

Reclamation of mine-degraded agricultural soils from metal mining: lessons from 4 years of monitoring activity in Korea

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Abstract The environmental issues associated with mining have damaged the industry's substantial global economic value. In particular, the mining industry has a negative legacy of contaminated land. The effective reclamation of contaminated soil is therefore required before former mining land can be further developed for residential and commercial purposes. The objective of this study was to technically evaluate the feasibility of reclamation techniques for agricultural soils contaminated with toxic elements (As, Cd, Cu, Pb, and Zn) associated with metal mining. The reclamation methods investigated were covering without stabilization, covering with stabilization, and exchange with stabilization. The thickness of the soil layer used in covering and exchange was in the range of 30–50 cm. Limestone, furnace slag, and a mixture of limestone and furnace slag were applied as soil amendments. After reclamation, the contamination level in surface tillage soils and crops was monitored regularly. Four years of monitoring data revealed that surface soil contamination levels could be maintained at acceptable levels, although at some sites, the metal levels in crops exceeded legislative limits. Soil reclamation at former mining sites in Korea has not yet been perfected, but the results of this study show that there is potential for safe agricultural operations on large sites in a cost-effective manner, as long as the appropriate control of surface soil contamination and adequate agronomic management is undertaken.

Keywords Covering · Exchange · Mined soil · Reclamation · Stabilization

Introduction

The mining industry makes a significant contribution to global civilization, but its benefits are offset by its considerable negative impacts on the environment and the public health of communities around mining sites (Cao 2007). The land surface is damaged and the waste rocks and tailings are often very unstable and will become sources of pollution (Wong 2003). The direct damage from such waste streams involves degraded land fertility for crop cultivation, forests, and grazing, which can ultimately result in abandonment of the land. The indirect damage involves air and water pollution, which eventually leads to losses in biodiversity, amenity, and economic wealth (Bradshaw 1997). As the public awareness of the impacts of toxic elements on animal and human health has grown, the need to develop guidelines and restoration methods for mitigating the risk of toxic elements in contaminated ecosystems has increased (Komárek et al. 2013).

The Ministry of Trade, Industry, and Energy of the Republic of Korea reported that the country currently has approximately 5400 mines, more than 85% of which have been abandoned without any minimum safety precautions, leading to contamination of surrounding ecosystems and communities (KMoTIE 2010). Thousands of unattended mining and refining facilities in Korea contain deposits of tailings and ore waste, along with other waste materials. Korean society has learned from past experience that mitigation of the risk from mining soil is urgently required to

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ensure environmental sustainability and protect public health.

There is no universally accepted terminology to describe the combination of reclamation, restoration, rehabilitation, and replacement of contaminated soil. Within the context of mining, reclamation often refers to the general process whereby a mined contaminated soil is returned to some form of beneficial use, whereas restoration refers to the reinstatement of the pre-mining ecosystem, including all of its structural and functional aspects (Cooke and Johnson 2002). Rehabilitation implies a progression toward reinstatement of the original ecosystem, and replacement is the creation of an alternative ecosystem in place of the original (Bradshaw 1997). In this study, reclamation is the most suitable terminology, because the ultimate aim was to achieve soil reuse for cultivation.

Various techniques can be used to decrease the risk posed by toxic elements in mined soils. The selection of specific technological methods is dependent upon the extent and nature of the contamination, type of soil, characteristics of the contaminated site, availability of materials, and relevant regulations. One major reclamation technique is the removal of the top layer of contaminated soil and its replacement with non-polluted soil (Bell 2001). This means that the contaminated soil is covered with a clean soil layer that is thick enough to restrict exposure to the toxic elements through hand-to-mouth or dermal contact. Replacement of the contaminated soil is a good solution, but it presents scientific, technical, and socio-economic issues regarding its feasibility, efficacy, durability, cost, and local perception (Basta and McGowen 2004; Lombi et al. 2002).

