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Indexing Concentrations of Heavy Metals

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Abstract: *Contamination of soil and water by Heavy metals (HMs) are major concerns for living beings. Samples of water or soil allow computation of ratios* $\frac{C_{ic}}{C_{i0}}$ *where* C_{ic} *denotes pooled average of concentration of the i-th HM for the current period and* C_{i0} *is the corresponding allowable concentration limit of the HM. Avoiding major limitations of existing methods, the paper suggests multiplicative aggregation of the relevant* $\frac{C_{ic}}{C_{i0}}$ *'s to get composite index* $CI_{HM-Soil}$ *or* $CI_{HM-Water}$ *reflecting overall HM concentration satisfying desired properties and facilitating ranking of regions, finding relative importance of each HM, minimizing substitution effects, enabling assessment of progress across time and undertaking parametric analysis. The indices can be used to summarize regional distributions. Based on theoretical advantages, the suggested multiplicative aggregation is an improvement over the existing methods of finding overall pollution status of HMs in soils and water and is recommended. Researchers and policy makers may derive benefits of the proposed* $CI_{HM-Soil}$ *and* $CI_{HM-Water}$ *to find overall concentration of HMs along with identification of critical HMs and critical regions for corrective mitigation actions.*

Keywords: *Heavy metals; Comprehensive assessment; Geometric mean, Chain indices, Time reversal test; Progress path.*

1. INTRODUCTION

Pollution by heavy metals is a pressing threat to nature and human well-being, with dire consequences for the environment, biodiversity, and public health. Contamination of soil and aquatic environment by Heavy metals (HMs) are major concerns for living beings (Singh et al. 2017; Liang et al. 2018). Desirable and permissible limits of concentrations of most of the toxic individual HMs are prescribed as per national and international standards. In practice, samples of water or soil contain a finite set of individual elements with different concentrations and corresponding allowable limits, which allow computation of ratios $\frac{C_{ic}}{C_{i0}}$ where C_{ic} denotes pooled average of concentration of the *i*-th HM for the current period and C_{i0} is the corresponding allowable concentration limit of the HM. Meaningful arithmetic aggregations of such ratios and parametric analysis are not possible (Valiente-Diaz et al. 2023). Composite index (CI) based on weighted sum assumes full compensability among the individual indicators and thus, poor performance in one aspect can be compensated by surplus in others (Mariani and Ciommi, 2022). Methodologically sound method of aggregation of such ratios $\frac{C_{ic}}{C_{i0}}$is needed to assess overall effect of the constituents to judge quality of soil or water.

However, toxicity and concentration of HMs are different. The former depend on degree of absorption (bioaccumulation), duration of exposure, ROS generation, weakening of theantioxidant defense, enzyme inactivation, and oxidative stress, etc. Even gender differences intoxicity of HMs have been reported (Vahter et al., 2007; Tchounwou et al., 2012). The International Agency for Research on Cancer (IARC) recommended arsenic; cadmium, chromium, and nickel are carcinogenic to humans(Forte et al. 2020). Trivalent inorganic arsenic compounds are more hazardous to human health than organic and pentavalent compounds (Medina-Pizzali et al. 2018).Estimating total HMs is not adequate to find their contamination and toxicity levels in the ambient environment (Kumar et al., 2020). The mechanisms of toxicity and dose dependent carcinogenicity of HMs are unclear (Balali-Mood et al. 2021) and is beyond the scope of the paper.

Avoiding major limitations of existing methods, the paper describes comprehensive assessment of concentrations of HMs by multiplicative aggregation of the relevant $\frac{C_{ic}}{C_{i0}}$ ‘s to get composite index for soil ($CI_{HM-Soil}$) or for water ($CI_{HM-Water}$) reflecting overall HM concentration satisfying desired properties and facilitating comparison of the indices for two regions or one region at two different time periods, finding relative importance of each indicator, minimizing substitution effects, enabling assessment of progress across time and undertaking parametric analysis.

