

Material Recovery and Environmental Impact by Informal E-Waste Recycling Site in the Philippines

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Abstract Waste electrical and electronic equipment (WEEE or e-waste) contains both valuable and hazardous substances and there is great demand for metal scrap. To better understand both material recovery and the environmental impacts by informal recycling of e-waste, and to find potential ways to improve the process, we carried out a field survey at an informal recycling site in the Philippines. We identified the Au recovery process used at the site and evaluated the layout of the recycling site. We collected 31 soil samples at the recycling site and analyzed the metal contents of each sample to clarify the metal distribution within the site. We determined that valuable substances (such as Au), as well as hazardous substances (such as Pb), were scattered throughout the soil at the informal recycling site. The results of our cluster analysis indicated that Au, Ag, Pb, and Sb were categorized in the same group. Improvements are needed in the metal recovery process and in hazardous substance emission control in the informal recycling.

Keywords E-waste • Informal recycling • Hazardous substances • Valuable substances

1 Introduction

Waste electrical and electronic equipment (WEEE or e-waste) is of great concern among researchers and policymakers because of its potential as a source of serious environmental pollution. Both domestically generated and imported wastes are of concern. E-waste contains both valuable and hazardous substances, however, and there is great demand for metal scrap. Oguchi [1, 2] showed the contents of toxic metals, precious metals, and other common metals in the printed circuit boards of

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21 and 24 types of WEEE, suggesting the importance of controlling these substances in recycling and waste management processes. Valuable substances such as precious and rare metals should be recovered effectively while at the same time the emissions of hazardous substances into the environment should be controlled.

Although the EU countries, as well as Japan, South Korea, and China, have introduced e-waste management systems [1, 3–6], China and other developing Asian countries still face environmental problems in informal recycling sectors. There is no clear definition of the concept of “informal,” and the term “informal” has often been characterized as being beyond the reach of different levels and mechanisms of official governance; lacking in regulation, structure, and institutionalization; and as non-registered and illegal [7].

Many studies of the health and environmental impacts of informal recycling of e-waste have been reported and reviewed. Sepúlveda et al. [8] examined improper recycling techniques such as dumping, dismantling, and inappropriate shredding, burning, and acid leaching; they also reviewed the related environmental impacts. Fujimori et al. [9] investigated the metal content of surface matrices such as soil and dust at both formal and informal recycling sites in the Philippines and determined that the health risks posed by toxic metals in dust were high for workers—even those at formal sites.

Although there have been many reports about the environmental impacts of hazardous substances in e-waste, very few studies have examined the fate of valuable substances at informal recycling sites. Keller [10] carried out excellent work for assessment of Au recovery processes in India, including cyanide leaching, mercury amalgamation, and Au stripping, but the smelting is common in the Philippines and other Southeast Asian countries. To better understand both material recovery and the environmental impacts by informal recycling of e-waste, and to find potential ways to improve the process, we carried out a field survey at an informal recycling site in the Philippines. We specifically focused on a precious metal recovery process and investigated the fate of metals as well as metal distribution on the surface soil at the site.

2 Materials and Methods

2.1 *Surveyed Recycling Site*

Field visit was carried out in January 2012. The recycling facility is situated in the City of Meycauayan, Bulacan, Philippines, and has been in operation for more than 30 years. It is located in a mixed residential and commercial neighborhood, and a perimeter fence effectively isolates it from the neighboring business establishments, residences, and a vacant lot near the back of the facility. The facility primarily buys e-waste from industrial facilities for dismantling. The parts are dismantled in a segregation area and then sold to junkshop owners, who usually

pick up the materials at the facility. The facility is also engaged in the transportation of hazardous wastes. Precious metals—specifically Au and Ag—are recovered in a smelting area. However, the precious metals recovery is usually done by outsiders who bring in raw materials sourced from elsewhere. The facility's owner occasionally decides to extract precious metals using terminals and integrated circuit (IC) chips of electronics gathered from within the facility, but this is done only when the supply of raw materials becomes abundant, because processing small amounts is normally not profitable. E-waste stock and various reagents and other input materials for precious metal recovery are stored in a stockyard area. The layout of the informal recycling site is shown in Fig. 1.