Another option is to stabilize toxic elements and reduce their mobility and availability (Adriano et al. 2004). Compared with other remediation methods, in situ stabilization is less expensive and may provide a long-term solution via reductions in the mobility and availability of toxic elements, either by precipitation or by sorption. In situ stabilization techniques also take advantage of the physicochemical and biological properties of toxic elements, do not generate by-products, and are less expensive than the alternatives. They are therefore suitable for reclamation of extensive areas of low-value land. However, this technique does not change the contamination level, and thus there are issues associated with long-term stability of the land. This type of reclamation can be difficult for cases of multi-element contamination, such as co-contamination with both As and heavy metals (Cd, Cu, Pb, etc.) (Lee et al. 2011a).

Another option is to use plants to remediate metal-contaminated soils (Cunningham et al. 1995). The phytoremediation of contaminated soils is the process of using plants to deliver the most cost-effective means of

mitigating the risks associated with a contaminated site. Phytoextraction is the use of plants to remove toxic elements from soil, which relies on the growth of high-biomass plants. Although it sounds promising, if plant growth prevents the subsequent use of the soil for agriculture, it cannot be viable. In addition, both phytoextraction and phytoremediation are relatively slow processes. Furthermore, the disposal of the contaminated biomass is also an issue (Sas-Nowosielska et al. 2004).

Soil reclamation practices have been widely applied in mining areas in Korea since 2006. In Korea, two kinds of sites require remediation and/or reclamation: (1) those exceeding the “trigger value” of soil contamination and (2) those exceeding the Korean legislative limit for crops, as authorized by the Ministry of Environment of the Republic of Korea. The methods used to deal with contaminated soil entail the physical covering and exchange of soil and chemical stabilization. There have been few studies that have investigated the effects of agricultural soil reclamation and its impact on the concentrations of toxic elements in crops and on soil quality. The objective of this study was to evaluate the technical feasibility and applicability of agricultural soil reclamation based on the covering and exchange of soil together with stabilization.

Materials and methods

Remediated mines and soil reclamation

In this study, a number of soil samples from nine mining locations were collected. One sample (a composite sample from five sites) from each area of farmland was obtained and analyzed. The details of the remediated mines are provided in Table 1.

Table 1 Details of reclaimed mining sites in Korea

Mine	Contaminants	Methods	Amendment
Yunhwa (YH)	As, Cd, Zn	E, CS	LS 3% + FS 2%
Dunjun (DJ)	As, Cd, Zn	C, CS	FS 1%
Eungog (EG)	Cd, Zn	C	–
Yonghwa (YW)	Cd, Cu, Zn	CS	LS 5%
Gupoong (GP)	Cd, Cu, Pb, Zn	ES	LS 5%
Munbak (MB)	Cd, Pb, Zn	CS	LS 5%
Gumjang (GJ)	Cd, Cu, Pb, Zn	ES	LS 5%
Yanggudong (YGD)	Cd, Cu, Pb, Zn	CS	LS 5% + FS 2%
Sanyang (SY)	As, Cd	CS	LS 5% + FS 2%

C Covering, CS covering and stabilization, ES exchanging and stabilization, LS limestone, FS furnace slag

The soil reclamation methods were (1) simple covering without stabilization (C), covering of contaminated soil with uncontaminated soil, (2) covering with stabilization (CS), i.e., controlling the bioavailability and/or mobility of contaminants in soil with amendments and covering the contaminated soil with uncontaminated soil, and (3) exchanging of contaminated soil and stabilization (ES), i.e., exchanging the contaminated soil with uncontaminated soil and stabilizing the remaining contaminated soil (Fig. 1). The thickness of the soil layers used for covering and exchange was 30–50 cm for both the covering and exchange methods. To stabilize the toxic metals in soil, three different amendments were applied: limestone, furnace slag, and a mixture of limestone and furnace slag. The soils used as a cover had a mostly sandy loam texture, and the contamination levels were below the Korean legislative trigger value for soil contamination.

