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Distribution and extent of heavy metal(loid) contamination in agricultural soils as affected by industrial activity

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Abstract

In Korea, rapid industrialization has often caused severe soil and water pollution near industrial complexes. Particularly, heavy metal(loid) contamination of agricultural lands could induce serious long-term problems for crop safety and productivity, requiring continual safety assessment. This study investigated heavy metal(loid) contamination of agricultural lands near fifteen industrial complexes. At each of industrial sites in Gyeongsangbuk-do, topsoils and subsoils were collected at two different distances from each site (0–500 m and 500–1000 m). For comparison, at each site, non-polluted soils were also collected more than 1000 m away from each industrial complex. With the exception of one sample, heavy metal(loid) concentration of all soils were lower than the Korean guidelines for soil contamination. However, the difference between the heavy metal(loid) concentrations of Cu, Pb and Zn in topsoil and subsoil increased the closer the samples were the industrial complexes, which implied that these elements were being generated by industrial activities and were freshly loaded on to near surface soils. While the heavy metal(loid) concentration in the studied sites did not exceed the Korean guideline, the geoaccumulation index of each soil indicated that the degree of Cd, Cu, and Pb contamination was heavily or extremely serious in more than twenty of the examined soils. The elevation of specific metals associated with industrial activity in soils in close proximity to industrial sites is of some concern and should be taken into consideration for the future management of agricultural soils around such complexes as well as the industrial complex operation itself.

Keywords: Geoaccumulation index, Heavy metal(loid), Industrial complexes, Soil contamination

Introduction

Environmental problems around industrial complexes have long been recognized as a national issue [1, 2]. Since many industrial facilities are concentrated in specific zoned areas, a multitude of industrial complexes can become a major point source of large industrial effluxes of sewage, waste, exhaust, and dust into the surrounding environment. For this reason, the soils around industrial complexes would be exposed to environmental pollutants more than the other areas [3]. While dust and wastewater released from industrial facilities can cause environmental pollution, exhausts generated during production and material transport to and from industrial sites are also a significant source of contamination [4–6]. In particular, heavy metal(loid)s contamination of soils near industrial complexes is a major issue worldwide [7]. For instance, Loska et al. [8] found the increased As, Cd, Hg, Pb and Sb, contents in soils near industrial regions in southern Poland. Likewise, high levels of heavy metals reaching 176, 725, 8469 and > 10,000 mg kg^{-1} for Cd, Cu, Pb and Zn were reported in soils around intense industrialized areas in Turkey [7].

Since the mobility of contaminants in soils is generally very low compared to that observed in water or air, contaminants remain in soils for a long time [9]. In addition,

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unlike organic contaminants, since heavy metal(loid)s do not decrease through decomposition or volatilization, discharged of such inorganic moieties from industrial complexes are likely to elevate soil concentrations continuously. Of increasing concern worldwide, is that due to increased industrialization many areas near industrial complexes have been used for agricultural crop production, which raises concerns of food chain contamination. For instance, in Korea, at least 472 industrial complexes are located in rural areas where paddy field and cultivated upland crops are grown [10]. This raises concerns that there is a likelihood of agricultural soil contamination by heavy metal(loid)s from the industrial complexes, which could subsequently affect crop's growth performance, food safety [11, 12] and ultimately cause widespread human health issues. Direct human ingestion of agricultural products grown on contaminated lands is a well-known cause of a wide variety of health issues and some diseases caused by heavy metal(loid) accumulation [13]. For example, Lee and Chon [14] conducted human health risk assessments which showed that the long-term intake of rice, cultivated from As-contaminated soil around an abandoned mine, increased the risk of cancer for the local people.