Eighty percent of the workers in the facility are men. They do the activities that require greater strength, such as lifting, hammering, and dismantling. Women generally do less strenuous activities such as cutting and stripping wires and dismantling and separating small metal parts. The facility employs an average of 20 workers, of whom 5 males are regular employees. The other workers are seasonal and work when there is a large amount of e-waste to be processed. Part-time workers are also employed to work in groups to process specific stocks of e-wastes and work anywhere from 1 to 6 months a year. Likewise, the smelters are called in to work only when needed.

The regular workers are paid US\$47 a week, and the seasonal workers are paid US\$35 a week for men and US\$25 a week for women. The smelters receive special rates because they are directly exposed to more harmful working conditions. In addition to wages, all workers are provided with free meals and snacks within the 8-h daily work period. The average age of the workers is 33 years, with the youngest at the time of our visit being 15 and the oldest 56.

Most of the workers reside within the city, and a few come from neighboring areas. The majority of the workers are elementary school graduates. Although some have attended high school, none of them has attended college. Ninety percent of the workers are married and support families with four or five members.

2.2 Gold Recovery Process

2.2.1 Process Overview

We reviewed two Au recovery processes in our field survey: a chemical (acid leaching) process and an IC burning process (called *sunog* IC in Filipino, where *sunog* means burning). Both processes utilize smelting after preprocessing, so the differences are primarily in the preprocessing and the types of materials to be applied.

The chemical Au recovery process is used for terminals, printed circuit boards, and jewelry, whereas IC chips are processed by using the IC burning process.

At the preprocessing stage, the chemical process uses cyanide to extract Au, whereas open burning is used to remove organic substances as part of the IC

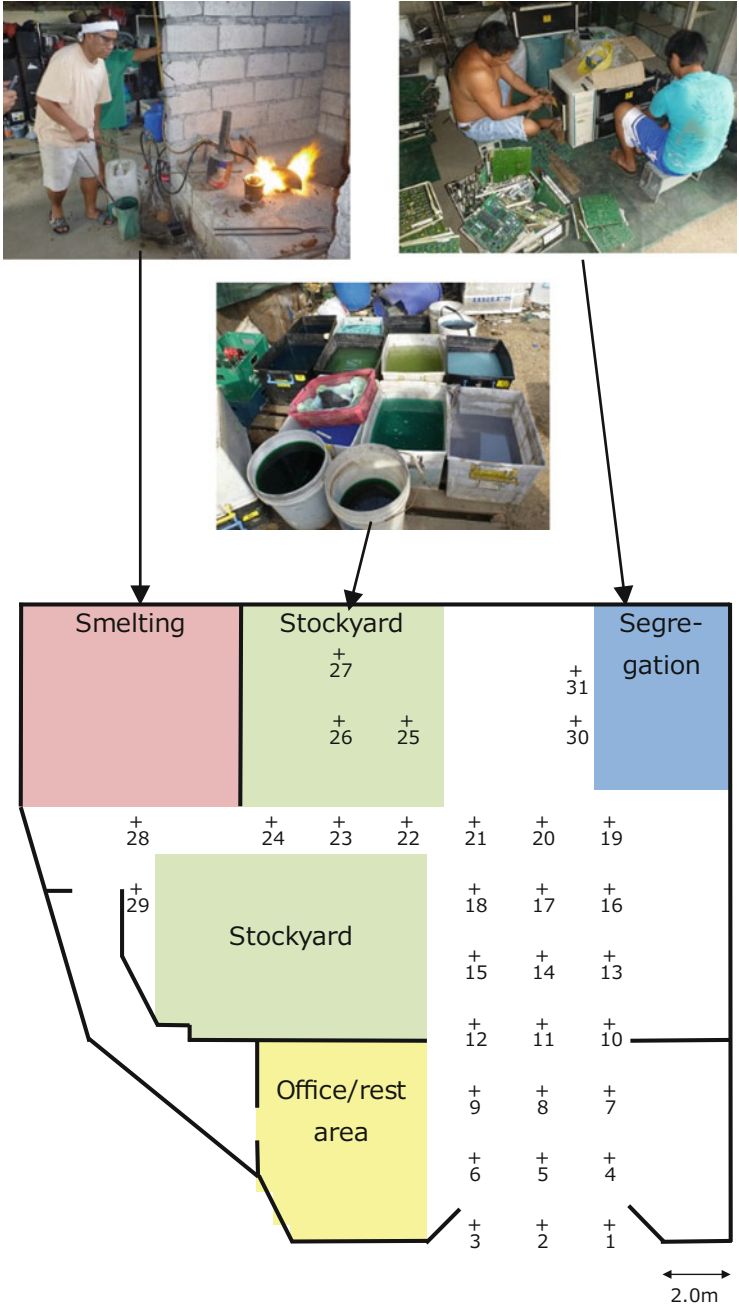


Fig. 1 Layout of the surveyed informal recycling site. Numbers with (+) refer to the location of soil samples

burning process. Open burning of ICs is strictly prohibited, however, because of environmental concerns. Therefore, the chemical process is almost exclusively used for Au recovery, and it was used in our test sample.