Soils and crops monitoring

After remediation, the contamination levels in both the surface tillage soils and crops were monitored once a year. Soil contamination levels were assessed using five composite samples from each area of reclaimed farmland. Soil samples were mixed, dried at room temperature, disaggregated, and passed through a 2-mm sieve. Samples were analyzed for the total concentrations of five major trace elements (As, Cd, Cu, Pb, and Zn). Pseudo-total trace element concentrations were determined using inductively

coupled plasma optical emission spectrometry (ICP-OES: Optima 7300, PerkinElmer, Waltham, MA, USA) after digesting the samples with aqua regia (HCl:HNO₃ = 3:1) in accordance with ISO 11466 (1995).

Harvested crops cultivated in reclaimed soils were rinsed with distilled water and dried at 80 °C for 48 h. The edible parts of the harvested crops were digested in hot nitric acid (> 80 °C) using a block digester, and the resultant solutions were filtered. The Cd and Pb concentrations in the edible parts were determined by ICP-OES. Spinach leaves were analyzed alongside a certified plant material (i.e., NIST 1570a) to ensure the quality of the plant digestion analyses. The average recoveries were 93–102 and 101–107% for Cd and Pb, respectively. Trace element concentrations expressed in mg kg⁻¹ fresh weight were compared with the Korean legislative values, which are given in Table 2.

Soil contamination assessment method

The assessment of soil contamination was conducted using the pollution index (PI), calculated using the modified formula below, based on the equation suggested by Hakanson (1980).

$$PI = \frac{C_n}{C_t}$$

where *C_n* is the concentration of the contaminant in soil and *C_t* is the trigger value for soil in Korea.



Fig. 1 A schematic diagram of soil reclamation methods for agricultural applications. **a** Stabilization and covering, **b** exchanging and covering

Table 2 Maximum acceptable metal concentrations in vegetables for human consumption (mg kg^{-1} fresh weight)

	Rice, wheat	Barely	Beans	Fruits	Leaf vegetables	Root vegetables	Red pepper
Cd	0.2	0.1	0.1	0.1	0.2	0.1	0.05
Pb	0.2	0.1	0.2	0.05	0.3	0.1	0.1

To compare the degree of contamination, soil contamination indices were divided into five grades according to their classification criteria (Table 3). The classifications used in the PI were adjusted based on the definitions given by Hakanson (1980).

Results and discussion

Under Korean law, reclaimed land should be returned to its original owners. In addition, the sustainability of reclamation largely depends on subsequent agricultural activities. After reclamation, comprehensive maintenance programs are conducted to monitor the stability and durability of the reclaimed soil by evaluating soil contamination levels in the surface soil (< 30 cm). Figure 2 shows the surface soil monitoring results from nine mining sites for the past 4 years, and Table 4 shows the PI value in the surface soils. In most cases, the contamination level of the surface soil did not change significantly over the past 4 years. Except for two mines (YH and DJ), all PI values indicated that the soils were uncontaminated. The monitoring results indicate that covering and/or exchanging with stabilization can decrease public exposure to contaminants and reduce the levels of toxic metals associated with ingested soil and dermal exposure.

Toxic element enrichment of reclaimed soils might be due to (1) contamination introduced by farmers during cropping, (2) the addition of contaminated soils or compost, (3) the use of contaminated water for irrigation, and (4) the remobilization of contaminants from stabilization layers (Douay et al. 2008). From such results, it can be inferred that reclaimed soils were not disturbed by

agronomic activities, at least from a soil contamination perspective.

Table 5 shows the contamination levels in crops found in this study. With regard to the Korean legislative limits, the residual concentrations in crops never exceed the limits for both Cd and Pb. While different from soil contamination, Cd and Pb concentrations in some crops (i.e., bean, Chinese cabbage, and rice) exceeded the recommended concentrations in food items. Crop contamination monitoring results revealed that the bioavailability of subsoil contaminants was not controlled efficiently in some cases. More attention should be given to soil management practices to prevent the potential hazard of exposure to toxic elements via the food chain.