Thus monitoring of heavy metal(loid) concentrations in the agricultural soils around industrial areas is necessary to accurately ascertain the present level of contamination and the consequential human health risk This information can be used to make better informed policy decisions on the management of industrial areas and nearby agricultural lands on a firm scientific basis. In this paper, the extent of heavy metal(loid) contamination in soils near industrial complexes was examined in three ways. Firstly, the relationship between the levels of metals in the soil and the distance from the industrial complex was evaluated with the hypothesis that if industrial activities had an influence on soil contamination, heavy metal(loid) concentration would be higher in the soils closer to industrial complexes. Secondly, a comparison of heavy metal(loid) content in topsoil and subsoil was conducted from the same geographic location to test the hypothesis that industrial contamination would lead to higher heavy metal(loid) contents in topsoils rather than subsoils. Thirdly, the geoaccumulation index was used to compare the level of contamination in notionally industrial affected regions relative to expected local background concentrations.

Materials and methods

Research areas

Fifteen large industrial complexes in Gyeongsangbuk-do, Korea were selected for this study. The scales of the selected complexes were extensive as each contained a large number of different businesses including those involved in electrical, electronics, food, fiber, machinery, nonferrous metal, steel, smelting and timber production (Table 1), which can all potentially be implicated with heavy metal(loid)s release. All of the lands surrounding the selected industrial sites were cultivated regions, used primarily as paddy fields and upland soils.

Table 1 Description of the fifteen agricultural industrial complexes investigated in the present study

Site	Area (m^2)	Types of industry
A	172	Food, fiber and clothes, timber and paper, petrochemistry, nonmetal, machinery, electrical electronics, manufacturing, non-manufacturing
B	129	Food, timber and paper, petrochemistry, nonmetal, machinery
C	224	Food, fiber and clothes, timber and paper, petrochemistry, manufacturing
D	720	Machinery, electrical electronics, transportation equipment
E	150	Fiber and clothes, steel, machinery, transportation equipment, manufacturing
F	146	Petrochemistry, steel, machinery, transportation equipment, manufacturing
G	330	Food, timber and paper, petrochemistry, nonmetal, steel, machinery, electrical electronics, transportation equipment, non-manufacturing
H	152	Fiber and clothes, timber and paper, petrochemistry, nonmetal, steel, transportation equipment
I	157	Food, fiber and clothes, timber and paper, petrochemistry, nonmetal, steel, machinery, electrical electronics, transportation equipment
J	120	Food, fiber and clothes, petrochemistry, nonmetal, steel
K	206	Food, timber and paper, petrochemistry, nonmetal, steel, machinery, electrical electronics
L	190	Fiber and clothes, petrochemistry, nonmetal, machinery, electrical electronics, manufacturing
M	176	Food, fiber and clothes, timber and paper, petrochemistry, steel, machinery, electrical electronics, manufacturing
N	486	Food, fiber and clothes, petrochemistry, steel, machinery, electrical electronics, transportation equipment, manufacturing
O	252	Food, fiber and clothes, petrochemistry, nonmetal, steel, machinery, manufacturing

Soil sample collection

For soil sampling, agricultural lands near each industrial complex were divided into two sections based on the distance from the boundary of the industrial area. The first section encompassed an area within 500 m of the industrial complex the boundary, while the second section encompassed the area between 500 and 1000 m of the boundary (Fig. 1). This demarcation allowed a determination of whether soils closer to the industrial area were more extensively influenced than soils further from the boundary. In each section, five distinct soil-sampling positions were selected which were evenly distributed within the section and at a similar distance from each other. From each sampling position topsoil (0–20 cm depth) and subsoil (20–40 cm depth) were sampled. For this, at each depth, three augur (7 cm diameter) soil samples taken ~ 20 m apart from each other were collected and composited to obtain a single sample for analysis. As a control, soil samples at > 1000 m from each industrial complex were also collected. Hence, 11 distinct topsoil- and subsoil-samples were collected from the agricultural soils near 15 industrial complexes, totaling 330 soil samples for analysis. The collected soil samples were air-dried and sieved through a 2 mm sieve prior to further analysis.

