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Article

Contamination and Spatial Distribution of Metal(loid)s in the Stream Sediment near the Greenhouse

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Abstract: As greenhouse horticulture continues to increase in South Korea, the effects of metal(loid)s from wastewater discharges on stream sediments were analyzed. A total of 106 samples were analyzed for cadmium (Cd), boron (B), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), chromium (Cr), manganese (Mn), and iron (Fe). The coefficient of variation (CV) of the metal(loid)s components was 96.58% for Mn, 93.40 for Zn, 62.47 for B, 58.90 for Pb, and 58.14 for Cd, suggesting anthropogenic sources. Correlation analysis suggested a cumulative source for Cd-Zn and B, and cluster analysis suggested an anthropogenic source for Cu, Pb, B, and Mn. The contamination factor (CF) suggested the need to trace the source of contamination to Pb (3.21 ± 1.89) and B (1.33 ± 0.83) and EF to Pb (3.30 ± 1.81) and B (1.44 ± 0.94). The analytical results identify anthropogenic sources of Pb, B, and Cd. The high contamination of Cd suggests the influence of mining areas, and Pb suggests the influence of traffic, fertilizers, pesticides, and fossil fuels in greenhouses, in addition to the influence of mines. B confirmed the impact of the facility's vegetable wastewater and suggested the need to further examine the cumulative impact of Mn, Fe, etc. By utilizing the facility horticulture wastewater for ecological restoration and other agricultural uses, we aim to prevent stream sediment pollution and realize a sustainable agricultural environment.

Keywords: heavy metal; facility horticulture; soil restoration; drainage water; soil; river

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1. Introduction

Republic of Korea's facility horticulture industry ranks among the top in the world, along with China, Spain, the Netherlands, and Japan, and is considered a "white revolution" due to the large income it generates [1–9]. For this reason, greenhouse horticulture in Korea continues to grow [10,11]. However, many environmental and ecological problems have been reported in facility horticulture, including land use change, groundwater depletion, waste disposal, and nonpoint pollution [12–17]. Among them, the use of nutrient solutions and the resulting wastewater discharge is a major problem [18–21]. Facility horticulture cultivation methods are broadly categorized into soilless cultivation-hydroponics (Figure 1A) and soil cultivation (Figure 1B). However, the application of nutrient solutions makes little difference. Crops grown in greenhouse horticulture include tomatoes, peppers, strawberries, cucumbers, peppers, lettuce, and Chinese cabbage [22–33]. Hydroponics cultivation dumps nutrients directly into streams (Figure 1C), while soil cultivation allows nutrients accumulated in the soil to leach out during rainfall (Figure 1D). There are cases of wastewater discharged from greenhouse crop cultivation and nutrients accumulated in the soil, contaminating groundwater, streams, rivers, and agricultural fields [34–37]. For this reason, countries such as the Netherlands, Spain, and Japan are developing recycling methods that do not discharge wastewater [38–42].

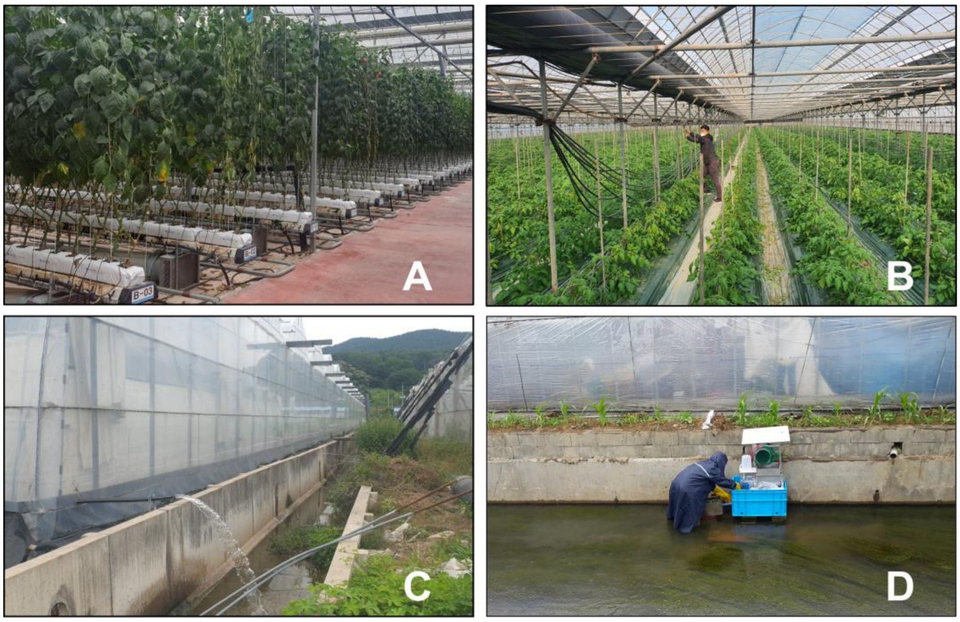

Figure 1. The location of study site in Buyeo County. (**A**) Soilless hydroponics cultivation. (**B**) Soil cultivation with nutrient solution. (**C**) Wastewater discharged from hydroponics cultivation. (**D**) Wastewater movement to the stream on rainy days in underground soil cultivation greenhouse.

Nutrients such as N, P, K, and Mg are the main fertilizer components of wastewater discharged from facility horticulture [43–46]. In addition, metal(loid)s, such as Mn, B, Fe, etc., are an essential requirement for hydroponics cultivation [47,48]. Since N, P, K, and Mg cause eutrophication of streams and rivers [49,50], various treatment methods are discussed to prevent water pollution. However, metal(loid)s were not discussed as a need for management because they are not used in large quantities. In the case of trace elements such as metal(loid)s, although they are not used in large amounts, metal(loid)s can cause problems if they continuously flow into rivers connected to the public's drinking water sources [51,52]. In particular, they are substances that accumulate in river and stream sediments and cause pollution, so they need to be recovered and reused or treated.

Soil contamination by metal(loid)s is a serious global environmental problem [53], especially in rivers and agricultural lands that utilize water, which is directly related to human health. Soil contamination with metal(loid)s is caused by a variety of sources, including natural weathering [54,55], mining activities [56,57], industrial activities [58,59], civil engineering, construction activities [60,61], and increased traffic [61–63].

In order to restore contaminated soils, it is necessary to identify the source of metal constituents through monitoring and to investigate the contamination characteristics (contamination level, spatial distribution, scope of influence, etc.) [64]. Therefore, many studies have investigated the contamination characteristics of surrounding soils from anthropogenic sources in various countries [65–69].

Soil that has exceeded the standard due to pollutant discharge is in urgent need for restoration measures in addition to blocking the source of pollution. In addition, stream and river soil and sediment are connected to drinking water and have a great impact on human health, so their management is important. Therefore, it is very important to review the pollution characteristics of stream sediments and to study their spatial distribution characteristics to block the inflow of pollutants.

Therefore, this study analyzed the impact of metal(loid)s in wastewater discharged from facility horticulture on streams. The target area for this analysis was Buyeo-gun, a county with a high concentration of greenhouse horticulture in Korea. We believe that this study can be used as a basis for the improvement of facility horticulture for sustainable agriculture, the need for water purification facilities, and the management of metal(loid) discharges.

2. Materials and Methods

A methodology to evaluate the metal(loid) content of stream sediments resulting from wastewater discharged from a facility horticulture complex was conducted in four phases. This study comprised the following stages: First, the study streams were selected based on the distribution of facility horticulture in Buyeo-gun, the study area (Section 2.1). Second, the sediment of the studied streams was sampled (Section 2.2). Third, the sampled stream sediments were analyzed for common metal(loid)s according to the Korean Process Test Method (Section 2.3). Fourth, information on the correlation of metal(loid)s in the study sites, sources, and pollution pathways was categorized through statistical analysis (Section 2.4). The detailed materials and methods are discussed below.

2.1. Study Sites

The target area is Buyeo-gun, Chungcheongnam-do, located in the center of Chungcheongnam-do, as shown in Figure 2. The total size of the target area is about 624.2 km^2, and forests (318.7 km^2), rice fields (148.8 km^2), and fields (45.8 km^2) are distributed in the following order [70].

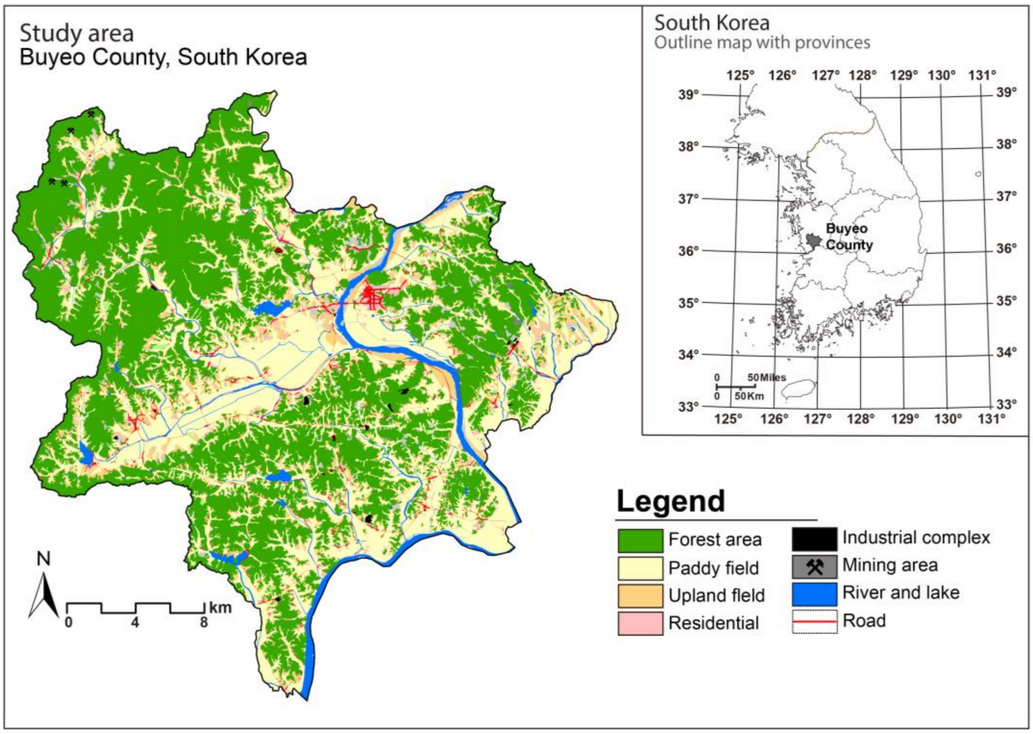

Figure 2. The location of study site in Buyeo County.

In the target area, agricultural land, which mainly includes rice paddies and fields, accounts for more than 30% of the total area, and excluding forests, the area of agricultural land is about 64%, so most of the land in the target area is used for agriculture. The soil types are classified as Inceptisols (56.2%), Ultisols (31.5%), Alfisols (6.5%), and Entisols (14%). The soil parent materials are metamorphic rocks (granite gneiss: 63.2%), acidic rocks (22.4%), and Holocene alluvial deposits (9.8%; primarily agricultural fields), and the soil textures are loam soil (91.2%), silt loam (4.3%), and sandy loam (3.2%), according to the Korean Soil Information System [70]. The total population of the target area is 71,143, of which the agricultural population is about 22,000, and more than 30% of the total population is engaged in agriculture [71]. Considering that the agricultural population in Korea is about 5% of the total population, the target area can be considered a typical agricultural area.