2.2.2 Mass Balance

The amounts of reagents used and the output (Au) obtained are largely affected by the quality and quantity of the material inputs. In this study, we used electrical conducting terminals containing Cu with Au coatings (also coated with Ni and Sn) as test samples to evaluate the mass balance for input and output of materials.

In order to confirm the mass balance of the process, we weighed each of the material used for and produced from the test run at the process: raw material (terminals before the cyanide treatment); borax; Pb nuggets; terminals after the cyanide treatment; two types of slag, one of which was borax-derived (slag A) and the other was lead-derived (slag B); and recovered Au nugget. We then sampled each of the input and output materials for the metal concentration analysis. Terminal and Au nugget samples were digested with aqua regia. Slag samples were pulverized and digested with aqua regia followed by alkali fusion with sodium carbonate and boric acid. Metal contents were determined by using inductively coupled plasma—optical emission spectroscopy (ICP–OES, SII NanoTechnology, SPS3500).

2.3 *Metal Content of Surface Soil at the Informal Recycling Site*

To better understand the distribution of metals at the study site, 31 grid-based surface soil samples were obtained for analysis for 63 metal elements (Ag, Al, As, Au, B, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Fe, Ga, Gd, Ge, Hf, Ho, In, Ir, K, La, Li, Lu, Mg, Mn, Mo, Na, Nb, Nd, Ni, Pb, Pd, Pr, Pt, Rb, Rh, Ru, Sb, Sc, Se, Si, Sm, Sn, Sr, Ta, Tb, Te, Ti, Tl, Tm, V, W, Y, Yb, Zn, Zr).

The soil samples were air dried at room temperature for at least 1 week. The dried soil samples were sieved with a 2-mm sieve. Approximately 1 g of the samples was digested with 20 mL of aqua regia on a hot plate. For silver analysis, the solvent was replaced with 6 M chloric acid after the aqua regia digestion to avoid silver chloride precipitation. Concentrations of metals were determined by using ICP–OES (Seiko Instruments, VISTA-PRO) and ICP – mass spectrometry (ICP–MS, Agilent Technologies, 7500cx).

Cluster analysis was then conducted to categorize the metals in the soil samples by using the Ward method.

3 Results

3.1 Gold Recovery Process

3.1.1 Process Overview

The chemical gold recovery process starts with dousing of the raw materials with a sodium cyanide solution prepared by dissolving 50 g of sodium cyanide powder and 75 g of a separating agent (85 % sodium m-nitrobenzene sulfonate and 15 % sodium carbonate) in 1 L of water (Fig. 2). Sodium cyanide is used to dissolve the Au, and the separating agent is used as a stripper to remove Ni from the base metal. The Au is plated over the Ni coating, so removing nickel therefore removes the Au from the base metal.

The raw material—cyanide mixture—is then heated and swirled continuously to facilitate the removal of Au (Reaction 1), as indicated by the loss of the gold color in the coating of the raw materials. The solid Cu materials, with their tin coatings exposed, are then separated from the solution and washed with water to ensure that all traces of cyanide are removed. The washings, however, are mixed with the Au–cyanide solution.