From the 4 years of monitoring results, it was evident that there were challenges faced in the reclamation of mined land, and some suggestions on how best to overcome them are given as follows. First, there is a need to develop soil reclamation guidelines and the optimal management practices for the use of reclaimed soil that can be easily understood and implemented by farmers; furthermore, the people responsible for the reclamation practices should be trained using appropriate manuals. Guidelines are required for various agricultural scenarios, and they should advise on how best to enable plant growth and to prevent accumulation of toxic elements, the transfer of contaminants from soil to the edible parts of the plant, and the dispersion of contaminants in the environment. The guidelines should adopt the principle of the phytoexclusion strategy, which includes the selection and breeding of low-accumulating cultivars, reduction of bioavailability in soil, and restriction of their potential uptake and translocation by plants (Tang et al. 2012). For further crop safety, the currently adopted reclamation methods such as covering with stabilization and exchanging with stabilization could be accompanied by a “capillary break” to prevent the upward migration of contaminants through the capillary movement of soil water (Mahimairaja et al. 2005).

Second, to improve the existing regulations, research should be conducted to strengthen the ecological and/or environmental risk assessment procedure, the criteria for assessing the success of reclamation, and the practical reclamation technologies used in former mining areas. The dynamics and restoration of soil-based ecosystem processes have received little systematic attention (Adriano

Table 3 Classification of soil contamination according to Hakanson (1980)

Index class	PI	Description of classes
I	$\text{PI} < 1$	Uncontaminated
II	$1 \leq \text{PI} < 3$	Moderately contaminated
III	$3 \leq \text{PI} < 6$	Considerably contaminated
IV	$6 \leq \text{PI} < 12$	Highly contaminated
V	$12 \leq \text{PI}$	Extremely contaminated

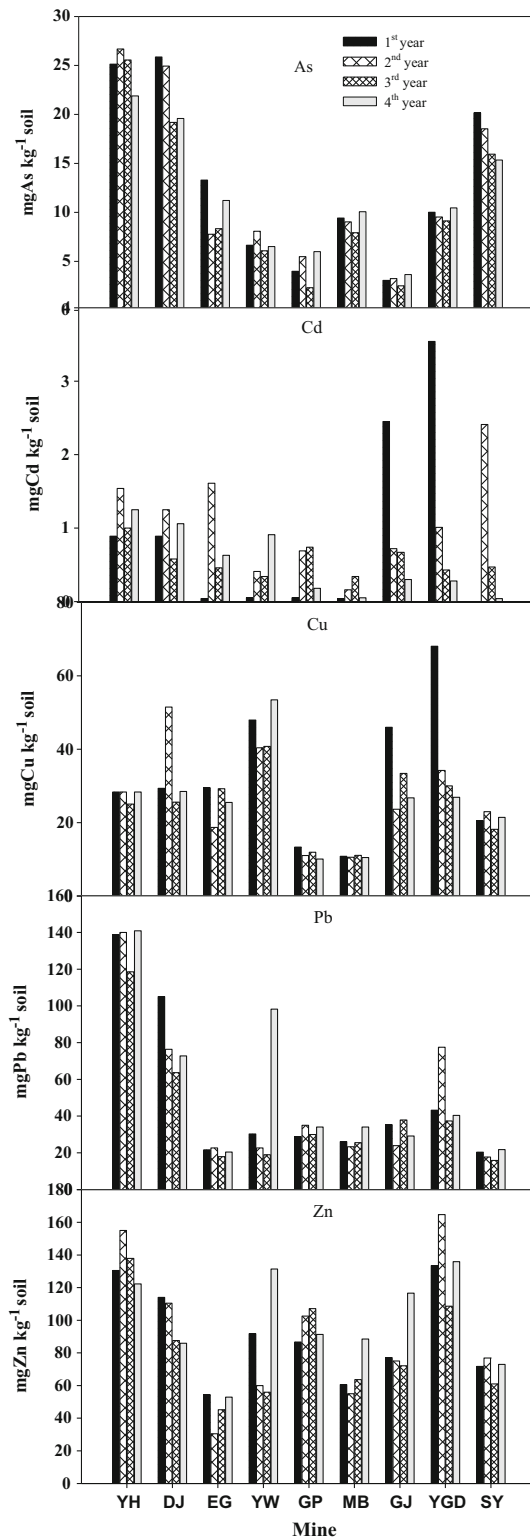


Fig. 2 Four years of monitoring data of surface soil contamination levels

et al. 2004; Mench et al. 2006). Chemical data alone are not sufficient to evaluate the toxic effects of pollutants and to characterize contaminated environments, because such data