2.2. Stream Sediment Sampling

Buyeo County is where the Geumgang River, the third largest river in Korea, flows through. For this reason, there is a good supply of water for agriculture, which is the reason for the high number of facility horticulture. As shown in Figure 3, we can see the distribution of facility horticulture centered on the Geumgang River [4,5]. The streams studied are concentrated in the Geumgang River. For the purpose of this study, sediment samples (stream sediment) were collected from a total of 106 points throughout the study area to determine whether metal(loid)s accumulated in stream sediment due to facility horticulture (Figure 3; black point). Reservoir was excluded, and top sediment (0~30 cm) was collected.

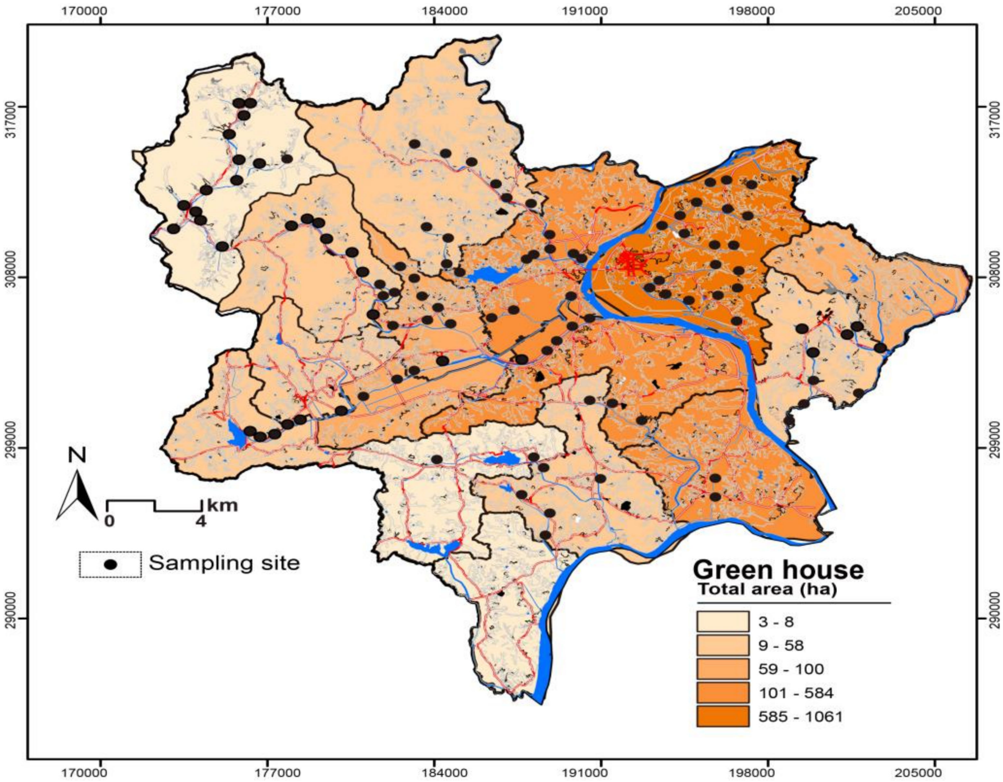

Figure 3. The area of greenhouse horticulture and sampling sites.

To obtain a representative sample from each sampling site, sediments from five sites were collected in a zigzag pattern and mixed evenly according to the Korean Ministry of Environment's Process Test Method [72]. Sediment samples were collected using a stainless steel hand auger and stored in polyethylene zipper bags for transportation to the indoor laboratory. The sediment was spread evenly on a steel pan to a uniform thickness and air-dried for one week before being passed through a 2 mm stainless steel sieve for chemical analysis.

2.3. Analysis of Metal(loid)s and Assessment of Stream Sediment

The pH (acidity) and EC (electrical conductivity) were basically analyzed, and the metal(loid)s included nine items: cadmium (Cd); boron (B); copper (Cu); nickel (Ni); lead (Pb); zinc (Zn); chromium (Cr); manganese (Mn); and iron (Fe). Based on physical, physiological, and chemical properties, 9 items have been classified: transition metals (Mn, Fe, Cu, Ni); post-transition metals (Zn, Cd, Pb); metalloids (As, B) [73]. The items analyzed in this study were subjected to hydrolysis according to the Korean Ministry of Environment process test method to analyze the total content of metal(loid) elements [74]. The pH and EC values were determined on a 1:5 (v/v): distilled water suspension by the Korean standard method. After passing the sediment through a 0.15 mm (No. 100) sieve and weighing 3 g quantitatively, it was placed in a test apparatus and purified by adding aqua regia (HCl [21 mL] + HNO3 [7 mL]) for 2 h. The temperature was then raised to reach reflux conditions and maintained in that state for 2 h. The digestate was filtered through filter paper (Advantec No. 2), and the total content of metal(loid)s was determined using ICP/OES (Optima 5300DV, Perkin Elmer, Columbus, OH, USA).

In order to evaluate the contamination level of stream sediment in the target area, the general indicators of contamination factor (CF) and enrichment factor (EF) were used. Contamination factor (CF) and enrichment factor (EF) are widely used to determine the contamination level of soil [75].

The contamination factor (CF) assesses the accumulation of metal(loid)s in sediments compared to pre-industrial reference levels [76]. It is calculated according to the following Equation (1):

$$CF = \frac{C_m}{C_b} \tag{1}$$

where Cm is the concentration of the trace element in the target area, and Cb is the background concentration of the trace element.

The contamination factor results are evaluated as CF < 1: low contamination; 1 < CF < 3: moderate contamination; 3 < CF < 6: significant contamination; and CF > 6: very high contamination [77].

The enrichment factor (EF) is a measure used to determine the effect of precipitation on metal(loid)s concentrations in sediments [78]. It is calculated according to the following Equation (2).

$$EF = \frac{(C_m/C_{Fe})_{sample}}{(C_m/C_{Fe})_{background}} \tag{2}$$

where (Cm/CFe) sample is the ratio of the trace element to the standardized element (Fe) in the stream sediment sample, and (Cm/CFe) background is the ratio of the trace element to the standardized element in the background concentration.

A pollution enrichment factor of less than 1.5 (EF < 1.5) indicates a natural environment with no anthropogenic influence. An EF greater than 1.5 (EF > 1.5) indicates anthropogenic pollution from air or water. EF < 1.5: no pollution; 1.5 < EF $\leq$ 3.0: low pollution; 3.0 < EF $\leq$ 5.0: moderate pollution; EF > 5.0: severe pollution [79].

2.4. Statistical Analysis

Statistical analysis was performed using SPSS 20.0 (IBM, Armonk, NY, USA). The statistical distribution of the data was tested for normality using the Kolmogorov–Smirnov test, with 95% confidence intervals around the mean. The correlations between metal components were examined using Pearson's correlation coefficient, considering the statistical significance level ($p < 0.01$). Correlations between metal(loid)s in sediment provide important information about their sources and pathways of contamination [80,81].

Multivariate statistical analyses (cluster analysis and principal component analysis) were performed to identify the sources of the metal(loid)s. In cluster analysis, data were standardized by Z-score and then clustered using Ward's method [82]. For principal component analysis, the rotation method was Verimax rotation with Kaiser normalization. This orthogonal rotation minimizes the number of variables with high loading values for each component to facilitate the interpretation of the results [83]. Here, only principal components with eigenvalues greater than 1 according to the Kaiser criterion were considered. This multivariate statistical analysis has been shown to be effective in discriminating between anthropogenic and natural sources of metal(loid)s in sediment [84,85].

GIS is an effective tool for analyzing the spatial variability of pollutants [86]. In this study, GIS mapping software (ArcGIS 10.2.2, ESRI, New York, NY, USA) was used to create spatial distribution maps for items with relatively high accumulation of metal(loid)s in the target area. To create this distribution map, spatial interpolation was performed using the inverse distance weighted (IDW) method. The inverse distance weighted method does not require any assumption of normality in the statistical distribution of the data and is widely used [87,88].

3. Results

3.1. Concentrations of Metal(loid)s in Stream Sediments at the Study Site

The total contents and descriptive statistics of nine metal(loid)s at the study site sediments are shown in Table 1. The table compares the background values (BV) of unpolluted Korean stream sediments with the results of the Ministry of Environment (MOE) [89] and the National Institute of Agricultural Science (NIAS) [90] for Cd, Cu, Ni, Pb, Zn, Cr, and Fe. B is compared to the United States Geological Survey (USGS) [91], and Mn is compared to the results from the global river sediments [92].

Table 1. Total content (mg/kg) and descriptive statistics of metal(loid)s in the study area *.

Item	Min	Max	Mean	SD	CV	BV	Skew.	Kurto.	p(K-S Test)
pH	4.54	8.13	6.07	0.76	12.47	6.3	0.074	−0.335	0.200
EC (dS/m)	0.17	4.86	1.11	0.90	81.03	0.16	1.980	4.548	0.000
Cd	0.05	1.47	0.42	0.24	58.14	0.40	1.835	5.416	0.000
B	9.13	131.07	43.87	27.40	62.47	33.0	1.170	0.611	0.000
Cu	3.50	42.50	18.46	8.71	47.17	14.0	0.464	−0.187	0.080
Ni	2.90	43.08	16.62	7.67	46.18	19.5	0.827	1.346	0.200
Pb	21.47	349.16	86.38	50.88	58.90	26.9	1.808	6.058	0.001
Zn	4.38	184.82	24.65	23.02	93.40	101.0	3.934	22.539	0.000
Cr	4.88	60.92	25.47	11.18	43.90	53.1	0.593	0.444	0.200
Mn	44.84	3215.00	378.57	365.63	96.58	770.0	4.795	34.142	0.000
Fe	3053.82	13279.18	7952.47	2558.93	32.18	-	0.159	−0.915	0.088

* SD: standard deviation; CV: coefficient of variation; BV: Background value (Background value of Cd, Cu, Ni, Pb, Zn, Cr, Fe in Korean river sediments—Ministry of Environment, 2011; National Institute of Environmental Research, 2011; Background value of B—USGS (1984); Background value of Mn—World average of river sediments, Lin et al., 2013); Skew.—skewness; Kurto.—kurtosis; p(K-S test), *p*-values of Kolmogorov–Smirnov test for normality of the raw data (for values higher than 0.05, the distribution is normal).

The ranges of concentrations of the nine items, including Cd, B, Cu, Ni, Pb, Zn, Cr, Mn, and Fe, are presented in Tables 1 and A1 in Appendix A. The means for each metal(loid)s were Cd 0.42 ± 0.24, B 43.87 ± 27.40, Cu 18.46 ± 8.71, Ni 16.62 ± 7.67, Pb 86.38 ± 50.88, Zn 24.65 ± 23.02, Cr 25.47 ± 11.18, Mn 378.57 ± 365.63, and Fe 7952.47 ± 2558.93 mg/kg (Table 1). The coefficient of variation (CV) for the concentrations of metal(loid)s was high order for Mn 96.58%, Zn 93.40%, B 62.47%, Pb 58.90%, Cd 58.14%, Cu 47.17%, Ni 46.18%, Cr 43.90%, and Fe 32.18%. Mn, Zn, B, Pb, and Cd have coefficients of variation greater than 50%. The background value (BV) for the assesses of contamination factor (CF) and enrichment factor (EF) was Cd 0.40, B 33.0, Cu 14.0, Ni 19.5, Pb 26.9, Zn 101.0, Cr 53.1, Mn 770.0. Fe is used as a reference element because of its high concentration in the Earth's crust and generally serves as the normalizing element to calculate the EF of metal(loid)s [93,94]. Therefore, Fe was a non-BV factor.