2. LITERATURE SURVEY

2.1. Concentration of Single HM

Yang et al. (2023) investigated concentrations of HMs using contaminant index (single factor) as $P_i = \frac{C_i}{S_i}$where C_i (mg/Kg) is the observed concentration and $S_i(mg/Kg)$ is the evaluation criterion of *i*-th HM; Nemerow pollution index as $P_N = \sqrt{\frac{{P_{i.Max}}^2 + {P_{i.ave}}^2}{2}}$ where $P_{i.Max}$ and $P_{i.ave}$ are maximum and arithmetic average of P_is respectively. The index $P_N$shares a strong positive relationship with Synthetic Pollution Index (SPI) (Gong et al. 2022) where$SPI = \sqrt{\frac{P_{i\,Max} + P_{i\,Min}}{2}}$.

Relative bioaccumulation index (RBI) of *i*-th HM is taken as *RBI*= $\frac{C_i}{C_{i_{Max}}}$(Proc et al. 2021)

Geo-accumulation index (I_{geo}) computed by $log_2[\frac{C_{Sample}}{1.5\times C_{Background}}]$ where C_{Sample} and $C_{Background}$ denotes respectively concentration of the single HM in the sample and the same in the background, taken from secondary data, which can vary with natural diagenesis (Muller, 1969). For geologically different regions, Alonso et al. (2004) determined $C_{Background}$ of Cd, Cr, Cu, Ni, Pb and Zn.

Enrichment Factor *(EF)* indicating enrichment degree of HMs is the average concentration of *i*-th HM in unit area of soil (or water)/ background level of the *i*-th HM. For soil, it is computed as $EF = \frac{(C_i/C_{Ref})_{Soil}}{(C_i/C_{Ref})_{Background}}$. However, it is difficult in practice to select a relevant background composition (Carleton et al. 2019).

Toxic risk index *(TRI)* of *i*-th HM considers probable effect level (PEL) and threshold effect level (TEL) and is calculated as$TRI_i = \sqrt{\frac{(C^i_{Soil}/C^i_{PEL})^2 + (C^i_{Soil}/C^i_{TEL})^2}{2}}$ where C^i_{Soil} denotes concentration of *i*-th HM in soil, C^i_{PEL} and C^i_{TEL} are the PEL and TEL of the HM (Smith et al. 1996).

Following major observations and limitations merit considerations:

- Units of concentration of HMs may vary. For example, HM concentrations in soil and plants are given in mgkg^{-1}and the same for fruits and vegetables are reported by µgg^{-1}. Of course, mgkg^{-1}can be converted to µgg^{-1} using 1mgkg^{-1}= 1000µgg^{-1} or 1 µgg^{-1} = 0.001 mgkg^{-1}. However, such conversions unnecessarily change standard deviation (SD) and consistency of the data (reflected by Coefficient of variation (CV)).
- P_i, P_N,*RBI*,I_{geo}, EF, TRI is unit free**.**
- $P_i$is a ratio and arithmetic average of ratios is not meaningful like average speed of number of moving cars is not equal to the arithmetic average of the speeds.
- Thus, $P_N$reflecting average pollution level in terms of average of various pollutants and computation of $P^2_{i.ave}$ are not meaningful.
- Two extreme values of SPI could be unrealistic outliers, which are data points deviating from the majority of the data points and mislead the nature of the association among the variables considered (Kim et al., 2015).

- Value of P_N or SPI depends heavily on data heterogeneity and suffer for ignoring other observations of the data. Better could be to find pooled average and pooled standard deviation (SD) of $P_i^{'}s$ based on all sample observations.
- EF between 0.5 to 1.5 indicates contribution of HM from natural source and $EF > 1.5$ implies anthropogenic origin of the HM (Nawrot et al., 2020)
- HM-wise concentrations follow different distributions which are unknown. One desirable point of good method of aggregation of individual concentration of HMs is to facilitate knowledge of the aggregated index.
- I_{geo} is essentially a logarithm of P_i.If geochemical background value = evaluation criterion of *i*-th HM, $I_{geo} = log_2[\frac{P_i}{1.5}]$, implies highly correlated I_{geo} and P_i.
- Threshold values for classification with respect to I_{geo}, $EF$and TRI are different. For example, “No pollution” is <0 for I_{geo}, < 1 for *EF* (“No enrichment”) and <5 for *TRI* (“No Toxic Risks”) (Zeng et al. 2019). Known and similar distribution of P_i, P_N,*RBI*,I_{geo}, EF, TRI say Normal distribution helps to find equivalent threshold values for each such index (Chakrabartty, 2021).
- Classification based on any of the above said indices needs to show efficiency of classification in terms of low value of within-group variance and high value of between-group variance, which presumes meaningful addition, multiplication, etc.

