Heavy metals contamination assessment in agricultural soil for shallot in Wanasari, Brebes Regency, Central Java Province

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Abstract. Assessment of heavy metals contamination was carried out in agricultural soil in Wanasari, Brebes Regency, Central Java Province. The study purposed to assess the level of metal contamination using statistical analysis, contamination factor (CF), geo-accumulation index (I-geo), and load pollution index (PLI). In this research, a total of 18 topsoil samples were taken and analyzed for the several metal concentrations are Pb, Cd, Cr, Co, and Ni. The concentration of heavy metals in soil samples was in the order of Cr > Ni > Pb > Co > Cd. The I-geo values of Pb, Co, Ni, and Cr indicated uncontaminated soils, while I-geo values for Cd showed level contamination in soil was uncontaminated to moderately contaminated. The values of pollution load index (PLI) indicated that these observed samples in agricultural soils for shallot in Wanasari, Brebes Regency were uncontaminated.

Keywords: Agricultural soil, geo-accumulation index, heavy metal contamination, pollution load index, shallot

1. Introduction

Heavy metals can be found both naturally in the soil (as parent materials) and effect from anthropogenic sources (industrial wastes, chemical fertilizers, agrochemicals, animal manure, soil amendments, and sewage sludge [1]–[5]. The use of pesticides and fertilizers has assisted the accumulation of metals soil [6]. Several pesticides and fertilizers are known to include heavy metals such as Pb, Zn, Hg, Cu, and Cd and excessive application of agricultural chemicals can increase the accumulation of metals in the soil [7], [8].

Brebes Regency is the largest shallot producer in Central Java and supplies around 37.8% of the national market demand for shallots [9]. Generally, farmers in Brebes planted shallots four times each year with other rotating crops such as chili, eggplant, corn and rice; or fallowed [10]. Most shallot farmers in Brebes Regency still use excessive fertilizers and pesticides to increase yields. This is one of the causes of environmental deterioration and decreasing of soil quality. Heavy metals not only induce soil contamination but also affect the yield, quality, and food safety [11].

Assessment and monitoring of heavy metal accumulation in soils are involved to evaluate the possible risk of agricultural land because of heavy metals contamination. The purpose of this research...
to risk assessment of heavy metals contamination in agricultural land for shallots in Brebes Regency, Central Java Province, Indonesia.

2. Material and Methods

A total of 18 soil samples with depth 0-20 cm taken from agricultural land for shallot in Wanasari Village, Brebes Regency, Central Java in August-October 2019. The research location is shown in Fig. 1. Coordinates point for soil sampling were recorded using GPS (global positioning system). The total contents of Pb, Cd, Ni, Cr, and Co in soil were analyzed with the wet ashing method using a strong acid mixture HNO₃ (65% pa) and HClO₄ (65% pa) and the extraction result were measured using AAS (Atomic Absorption Spectrometry). The accuracy of the heavy metal measurement data is carried out by verifying them with the standard solution from Merck. Analysis of soil was implemented at the Integrated Laboratory of Indonesian Agricultural Environment Research Institute (IAERI) which has been accredited with ISO / IEC 17025: 2017 in Jakenan, Central Java.

![Figure 1. The map of soil sampling location in Wanasari, Brebes Regency](image)

2.1. Degree of soil contamination

The index of potential risk can be used as an effective tool for assigned the level of contamination in soil with I-geo (geo-accumulation index), CF (contaminant factor) and PLI (pollution load index) [12]–[14].
2.1.1. I-geo (Geo-accumulation index)
The I-geo for the metals were introduced by Müller [15]. The score of the geo-accumulation index is described with equation
\[
I_{\text{geo}} = \log_2 \left( \frac{C_n}{1.5 B_n} \right)
\]
Where Cn is the metal concentration in the soil, Bn is the geochemical background concentration factor and 1.5 is the background matrix correction factor due to lithogenic issues.

2.1.2. CF (Contamination factor)
The degree of soil contamination by metal is determined in terms of CF value, which is calculated with equation below:
\[
CF = \frac{C_{\text{metal}}}{C_{\text{background}}}
\]
Where C-metal and C-background are the metal concentration in soil and the background value of metal, respectively.