The statistical distribution of the data was tested for normality using the Kolmogorov–Smirnov test. The results showed that the datasets for metal(loid)s in sediment were not normally distributed. The distributions of the metal(loid)s were mostly strongly positive with skewness values greater than 1, and their kurtosis was very sharp. Moreover, even after logarithmic transformation, the distributions of the metal(loid)s components did not show normality.

3.2. Correlation between Metal(loid)s and Geographic Coordinates

Table 2 shows the Pearson correlation coefficients between the nine metal(loid)s. Significant positive correlations between constituents can be interpreted as indicating that their sources and transport pathways are the same. Based on the CVs reviewed in Section 3.1, the correlations of Mn, Zn, B, Pb, and Cd, which are related to anthropogenic pollution sources, were analyzed. Mn was correlated with B, Cu, Ni, Pb, and Cr. Zn showed a strong positive correlation with Cd (r = 0.728 **), and B showed a positive correlation with Cu (r = 0.528 **). The highest correlations among metal(loid)s are Cr-Ni (r = 0.965 **), Zn-Cd (r = 0.728 **), Ni-Cu (r = 0.721 **), and Cr-Cu (r = 0.715 **). Based on the results of this correlation analysis and CV evaluation, it is necessary to investigate the source tracking and occurrence of the items mentioned as anthropogenic pollutants. Furthermore, it is necessary to analyze whether these results are consistent with the results of the multivariate statistical analysis (Section 3.3) and high correlation assessment of stream sediment of study areas (Section 4.1).

Table 2. Pearson's correlation matrix of the metal(loid) contents in the study area.

	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
Cd	1								
B	−0.105	1							
Cu	0.183	0.528 **	1						
Ni	0.297 **	0.352 **	0.721 **	1					
Pb	0.011	0.479 **	0.690 **	0.397 **	1				
Zn	0.728 **	0.276 **	0.250 **	0.206 *	0.131	1			
Cr	0.260 **	0.444 **	0.715 **	0.965 **	0.413 **	0.231 *	1		
Mn	0.183	0.405 **	0.406 **	0.321 **	0.366 **	0.148	0.267 **	1	
Fe	0.015	0.067	0.516 **	0.678 **	0.386 **	−0.178	0.637 **	0.346 **	1

** Highly significant at p = ** < 0.01, * < 0.05.

Figure 4 is a graphical representation of the recognized correlations in Table 2, such as B, Cd, Zn, Cr, Ni, and Fe. This figure additionally shows the correlations for geographic coordinates (X coordinates, TM).

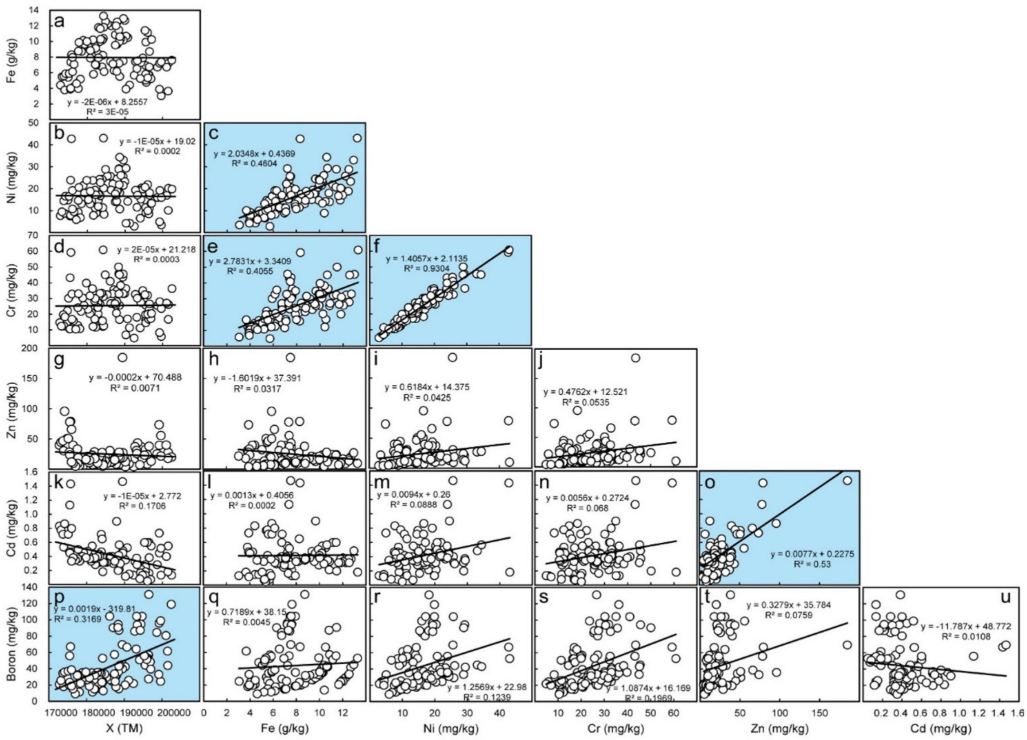

Figure 4. Correlation between metal(loid)s in sediment and geographic coordinates. (**a**–**u**) graphical presentation of correlation analysis result. (**Background color**) blue: high correlation, white: low correlation.

Correlations between them are known to provide information about sources and migration pathways. Boron (B) was analyzed with an increasing trend with increasing x-coordinate (TM) (Figure 4p).

The remaining metal(loid) entries (Figure 4a,b,d,g,k) were not associated. This may indicate the presence of anthropogenic factors at the location that are associated with boron accumulation in the sediment. In this study, we would like to examine boron assessing streambed sediment contamination (Section 3.4) and the impact of the facility greenhouse (Section 4.2).

3.3. Multivariate Statistical Analysis between Metal(loid) Items

Figure 5 shows a dendrogram of the results of the clustering analysis and principal component analysis.

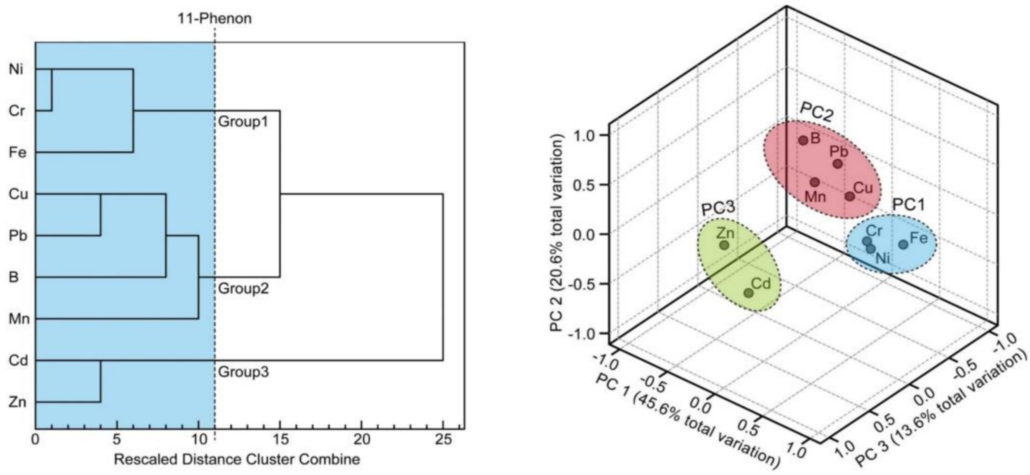

Figure 5. Hierarchical dendrogram showing clustering of metal(loid)s, according to Ward's methods, using squared Euclidean distance and factor loadings for principal component analysis (PCA) after varimax rotation.

Distance clustering indicates the degree of association between the components, and the smaller the value for distance clustering, the more significant the association. Therefore, components classified in the same group in the clustering analysis can be interpreted as having similar characteristics. In Figure 5, the metallic components and the principal component analysis (PCA) can be divided into three groups: Ni, Cr, and Fe in the first group (Group 1; PC1); Cu Pb, B, and Mn in the second group (Group 2; PC2); and Cd and Zn in the third group (Group 3; PC3). In Group 1, Ni, Cr, and Fe are considered to be a common group due to their natural occurrence, as described in Table 2 and Figure 4.

3.4. Assessing Streambed Sediment Contamination (CF and EF)

The contamination factor (CF) and enrichment factor (EF) were used to evaluate the contamination level of stream sediments in the target area (Figure 6). CF and EF showed similar distribution trends.

The mean values of CF showed a decreasing order of Pb (3.21 ± 1.89) > B (1.33 ± 0.83) > Cu (1.32 ± 0.62) > Cd (1.04 ± 0.60) > Fe (1.00 ± 0.32) > Ni (0.85 ± 0.39) > Mn (0.49 ± 0.47) > Cr (0.48 ± 0.21) > Zn (0.249 ± 0.23). Pb is more than 80% of the surveyed sites, and B is more than 40% of the surveyed sites, with 1.5 < EF (anthropogenic influence), which means that these two elements have been accumulated relatively more than others by anthropogenic sources. More than 60% of Pb and 15% of B had 3.0 < EF (moderate contamination), and especially Pb had 5.0 < EF (extreme contamination).

The results for CF were the same for EF due to similar definitions. The EF values of the investigated samples ranged from 0.038 to 10.273. The descending order of the average EF values was Pb (3.30 ± 1.81) > B (1.44 ± 0.94) > Cu (1.34 ± 0.58) > Cd (1.15 ± 0.82) > Fe

(1.00 ± 0.00) > Ni (0.85 ± 0.29) > Mn (0.48 ± 0.38) > Cr (0.48 ± 0.16) > Zn (0.29 ± 0.30). Compared to the group in Figure 5, Pb, B, and Cu are in the same group, which suggests anthropogenic contamination based on the previous discussion.