Analysis

Soil heavy metal(loid) concentration was analyzed following the Korean Standard Method for Examination of Soil [15]. Dried and sieved (< 2 mm) soil samples were ground to a fine powder using a mortar and pestle prior to acid digestion. A subsample of the dried and ground soil (2 g) was digested with aqua regia (9 mL), a mixture of nitric acid and hydrochloric acid (1:3), using a graphite digestion system (OD-98-001, ODLAB, Korea) for 1.5 h at 160 °C. The digested solutions were thereafter diluted to 50 mL in a volumetric flask and filtered using a 0.45μm disposable syringe filter prior to quantitative analysis. The concentration of As, Cd, Cu, Ni, Pb, and Zn in the filtered solution was measured by ICP-OES (8300DV, Perkin Elmer, USA). Mercury was analyzed using a Direct

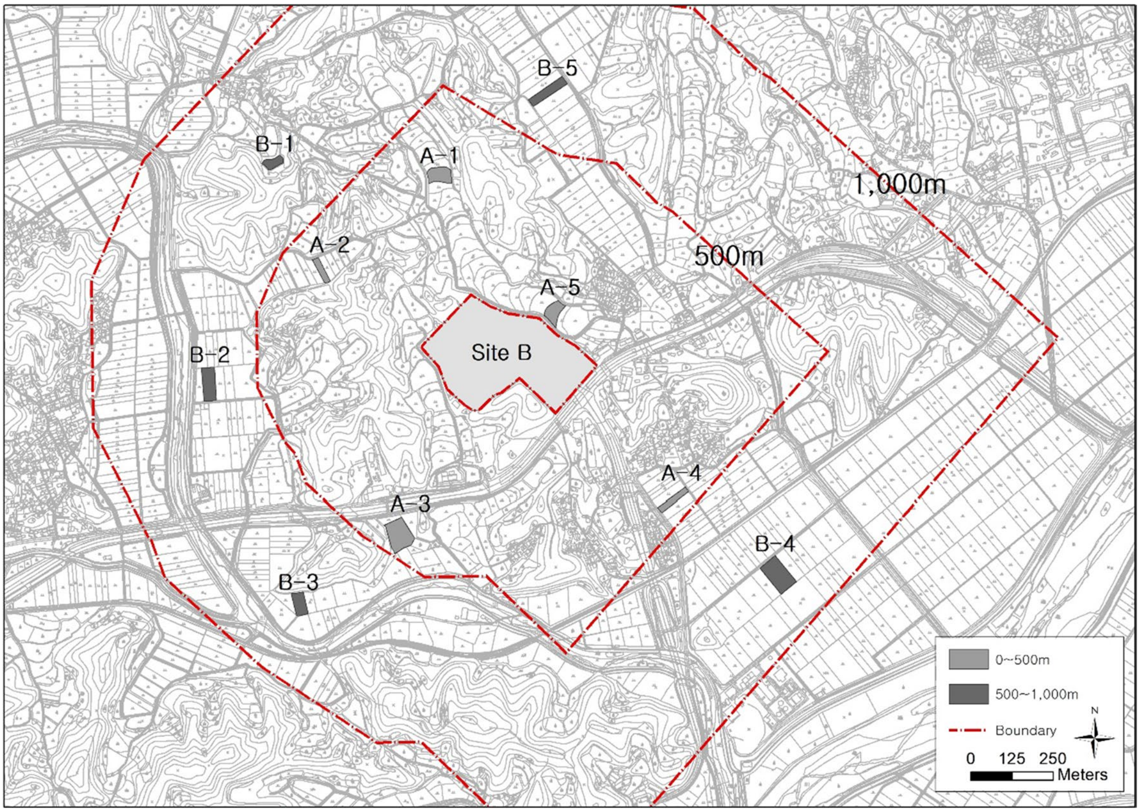

Fig. 1 Example of the general sampling strategy around an industrial complex (site B). The inner line denotes the boundary of the industrial complex, while the middle and outer lines are respectively 500 and 1000 m from the inner boundary. A and B indicate the specific sampling positions at 0–500 and 500–1000 m, respectively. Within each section, five sampling positions were selected having an even distribution within each section encompassed

Table 2 Degree of soil contamination based on geoaccumulation index (I_{geo}) (Huang et al. 2017)