Studies focusing on one specific HM cannot satisfy meaningful 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 (Yoo et al. 2018). Better assessment of overall contamination of soil, plants and fish for better evaluation of the risk associated with them need composite index (CI) of pollution indices observed from different sectors for a nation (Ahirvar et al. 2023).However, Xie et al. (2021) observed no agreed standard for evaluation environmental quality of soil.

2.2. Combined Concentration of HMs

Tamasi and Cini (2004) obtained Mean Metal Index (MI) as $MI = \sum_{i=1}^{n} \frac{C_i}{MAC_i}$ where $MAC_i$is the maximum allowed concentration of the *i*-th HM.

Individual bioaccumulation index (*IMBI*) combines *RBIs* of *n*-number of HMs as $IMBI = \frac{1}{n}[\sum_{i=1}^{n}(\frac{C_i}{C_{i_{Max}}})]$, even if $C_{i_{Max}} \neq C_{j_{Max}}$ for $i \neq j$ and thus, $\frac{C_i}{C_{i_{Max}}} + \frac{C_j}{C_{j_{Max}}}$ is not meaningful

Based on maximumI_{geo} value (I_{geoMax}) and average of I_{geo} value (I_{geoAv}.) of eight different HMs in a sample, Santos-Francés et al. (2017)calculated mean, SD, Median and Geometric mean for I_{geo} and Nemerow index $I_{IN} = \sqrt{\frac{P_{geoMax} + P_{geoAv}}{2}}$separately for each HM without specifying most suitable averaging procedure.

The toxic risk index (*TRI*)is taken as the sum of all the $TRI_i s$ i.e. $TRI = \sum_{i=1}^{n} TRI_i$

Metal Pollution Index *(MPI).* The overall concentration of Pb, Cd, As, and Hg in vegetable and fruit samples is estimated by *MPI* for *n*-samples as$\sqrt[n]{(Cf_1 x Cc_2 x \ldots \ldots . Cf_n x)}$where Cf_n is the concentration of HM in the *n*-th sample (Teodorovic et al. 2000).

Heavy metal pollution index *(HPI)* is used find contamination level of HMs as a weighted sum approach where weights are inversely proportional to the standard value recommended to every adjacent element (Mohan et al., 1996) and is given by *HPI*=$\frac{\sum_{i=1}^{n} w_i Q_i}{\sum_{i=1}^{n} w_i}$ where $w_i$is the weight to *i*-th parameter *i*=1. 2. …..,*n* andQ_i denotes the sub-index for the *i*-th parameter= $\sum_{i=1}^{n}[\frac{M_i - l_i}{S_i - l_i}] * 100$ where $M_i, l_i$and S_i denote respectively assessed value, tolerable value and allowable value.

2.3. Major Limitations

– I_{geo} is a relative measure and depends on value of $C_{Background}$. For a given value of $C_{Background}$, $\frac{I_{geo\,(1)}}{I_{geo\,(2)}} \neq \frac{C_{Sample\ (1)}}{C_{Sample\ (2)}}$. Chakrabartty (2023) gave an example where for the X-th HM, $C_{Background\ X}$=36.0 mg·kg^{-1}, $C_{Sample\ (1)}$ = 37 units and $C_{Sample\ (2)}$ = 38 units implying $\frac{C_{Sample\ (1)}}{C_{Sample\ (2)}} = \frac{37}{38}$ = 0.973684. But, $I_{geo\,(1)} = log_2(0.685185)$ = -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} due to unit increase in C_{Sample} is different at different range of values i.e. $\frac{\Delta I_{geo}}{\Delta C_{Sample}}$is not constant. If concentration of X in $C_{Sample\ (3)}$and $C_{Sample\ (4)}$ are taken as 74 and 75 respectively and$C_{Background}$ for X is 36.0, $\frac{I_{geo\,(3)}}{I_{geo\,(4)}} = 0.959274 \neq \frac{I_{geo\,(1)}}{I_{geo\,(2)}}$