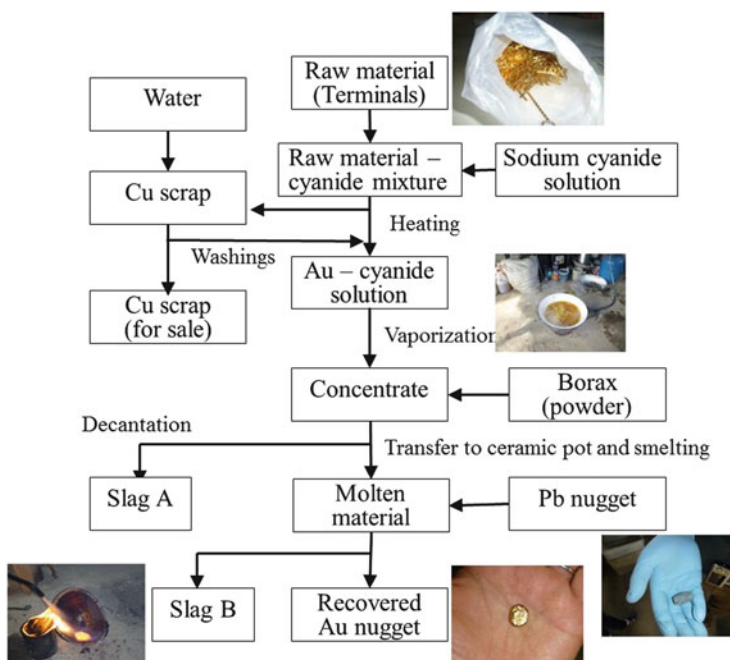
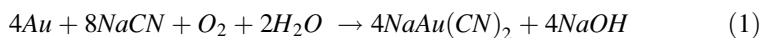
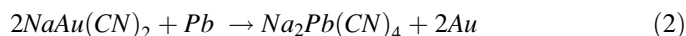


Fig. 2 Schematic of the chemical gold recovery process at the surveyed informal recycling site



The water in the Au–cyanide solution is then boiled off. When the final solution thickens, borax powder is added and mixed in. The resulting paste is then transferred to a ceramic pot or crucible and subjected to firing. Pb nugget (in the form of a Pb bar) is added to the molten material to obtain metallic Au (Reaction 2). Non-gold components are removed by decantation, and the recovered Au is transferred to a different container, where it cools and solidifies.



3.1.2 Mass Balance

We identified 0.25 L of cyanide solution, 1.75 L of water, 274 g of borax powder, and 228 g of Pb nugget as input materials for the 989 g of terminals used as our test sample.

The results of the analysis are shown in Table 1. Au and Cu contents of the input terminals were 0.92 % and 89.1 %, respectively. The Au content of the recovered Au nugget was 85.2 %. Although the Pb content of the Pb nugget used as an input was not analyzed, the Pb content was determined to be 51.4 % in slag B, and the Pb concentration was 2.9 % in the recovered Au.

On the basis of the results for the output materials, 5.5 g of Au was recovered as Au nugget and 1.5 g of Au remained on the processed terminals, suggesting approximately 80 % of Au was recovered from the raw materials by using this process. Pb was mostly transferred to slag; however, a small portion (0.1 %) was contained in the recovered Au nugget.

Table 1 Metal contents of the input and output materials in our test sample (mg/kg-dry)

	Input	Output	Recovered Au nugget (output material)	Slag A (from borax addition)	Slag B (from Pb addition)
	Terminal (original input material)	Terminal (after sodium cyanide solution treatment)			
Au	9200	1500	852,000	20	40
Cu	891,000	867,000	93,800	2400	29,300
Sn	71,100	67,700	<10	4300	24,700
Ni	13,800	13,400	20	1200	74,200
Pb	<10	20	29,200	124,000	514,000
Si	90	90	80	136,000	36,300
Mass (g)	989	969	6.4	194	217

Table 2 Metal contents of the input and output materials in our test sample (mg/kg-dry)