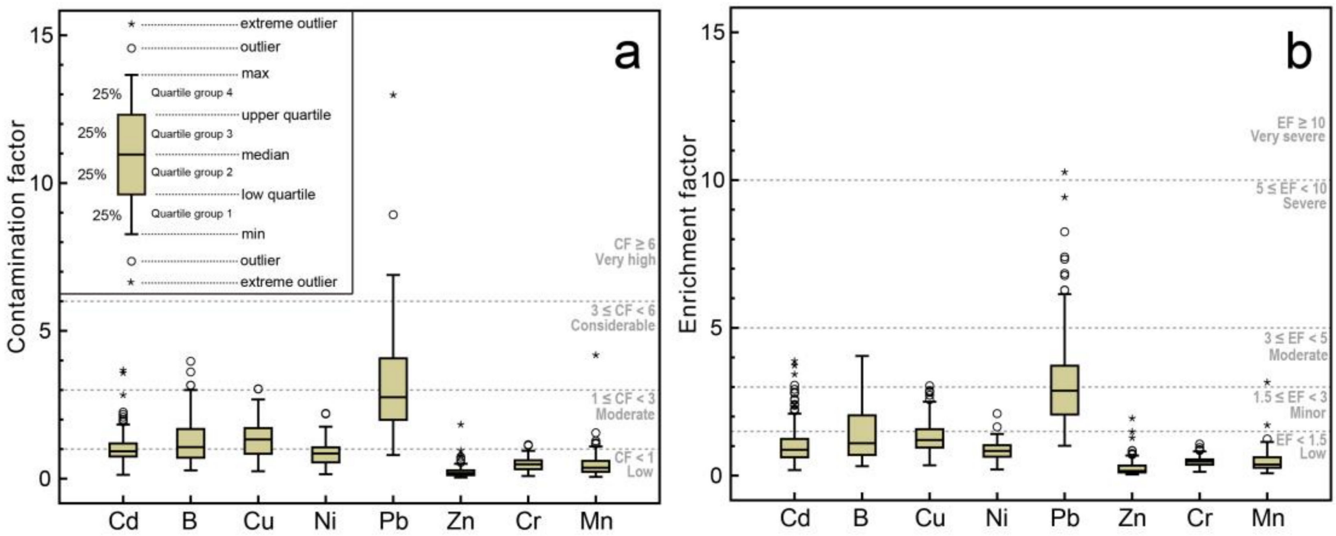

Figure 6. The statistical description of contamination factor (CF; (**a**)) and enrichment factor (EF; (**b**)) in studied sediment samples.

4. Discussion

Greenhouse horticulture in the Republic of Korea continues to grow, and wastewater discharge has become a major problem. In this study, we analyzed the effects of metal(loid)s from facility horticulture wastewater on stream sediments. It was concluded that this study could be used as a basis for improving facility horticulture for sustainable agriculture, the need for water purification facilities, and the management of metal(loid) discharges. The study site was located in Buyeo-gun, a county with a high concentration of greenhouse horticulture in Korea. Soil samples (stream sediment) were collected from a total of 106 points, and nine items, including cadmium (Cd), boron (B), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), chromium (Cr), manganese (Mn), and iron (Fe), were analyzed. We reviewed the concentrations of metal(loid)s in the stream sediments of the study sites and conducted correlations between metal(loid)s and geographic coordinates, multivariate statistical analysis among metal(loid) items, and stream sediment contamination evaluation (CF and EF) to analyze the sources of contaminated components.

4.1. General Characteristics of Stream Sedimentary Soil Pollutants

The CV (Table 1) of the analyzed metal(loid)s was high for Mn (96.58%), Zn (93.40), B (62.47), Pb (58.90), Cd (58.14), Cu (47.17), Ni (46.18), Cr (43.90), and Fe (32.18). A CV greater than or equal to 50% may indicate a possible origin from anthropogenic activities [95]. Since these coefficients of variation are reliable indicators for distinguishing between constituents derived from anthropogenic activities [83,96], it is considered necessary to investigate the anthropogenic sources of Mn, Zn, B, Pb, and Cd in the streams in the study area. The Kolmogorov–Smirnov test results (Table 1) showed that the datasets for metal(loid)s in sediment were not normally distributed. Among multivariate statistical methods, cluster analysis and principal component analysis are classified as non-parametric methods that do not require normality assumptions for data [97,98]. Therefore, in this study, the total concentration values of metal components were used as the dataset for multivariate statistical analysis without data transformation to ensure normality. Based on the above descriptive statistics and CV evaluation, it is necessary to identify anthropogenic sources of Mn, Zn, B, Pb, and Cd in streams.

Correlation analysis (Table 2) among the nine metal(loid)s showed that Mn was correlated with B, Cu, Ni, Pb, and Cr. Zn showed a strong positive correlation with Cd (r = 0.728 **), and B showed a positive correlation with Cu (r = 0.528 **). The highest correlations were Cr-Ni (r = 0.965 **), Zn-Cd (r = 0.728 **), Ni-Cu (r = 0.721 **), and Cr-Cu (r = 0.715 **). Correlations were further plotted against geographic coordinates (Figure 4). Cr and Ni commonly show a high correlation with Fe (Figure 4a,e), which may indicate that Cr and Ni are of natural origin. In fact, naturally occurring metal(loid)s are highly correlated with Fe because Fe adsorbs large amounts of metal(loid)s during soil formation [99]. Fe is commonly used as an indicator to distinguish between naturally occurring components [100,101]. Zn and Cd are highly correlated with each other (Figure 4o) and are known to behave together in soil because they have similar ionic radii and are sulfophilic elements [102,103]. In the case of boron (B), it was analyzed as an increasing trend as the X coordinate (TM) increased (Figure 4p). Boron(B) is one of the seven essential metal(loid)s in nature and is closely related to the survival of living things [104]. In agriculture, boron is used in products such as fertilizers, herbicides, and pesticides [105,106]. Based on the results of the anomalous correlation analysis, it is judged that the need to investigate the cause of the occurrence of Cd-Zn and the accumulation of B is recognized.

The dendrogram of the results of the cluster analysis showed that Ni, Cr, and Fe were in Group 1; Cu, Pb, B, and Mn were in Group 2, and Cd and Zn were in Group 3. In general, they have a coefficient of variation (CV) of less than 50%, such as Ni, Cr, and Fe (Table 1). Therefore, the elements in this group tend to be geologically related to each other due to their similar chemical properties [107,108]; Ni, Cr, and Fe in group 1 were determined to be derived from natural sources, such as soil parent rocks, in the target area. The coefficients of variation for Cu, Pb, B, and Mn in Group 2 were relatively high compared to those in Group 1, with coefficients of variation greater than 50%. Furthermore, B and Mn are metal(loid)s that are commonly used in fertilizers and hydroponics in agriculture [44,104,105,109]. Group 3, Zn and Cd, includes groups 1 and 2, but their content is composed separately. Therefore, they can be categorized as natural and anthropogenic composite sources. Therefore, it is necessary to identify anthropogenic sources of Cu, Pb, B, and Mn and to explore ways to reduce them.

The contamination factor (CF) and enrichment factor (EF) were used to evaluate the contamination levels of the target stream sediments (Figure 6). CF and EF showed similar distribution trends. The mean values of CF were Pb (3.21 $\pm$ 1.89) > B (1.33 $\pm$ 0.83) > Cu (1.32 $\pm$ 0.62) > Cd (1.04 $\pm$ 0.60) > Fe (1.00 $\pm$ 0.32) > Ni (0.85 $\pm$ 0.39) > Mn (0.49 $\pm$ 0.47) > Cr (0.48 $\pm$ 0.21) > Zn (0.249 $\pm$ 0.23). Pb is more than 80% of the surveyed spots; B is more than 40% of the surveyed spots, and EF is more than 1.5. This indicates anthropogenic pollution. In addition, more than 60% of B and 15% of B are above EF 3.0. In particular, Pb is above 20%, with an EF of 5.0. The results for CF were the same for EF due to similar definitions. The variability of Pb, B, and Cu was largely based on EF, and EF is a widely applied proxy to estimate geochemical anomalies and the impact of human behavior on sediment chemistry [74,75]. Based on the analytical results of EF, the need to trace anthropogenic sources of Pb, B, and Cu was recognized.

In the distribution results of CF and EF, it can be said that the need to find the anthropogenic sources of Pb and B and to find ways to reduce and restore them has been recognized. Based on the results of the analysis, the need to track anthropogenic sources of Pb, B, and Cu was suggested.

4.2. Trace Element Distribution Characterization and Source Analysis

Based on the previously analyzed descriptive statistics (Section 3.1), it was necessary to identify the anthropogenic occurrence characteristics of Mn, Zn, B, Pb, and Cd; the source of Cd-Zn and the cumulative source of B through correlation analysis (Section 3.2); the anthropogenic source of Cu, Pb, B, and Mn through multivariate statistical analysis (Section 3.3); and the anthropogenic source of Pb, B, and Cu through CF and EF evaluation of stream sediments. Based on these results, it was determined that Cd and B were the most common items that needed to be identified and that the occurrence of Pb, which is the most highly contaminated, needed to be investigated.

As mentioned earlier, Cd is known to share the same behavior as Zn. In the case of natural and anthropogenic combined sources of Cd and Zn, most of the contamination levels were low, with EF < 1.5, but relatively high concentrations and EF distributions can be seen in certain areas (left–up) (Figure 7). This is a famous mining area in Buyeo-gun and near the city of Chungyang-gun, and Cd and Zn can be judged to be contaminated by mining. In fact, many papers have confirmed that Cd and Zn, as well as Pb and Cu, are the main causes of soil contamination by mines [110–112].

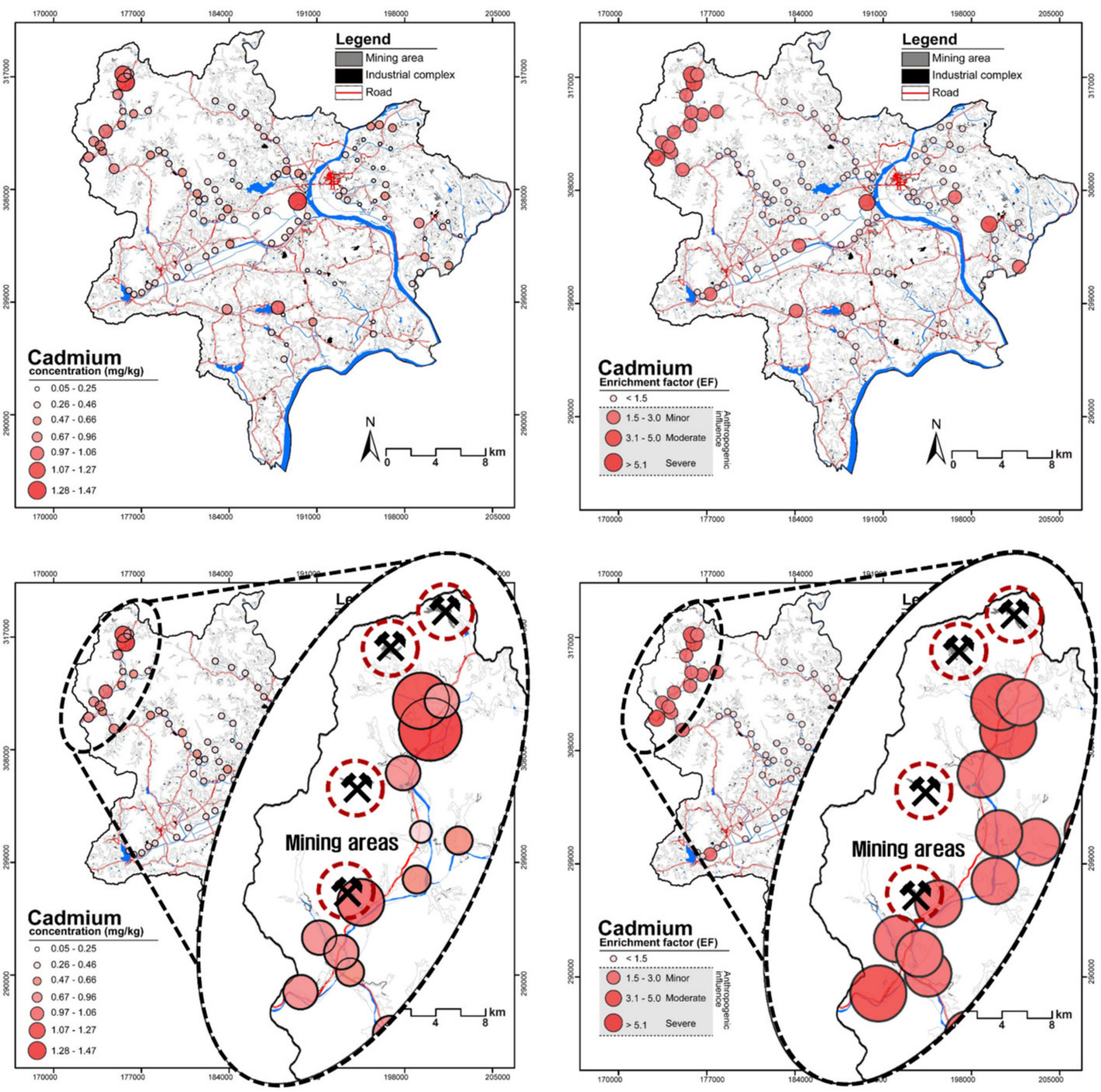

Figure 7. Distribution map of Cd contamination and EF in the study area.