- C_{Sample} could be better calculated as pooled mean
- $C_{Background}$ may not be available for all areas, especially for areas considered as free from HM pollutants and geochemical baseline map is under progress. For example, information on$C_{Background}$ at Brahmaputra valley, North East India was not available (Dutta et al. 2021).
- *HPI* uses a rating scale for each selected parameter and weights are assigned. Scoring systemsof subjective rating scale are rather arbitrary ranging between 0 and 1. Selection of weights is not beyond questions (Greco, et al. 2019).
- Calculation of Q_i to evaluate *HPI* may give wrong results. Eldaw et al. (2020) gave an example where observed concentration of zinc in sample A ($C_{Zn,A}$) exceeded the same for sample B ($C_{Zn,\boldsymbol{B}}$) and highest desirable limit value of zinc was in between $C_{Zn,A}$and $C_{Zn,B}$, gave result which contradicted the reality. In addition, sum of Q_i equaled the overall index even when $M_i < l_i$.
- In case the relative weight of a HM is zero, influence of the HM will be absent, despite $M_i > S_i$
- For *HPI*, standard value ($\boldsymbol{S_i}$) could be the permissible or desirable value of the parameter and value of the $\boldsymbol{l_i}$ is not fixed for the calculation and may be manipulated (Singh et al., 2019).
- Despite non-meaningfulness of arithmetic mean of *HPI*for given values of $\boldsymbol{l_i}$and$\boldsymbol{S_i}$**,** researchers considered mean *HPI* **(**Chiamsathit et al. 2020).
- *MPI* expressed in *(mg perkg)* is not unit free.
- *HPI* and *MI* showed identical results for assessment of groundwater pollution (Salah et al., 2015). For a discussion on limitations of combined indices see Ahirvar et al. (2023).

Most of the above said combined concentration of HMs methods lack theoretical basis without knowledge of distribution of the aggregated measures to undertake statistical inferences, produce different results and fail to assess relative importance of individual HMs (Eldaw et al. 2020).

3. SUGGESTED METHOD

In line with Metal Pollution Index (*MPI*), assessment of overall concentration of HMs is suggested as multiplicative aggregation of individual concentrations of each of *n*-number of HMs (indicators) like Cr, Cd, Hg, Pb, As, etc. causing adverse health implications even at lower concentration (Sahoo and Sahu, 2022) in soil as

$$CI_{HM-Soil} = \sqrt[n]{\frac{C_{1c}.C_{2c}\ldots\ldots C_{nc}}{C_{10}.C_{20}\ldots\ldots C_{n0}}} \tag{1}$$

or ignoring the *n*-th root by $CI_{HM-Soil} = \frac{C_{1c}.C_{2c}\ldots\ldots C_{nc}}{C_{10}.C_{20}\ldots\ldots C_{n0}}$ (2)

where C_{1c} and C_{10} denote respectively the obtained concentration of the 1st HM in the current period (pooled average of concentration of the HM taken over the sampling units) and the corresponding values of evaluation criteria of the HM. (C_{2c}, C_{20}) and other (C_{jc}, C_{j0})'s are defined accordingly.

Clearly, unit free ratio $\frac{C_{jc}}{C_{j0}}>1$ indicates concentration of the j-th indicator at current period exceeded the evaluation criterion. Without loss of generality, let us assume $C_{jc} > 0$ and $C_{j0} > 0 \forall\, j = 1, 2, \dots\dots, n$

$CI_{HM-Water}$ for m-number of HMs can be similarly computed as

$$CI_{HM-Water} = \sqrt[m]{\frac{C_{1c}.C_{2c}\dots\dots C_{mc}}{C_{10}.C_{20}\dots\dots C_{m0}}} \tag{3}$$

or equivalently as $CI_{HM-Water} = \frac{C_{1c}.C_{2c}\dots\dots C_{mc}}{C_{10}.C_{20}\dots\dots C_{m0}}$ (4)

Multiplicative aggregation in (1) to (4) can be applied for all types of concentration measures including those in percentages, irrespective of their score ranges, distributions and sample size.