I_{geo} value	Degree of soil contamination
$I_{geo} \leq 0$	Practically uncontaminated
$0 < I_{geo} < 1$	Uncontaminated to moderately contaminated
$1 < I_{geo} < 2$	Moderately contaminated
$2 < I_{geo} < 3$	Moderately to heavily contaminated
$3 < I_{geo} < 4$	Heavily contaminated
$4 < I_{geo} < 5$	Heavily to extremely contaminated
$5 < I_{geo}$	Extremely contaminated

Mercury Analyzer (DMA80, Mileston&T CI, Ltd). For quality assurance and quality control, the recovery rate of Montana soil SRM 2711a (National Institute of Standards & Technology) was also examined.

Data analysis

The effect of industrial activities on heavy metal(loid) accumulation in soils was investigated by comparing heavy metal(loid) concentrations of topsoil and subsoil at each sampling position. The value of subsoil was subtracted from the value of topsoil, and the differences were averaged in three distance groups (0–500 m, 500–1000 m, and > 1000 m) for comparison. Standard errors within each group were also calculated.

To assess the contamination level at the investigation sites, the geoaccumulation index (I_{geo}) was calculated (Eq. 1).

$$I_{geo} = \log_2 [C_n/(1.5 \times B_n)]. \tag{1}$$

where C_n is the concentration of each metal 'n' in the tested soil, and B_n is the background level of

Table 3 Summary of selected heavy metal(loid) concentrations in the agricultural soils around fifteen industrial complexes in Gyeongsangbuk-do, Korea

	As (mg kg^{-1})	Cd (mg kg^{-1})	Cu (mg kg^{-1})	Ni (mg kg^{-1})	Pb (mg kg^{-1})	Zn (mg kg^{-1})	Hg (mg kg^{-1})
Topsoil							
Maximum	57.08	2.95	48.49	25.19	197.89	128.32	0.17
Median	3.57	0.06	12.68	7.96	3.99	37.73	0.03
Minimum	0.17	0.00	0.00	1.23	0.00	19.53	0.01
Average	4.80	0.14	14.82	8.24	7.70	41.10	0.03
Subsoil							
Maximum	42.73	2.09	39.69	33.73	176.14	118.64	0.11
Median	3.89	0.05	11.85	7.62	3.75	35.15	0.02
Minimum	0.34	0.00	0.00	0.94	0.00	18.24	0.01
Average	4.73	0.14	13.24	8.09	6.75	38.20	0.03
Korean guideline	25	4	150	100	200	300	4
No. of samples > Korean guideline	1	0	0	0	0	0	0
Korean natural background	6.83	0.29	15.26	17.68	18.43	54.27	0.06

Table 4 The numbers of soil samples which exceeded the Korean natural background of heavy metal(loid)s

Distance	As	Cd	Cu	Ni	Pb	Zn	Hg
0–500 m							
Topsoil	16	16	36	1	6	8	4
Subsoil	18	12	38	2	5	9	4
Total	34	28	74	3	11	17	8
500–1000 m							
Topsoil	14	7	35	3	0	6	6
Subsoil	13	12	32	2	1	5	3
Total	27	19	67	5	1	11	9
> 1000 m							
Topsoil	4	3	5	0	0	0	0
Subsoil	5	4	6	0	0	0	0
Total	9	7	11	0	0	0	0