do not indicate the effects of chemicals on organisms or the interactions among contaminants, the soil matrix, and biota (Brown et al. 2004; Leitgib et al. 2007). The recovery of derelict soils should be assessed not only using the soil chemical characteristics determined by conventional analytical tests and extraction procedures, but also by assays that measure the restoration of soil habitat function (Brown et al. 2004; Lee et al. 2011b). In situ chemical stabilization relies on decreasing the bioavailability of toxic elements, because it is designed to remediate soils, not by removing toxic elements from soil, but by fixing them in more stable forms (Hamon et al. 2002). Therefore, the identification of the chemical form of an element or a combination of toxic elements as the bioavailable fraction is fundamental for improving remediation (Peijnenburg et al. 2007). As in Korea, in other countries, the guidelines for the remediation of contaminated soil are based on the total concentrations of specific elements. Thus, the development of monitoring tools that measure the availability of trace elements is crucial.

Third, innovative, cost-effective, low-input technologies are needed to manage the reclamation of contaminated agricultural soils and to encourage public awareness for community acceptance. As previously mentioned, some physicochemical methods such as covering, exchanging, and washing are very effective at lowering the risks posed by contaminants, but they may be expensive to implement and are not environmentally benign. Furthermore, it may be difficult to obtain non-polluted soil to replace excavated soil. In terms of stabilization, the technologies available to treat As and other metal contaminants simultaneously are insufficient. Various in situ stabilization techniques are available for cationic metals, but feasible treatment options for As-polluted environments are limited (Lombi et al. 2000). Thus, the development of stabilization methods for multi-element contamination is essential.

Conclusions

The reclamation of soils in former mining areas in Korea has not yet been perfected, but there is the potential for safe agricultural activity on large sites in a cost-effective manner, if the appropriate control of surface soil contamination and adequate agronomic management can be attained. There are several challenges to be faced in the reclamation of mined land in Korea. First, there is the need for the development of reclamation guidelines and the optimal management practices for the use of reclaimed land. Second, research should be conducted on the ecological and/or environmental risk assessment criteria for assessing the success of reclamation. Finally, innovative, low-cost, and low-input technologies are needed to manage