In addition, there is a survey result that this area is a concentrated area of many abandoned mines such as coal, metal, and minerals [113]. In addition, doubts about EF were evaluated in the northeast and additional areas. This can also be estimated as the influence of the concentration of many abandoned mines in Buyeo-gun [64,113].

Pb, which was previously mentioned as an anthropogenic source, was found to have a high overall contamination level (EF > 1.5) and was analyzed to be relatively high in Buyeo IC, a high-traffic area, and areas with the highest number of tourists (Gungnamji, Gudrae, etc.; Figure 8), in addition to the impact of mines [114].

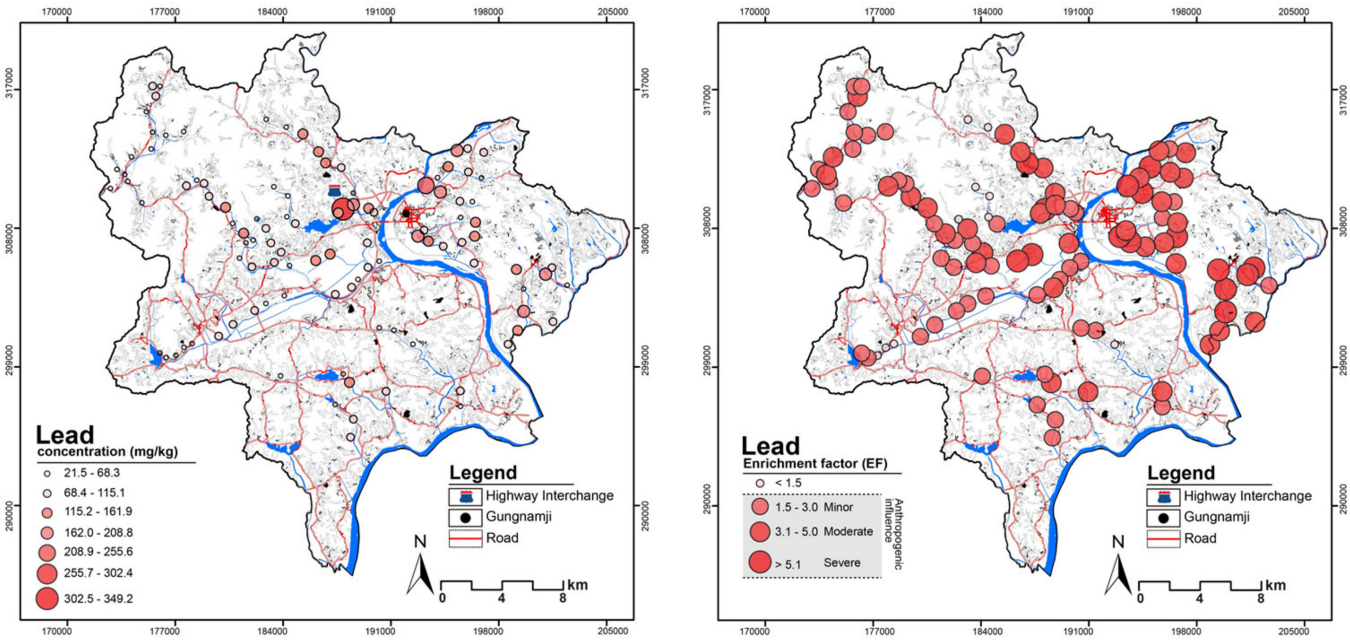

Figure 8. Distribution map of Pb contamination and EF in the study area.

Pb has been suggested as a source of soil contamination in high-traffic areas [115]. The accumulation of Pb in stream sediments in the target area may have been influenced by mines located in the area, and atmospheric deposition of lead from the combustion of fossil fuels (vehicles, etc.) may have accumulated. In addition, the impact of fertilizers and pesticides used in the past at the site can be considered [116–118]. In addition, it can be hypothesized that fossil fuels used in the facility's greenhouses could also have contributed. The impact of existing mines and tourism can be seen in the target area. However, it is difficult to attribute pollution to greenhouses alone. Therefore, a follow-up study is needed to determine whether fossil fuels used in greenhouses contribute to stream soil pollution. This study would need to be conducted in an independent, non-mined area. We suggest this as an area for further research.

Among metal(loid)s in facility horticulture, boron is an essential element for crops and is second only to Fe in agricultural fertilizers [44,109]. Boron in the stream sediments of the target area was analyzed to be the highest in Buyeo-gun (Figure 9).

This highly contaminated space has the largest facility vegetable area (1061 ha) among the target area (2855 ha) (about 60% of the agricultural land). To confirm this, we overlaid the distribution of boron in stream sediments and the area of facility cultivation to investigate the relationship (Figure 9–down). Among the components in the drainage directly discharged from the actual nutrient solution cultivation farms, tomato-based B was found to discharge 1.97 kg/ha in one year [109]. Soil and sediment accumulation of boron is not common in Korea. The investigation of the stream sediment confirmed the high level of boron contamination and confirmed that it is an effect of wastewater from the facility greenhouse. It was confirmed that the nutrient solution used in facility horticulture contains a large amount of (Boric Acid, H_3BO_3) [109]. However, it also supplies high concentrations of Mn and Fe, but the extent of the impact was not confirmed in this study.

Therefore, it is necessary to investigate whether the facility's greenhouse operation affects the accumulation of Mn, Fe, etc., in the surrounding stream sediments. The results of B contamination and EF evaluated in this study show that the contamination level is not high, but the CV is 96.58%. It is judged that it is necessary to further examine this site by adding the general Mn standard of Korean rivers and streams. It is also recognized that it is necessary to investigate whether Mn contained in nutrient solutions used in greenhouses accumulates in streams through further research.

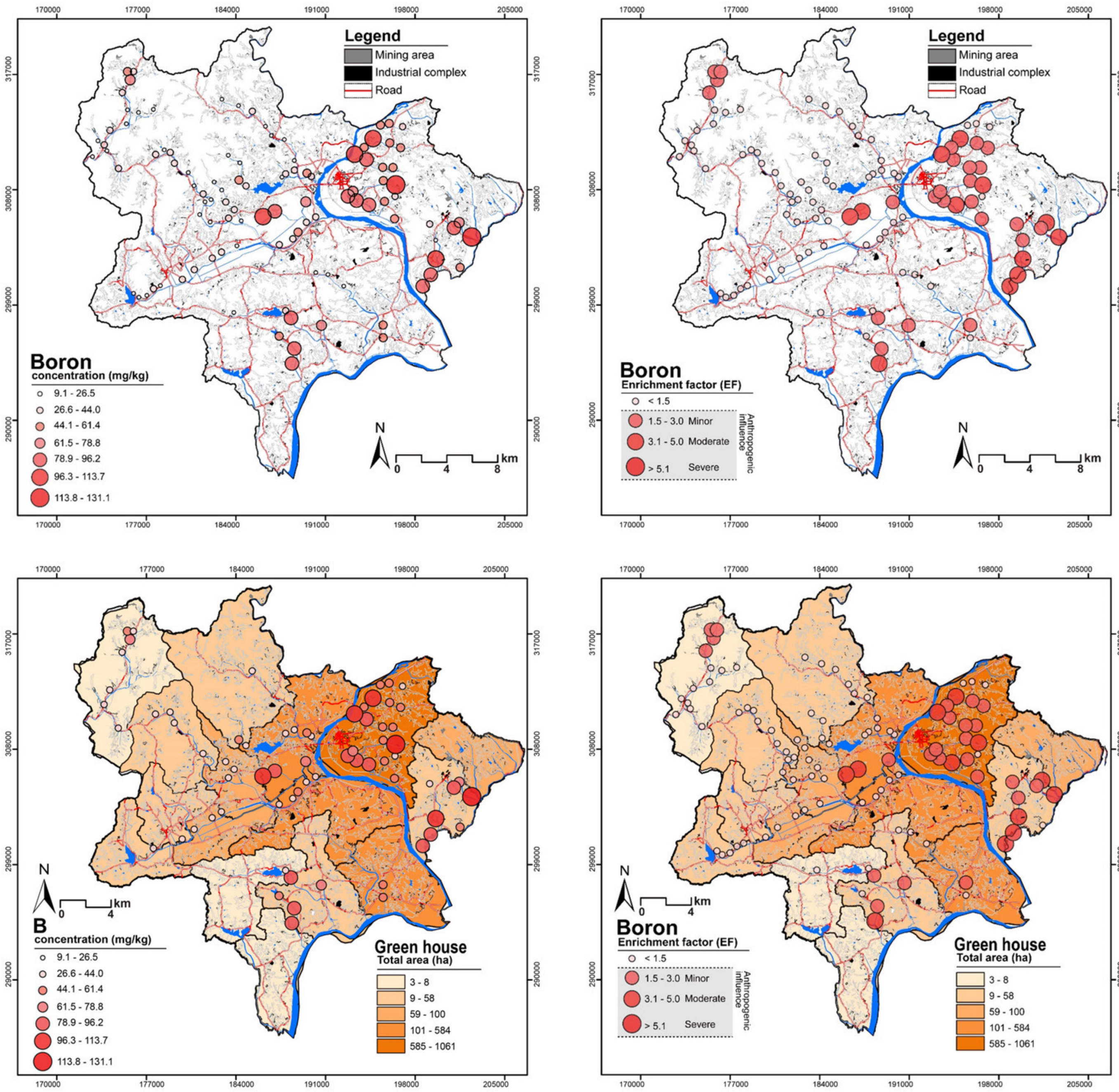

Figure 9. Distribution map of B contamination and EF in the study area (**Up**), Overall map of B contamination and EF in the greenhouse total area (**Down**).

5. Conclusions

The high contamination of Cd was confirmed to be the effect of the famous mining area in Buyeo-gun. Pb was analyzed as an effect of high traffic in addition to the mining area. In addition, it was hypothesized that fertilizers and pesticides used in the past in the target area and fossil fuels used in the facility's greenhouse could also have contributed. There were some low EF areas; they may be analyzed in detail. Therefore, it was suggested that this Pb impact study be more studied in an independent, non-mined area. Boron, which is an essential element for horticultural crops, was analyzed to be highly distributed in the sediments of the stream in the target area. It was confirmed that this contamination was due to the greenhouse fertilizer Boric Acid wastewater. And it was suggested that the cumulative effects of Mn, Fe, etc., should be further examined.

