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Efficiency of the Air-Pollution Control System of a Lead-Acid-Battery Recycling Industry

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Abstract: The air-pollution control system of a lead-acid-battery recycling industry was studied. The system comprised two streams with gravity settlers followed by filter bags for the factory indoor air and the metal-recycling furnace, respectively. Efficiency in particle removal according to mass was found to be 99.91%. Moreover, filter bags and dust from the gravity settlers were analyzed for heavy metals by Wavelength Dispersive X-Ray Fluorescence. The results showed high concentrations of Pb and Na in all cases. In the filter bag samples from the indoor atmosphere stream, Ca, Cu, Fe, and Al were found in concentrations higher than that in the filter bag samples from the furnace stream. The opposite was found for Na. Tl and K were only found in furnace stream bag filters. The elemental concentration of the dust from the furnace fumes stream contained mainly Fe, Na, Cd, Pb, Sb, and Cl, while the indoor main stream contained mainly P, Fe, Na, Pb, and Sb. In all cases, impurities of Nd, Ni, Rb, Sr, Th, Hg, and Bi were found. The high efficiency of the air-pollution control system in particle removal shows that a considerable reduction in emissions was achieved.

Keywords: air-pollution control; battery recycling; heavy metals; control system efficiency; chemical analysis

1. Introduction

Battery recycling is the activity that aims to reduce the number of batteries being disposed as solid waste. Batteries contain heavy metals and toxicants that can contaminate soil and water. Spent batteries from cars, motorcycles, uninterruptible power source UPS, marine applications, fork-lift trucks, electric vehicles, and many others represent an important secondary source of metals (mainly lead) in very high concentration levels, sometimes higher than in natural sources; these metals sometimes are very expensive. Lead recycling saves energy since it is far more energy-efficient to recycle than it is to produce lead from mining and processing ores. Therefore, the battery-recycling approach is a sustainable process for both natural and economic resources.

Outdated recycling processes in some developing countries result in large amounts of lead-dust fumes, dust, and hazardous wastes that cause serious problems to human's health [1–3]. Dust particles emitted indoors by furnaces and metallurgical smelters are enriched in metallic compounds, and this may be deleterious to workers. Heavy metals cause serious problems to human' health [4] and should be kept out of the waste stream. Consequently, the operation of an appropriate pollution control system in such industries is essential. There are only a few studies in the field of battery recycling that are occupied with its air pollution control system [5,6]. Rada et al. [5] and Pan et al. [6] proposed a new recycling method that fulfilled the requirements of an eco-innovative technology controlling air pollution. Due to concerns that pollutants are captured by the individual parts of an air-pollution control system, there have been several studies [7–20]. Only the studies conducted by Sobanska et al. [19] and Spear et al. [20] investigated the fly ash and dust emitted by primary lead smelters. Limited information is available on the quantitative distribution of heavy metals emitted by the battery recycling process and captured by an air-pollution control system. According to the literature, only Ettler et al. [21] investigated the mineralogy of air-pollution control system residues from a car-battery recycling center in the Czech Republic.

The objective of this study was to investigate the air-pollution control-system efficiency of a lead-battery recycling factory and to analyze the heavy-metal concentrations from different origins in the air-pollution control system, such as in the bag house filters and the dust from the tanks that are gathered by gravity settlers and bag houses.

A lead-acid battery recycling factory, "Sunlight", in Greece was investigated. Sunlight recycling is the lead-acid battery recycling branch of systems SUNLIGHT S.A., which has an area of 4.2 hectares and it is located in the Industrial Area of Komotini, Thrace, Northern Greece. Lead-acid batteries are mainly car and motorcycle batteries, industrial fork-lift batteries, and UPS batteries. The recycling cycle consists of the collection of the batteries, breaking batteries into smaller pieces, neutralization of the acid content, separation of the metal (Pb) grid from the plastic casing, cleaning of the metal grid, and melting the metal grid in a high temperature furnace for recycling or further metallurgical processes. The factory is able to recycle 25,000 tons of spent batteries per year, resulting in financial and environmental benefits.

2. Materials and Methods

Air-Pollution Control-System Description

In Figure 1, the Sunlight air-pollution control system is depicted. The furnace KL-710 and its produced fumes are under ventilation by Fan U-720, which regulates the flow rate up to 40,000.00 m textsuperscript3/h. Polluted air from the furnace is first brought through flue-dust gravity settling chamber MC 720 and then through bag-house PK-720. The latter consists of 792 filtering bags, Dn 123/3100 mm made of acrylic co-polymers with a waterproof membrane of 600 g/m^2, suitable for continuous operation up to 130 °C.