Replacing the evaluation criteria by the figures of the previous year will give progress of $CI_{HM-Soil}$ or $CI_{HM-Water}$ on year-to-year basis in macro level and progress (increase/decline) of each of individual HMs like CI_{Cd}, CI_{Hg}, CI_{Pb}, CI_{As} at micro level.

In line with general convention of index value = 100, $CI_{HM-Soil}$*100 and $CI_{HM-Water} * 100$ may be taken to reflect percentage changes.

$CI_{HM-Soil} > 1 \implies$ Overall increase of concentrations of the HMs from the evaluation criteria in soil. Same can be used for $CI_{HM-Water}$.

$\frac{CI_{HM-Soil\ t}}{CI_{HM-Soil\ (t-1)}} < 1$ indicates decline in overall concentrations in the$t$-thyear over (t-1)-th year.

CI_{Cd} becomes critical HM in the t-th year, if $\frac{CI_{\text{Cd}\ t}}{CI_{\text{Cd}\ (t-1)}}>1$. Similarly, critical HMs can be found in a year over the previous year, separately for soil and water. Such critical HMs draw the attention of the policy makers to initiate appropriate corrective actions.

Taking logarithm on both sides of (2) and (4) we get $\log(CI_{HM-Soil}) = \sum_{i=1}^{n} \log(\frac{C_{ic}}{C_{i0}})$and

Log $(CI_{HM-Water}) = \sum_{i=1}^{m} \log(\frac{C_{ic}}{C_{i0}})$ which become additive models, each following lognormal distribution since $\log(GM)$ approaches lognormal distribution (Alf and Grossberg, 1979). The logarithm of the aggregate function is continuous and differentiable with $\frac{d}{dx} log(Aggregate\, function)> 0$

3.1. Properties

For a given region, $CI_{HM-Soil}$ or $CI_{HM-Water}$ depicts overall increase/decline of pollution by HMs in the current year with respect to the target values or previous year by a unit free continuous function satisfying the following desired properties:

1. Independent of change of units
2. Reduced level of substitutability among the component indices
3. Produces no bias for polluted or non-polluted regions since $CI_{HM-Soil}$ or $CI_{HM-Water}$ is least affected by outliers.
4. Satisfaction of Time reversal test since $CI_{HM-Soil\ c0} * CI_{HM-Soil\ 0c} = 1$ and $CI_{HM-Water\ c0} * CI_{HM-Water\ 0c} = 1$
5. Possible to form chain indices like $CI_{HM-Soil\ 20} = CI_{HM-Soil\ 21} . CI_{HM-Soil\ 10}$
6. Enables to draw graph of $CI_{HM-Soil\ c0}$ or $CI_{HM-Water}$ across time showing path of improvement or decline for a region over successive time periods. Regions can also be compared with respect to such paths.

7. Taking log on both sides of equation (1), $logCI_{HM-Soil\ c0} = \frac{1}{n}\sum_{i=1}^{n} log[\frac{C_{ic}}{C_{i0}}]$

Thus, SD of$I_{HM-Soil\ c0}$ can be obtained as Geometric standard deviation (GSD) since

log(GSD of$X_1, X_2, \ldots\ldots, X_n$)) = usual SD of log$X_1$, log$X_2$, ………, log$X_n$, which in turn can be used to compute coefficient of variation (CV) to indicate consistency of the data.