In conclusion, it was determined that facility horticulture wastewater contributes to metal(loid)s in stream sediments. We propose wastewater treatment to prevent this, but it is also necessary to find other ways to utilize it. Water treatment is very costly. If ecological restoration and other agricultural uses are possible, there is no need to incur the cost of water treatment. Research can be utilized to manage wastewater discharges from facility horticulture and prevent contamination of river and stream sediments to achieve a sustainable agricultural environment.

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Data Availability Statement: Data are contained within the article. Additional data can be obtained by contacting the corresponding author of the article.

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Conflicts of Interest: Author Jinkwan Son was employed by the company of Research of Institute of Environment Ecology & Green Space Sanglimwon Co., Ltd. from 1 January 2024. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships.

Appendix A

Table A1. The analysis result of survey 106 stream sediment samples.

No.	pH	EC	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
1	6.86	0.55	0.83	20.88	9.22	7.72	26.12	40.07	14.78	176.18	4425.37
2	6.22	0.72	0.77	29.63	12.23	14.77	45.35	50.92	22.25	241.42	5413.45
3	6.76	0.41	0.53	18.05	8.05	8.82	26.67	30.62	12.52	236.33	3800.07
4	7.14	0.37	0.78	25.55	9.93	9.52	36.13	44.59	12.47	288.28	5507.15
5	8.13	0.80	0.72	34.55	12.88	14.52	57.98	46.88	19.73	319.72	5595.44
6	7.23	1.10	0.87	35.87	25.88	16.63	65.18	95.83	18.37	453.85	5891.40
7	5.79	0.43	0.47	19.50	6.52	8.65	29.85	39.95	12.28	78.48	4237.29
8	6.97	0.30	0.45	20.23	7.85	9.37	31.37	33.65	12.97	111.63	3977.90
9	6.66	0.31	0.47	19.85	5.70	7.50	25.30	28.35	10.62	255.27	3901.81
10	7.18	0.37	0.45	23.33	5.97	7.92	27.12	23.93	10.70	105.02	4011.43
11	6.89	0.44	0.72	37.05	12.92	16.55	52.88	50.77	30.15	145.12	5901.57
12	6.93	0.43	1.43	66.98	19.75	42.80	86.30	79.32	59.25	918.17	8315.03
13	6.60	0.52	1.13	55.80	20.32	23.90	72.95	78.22	43.22	444.12	7372.68
14	5.89	1.98	0.80	38.72	18.95	19.25	53.32	66.05	32.90	259.45	6133.60
15	6.44	1.03	1.47	69.60	29.97	25.60	109.53	184.82	43.55	543.33	7509.20

Table A1. *Cont.*

No.	pH	EC	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
16	5.85	2.56	0.30	90.37	28.47	19.13	124.24	21.77	35.67	594.50	6290.98
17	6.33	1.03	0.40	104.33	37.20	25.70	149.40	30.92	40.98	747.33	7446.87
18	7.40	1.55	0.55	44.02	27.32	20.27	98.92	27.95	26.25	472.07	7228.90
19	5.61	1.47	0.28	84.48	19.97	15.98	109.10	25.48	25.50	402.07	6729.77
20	7.25	0.68	0.45	96.02	27.20	19.25	122.07	39.65	28.00	1192.00	7213.50
21	5.48	1.36	0.13	119.06	16.47	19.87	63.88	20.18	36.32	217.07	7616.57
22	4.93	4.25	0.18	61.55	19.58	10.12	77.82	18.13	16.77	279.45	3662.03
23	5.63	2.75	0.40	80.70	33.37	16.97	168.32	40.18	27.15	211.83	6723.33
24	4.86	2.79	0.52	98.88	37.50	16.00	183.38	55.75	23.62	491.70	7401.82
25	6.68	0.52	0.13	24.53	4.20	3.48	64.78	22.10	5.87	69.40	3053.82
26	5.91	0.77	0.73	33.10	9.12	4.97	135.83	73.75	8.33	146.33	3908.68
27	6.00	0.88	0.37	94.37	27.25	29.23	71.25	20.98	45.67	303.33	7217.20
28	5.81	0.89	0.22	87.17	23.92	18.07	89.40	22.45	25.67	911.56	10,822.68
29	6.28	1.22	0.33	57.33	16.55	14.06	61.13	18.27	27.16	293.25	10,870.95
30	6.16	1.47	0.28	53.32	16.25	14.71	57.40	15.33	25.80	370.70	9930.35
31	6.67	1.04	0.25	52.70	16.70	11.30	88.74	17.22	16.35	592.75	6921.60
32	5.26	0.52	0.13	21.97	3.50	2.90	26.74	4.45	4.88	44.84	5693.93
33	6.52	0.65	0.15	25.91	6.57	4.35	41.70	9.27	11.52	178.15	4441.45
34	6.78	3.17	0.12	20.15	5.00	4.27	39.04	4.38	6.95	139.77	3922.79
35	5.27	1.61	0.53	66.49	18.49	14.23	107.12	17.67	17.07	3215.00	10,514.78
36	4.74	1.78	0.20	90.69	31.51	28.90	126.15	24.33	50.13	317.25	11,471.37
37	7.75	0.85	0.90	31.53	18.78	24.53	55.45	13.17	34.90	335.00	9652.59
38	5.77	1.36	0.72	19.70	11.75	10.63	39.98	7.02	15.00	132.27	6772.78
39	5.95	1.43	0.60	27.42	32.37	17.09	140.03	14.33	23.03	110.92	6748.86
40	6.49	0.98	0.15	47.73	14.40	10.00	76.75	9.28	16.33	136.03	4759.00
41	5.69	1.35	0.17	56.97	11.92	11.12	72.03	11.70	20.35	280.72	5720.68
42	5.84	1.43	0.22	94.72	22.12	20.42	86.32	37.50	31.88	521.33	7593.22
43	6.73	0.72	0.30	93.78	32.52	19.37	157.07	43.58	30.55	556.67	7563.40
44	6.85	2.15	0.33	86.04	30.53	19.35	170.17	37.57	32.68	516.10	7427.05
45	5.35	2.43	0.38	64.13	12.40	11.57	79.20	31.08	18.12	349.42	6531.78
46	7.11	1.87	0.18	44.35	9.12	9.05	48.05	32.60	14.53	270.10	5059.82
47	6.44	1.59	0.38	131.07	23.00	18.20	126.15	38.18	26.67	807.67	8726.93
48	6.29	0.72	0.08	52.80	10.45	8.56	54.57	20.87	16.88	160.37	5709.49
49	6.48	0.77	0.13	55.45	19.11	11.47	70.92	19.88	19.78	174.08	5826.72
50	6.73	0.74	0.17	87.19	20.79	15.31	168.98	22.48	27.52	458.74	7305.10
51	5.43	1.31	0.07	48.42	10.18	13.29	61.60	13.57	17.30	160.23	5244.75
52	5.07	1.35	0.05	53.12	10.72	10.00	70.43	8.63	20.00	97.23	5359.92
53	4.85	3.20	0.13	104.38	21.47	19.88	158.28	17.73	35.05	405.65	7902.47
54	5.89	0.97	0.08	50.88	6.43	6.60	59.55	7.73	11.35	283.82	5265.42
55	5.78	1.18	0.28	104.38	32.52	17.13	240.32	26.18	30.25	905.83	7540.02

Table A1. *Cont.*

No.	pH	EC	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
56	6.49	1.07	0.55	39.47	17.17	13.38	105.02	13.40	20.50	317.80	10,261.73
57	6.43	0.99	0.60	51.37	24.15	21.90	92.07	24.22	32.15	577.50	11,146.62
58	6.24	1.20	0.60	55.78	25.92	20.74	185.38	23.55	32.51	985.17	11,417.83
59	4.83	1.02	0.42	15.30	13.83	10.78	57.67	8.42	16.62	91.87	5526.75
60	5.60	0.95	0.35	17.77	13.73	14.30	70.53	8.77	20.80	127.87	6183.40
61	5.51	1.69	0.40	24.71	23.48	22.22	140.82	11.91	33.01	100.83	7135.90
62	5.09	2.69	0.38	26.42	21.83	19.47	126.22	11.78	26.90	209.22	12,013.00
63	5.70	1.00	0.32	29.22	17.40	25.85	129.64	9.73	32.49	212.58	12,254.50
64	6.56	0.47	0.32	13.76	7.42	8.88	35.67	5.07	12.27	270.40	10,416.20
65	6.29	0.57	0.37	23.58	13.47	14.82	58.52	10.22	20.28	444.48	11,799.60
66	7.18	0.60	0.37	41.65	24.53	23.28	92.67	20.20	33.17	602.50	12,700.97
67	6.88	0.67	0.48	45.07	42.37	33.08	116.30	27.42	45.50	912.17	12,957.73
68	5.77	1.20	0.37	32.83	22.93	18.90	349.16	17.05	27.97	458.88	12,508.22
69	6.20	0.97	0.33	24.43	21.17	17.10	149.80	17.03	25.83	285.70	12,023.85
70	6.34	0.62	0.32	33.42	27.17	28.87	87.53	14.30	45.07	373.83	12,548.53
71	6.28	0.22	0.18	52.92	13.73	43.08	60.47	10.18	60.92	249.22	13,279.18
72	6.54	0.60	0.32	23.87	20.97	20.32	53.57	10.08	31.17	289.77	11,838.88
73	6.38	0.57	0.32	20.60	14.48	16.80	44.43	7.40	26.58	230.15	11,650.22
74	5.49	0.98	0.47	34.17	29.35	24.35	111.85	18.33	33.45	276.02	10,176.38
75	6.15	0.65	0.43	31.02	23.90	24.35	89.98	16.87	31.73	929.33	9973.82
76	6.02	1.98	0.37	20.03	25.38	17.49	95.22	15.88	27.35	170.15	8408.53
77	6.13	0.26	0.28	14.92	9.77	8.92	27.52	9.75	11.68	263.97	6968.23
78	5.87	0.23	0.32	12.93	8.12	8.35	37.22	6.87	12.65	96.60	6148.83
79	5.94	4.29	0.40	35.37	26.95	22.83	109.63	21.62	33.27	484.78	10,293.93
80	4.63	2.78	0.43	26.35	21.53	19.40	78.58	16.40	28.53	272.80	9414.25
81	5.50	0.34	0.33	19.48	13.12	14.58	43.65	10.30	21.12	200.65	8150.08
82	4.76	0.82	0.37	11.50	8.17	7.87	29.50	7.83	12.28	53.78	5814.17
83	5.35	0.67	0.37	15.05	11.87	11.35	39.98	28.77	16.98	104.27	7061.73
84	5.11	1.38	0.55	35.00	28.62	23.80	117.78	23.38	36.30	334.72	10,223.13
85	5.10	1.16	0.40	18.68	14.50	13.08	61.03	11.17	20.38	201.72	7879.78
86	5.45	4.86	0.48	26.43	17.00	15.03	150.45	21.60	23.23	761.75	9337.00
87	6.75	0.30	0.45	28.58	18.67	18.00	107.67	19.82	25.73	625.83	9641.20
88	5.60	0.82	0.40	35.20	23.58	21.45	93.58	18.73	30.38	422.00	10,161.83
89	5.61	0.17	0.35	22.15	12.55	13.20	66.93	12.07	21.82	239.58	8721.18
90	6.43	0.46	0.50	32.72	20.05	20.65	108.48	19.18	33.07	473.60	9897.60
91	4.70	0.27	0.35	42.35	26.83	29.40	63.57	12.38	36.40	447.48	10,701.77
92	4.96	0.75	0.45	39.75	23.35	21.52	97.33	18.92	33.13	529.67	10,175.70
93	5.61	1.08	0.30	44.30	20.97	26.12	65.18	19.15	38.37	353.82	10,376.92
94	6.52	0.38	0.42	36.23	25.78	21.88	109.15	21.13	33.02	343.75	9989.07
95	4.80	0.61	0.33	33.72	18.25	17.68	78.05	15.72	26.82	299.40	9546.33

