1$ or $\frac{X_{it}}{X_{i(t-1)}} < 1$.

Improvement in an index and the corresponding gain in I_{HMc0} are linearly related since 1% increase in $\frac{X_{it}}{X_{i(t-1)}} \Rightarrow 1\%$ increase in $I_{HMt(t-1)}$ if all others are unchanged

Applications

The proposed method can be applied even for skewed data and irrespective of correlations between a pair of indices and help in meaningful comparison of a set of regions of a country.

For comparison of properly defined sub-areas say agricultural lands, river basins, lands in vicinity of industrial activities like mining, production, highways, solid waste dump sites, etc., the proposed I_{HM} index can also be applied with pre-selected pollutants and indices.

Can be used for classification of regions with respect to overall pollution status of HMs.

The graph of progress/decline of $I_{HMt(t-1)}$, using the Chain indices for a region will help to assess impact of various mitigation measures across time

Limitations

- Introduction of a new index requires value of that index in the base period and at different time periods.

- The method fails if an indicator takes negative or zero value.

- Assumes no missing data

RESULTS AND DISCUSSION

Limitations of existing methods of overall pollution status of HMs were reviewed. The proposed I_{HM} in terms of GM, avoiding scaling and finding weights considers all chosen indices to depict comprehensive improvement or decline of soil pollution by HMs in a region in the current year with respect to base year or previous year and helps in meaningful comparison of a group of regions/countries. The proposed I_{HM} considers GM of the ratios $\frac{x_{ic}}{x_{i0}}$ without normalization of individual indices like geometric aggregation adopted in Human Development Index, 2010 [37].

Important features of the proposed measure include:

- Reduction of substitutability among the component indices.

- Produces no bias for polluted or non-polluted regions since effect of outliers is not much.

- Unit-free monotonic continuous function.

- Facilitates formation of chain indices and satisfies time reversal test.

- Possible to find distribution of I_{HM} and compute mean and variance for a sample of stations.

- Allows ranking and classification of regions in terms of I_{HM} values and assesses paths of improvement registered by a region over time.

- Meaningful comparison of countries can be made in terms of progress made on Year-to-Year basis, despite different base period figures across countries,

GM approach is preferred for its additional properties like formation of chain indices, satisfaction of time reversal test, easy computation of contribution of the indices to I_{HM}, linearity between improvement in a ratio of an index and the corresponding gain in I_{HMc0}. Distance of a country from the SDG goals at a particular time period can be found by replacing the base period

data with the SDG goals adopted by the country. Besides, the measure can be used for statistical testing of $H_0 : I_{HM\,i} = I_{HM\,j}$ for the i-th and j-th region at a particular time or $H_0 : I_{HM(t)} = I_{HM(t+1)}$ for a particular region.

I_{HM} may be computed separately for biogenic and physicogenic aggregates to see which one is more likely to lead to soil improvement.

CONCLUSION

The proposed aggregation by GM is an improvement over the existing methods of finding overall pollution status of HMs (I_{HM}) in soils and is recommended. Researchers and stakeholders may derive benefits of the proposed I_{HM} to find overall results of soil use and management along with identification of critical areas requiring managerial attentions. Future studies to assess impact of I_{HM} in soils on vegetables, fruits, fish, shellfish, crustaceans, etc. may be undertaken.

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Data availability

Not applicable. No dataset used in the study.

Ethics approval

Not required for this methodological paper.

Ethical responsibilities of authors

All authors have read, understood, and have complied as applicable with the statement on ethical responsibilities of authors.

Informed consent

No data collected from individuals.

Author's contribution statement

Conceptualization, methodology, writing-original draft preparation, writing-reviewing and editing, by the single author.

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