Soil sample number	Metal contents													
	Ag	Al	As	Au	Ba	Ca	Cd	Ce	Co	Cr	Cu			
1	2.0	37,049	2.3	0.15	157	31,882	5.0	19	27	27	120			
2	1.8	37,573	1.7	0.22	114	33,303	0.7	12	30	36	91			
3	0.5	44,549	1.6	0.05	64	40,523	0.5	12	36	13	110			
4	1.9	35,031	2.0	0.27	121	32,139	1.2	14	33	27	121			
5	2.9	38,114	1.8	0.40	85	36,604	2.2	12	30	22	128			
6	3.5	40,082	1.7	0.17	112	33,908	1.1	13	33	36	109			
7	17.6	37,730	2.2	4.59	153	39,620	2.7	17	31	29	1230			
8	3.4	41,703	1.8	0.20	89	35,971	1.2	14	34	16	106			
9	4.0	39,752	1.8	0.52	105	33,995	1.3	16	34	17	126			
10	5.0	43,121	1.6	0.96	51	39,227	3.0	10	36	16	310			
11	4.3	40,153	1.9	1.30	94	36,548	1.2	14	33	17	556			
12	3.0	44,959	1.7	5.00	120	38,028	2.0	16	35	13	130			
13	6.4	41,597	2.1	0.21	130	33,005	9.0	15	34	16	306			
14	2.5	39,920	2.2	0.47	153	30,917	1.3	21	29	16	138			
15	5.9	41,777	2.3	0.27	155	30,310	1.3	22	29	16	102			
16	2.0	45,270	2.1	0.17	127	34,344	2.0	17	34	14	160			
17	10.9	37,898	3.5	3.27	197	32,517	2.7	25	34	27	234			
18	5.0	43,251	3.1	0.75	350	29,955	3.0	40	37	18	270			
19	16.0	37,147	2.9	1.26	153	39,724	18.0	19	35	26	1100			
20	17.8	38,423	2.7	1.95	124	37,355	9.6	17	30	33	336			
21	20.0	26,813	30.6	5.00	329	82,988	40.0	46	43	160	1000			
22	75.7	21,783	6.4	7.91	504	26,581	214.8	17	244	262	2444			
23	160.0	48,463	3.8	2.00	174	14,620	35.0	72	150	49	1900			
24	26.0	57,560	3.3	9.00	885	12,581	9.0	75	47	39	7300			
25	3.1	85,114	8.6	1.95	523	7693	22.8	43	52	47	2513			

26	2.0	65,115	3.1	0.63	260	9340	36.0	63	34	41	310
27	3.1	65,244	3.1	0.80	297	9595	2.9	64	44	42	516
28	240.0	18,166	14.8	21.00	159	32,147	96.0	13	78	111	3600
29	150.0	49,329	4.6	5.00	216	11,482	7.0	65	48	40	2100
30	14.0	64,889	3.9	1.00	1056	12,004	18.0	45	56	78	630
31	3.0	73,432	3.8	0.29	514	11,201	1.0	40	52	67	290
Geometric mean	7.1	42,484.6	3.0	0.9	181.6	26,234.6	4.6	23.2	41.1	31.5	390.9
	Dy	Fe	Mn	Nd	Ni	Pb	Sb	Si	Sn	Zn	
1	3.4	69,441	1470	9.5	37	120	3.5	705	20	255	
2	3.5	68,421	1299	7.5	23	55	0.25	423	12	156	
3	3.9	83,776	1522	7.4	18	28	1.3	553	4	153	
4	3.2	72,361	1313	7.5	32	122	1.1	285	38	243	
5	3.4	71,258	1387	7.2	23	111	1.4	284	23	207	
6	3.8	74,321	1437	7.8	21	105	0.8	302	23	178	
7	4.1	67,706	1701	9.7	96	164	1.4	333	86	281	
8	3.8	78,519	1480	7.7	24	131	1.3	455	157	189	
9	3.7	73,868	1492	8.6	43	183	1.0	580	33	162	
10	4.0	80,178	1408	7.4	30	140	2.2	927	94	244	
11	3.7	76,874	1463	8.0	29	386	2.8	285	34	171	
12	3.8	77,383	1554	8.4	41	130	2.3	524	48	147	
13	3.6	75,106	1544	8.1	46	55	0.25	532	155	150	
14	3.9	67,065	1410	10.9	18	166	0.6	334	242	143	
15	3.5	66,737	1439	9.9	25	57	0.25	235	75	139	
16	3.7	76,728	1537	8.9	3918	41	1.1	585	47	173	
17	3.5	73,973	1783	10.1	76	533	5.1	325	648	231	
18	3.9	70,826	2284	13.5	26	230	4.0	604	77	178	
19	3.3	74,858	1448	9.3	341	450	12.5	641	220	779	

(continued)

Table 2 (continued)

Soil sample number	Metal contents										
20	3.4	69,778	1335	8.6	59	412	3.7	262	71	307	
21	3.2	67,331	1450	22.9	188	1400	83.2	554	122	1232	
22	2.0	142,493	3465	15.2	500	4840	51.6	284	1150	3031	
23	5.0	58,249	2268	27.8	125	2000	41.2	817	328	422	
24	25.4	60,348	3082	29.1	336	1200	20.3	994	191	218	
25	2.4	90,631	1040	12.2	88	5066	40.3	426	888	300	
26	5.5	61,665	2213	29.7	33	91	2.1	711	47	169	
27	5.5	74,052	2829	26.5	43	225	2.6	1093	239	326	
28	2.1	56,027	724	12.4	534	9000	179.3	548	1098	1062	
29	4.8	54,120	2497	26.9	158	4300	65.2	663	362	313	
30	2.0	125,251	2315	10.4	229	800	16.2	716	1178	636	
31	2.2	116,547	1924	11.4	128	300	0.5	386	361	654	
Geometric mean	3.7	74825.2	1657.8	11.4	71.7	299.0	3.5	483.8	110.6	284.1	