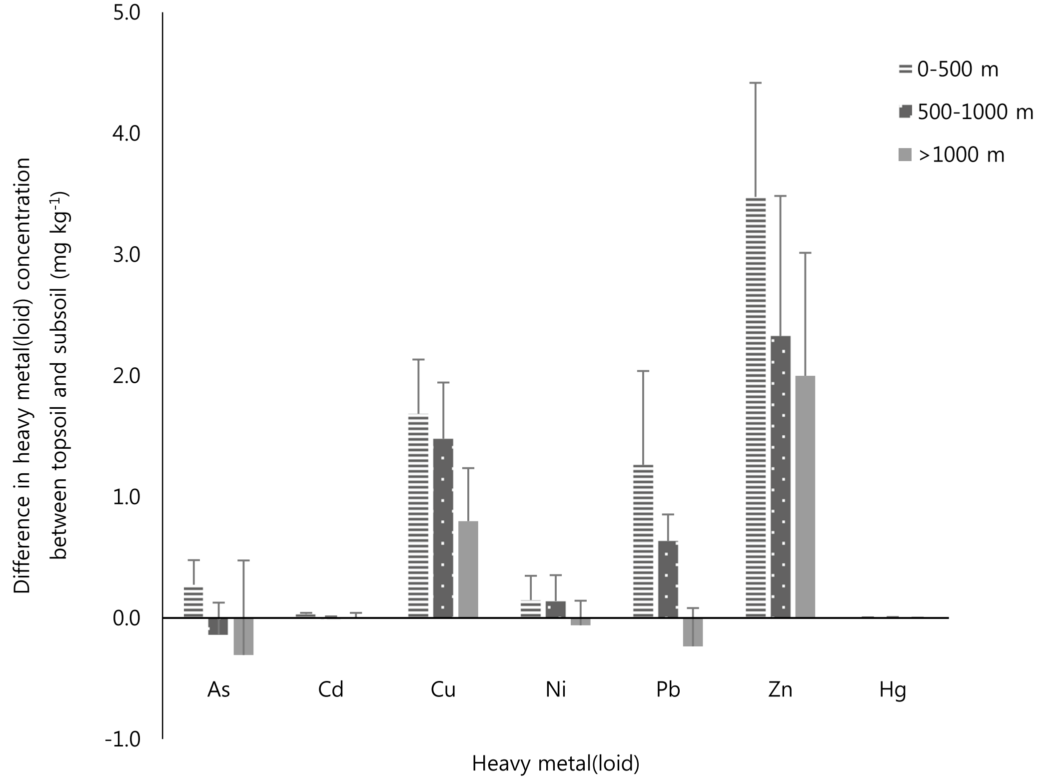

Fig. 2 Averaged difference between heavy metal(loid) concentration of topsoil and subsoil at 0–500 m, 500–1000 m, and > 1000 m from industrial complexes. Differences were determined by subtracting subsoil values from topsoil values. Error bars are standard errors

corresponding metal 'n' in uncontaminated soils. The constant 1.5 is to correct the fluctuation of the background level of 'n' [16]. In this study, the concentration of heavy metal(loid)s in the soils collected at > 1000 m from each industrial complexes were used for B_n. I_{geo} is a widely-used parameter for quantifying the relative levels of heavy metal(loid) contamination of soils and sediments in comparison to expected concentrations in uncontaminated soils [16–20], where I_{geo} classifies the degree of soil contamination into 7 classes (Table 2).

For each heavy metal(loid), I_{geo} values of topsoil and subsoil at each distance group were averaged to compare the general degree of contamination by distances and depths. Standard errors were calculated for each group and the number of soil samples in each I_{geo} range was also counted.

Results and discussion

Distribution of heavy metal(loid) concentration

The broad distribution of soil heavy metal(loid) concentration around the 15 industrial areas is shown in Table 3 relative to soil background concentrations previously determined by Kim et al. [21] for Hg and by Yoon et al. [22] for all other heavy metal(loid)s. Since most values did not exceed the Korean guidelines for heavy metal(loid) concentration in soils, this indicated that almost all of the investigated soils could be legally classified as being non-contaminated. All of the mean values were also under the Korean natural background.

However, while on average the results indicated no significant initial cause for concern this was not true for all of the samples analyzed. As indicated by the maximum values observed for each element needs, a closer examination of the data set indicated that for some soils the heavy metal(loid) concentrations were very high. For example, one soil sample contained 57.1 mg kg^{-1} As which was 9 times higher than the Korean natural background and well above the As guideline value of 6.8 mg kg^{-1}. Likewise, the maximum values of Cd and Pb in topsoil were almost ten times higher than the respective Korean natural background levels. For As, Cd, Cu, Pb, and Zn, the number of soil samples which had concentrations above the Korean natural background values increased the closer the samples were to the industrial complexes (Table 4), which implied that these metal(loid) levels were highly influenced by industrial activities.