2.1.3. PLI (Pollution load index)
Pollution load index was determined as the n root of CF values for number of metals researched (n) which the formula specified below:
\[
PLI = \sqrt[n]{(CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n)}
\]

| Value | Soil contamination classification |
|-------|----------------------------------|
| I-geo < 0 | Uncontaminated |
| 0 - 1 | Uncontaminated to moderately contaminated |
| 1 - 2 | Moderately uncontaminated |
| 2 - 3 | Moderately to heavily contaminated |
| 3 - 4 | Heavily contaminated |
| 4 - 5 | Heavily to extremely contaminated |
| I-geo > 5 | Extremely contaminated |

| CF < 1 | Weak contaminated |
| 1 ≤ CF < 3 | Moderate contaminated |
| 3 ≤ CF < 6 | High contaminated |
| CF > 6 | Very high contaminated |

| PLI < 1 | Uncontaminated |
| PLI > 1 | Contaminated |

3. Results and Discussion
The standard statistics (such as minimum, maximum, mean, standard deviation, coefficient of variation, skewness and kurtosis value), background value, and critical value of heavy metals content in soil are shown in Table 2. The concentration of Pb, Cd, Co, Ni, and Cr in soil samples ranged 10.42-18.94, 0.99-2.31, 3.06-7.94, 10.61-20.71, and 16.26-45.88 mg kg\(^{-1}\) respectively with mean values of 14.65, 1.55, 5.71, 16.52, and 28.37 mg kg\(^{-1}\) respectively. The maximum content of Pb, Cd, C, Ni, and Cr lower than the critical value of 400, 8, 50, 100, and 100 mg kg\(^{-1}\) respectively.
Table 2. Descriptive statistics of several heavy metals in agricultural soil in Wanasari Village, Brebes Regency.

| Content | Pb  | Cd  | Co  | Ni  | Cr  |
|---------|-----|-----|-----|-----|-----|
| n       | 18  | 18  | 18  | 18  | 18  |
| Minimum | 10.42 | 0.99 | 3.06 | 10.61 | 16.26 |
| Maximum | 18.94 | 2.31 | 7.94 | 20.71 | 45.88 |
| Mean    | 14.65 | 1.55 | 5.71 | 16.52 | 28.37 |
| Standard deviation | 3.11 | 0.43 | 1.57 | 2.96 | 8.60 |
| Coefficients of variation (%) | 21.22 | 27.96 | 27.43 | 17.94 | 30.30 |
| Skewness | 0.09 | 0.75 | -0.43 | -0.24 | 0.64 |
| Kurtosis | -1.66 | -0.88 | -1.06 | -0.75 | -0.41 |
| Background value [19] | 17 | 0.3 | 19 | 68 | 95 |
| Critical value [20] | 400 | 8 | 50 | 100 | 100 |

The anthropogenic result for heavy metals contamination was assessed by using contaminant factor (CF) show in Table 3. The CF of Co, Ni, and Cr was found to be smaller than one, indicating weak contamination by several metals in soils. The CF value for Pb in soil amount of 67% means considerable contamination and 33% indicating moderate contamination in agricultural soil for shallot in Wanasari, Brebes Regency. For Cd, the CF value mean is considerable contamination (72%) and very high contamination (28%) in soil samples. Heavy metals are found in different natural media in concentration more than background values which derive from anthropogenic sources. Contamination of heavy metals in soil not only the agricultural activities but also due to industrial activities and road traffic that contribute to metals contamination.

Table 3. CF (Contaminant factor) and PLI (pollution load index) value for several metals in soil in Wanasari, Brebes Regency.

| No  | Coordinate | Contaminant Factor | PLI |
|-----|------------|--------------------|-----|
|     | X          | Y                  | Pb  | Cd  | Co  | Ni  | Cr  |     |
| 1   | 108.99262  | -6.94469           | 1.04 | 6.77 | 0.33 | 0.23 | 0.22 | 0.65 |
| 2   | 109.00625  | -6.93118           | 0.91 | 4.16 | 0.34 | 0.18 | 0.25 | 0.57 |
| 3   | 109.01982  | -6.93124           | 0.81 | 7.56 | 0.16 | 0.21 | 0.21 | 0.54 |
| 4   | 109.01987  | -6.91768           | 1.09 | 4.36 | 0.20 | 0.27 | 0.33 | 0.61 |
| 5   | 108.99279  | -6.90401           | 0.71 | 4.19 | 0.34 | 0.26 | 0.17 | 0.54 |
| 6   | 109.00636  | -6.90406           | 0.69 | 5.90 | 0.31 | 0.30 | 0.39 | 0.68 |
| 7   | 109.01993  | -6.90412           | 0.89 | 3.60 | 0.27 | 0.24 | 0.22 | 0.54 |
| 8   | 109.00642  | -6.89050           | 0.63 | 4.01 | 0.27 | 0.28 | 0.26 | 0.55 |
| 9   | 109.01999  | -6.89056           | 0.75 | 4.56 | 0.19 | 0.16 | 0.19 | 0.46 |
| 10  | 109.00647  | -6.87694           | 0.61 | 4.01 | 0.17 | 0.23 | 0.33 | 0.50 |
| 11  | 109.02004  | -6.87700           | 0.95 | 7.50 | 0.22 | 0.30 | 0.46 | 0.74 |
| 12  | 108.99296  | -6.86332           | 1.10 | 7.72 | 0.34 | 0.22 | 0.24 | 0.69 |
| 13  | 109.00653  | -6.86338           | 0.67 | 4.77 | 0.38 | 0.27 | 0.25 | 0.61 |
| 14  | 109.02010  | -6.86344           | 0.77 | 3.29 | 0.35 | 0.28 | 0.48 | 0.66 |
| 15  | 108.99302  | -6.84976           | 1.09 | 4.51 | 0.41 | 0.21 | 0.29 | 0.65 |
| 16  | 109.00659  | -6.84982           | 0.65 | 4.88 | 0.42 | 0.20 | 0.33 | 0.61 |
| 17  | 108.99308  | -6.83620           | 1.11 | 6.52 | 0.31 | 0.30 | 0.40 | 0.78 |
| 18  | 109.00664  | -6.83626           | 1.04 | 4.52 | 0.37 | 0.23 | 0.34 | 0.67 |
Pollution level and its difference of the areas were determined with the use of pollution load index value. This index is a rapid tool which is very convenient to compare the contamination level on the other areas. The PLI values for all metals in Table 3 show low value because less than 1. All soil samples indicated that agricultural soil for shallot in Wanasari Village, Brebes Regency, Central Java is uncontaminated by several metals (Pb, Cd, Co, Ni, and Cr).