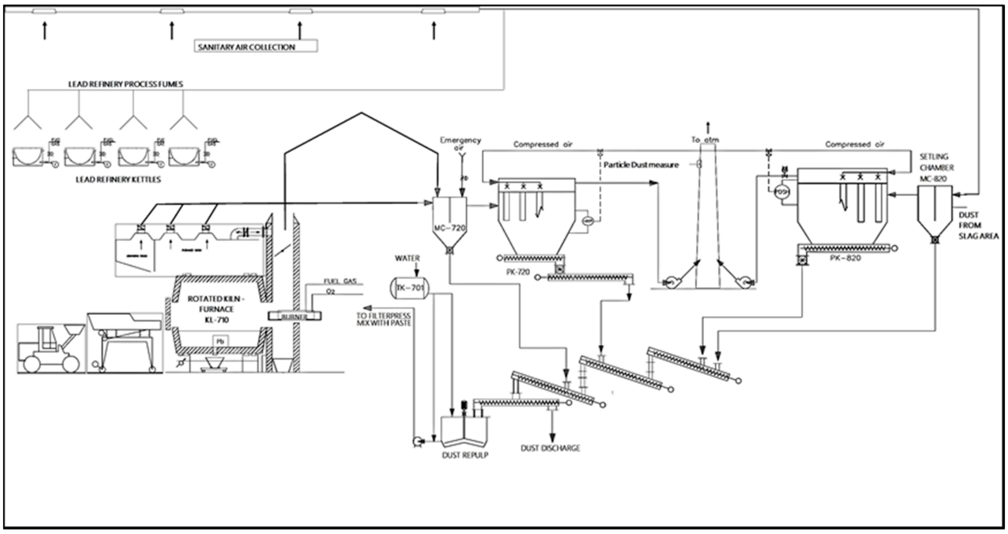


Figure 1. Sunlight air-pollution control system.

Moreover, the sanitary air from the lead refinery kettles and the surmounted space is ventilated by a U-820 fan with a flow rate up to 120,000.00 m^3/h. The collected stream is first brought through another flue-dust gravity settling chamber, MC 820, and then sent to a PK820 bag house that consists of 1188 filtering bags, Dn 123 × 4000 mm, of acrilyc needle felt with surface treatment anti-block waterproof PTFE 600 g/m^2, suitable for continuous operation up to 140 °C.

As dust collects on the filters, a dust layer builds up that increases the pressure inside the bag, hence increasing the energy requirements to move the gas through the dust cake. Eventually, the dust layer becomes so thick that the pressure threshold is exceeded and the dust cake needs to be removed. The type of dust-cake removal (cleaning) in our case is pulse-jet self-cleaning. The air supply used for cleaning is almost 600 kPa and is applied when the pressure inside the bag exceeds 2 kPa.

The flue dust selected at the bottom of the two bag houses is transported by the Archimedes screw conveyor systems into the flue-dust tank V-710. The dust from the bottom of the two gravity settling chambers is also discharged to the same tank. The dust is suspended in the tank with water and then it is filtered to a paste. The paste is sent back to the furnace in order to reduce the volume and the amount of fine dust in the working area.

The "free"-of-particles air stream after the bag houses (up to 160,000.00 m^3/h) is emitted through a self-lifting stack into the atmosphere. The stack dimensions are 20 m height and 2.5 m diameter.

3. Results/Discussion

3.1. Air-Pollution Control-System Efficiency in Particle Removal

The calculation of the system's efficiency was realized for a time period of about three months from April 2018, when the bag house filters were replaced by clean new filters, to June 2018, when samples from the bag filters were selected and analyzed. The bags are cleaned by intermittent jets of compressed air that flow into the inside of the bag to blow the cake off. Often, these bag houses are cleaned while they are in service; the internal pulse causes many of the collected solids to fall to the hopper, but some remain in the fabric of the filter cloth.

According to the manufacturer's specifications, the total flow rate in the stack was up to 160,000.00 m textsuperscript3/h. The flow rates reaching the stack after PK720 and PK820 were measured, and the real flow rate in the stack was calculated by their addition. There were continuous measurements for the whole time period that we conducted this study. There were also data from the particle concentration (mg/m^3) measured at the same time in the stack. The mass of the particles that were not captured by the control system m_{out} was obtained by multiplying the real flow rates with particle concentration at every moment. The total m_{out} was found by adding all these values for the period of three months and was 167.7 kg. Furthermore, the particles captured by the flue dust system at the same period was gathered in the tank and was weighed (m_{dust}). It was 190,862 kg. The m_{filter} is the mass captured by the filter and obtained by gravimetry. The bag filters that were used for heavy-metal analysis were weighed. The blanks were also pre-weighed. Subtracting the blank from the bag filter sample, the weight of the captured dust was found. There were six filters (each one 12.56 cm^2) from every bag house, and we calculated the mean dust captured from these filters. It was 0.14095 gr for PK820 and 0.1483 for PK720. As the total filtering surface was 1820 and 964 m^2 for PK820, and PK720, respectively, and also considering homogeneity to the whole filter, we calculated 209 kg of captured dust for PK820, and 114 kg for PK720. Adding these amounts, we found the total dust captured by the bag filters $\left(m_{filter}\right)$ to be 232 kg. The sum of m_{dust} and m_{filter} gives the total mass captured by the control system.