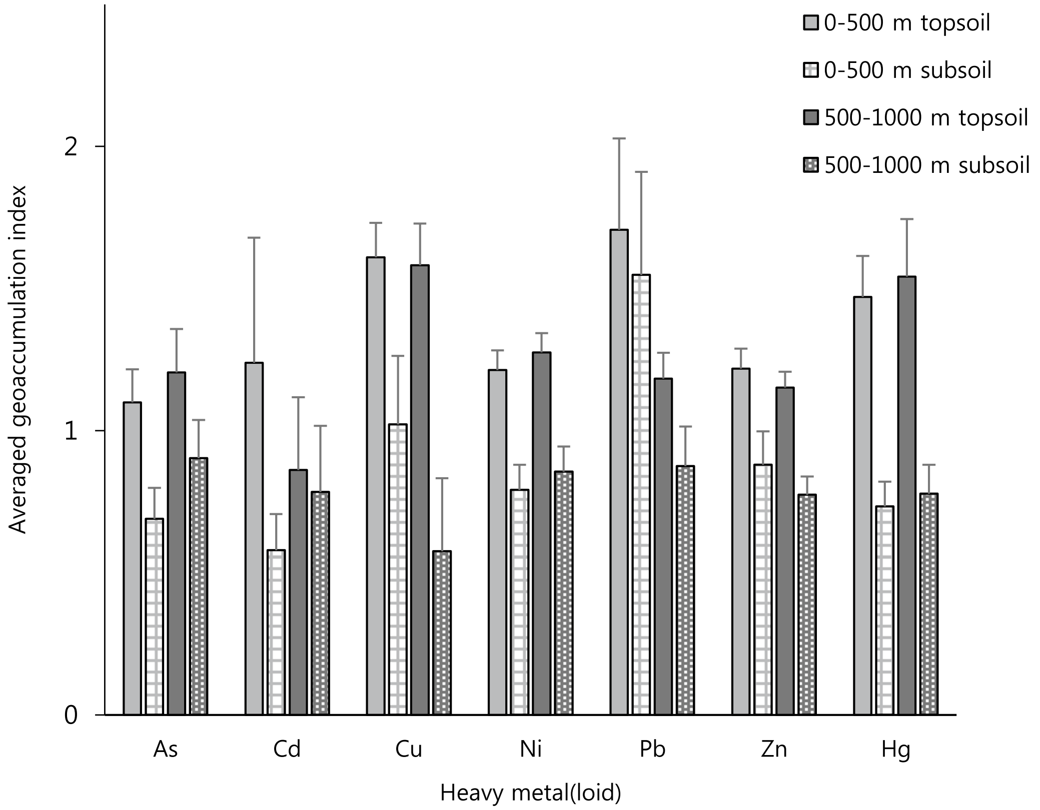

Fig. 3 Averaged I_{geo} values of the soils at two depths and two distances from the industrial complexes for each heavy metal(loid). Error bars are standard errors

Elevated soil concentration of heavy metal(loid)s by industrial complexes

Figure 2 shows the mean difference between heavy metal(loid) concentrations in the topsoil and subsoil. If the value was negative, it indicated that the topsoil had a lower concentration than the subsoil, which would indicate that the original heavy metal(loid)s in the soil were transported from topsoil to subsoil by some mechanism such as precipitation. If the value was positive, the topsoil had a higher metal content than the subsoil, which would indicate that there was an increase of heavy metal(loid) load on the soil from the external environment. In the present study, most of the differences were positive, indicating that the topsoil had higher heavy metal(loid) content than subsoil. While the concentrations of As, Cd, Ni, and Hg were not much different by depth and distance, the deducted values of Cu, Pb, and Zn tended to be highest at 0-500 m soils and lowest at > 1000 m. The most significant differences were observed for Pb which were positive for both distances between 0–1000 m but were negative at > 1000 m. For Cu and Zn, using composts can be a reason that their concentrations in topsoils are higher than subsoils. However, in agreement with the results of Table 4, the load of Cu, Pb, and Zn on the topsoil increased with closeness to the industrial complexes. This strongly supported the notion that industrial activities were closely related to the accumulation of these elements in the nearby agricultural lands.