Table A1. *Cont.*

No.	pH	EC	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
96	7.74	0.48	0.65	23.40	16.42	11.42	59.80	27.74	15.84	404.00	8248.65
97	5.55	0.50	0.40	35.49	13.32	11.97	63.75	10.45	24.27	250.83	7919.05
98	6.25	0.30	0.38	31.97	19.42	17.28	75.32	17.95	29.98	312.87	9149.78
99	4.54	1.14	0.37	28.77	19.48	18.90	79.58	15.07	31.77	173.70	8851.60
100	6.06	0.46	0.38	35.98	23.20	23.88	87.97	16.43	39.80	320.67	10,005.67
101	6.65	0.19	0.33	11.43	5.98	6.83	21.47	6.25	10.85	142.48	5194.95
102	6.56	0.25	0.37	31.81	11.72	14.68	41.88	9.70	24.22	242.82	8775.71
103	6.39	0.28	0.35	9.13	5.83	5.92	21.47	5.12	10.51	108.03	4617.75
104	6.50	0.25	0.38	15.20	11.57	9.52	43.95	10.07	15.87	143.28	7084.77
105	5.34	0.54	0.33	18.85	10.55	10.83	58.35	12.18	16.88	134.83	7973.60
106	5.54	1.39	0.57	43.05	42.50	34.42	176.77	28.32	45.18	840.17	10,629.27

Table A2. The CF assessment result of 106 stream sediment samples.

No.	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
1	3.74	1.14	1.18	0.71	1.74	0.71	0.50	0.41	1.00
2	2.82	1.32	1.28	1.11	2.48	0.74	0.62	0.46	1.00
3	2.79	1.14	1.20	0.95	2.07	0.63	0.49	0.64	1.00
4	2.81	1.12	1.02	0.70	1.94	0.64	0.34	0.54	1.00
5	2.55	1.49	1.31	1.06	3.06	0.66	0.53	0.59	1.00
6	2.92	1.47	2.50	1.15	3.27	1.28	0.47	0.80	1.00
7	2.19	1.11	0.87	0.83	2.08	0.74	0.43	0.19	1.00
8	2.25	1.23	1.12	0.96	2.33	0.67	0.49	0.29	1.00
9	2.38	1.23	0.83	0.78	1.92	0.57	0.41	0.68	1.00
10	2.23	1.40	0.84	0.80	2.00	0.47	0.40	0.27	1.00
11	2.41	1.51	1.24	1.14	2.65	0.68	0.77	0.25	1.00
12	3.43	1.94	1.35	2.10	3.07	0.75	1.07	1.14	1.00
13	3.06	1.82	1.57	1.32	2.92	0.84	0.88	0.62	1.00
14	2.59	1.52	1.75	1.28	2.57	0.85	0.80	0.44	1.00
15	3.88	2.23	2.27	1.39	4.31	1.94	0.87	0.75	1.00
16	0.95	3.46	2.57	1.24	5.84	0.27	0.85	0.98	1.00
17	1.07	3.38	2.84	1.41	5.93	0.33	0.82	1.04	1.00
18	1.51	1.47	2.15	1.14	4.05	0.30	0.54	0.67	1.00
19	0.84	3.03	1.69	0.97	4.79	0.30	0.57	0.62	1.00
20	1.24	3.21	2.14	1.09	5.00	0.43	0.58	1.71	1.00
21	0.35	3.77	1.23	1.06	2.48	0.21	0.71	0.29	1.00
22	1.00	4.05	3.04	1.13	6.28	0.39	0.69	0.79	1.00
23	1.18	2.89	2.82	1.03	7.40	0.47	0.60	0.33	1.00
24	1.39	3.22	2.88	0.88	7.32	0.59	0.48	0.69	1.00
25	0.87	1.94	0.78	0.47	6.27	0.57	0.29	0.23	1.00

Table A2. *Cont.*

No.	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
26	3.73	2.04	1.32	0.52	10.27	1.49	0.32	0.39	1.00
27	1.03	3.15	2.14	1.65	2.92	0.23	0.95	0.43	1.00
28	0.40	1.94	1.26	0.68	2.44	0.16	0.36	0.87	1.00
29	0.61	1.27	0.86	0.53	1.66	0.13	0.37	0.28	1.00
30	0.57	1.29	0.93	0.60	1.71	0.12	0.39	0.39	1.00
31	0.72	1.83	1.37	0.67	3.79	0.20	0.35	0.88	1.00
32	0.47	0.93	0.35	0.21	1.39	0.06	0.13	0.08	1.00
33	0.67	1.41	0.84	0.40	2.78	0.16	0.39	0.41	1.00
34	0.59	1.24	0.72	0.44	2.94	0.09	0.27	0.37	1.00
35	0.99	1.52	1.00	0.55	3.01	0.13	0.24	3.16	1.00
36	0.35	1.90	1.56	1.03	3.25	0.17	0.65	0.29	1.00
37	1.85	0.79	1.11	1.04	1.70	0.11	0.54	0.36	1.00
38	2.10	0.70	0.99	0.64	1.75	0.08	0.33	0.20	1.00
39	1.77	0.98	2.72	1.03	6.13	0.17	0.51	0.17	1.00
40	0.63	2.42	1.72	0.86	4.77	0.15	0.51	0.30	1.00
41	0.58	2.40	1.18	0.79	3.72	0.16	0.53	0.51	1.00
42	0.57	3.01	1.65	1.10	3.36	0.39	0.63	0.71	1.00
43	0.79	2.99	2.44	1.04	6.14	0.45	0.60	0.76	1.00
44	0.88	2.79	2.34	1.06	6.77	0.40	0.66	0.72	1.00
45	1.15	2.37	1.08	0.72	3.58	0.37	0.42	0.55	1.00
46	0.72	2.11	1.02	0.73	2.81	0.51	0.43	0.55	1.00
47	0.87	3.62	1.50	0.85	4.27	0.34	0.46	0.96	1.00
48	0.29	2.23	1.04	0.61	2.83	0.29	0.44	0.29	1.00
49	0.45	2.29	1.86	0.80	3.60	0.27	0.51	0.31	1.00
50	0.45	2.88	1.62	0.85	6.84	0.24	0.56	0.65	1.00
51	0.27	2.22	1.10	1.03	3.47	0.20	0.49	0.32	1.00
52	0.19	2.39	1.14	0.76	3.88	0.13	0.56	0.19	1.00
53	0.34	3.18	1.54	1.03	5.92	0.18	0.66	0.53	1.00
54	0.31	2.33	0.69	0.51	3.34	0.12	0.32	0.56	1.00
55	0.75	3.34	2.45	0.93	9.42	0.27	0.60	1.24	1.00
56	1.07	0.93	0.95	0.53	3.03	0.10	0.30	0.32	1.00
57	1.07	1.11	1.23	0.80	2.44	0.17	0.43	0.54	1.00
58	1.04	1.18	1.29	0.74	4.80	0.16	0.43	0.89	1.00
59	1.50	0.67	1.42	0.80	3.08	0.12	0.45	0.17	1.00
60	1.13	0.69	1.26	0.94	3.37	0.11	0.50	0.21	1.00
61	1.11	0.83	1.87	1.27	5.83	0.13	0.69	0.15	1.00
62	0.63	0.53	1.03	0.66	3.11	0.08	0.34	0.18	1.00
63	0.51	0.57	0.81	0.86	3.13	0.06	0.40	0.18	1.00
64	0.60	0.32	0.40	0.35	1.01	0.04	0.18	0.27	1.00
65	0.62	0.48	0.65	0.51	1.47	0.07	0.26	0.39	1.00

Table A2. *Cont.*

No.	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
66	0.57	0.79	1.10	0.75	2.16	0.13	0.39	0.49	1.00
67	0.74	0.84	1.86	1.04	2.65	0.17	0.53	0.73	1.00
68	0.58	0.63	1.04	0.62	8.25	0.11	0.33	0.38	1.00
69	0.55	0.49	1.00	0.58	3.68	0.11	0.32	0.25	1.00
70	0.50	0.64	1.23	0.94	2.06	0.09	0.54	0.31	1.00
71	0.27	0.96	0.59	1.32	1.35	0.06	0.69	0.19	1.00
72	0.53	0.49	1.01	0.70	1.34	0.07	0.39	0.25	1.00
73	0.54	0.43	0.71	0.59	1.13	0.05	0.34	0.20	1.00
74	0.91	0.81	1.64	0.98	3.25	0.14	0.49	0.28	1.00
75	0.86	0.75	1.36	1.00	2.67	0.13	0.48	0.96	1.00
76	0.87	0.57	1.71	0.85	3.35	0.15	0.49	0.21	1.00
77	0.81	0.52	0.80	0.52	1.17	0.11	0.25	0.39	1.00
78	1.02	0.51	0.75	0.55	1.79	0.09	0.31	0.16	1.00
79	0.77	0.83	1.49	0.90	3.15	0.17	0.48	0.49	1.00
80	0.92	0.67	1.30	0.84	2.47	0.14	0.45	0.30	1.00
81	0.81	0.58	0.91	0.73	1.58	0.10	0.39	0.25	1.00
82	1.25	0.48	0.80	0.55	1.50	0.11	0.32	0.10	1.00
83	1.03	0.51	0.95	0.66	1.67	0.32	0.36	0.15	1.00
84	1.07	0.82	1.59	0.95	3.41	0.18	0.53	0.34	1.00
85	1.01	0.57	1.05	0.68	2.29	0.11	0.39	0.26	1.00
86	1.01	0.68	1.03	0.66	4.76	0.18	0.37	0.84	1.00
87	0.93	0.71	1.10	0.76	3.30	0.16	0.40	0.67	1.00
88	0.78	0.83	1.32	0.86	2.72	0.15	0.45	0.43	1.00
89	0.80	0.61	0.82	0.62	2.27	0.11	0.37	0.28	1.00
90	1.00	0.80	1.15	0.85	3.24	0.15	0.50	0.49	1.00
91	0.65	0.95	1.42	1.12	1.76	0.09	0.51	0.43	1.00
92	0.88	0.94	1.30	0.86	2.83	0.15	0.49	0.54	1.00
93	0.57	1.03	1.15	1.03	1.86	0.15	0.55	0.35	1.00
94	0.83	0.87	1.47	0.89	3.23	0.17	0.49	0.36	1.00
95	0.69	0.85	1.09	0.76	2.42	0.13	0.42	0.32	1.00
96	1.57	0.68	1.13	0.56	2.14	0.26	0.29	0.51	1.00
97	1.00	1.08	0.96	0.62	2.38	0.10	0.46	0.33	1.00
98	0.83	0.84	1.21	0.77	2.43	0.15	0.49	0.35	1.00
99	0.82	0.78	1.25	0.87	2.66	0.13	0.54	0.20	1.00
100	0.76	0.87	1.32	0.97	2.60	0.13	0.60	0.33	1.00
101	1.28	0.53	0.65	0.54	1.22	0.09	0.31	0.28	1.00
102	0.83	0.87	0.76	0.68	1.41	0.09	0.41	0.29	1.00
103	1.51	0.48	0.72	0.52	1.37	0.09	0.34	0.24	1.00
104	1.08	0.52	0.93	0.55	1.83	0.11	0.34	0.21	1.00
105	0.83	0.57	0.75	0.55	2.16	0.12	0.32	0.17	1.00
106	1.06	0.98	2.27	1.32	4.92	0.21	0.64	0.82	1.00

Table A3. The EF assessment result of 106 stream sediment samples.