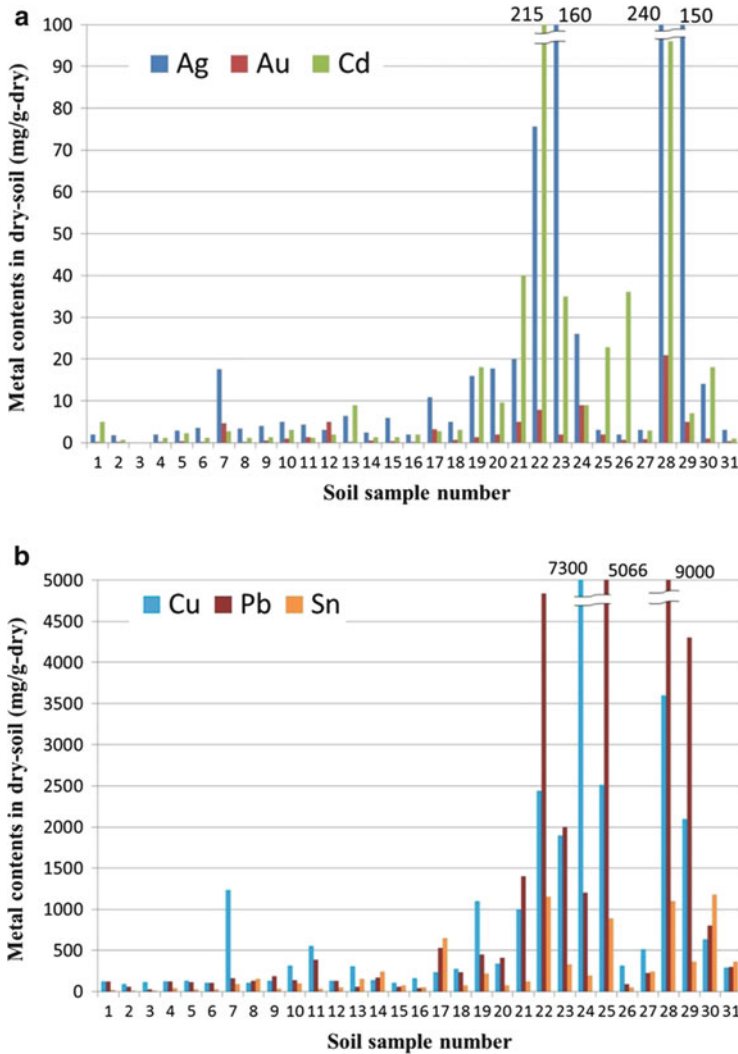


Fig. 3 Selected metal contents in surface soil samples at the surveyed recycling site (a) Ag, Au, Cd (b) Cu, Pb, Sn

3.2 Metal Contents of Soil Samples

The metal contents of soil sampled at the surveyed recycling site are shown in Table 2. Figure 3 shows the results for selected metals (Ag, Au, Cd, Cu, Pb, and Sn). The metal contents of Au, Pb, and Cu are shown by sample location in Fig. 4.

The highest concentration of Au—21 mg/kg-dry—was found in the soil closest to the smelting area; surprisingly, this concentration was higher than the level in gold ore (about 5 mg/kg) [11].

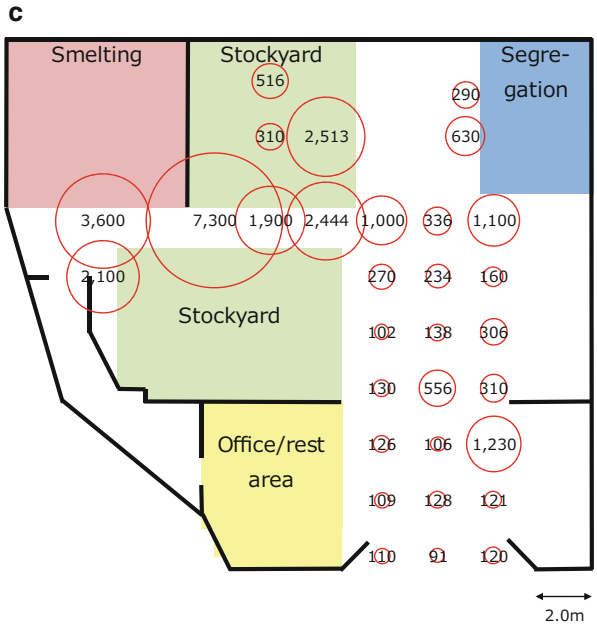


Fig. 4 (continued)