Geo-accumulation index value was also determined to assess the pollution status of heavy metals in soil (Table 4). Geochemical background concentration was multiplied each time by a matrix correction factor 1.5 in order to give content variation of heavy metals in the environment with very low anthropogenic effect [21]. Geo-accumulation index values of Pb, Co, Ni, and Cr indicated uncontaminated soils, but I-geo values for Cd showed level contamination in soil was uncontaminated to moderately contaminated.

**Table 4.** I-geo values for several metals in soil in Wanasari, Brebes Regency, Central Java Province

| No | Coordinate | I-geo | Pb | Cd | Co | Ni | Cr |
|----|------------|-------|----|----|----|----|----|
|    | X          | Y     |    |    |    |    |    |
| 1  | 108.99262  | -6.94469 | -0.16 | 0.65 | -0.66 | -0.81 | -0.83 |
| 2  | 109.00625  | -6.93118 | -0.22 | 0.44 | -0.64 | -0.92 | -0.77 |
| 3  | 109.01982  | -6.93124 | -0.27 | 0.70 | -0.97 | 0.85 | -0.85 |
| 4  | 109.01987  | -6.91768 | -0.14 | 0.46 | -0.89 | -0.74 | -0.65 |
| 5  | 108.99279  | -6.90401 | -0.32 | 0.45 | -0.65 | -0.76 | -0.94 |
| 6  | 109.00636  | -6.90406 | -0.33 | 0.59 | -0.69 | -0.70 | -0.58 |
| 7  | 109.01993  | -6.90412 | -0.22 | 0.38 | -0.75 | -0.80 | -0.83 |
| 8  | 109.00642  | -6.89050 | -0.38 | 0.43 | -0.74 | -0.73 | -0.75 |
| 9  | 109.01999  | -6.89056 | -0.30 | 0.48 | -0.89 | -0.98 | -0.90 |
| 10 | 109.00647  | -6.87694 | -0.39 | 0.43 | -0.95 | -0.81 | -0.66 |
| 11 | 109.02004  | -6.87700 | -0.20 | 0.70 | -0.83 | -0.70 | -0.51 |
| 12 | 108.99296  | -6.86332 | -0.14 | 0.71 | -0.64 | -0.84 | -0.79 |
| 13 | 109.00653  | -6.86338 | -0.35 | 0.50 | -0.59 | -0.74 | -0.78 |
| 14 | 109.02010  | -6.86344 | -0.29 | 0.34 | -0.63 | -0.73 | -0.49 |
| 15 | 108.99302  | -6.84976 | -0.14 | 0.48 | -0.56 | -0.86 | -0.72 |
| 16 | 109.00659  | -6.84982 | -0.36 | 0.51 | -0.56 | -0.88 | -0.66 |
| 17 | 108.99308  | -6.83620 | -0.13 | 0.64 | -0.66 | -0.69 | -0.58 |
| 18 | 109.00664  | -6.83626 | -0.16 | 0.48 | -0.60 | -0.81 | -0.64 |

Correlation analysis is an effective method to explain the relationships between multiple variables. Some metals in soil usually have complicated relationship. A significant positive correlation among metals in soil shown that the accumulated heavy metal concentrations come from the same pollutant sources [22]. Pearson’s correlation result and their significance are reported in Table 5. Ni concentration has a positive significant (P<0.05) correlation with Cr (0.596), indicates that Ni likely originated from the same pollutant source with Cr.
Table 5. Pearson’s correlation matrix for the several metal concentrations

| Parameter | Pb  | Cd  | Co  | Ni  | Cr  |
|-----------|-----|-----|-----|-----|-----|
| Pb        | 1   |     |     |     |     |
| Cd        | 0.384 | 1   |     |     |     |
| Co        | 0.145 | -0.145 | 1   |     |     |
| Ni        | 0.003 | 0.111 | -0.015 | 1  |     |
| Cr        | 0.1  | 0.039 | 0.073 | 0.596 | 1  |