No.	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
1	2.08	0.63	0.66	0.40	0.97	0.40	0.28	0.23	0.56
2	1.92	0.90	0.87	0.76	1.69	0.50	0.42	0.31	0.68
3	1.33	0.55	0.58	0.45	0.99	0.30	0.24	0.31	0.48
4	1.94	0.77	0.71	0.49	1.34	0.44	0.23	0.37	0.69
5	1.79	1.05	0.92	0.74	2.16	0.46	0.37	0.42	0.70
6	2.17	1.09	1.85	0.85	2.42	0.95	0.35	0.59	0.74
7	1.17	0.59	0.47	0.44	1.11	0.40	0.23	0.10	0.53
8	1.13	0.61	0.56	0.48	1.17	0.33	0.24	0.14	0.50
9	1.17	0.60	0.41	0.38	0.94	0.28	0.20	0.33	0.49
10	1.13	0.71	0.43	0.41	1.01	0.24	0.20	0.14	0.50
11	1.79	1.12	0.92	0.85	1.97	0.50	0.57	0.19	0.74
12	3.58	2.03	1.41	2.19	3.21	0.79	1.12	1.19	1.05
13	2.83	1.69	1.45	1.23	2.71	0.77	0.81	0.58	0.93
14	2.00	1.17	1.35	0.99	1.98	0.65	0.62	0.34	0.77
15	3.67	2.11	2.14	1.31	4.07	1.83	0.82	0.71	0.94
16	0.75	2.74	2.03	0.98	4.62	0.22	0.67	0.77	0.79
17	1.00	3.16	2.66	1.32	5.55	0.31	0.77	0.97	0.94
18	1.38	1.33	1.95	1.04	3.68	0.28	0.49	0.61	0.91
19	0.71	2.56	1.43	0.82	4.06	0.25	0.48	0.52	0.85
20	1.13	2.91	1.94	0.99	4.54	0.39	0.53	1.55	0.91
21	0.33	3.61	1.18	1.02	2.37	0.20	0.68	0.28	0.96
22	0.46	1.87	1.40	0.52	2.89	0.18	0.32	0.36	0.46
23	1.00	2.45	2.38	0.87	6.26	0.40	0.51	0.28	0.85
24	1.29	3.00	2.68	0.82	6.82	0.55	0.44	0.64	0.93
25	0.33	0.74	0.30	0.18	2.41	0.22	0.11	0.09	0.38
26	1.83	1.00	0.65	0.25	5.05	0.73	0.16	0.19	0.49
27	0.93	2.86	1.95	1.50	2.65	0.21	0.86	0.39	0.91
28	0.54	2.64	1.71	0.93	3.32	0.22	0.48	1.18	1.36
29	0.83	1.74	1.18	0.72	2.27	0.18	0.51	0.38	1.37
30	0.71	1.62	1.16	0.75	2.13	0.15	0.49	0.48	1.25
31	0.63	1.60	1.19	0.58	3.30	0.17	0.31	0.77	0.87
32	0.33	0.67	0.25	0.15	0.99	0.04	0.09	0.06	0.72
33	0.38	0.79	0.47	0.22	1.55	0.09	0.22	0.23	0.56
34	0.29	0.61	0.36	0.22	1.45	0.04	0.13	0.18	0.49
35	1.31	2.01	1.32	0.73	3.98	0.17	0.32	4.18	1.32
36	0.50	2.75	2.25	1.48	4.69	0.24	0.94	0.41	1.44
37	2.25	0.96	1.34	1.26	2.06	0.13	0.66	0.44	1.21
38	1.79	0.60	0.84	0.55	1.49	0.07	0.28	0.17	0.85
39	1.50	0.83	2.31	0.88	5.21	0.14	0.43	0.14	0.85
40	0.38	1.45	1.03	0.51	2.85	0.09	0.31	0.18	0.60

Table A3. *Cont.*

No.	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
41	0.42	1.73	0.85	0.57	2.68	0.12	0.38	0.36	0.72
42	0.54	2.87	1.58	1.05	3.21	0.37	0.60	0.68	0.95
43	0.75	2.84	2.32	0.99	5.84	0.43	0.58	0.72	0.95
44	0.82	2.61	2.18	0.99	6.33	0.37	0.62	0.67	0.93
45	0.94	1.94	0.89	0.59	2.94	0.31	0.34	0.45	0.82
46	0.46	1.34	0.65	0.46	1.79	0.32	0.27	0.35	0.64
47	0.96	3.97	1.64	0.93	4.69	0.38	0.50	1.05	1.10
48	0.21	1.60	0.75	0.44	2.03	0.21	0.32	0.21	0.72
49	0.33	1.68	1.36	0.59	2.64	0.20	0.37	0.23	0.73
50	0.42	2.64	1.49	0.78	6.28	0.22	0.52	0.60	0.92
51	0.18	1.47	0.73	0.68	2.29	0.13	0.33	0.21	0.66
52	0.13	1.61	0.77	0.51	2.62	0.09	0.38	0.13	0.67
53	0.33	3.16	1.53	1.02	5.88	0.18	0.66	0.53	0.99
54	0.21	1.54	0.46	0.34	2.21	0.08	0.21	0.37	0.66
55	0.71	3.16	2.32	0.88	8.93	0.26	0.57	1.18	0.95
56	1.38	1.20	1.23	0.69	3.90	0.13	0.39	0.41	1.29
57	1.50	1.56	1.73	1.12	3.42	0.24	0.61	0.75	1.40
58	1.50	1.69	1.85	1.06	6.89	0.23	0.61	1.28	1.44
59	1.04	0.46	0.99	0.55	2.14	0.08	0.31	0.12	0.70
60	0.88	0.54	0.98	0.73	2.62	0.09	0.39	0.17	0.78
61	1.00	0.75	1.68	1.14	5.23	0.12	0.62	0.13	0.90
62	0.96	0.80	1.56	1.00	4.69	0.12	0.51	0.27	1.51
63	0.79	0.89	1.24	1.33	4.82	0.10	0.61	0.28	1.54
64	0.79	0.42	0.53	0.46	1.33	0.05	0.23	0.35	1.31
65	0.92	0.71	0.96	0.76	2.18	0.10	0.38	0.58	1.48
66	0.92	1.26	1.75	1.19	3.44	0.20	0.62	0.78	1.60
67	1.21	1.37	3.03	1.70	4.32	0.27	0.86	1.18	1.63
68	0.92	0.99	1.64	0.97	12.98	0.17	0.53	0.60	1.57
69	0.83	0.74	1.51	0.88	5.57	0.17	0.49	0.37	1.51
70	0.79	1.01	1.94	1.48	3.25	0.14	0.85	0.49	1.58
71	0.46	1.60	0.98	2.21	2.25	0.10	1.15	0.32	1.67
72	0.79	0.72	1.50	1.04	1.99	0.10	0.59	0.38	1.49
73	0.79	0.62	1.03	0.86	1.65	0.07	0.50	0.30	1.47
74	1.17	1.04	2.10	1.25	4.16	0.18	0.63	0.36	1.28
75	1.08	0.94	1.71	1.25	3.35	0.17	0.60	1.21	1.25
76	0.92	0.61	1.81	0.90	3.54	0.16	0.52	0.22	1.06
77	0.71	0.45	0.70	0.46	1.02	0.10	0.22	0.34	0.88
78	0.79	0.39	0.58	0.43	1.38	0.07	0.24	0.13	0.77
79	1.00	1.07	1.93	1.17	4.08	0.21	0.63	0.63	1.29
80	1.08	0.80	1.54	0.99	2.92	0.16	0.54	0.35	1.18

Table A3. *Cont.*

No.	Cd	B	Cu	Ni	Pb	Zn	Cr	Mn	Fe
81	0.83	0.59	0.94	0.75	1.62	0.10	0.40	0.26	1.02
82	0.92	0.35	0.58	0.40	1.10	0.08	0.23	0.07	0.73
83	0.92	0.46	0.85	0.58	1.49	0.28	0.32	0.14	0.89
84	1.38	1.06	2.04	1.22	4.38	0.23	0.68	0.43	1.29
85	1.00	0.57	1.04	0.67	2.27	0.11	0.38	0.26	0.99
86	1.19	0.80	1.21	0.77	5.59	0.21	0.44	0.99	1.17
87	1.13	0.87	1.33	0.92	4.00	0.20	0.48	0.81	1.21
88	1.00	1.07	1.68	1.10	3.48	0.19	0.57	0.55	1.28
89	0.88	0.67	0.90	0.68	2.49	0.12	0.41	0.31	1.10
90	1.25	0.99	1.43	1.06	4.03	0.19	0.62	0.62	1.24
91	0.88	1.28	1.92	1.51	2.36	0.12	0.69	0.58	1.35
92	1.13	1.20	1.67	1.10	3.62	0.19	0.62	0.69	1.28
93	0.75	1.34	1.50	1.34	2.42	0.19	0.72	0.46	1.30
94	1.04	1.10	1.84	1.12	4.06	0.21	0.62	0.45	1.26
95	0.83	1.02	1.30	0.91	2.90	0.16	0.51	0.39	1.20
96	1.63	0.71	1.17	0.59	2.22	0.27	0.30	0.52	1.04
97	1.00	1.08	0.95	0.61	2.37	0.10	0.46	0.33	1.00
98	0.96	0.97	1.39	0.89	2.80	0.18	0.56	0.41	1.15
99	0.92	0.87	1.39	0.97	2.96	0.15	0.60	0.23	1.11
100	0.96	1.09	1.66	1.22	3.27	0.16	0.75	0.42	1.26
101	0.83	0.35	0.43	0.35	0.80	0.06	0.20	0.19	0.65
102	0.92	0.96	0.84	0.75	1.56	0.10	0.46	0.32	1.10
103	0.88	0.28	0.42	0.30	0.80	0.05	0.20	0.14	0.58
104	0.96	0.46	0.83	0.49	1.63	0.10	0.30	0.19	0.89
105	0.83	0.57	0.75	0.56	2.17	0.12	0.32	0.18	1.00
106	1.42	1.30	3.04	1.76	6.57	0.28	0.85	1.09	1.34

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