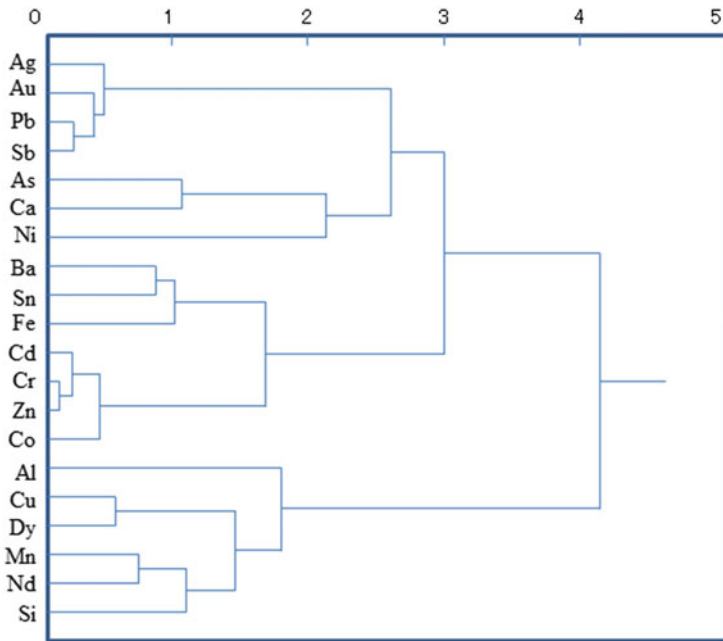


Fig. 5 Dendrogram of metal characterization based on the concentrations of metals in soil samples

The Pb concentration ranged from 28 to 9000 mg/kg-dry, with a geometric mean of 299 mg/kg-dry. The Pb content criterion in soil considering direct ingestion risk, as specified by the Japanese Soil Contamination Countermeasures Act, is 150 mg/kg-dry, and 18 of the 31 (58 %) sampling points exceeded this criterion. Higher concentrations (>1000 mg/kg-dry) were found near the smelting area and stockyard.

The Cu concentration ranged from 91 to 7300 mg/kg-dry, with a geometric mean of 391 mg/kg-dry. High concentrations of more than 1000 mg/kg-dry were found near the stockyard and smelting areas, as well as in the segregation area and the nearby entrance.

A dendrogram resulting from the cluster analysis is shown in Fig. 5. It shows that Ag, Au, Pb, and Sb are grouped into the same category, whereas common metals such as Cu and Al are grouped into another category.

4 Discussion

4.1 Gold Recovery Process

We investigated a chemical Au recovery process at an informal recycling facility in the Philippines. The process can be divided into a preprocessing stage and a smelting stage. In the preprocessing stage, sodium cyanide solution was used to extract Au. We have observed different reagents being used in other field visits—for example, nitric acid and hydrogen peroxide, nitric acid, and aqua regia at sites in Vietnam and China. It seemed that this was related with informal recyclers' attempt for a suitable combination of materials and reagents in each local community.

The results of the mass balance analysis showed that Pb from input additives was shifted into output slag and even the recovered Au nugget. Dust containing Pb was also most likely scattered to areas surrounding the smelting area.

The environmental impacts of discharges and residues from the recovery process need to be addressed. Gaseous discharges other than lead-containing dust are also produced. During the heating of the raw material—cyanide mixture—and the boiling of the resulting Au–cyanide solution, water vapor is discharged openly and directly to the atmosphere. The vapors also contain cyanide and NO₂ and are invariably toxic. Notwithstanding this hazard, the facility is operated without safety equipment—not even exhaust fans.

Liquid discharges are also produced. The chemical process produces a cyanide-laced dilute muriatic acid solution used by the processors to wash their hands to remove cyanide solution. These liquids are discharged into canals and septic tanks or poured directly on the ground.