Bold italic values are significance at P< 0.05

4. Conclusion
The heavy metals concentration in soil samples vary widely as follow: Cr > Ni > Pb > Co > Cd. The contamination status in the study area indicates the possibility comes from anthropogenic results such as industrial and agricultural activities. The levels of soil contamination were measured using PLI (pollution load index) and I-geo (geo-accumulation index). The I-geo value in the Cd parameter indicates the soils category uncontaminated to moderately contaminated. the soils in all studied samples were uncontaminated with respect to Pb, Co, Ni, and Cr. The PLI value is very low and varies between 0.46-0.78, this indicates that the agricultural soil for shallots in Wanasari Village, Brebes Regency, Central Java Province is classified as uncontaminated.

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6. References
[1] Naveedullah, Hashmi, M.Z., Yu, C., Shen, H., Duan, D., Shen, C., Lou, L., & Chen, Y. 2013 *BioMed Research International* **2013** 1-10.
[2] Fei, X., Xiao, R., Chrisakos, G., Langousis, Ren, Z., Tian, Y., & Lu, X. 2019 *Ecological Indicators* **106** 1-9.
[3] Alloway, B.J. 2013 Sources of heavy metals and metalloids in soils. *Heavy metals in soils*. In: Alloway, B.J. (Ed.), Trace Metals and Metalloids in Soils and Their Bioavailability, third ed. Springer pp. 11–50.
[4] Zoffoli H.J.O., Amarel-Sobrinho, N. M. B., Zonta, E., Luisi, M.V., Marcon, & G. Tolon-Becerra 2013 *Environ Monit Assess* **185** 2423–2437.
[5] Li, X., Liu, L., Wang, Y., Luo, G., Chen, X., Yang, X., Hall, M.H.P., Guo, R., Wang, H., Cui, J., & He, X. 2013 *Geoderma* **192** 50-58.
[6] Atafar, Z., Mesdaghinia, A., Nouri, J., Homae, M., Yunesian, M., Ahmadi Moghadam, M., & Mahvi, A.H. 2010 *Environ Monit Assess* **160** 83-89.
[7] Karisma, B & Prasad, S 2014 *IOSR Journal of Environmental Science, Toxicology and Food Technology (IOSR-JESTF)* **8** 46-54.
[8] Cui X, Sun X L, Hu P J, Yuan C, Luo Y M, Wu L H, & Christie P 2015 *Pedosphere.** **25** 878–887.
[9] Susanti, A and Waryanto, B 2017 *Agricultural Statistics 2017* Ministry of Agriculture. Indonesia.
[10] Muliana, Hartono, A., Anwar, S., Susila, A.D., & Sabiham, S 2018 AGRIVITA Journal of Agricultural Science 40 515-526.
[11] Edelsteina, M & Ben-Hur, M 2018 Scientia Horticulturae 234 431-444.
[12] Keshavarzi, A. & Kumar, V 2020 Geology, Ecology, and Landscapes 4 87-103.
[13] Navarrete-Rodriguez, G., Castaneda-Chavez, M.R., & Lango-Reynoso, F 2020 Int. J. Environ. Res. Public Health 17 969.
[14] Martinez-Mera, E.A., Torregoza-Espinosa, A.C., Crissien-Borrero, T.J., Marrugo-Negrete, J.L. & González-Marquez, L.C 2019 Heliyon 5 1-9.
[15] Müller, G 1988 Chemical Decontamination of Dedged Materials, Combustion Residues, Soil and Other Materials Contaminated with Heavy Metals, In: W. Wolf, J. Van deBrink & F. J. Colon, Eds., 2nd International TNO/BMFT Conference on Contaminated Soil 2 pp. 1439-1442.
[16] Hakanson, L 1980 Water Research 14 975–1001.
[17] Sutherland, R.A 2000 Environmental Geology 39 611–627.
[18] Tomlinson, D.C., Wilson, J.G., Harris C.R., & Jeffrey, D.W 1980 Helgoland Marine Research 33 566-575.
[19] Wedepohl, K.Henes., & Hitler 1995 Geochimica et Cosmochimica Acta 59 1217–1232
[20] Alloway, B. J 1995 Soil processes and the behaviour of metals. New York: Wiley.
[21] Vareda, J.P., Valente, A.J.M., & Duares, L 2019 Journal of Environmental Management 246 101-118.
[22] Micó C, Recatal´a L, Peris M, & S´anchez J 2006 Chemosphere 65 863-872.