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

8. 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.
9. Relative importance and contribution of *j*-th HM is given by$\frac{C_{jc}/C_{j0}}{CI_{HM-Soil}}$ and $\frac{C_{jc}/C_{j0}}{CI_{HM-Water}}$
10. Improvement in *k*-th HM and the corresponding gain in $CI_{HM-Soil}$ are linearly related since 1% increase in $\frac{C_{kc}}{C_{k0}}$⟹1% increase in $CI_{HM-Soil\ t\ (t-1)}$ if all others are unchanged
11. Distribution of GM approaches lognormal facilitating computation of mean and SD and testing $H_0: CI_{HM-Soil\ c0_i} = CI_{HM-Soil\ c0_j}$ for two regions *i*≠*j* or

$H_0: CI_{HM-Soil\ c0_{ti}} = CI_{HM-Soil\ c0_{(t-1)i}}$ for the *i*-th region using conventional *t*-tests on the logarithms of the observations. Testing of equality of each of $CI_{\mathrm{Cd}}, CI_{Hg}, CI_{Pb}, CI_{As}$ can also be undertaken over time and space.

4. DISCUSSION

The suggested indices $CI_{HM-Soil}$ or $CI_{HM-Water}$ are simple to calculate, easy to interpret and can be computed for any finite number of HMs. Each index satisfies desired properties from measurement angle and follows Normal distribution for large sample size. The indices can be used to summarize regional distributions.

Instead of ranking the HMs in terms of measured concentration values like $P_b > C_d > C_r$ etc. the regions can be ranked with respect to $CI_{HM-Water}$ like $Region_1 > Region_2 > Region_3$ etc. Similarly, ranking of regions with $CI_{HM-Soil}$ can be made with added advantage of testing statistical hypothesis $H_0: \overline{CI_{HM-Water}}_{ith\ region} = \overline{CI_{HM-Water}}_{jth\ region}$

Following similar approach, overall concentration levels for air CI_{HM-Air}, commericial fish/ crustaceans $CI_{HM-Fish}$, fruits and vegetables $CI_{HM-Plant}$ can be computed to cover the routes (like oral, dermal and inhalation routes) of absorption of HMs and their compounds in human body.

Implications of contamination of soil and water by HMs for agricultural productivity and the resultant food security can be studied using $CI_{HM-Soil}\ and CI_{HM-Water}$ with action initiated for mitigation of adverse impacts.

Appropriate measure of toxicity, carcinogenicity, allergenicity of HMs on human life $CI_{Toxicity}$ can be predicted as a linear combination of $CI_{HM-Soil}$, $CI_{HM-Water}$, CI_{HM-Air}, $CI_{HM-Fish}$, $CI_{HM-Plant}$ along with other relevant measures indicating level of exposures and degree of consumption of plant products and fish containing toxic HMs, provided assumptions of multiple linear regression are satisfied.

Empirical relationship between $CI_{HM-Soil}$ and $CI_{HM-Water}$ can throw insight of anthropogenic sources of HMs which can contaminate both soil and water environment through waste waters flowing from mines and waste storage, runoff of pesticides from agricultural land, metal-containing sewage sludge applied to agricultural fields, etc.causing contamination of soils, waters and crops.

5. LIMITATIONS

- The suggested method fails if any $C_{jc} = 0$ or $C_{j0} = 0$
- It is better to test normality of $CI_{HM-Soil}$ and $CI_{HM-Water}$ by say Anderson-Darling test of normality.
- Integration of organic compounds like pesticides, PAHs, PCBs may be done in future studies.

6. CONCLUSIONS

Based on theoretical advantages, the suggested multiplicative aggregation is an improvement over the existing methods of finding overall pollution status of HMs in soils and water and is recommended. Researchers and policy makers may derive benefits of the proposed $CI_{HM-Soil}$ and $CI_{HM-Water}$ to find overall concentration of HMs along with identification of critical HMs and critical regions for corrective mitigation actions. Future studies may be undertaken to investigate toxicity of concentrations of HMs in soils and water and their uptake by plants, fish and estimating empirical relationship between $CI_{HM-Soil}$ and $CI_{HM-Water}$.

ETHICAL RESPONSIBILITIES OF AUTHORS

All authors have read, understood, and have complied as applicable with the statement on Ethical responsibilities of Authors: as found in the Instructions for Authors

AUTHORS' CONTRIBUTION STATEMENT

Conceptualization, Methodology, Writing- Original draft preparation, Writing- Reviewing and editing, by the single Author

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