In the chemical process, Cu scraps are obtained after the Au plating has been removed. These materials are placed in sacks and sold to junkshop buyers who, in turn, sell them to traders for export to other countries. During the firing process,

slags are produced from the solidification of the molten materials. These slags are normally left in the hearth or buried underground once a sufficiently large amount has been produced. The processes also generate used ceramic pots. Because these pots still contain minute quantities of precious metals, they are stored in the facility until there are enough of them to make Au extraction feasible. It may take several months to accumulate enough pots, and the recovery rate is normally very low.

4.2 Distribution of Metals Within the Recycling Site

The results of the soil analyses indicated that valuable substances (such as Au) as well as hazardous substances (such as Pb) were scattered within the informal recycling site. Taking into consideration that Pb is used as an input in the smelting process, the Pb and Au may have been emitted from the smelting process.

Fujimori et al. [12] studied the distribution of heavy metals (Pb, Cu, and Zn) in surface soil samples at an e-waste recycling site in the Philippines by using field-portable X-ray fluorescence. The results indicated that heavy metals had a relatively “small” pollution scale and largely remained inside the original workshop, as compared with toxic organohalogen compounds. Our results are supported by these findings.

Our cluster analysis showed that Ag, Au, Pb, and Sb were categorized into the same close group. As discussed in Sect. 3.1, Pb is an important input for Au recovery. Ag might be present in the printed circuit boards treated for Au recovery, or Ag recovery treatments might also be implemented in the same smelting area. Some kinds of the printed circuit boards may contain Sb as flame retardant substances, and Sb may have also been emitted from the Au recovery process.

In a cluster analysis by Fujimori et al. [9], As, Co, Fe, Mn, and Ni in soil samples from other informal recycling facilities were categorized as crust-derived metals. At those informal sites, physical activities such as dismantling and crushing were mainly conducted; the chemical Au recovery process was not. The crust-derived metals were categorized into groups other than the Au group in our study. For clarification, we need to better understand the contents of substances and the activities at stockyard and segregation areas.

4.3 Health Risks

The recycling activities expose the workers to several health risks. Not only are workers exposed to hazardous substances in the soil; they also inhale metallic dust from smelting activities and from activities in the stockyard and segregation areas. The handling of batteries and other equipment also exposes workers to corrosive liquids and vapors. Extraction of metals, which generates toxic vapors, also exposes them to health risks, especially from the smelters themselves. Although most

workers wear long-sleeved shirts and long pants, they do not use any personal protective equipment (PPE) such as masks and gloves. It is particularly worrisome to see the workers extract the metal without using PPEs, despite the fact that they are handling highly toxic and hazardous cyanide and Pb.

According to worker interviews, the most common health problems, experienced by half of the workers, are the occasional coughs and colds. Cigarette smokers, who comprise 40 % of the worker population, and alcohol drinkers, who make up 70 % of the worker population, are more prone to these conditions. A few workers have skin allergies, and one worker doing metal recovery has visual impairment.

5 Conclusion

We carried out a field survey at an informal e-waste recycling site in the Philippines. We identified the Au recovery process used at the site and evaluated the layout of the recycling site. We evaluated the whole Au recovery process, including the metal content of all the input and output materials. Pb from inputs was recovered from output slag and even from recovered Au nugget.

We collected 31 soil samples at the recycling site and analyzed the metal contents of each sample to clarify the metal distribution within the site. Pb contents ranged from 28 to 9000 mg/kg, whereas Au ranged from <0.1 to 21 mg/kg. The highest contents for both were measured at the points closest to the smelting area; high contents were generally found around the smelting area and stockyard. We determined that valuable substances (such as Au), as well as hazardous substances (such as Pb), were scattered throughout the soil at the informal recycling site. The results of our cluster analysis indicated that Au, Ag, Pb, and Sb were categorized in the same group. Given that Pb is used as an input in the smelting process, it is likely the hazardous substance (Pb) and the valuable substance (Au) were emitted from the same smelting process.

Improvements are needed in the metal recovery process and in hazardous substance emission control. Establishing closed system of the smelting process and treatment of flue gases are recommended. Protective measures for workers are also needed. Many informal workers do not understand well the health impact by recycling activities or think lightly of that. Thus, workers' awareness raising of the need for protective measures is significant. If these types of measures are implemented, formalization of the informal recycling sector is expected to proceed.

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