Table 4 Soil contamination levels expressed by PI value

Mine	Pollution index ^a																			
	As				Cd				Cu				Pb				Zn			
	1st	2nd	3rd	4th	1st	2nd	3rd	4th	1st	2nd	3rd	4th	1st	2nd	3rd	4th	1st	2nd	3rd	4th
YH	1.01 (II)	1.07 (II)	1.02 (II)	0.88 (I)	0.22 (I)	0.39 (I)	0.25 (I)	0.31 (I)	0.28 (I)	0.28 (I)	0.25 (I)	0.28 (I)	0.69 (I)	0.70 (I)	0.59 (I)	0.70 (I)	0.43 (I)	0.52 (I)	0.46 (I)	0.41 (I)
DJ	1.03 (II)	1.00 (II)	0.77 (I)	0.78 (I)	0.22 (I)	0.31 (I)	0.15 (I)	0.27 (I)	0.29 (I)	0.51 (I)	0.26 (I)	0.28 (I)	0.53 (I)	0.38 (I)	0.32 (I)	0.36 (I)	0.38 (I)	0.37 (I)	0.29 (I)	0.29 (I)
EG	0.53 (I)	0.31 (I)	0.33 (I)	0.45 (I)	0.01 (I)	0.40 (I)	0.12 (I)	0.16 (I)	0.30 (I)	0.19 (I)	0.29 (I)	0.26 (I)	0.11 (I)	0.11 (I)	0.09 (I)	0.10 (I)	0.18 (I)	0.10 (I)	0.15 (I)	0.18 (I)
YW	0.27 (I)	0.32 (I)	0.24 (I)	0.26 (I)	0.01 (I)	0.10 (I)	0.09 (I)	0.23 (I)	0.48 (I)	0.40 (I)	0.41 (I)	0.53 (I)	0.15 (I)	0.11 (I)	0.09 (I)	0.49 (I)	0.31 (I)	0.20 (I)	0.19 (I)	0.44 (I)
GP	0.16 (I)	0.22 (I)	0.09 (I)	0.24 (I)	0.01 (I)	0.17 (I)	0.19 (I)	0.05 (I)	0.13 (I)	0.11 (I)	0.12 (I)	0.01 (I)	0.14 (I)	0.17 (I)	0.15 (I)	0.17 (I)	0.29 (I)	0.34 (I)	0.36 (I)	0.30 (I)
MB	0.38 (I)	0.36 (I)	0.32 (I)	0.40 (I)	0.01 (I)	0.04 (I)	0.09 (I)	0.01 (I)	0.11 (I)	0.11 (I)	0.11 (I)	0.02 (I)	0.13 (I)	0.12 (I)	0.13 (I)	0.17 (I)	0.20 (I)	0.18 (I)	0.21 (I)	0.29 (I)
GJ	0.12 (I)	0.13 (I)	0.10 (I)	0.15 (I)	0.61 (I)	0.18 (I)	0.17 (I)	0.08 (I)	0.46 (I)	0.24 (I)	0.33 (I)	0.07 (I)	0.18 (I)	0.12 (I)	0.19 (I)	0.15 (I)	0.26 (I)	0.25 (I)	0.24 (I)	0.39 (I)
YGD	0.40 (I)	0.38 (I)	0.37 (I)	0.42 (I)	0.89 (I)	0.25 (I)	0.11 (I)	0.07 (I)	0.68 (I)	0.34 (I)	0.30 (I)	0.07 (I)	0.22 (I)	0.39 (I)	0.19 (I)	0.20 (I)	0.44 (I)	0.55 (I)	0.36 (I)	0.45 (I)
SY	0.81 (I)	0.74 (I)	0.64 (I)	0.61 (I)	0.00 (I)	0.06 (I)	0.12 (I)	0.01 (I)	0.01 (I)	0.23 (I)	0.18 (I)	0.01 (I)	0.10 (I)	0.09 (I)	0.08 (I)	0.11 (I)	0.24 (I)	0.26 (I)	0.20 (I)	0.24 (I)

The number in parentheses is the classification of soil contamination according to Hakanson (1980)

^aThe pollution index (PI) of soil was determined using the trigger value for soil contamination in Korea. Trigger values for soil contamination in Korea are 25, 4, 100, 200, and 300 mg kg⁻¹ for As, Cd, Cu, Pb, and Zn, respectively

Table 5 Contamination levels in crops grown in reclaimed soil (mg kg⁻¹ fresh weight)

Mine	Crop	2011		2012		2013	
		Cd	Pb	Cd	Pb	Cd	Pb
YH	Bean	0.12	0.33	< 0.005	< 0.005	0.21	0.29
	Red pepper	–	–	< 0.005	< 0.005	–	–
DJ	Chinese cabbage	0.41	0.79	< 0.005	< 0.005	0.49	< 0.005
EG	Red pepper	0.08	< 0.005	–	–	–	–
	Chinese cabbage	–	–	< 0.005	0.21	–	–
YW	Bean	–	–	–	–	< 0.005	0.29
	Rice	< 0.005	< 0.005	< 0.005	< 0.005	0.08	0.04
GP	Lettuce	–	–	–	–	0.15	< 0.005
	Rice	< 0.005	< 0.005	< 0.005	< 0.005	0.09	< 0.005
MB	Chinese cabbage	–	–	–	–	0.08	0.14
	Rice	< 0.005	0.05	< 0.005	< 0.005	< 0.005	< 0.005
GJ	Rice	0.02	0.05	< 0.005	0.03	< 0.005	0.02
YGD	Rice	0.09	0.34	< 0.005	< 0.005	0.03	< 0.005
SY	Rice	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	0.09
	Bean	< 0.005	< 0.005	–	–	< 0.005	0.23

Bold letters indicate over the maximum limit for Cd and Pb in food items in Korea

the reclamation of contaminated agricultural soils and encourage community acceptance.

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