Soil contamination assessment

Averaged I_{geo} for each heavy metal(loid) were shown in Fig. 3. In topsoil, the averaged I_{geo} were mainly between 1 and 2 (moderately contaminated), while the I_{geo} of the subsoil were mostly under 1 (uncontaminated to moderately contaminated). For all elements, the mean I_{geo} tended to be higher in topsoil than subsoil at each distance. While As, Cd, Ni, and Hg showed little difference between topsoil and subsoil concentrations (Fig. 2), the contamination levels of these elements in topsoil was consistently higher than subsoil (Fig. 3), also indicating some potential influence from industrial activities. For Cd and Pb, the difference between I_{geo} (0–500 m topsoil) and I_{geo} (500–1000 m topsoil) was larger than all other elements and higher for 0–500 m. This implied that the load of these elements were much more highly influenced by industrial activities.

Table 5 The number of soil samples at each I_{geo} range and the total number of soils whose I_{geo} value is over 3

I_{geo} value	Distance (m)	As	Cd	Cu	Ni	Pb	Zn	Hg
$I_{geo} \leq 0$	0–500	6	79	8	4	11	7	6
	500–1000	7	80	5	4	13	6	7
$0 < I_{geo} < 1$	0–500	96	41	71	89	82	81	79
	500–1000	90	43	77	85	79	85	85
$1 < I_{geo} < 2$	0–500	36	12	41	44	24	50	50
	500–1000	33	9	44	46	36	52	37
$2 < I_{geo} < 3$	0–500	5	9	16	9	15	6	8
	500–1000	11	6	14	12	15	6	10
$3 < I_{geo} < 4$	0–500	5	4	9	4	4	3	3
	500–1000	3	5	4	2	4	1	5
$4 < I_{geo} < 5$	0–500	1	0	0	0	2	2	3
	500–1000	2	1	0	1	2	0	5
$5 < I_{geo}$	0–500	1	5	5	0	12	1	1
	500–1000	4	6	6	0	1	0	1
Total no. of sites $I_{geo} > 3$		16	21	24	7	25	7	18

Overall while the averaged I_{geo} values show that the investigated areas were uncontaminated or only moderately contaminated. However, the I_{geo} values for each soil sample showed that there were some heavily or extremely contaminated areas (Table 5). This indicated that although most soils analyzed did not exceed the Korean guideline for heavy metal(loid) (Table 3), the soils near industrial complexes were undergoing contaminated. The total numbers of samples whose I_{geo} values were > 3 were highest for Pb, followed by Cu and Cd. This suggested that Pb, Cu, and Cd were the most-released elements from the associated industrial complexes. This result was similar to the previous research by Kim et al. [23] who had shown that increases in soil Pb, Cu, Cd, and Zn near industrial areas in Taejon, Korea. The main source of Pb was reported to be from fuel [23]. While the presence of Cu and Cd were thought to be most highly associated with nonferrous metal industries and automobiles [4]. Taking a precautionary approach and considering the potential for the future elevation of heavy metals in agricultural soils in the vicinity of industrial complexes, while there seems to be no immediate cause for alarm, future management of agricultural soils should consider this potential as well as continued assessment of local soil quality and the testing of food produced from such regions.

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Authors' contributions

HK and ML carried out most of the experimental works and wrote the manuscript. JHL analyzed and interpreted geoaccumulation index. KHK and KRK contributed in building an object, establishing analytical methods, and supervising all the steps of this study. GO was a major advisor in writing this manuscript including English-writing. All authors read and approved the final manuscript.

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Availability of data and materials

The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Competing interests

The authors declare that they have no competing interests.

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