As a result, the particle mass reaching the air-pollution control system in Equation (1) was found to be:

$$m_{in} = m_{dust} + m_{filter} + m_{out} \tag{1}$$

According to Equation (2), the total efficiency of the system was:

$$n = \frac{m_{in} - m_{out}}{m_{in}} \tag{2}$$

For

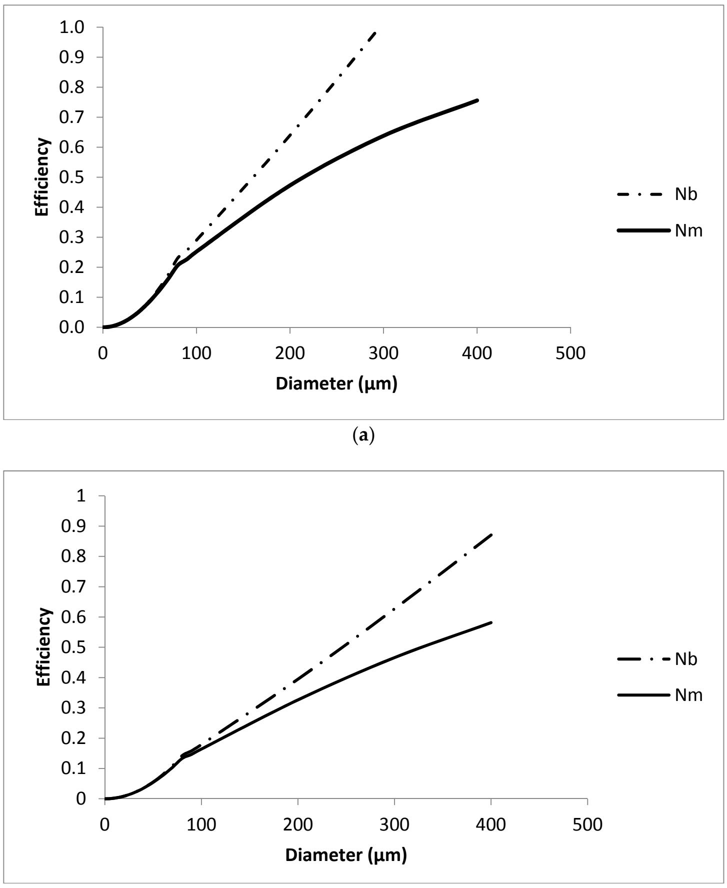
m_{dust} = 190,862 kg

m_{filter} = 323 kg
m_{out} = 167.7 kg

The total n was calculated to be 99.91%.

According to Liu et al. [22], the weight efficiency for the total dust in membrane filters in bag houses is 99.9% and this is in agreement with our results.

Moreover, the efficiency–diameter relation for the two gravity settlers for both the blocked (n_b) (Laminar flow) and mixed (n_m) flow (Turbulent flow) models, assuming Stoke's law, was calculated. We assumed particle diameters ranging from 0.5 to 400 µm. In Figure 2a,b the efficiency–diameter relations for gravity settling of MC720 and MC820, respectively, are indicated. The flow rate from MC720 was assumed to be 40,000 m^3/h and from MC820, 120,000 m^3/h. In both cases, for small particles, for which the calculated collection efficiencies are small, the mixed and blocked flow models gave practically the same answers. For larger particles, the efficiencies became larger, and the two models gave different answers. For MC720, collection efficiency was more than 50% for particles larger than 200 µm in both flow models and reached the maximum of 80% for particles with a diameter larger than 400 µm mainly for a mixed flow. For MC820, the trend was almost the same. It reached the maximum collection efficiency of 60% for particles with a diameter larger than 400 µm for a mixed flow and almost 90% for block flow. Consequently, these two devices only showed higher efficiency for coarse particles larger than 200 µm and this results in the need for the bag houses after the gravity filters. Generally, gravity settlers are old unsophisticated devices that are used in industries treating very dirty gases in order to remove, as a first step, the large particles [23,24]. They are followed, in most cases, by bag filters.



(**a**)

(**b**)

Figure 2. Efficiency-diameter relation for gravity settling in **(a)** MC720 (40,000 m^3/h) and (**b**) MC820 (120,000 m^3/h).

3.2. Particles in the Atmosphere

The suspended particles in the stack from April 2018 to 21 June 2018 were measured with a Laser Dust Monitor (NEOM), NEO Monitors AS, Norway. This is an optical instrument based on transmitting visible laser light from a transmitter unit on one side of the stack to a receiver unit on the diametrically opposite side of the stack. The measuring technique is based on measuring absorption and the scattering of light created by the dust particles present in the stack. The measurement signal corresponds to the integrated dust concentration over the entire optical path (stack). However, the instrument does not determine particles below "Aitken" nuclei. This is not significant for the total mass, but their presence in the atmosphere is important for human health. Particle concentration is indicated in Figure 3. According to the European Commission implementing EU Decision 2016/1032, establishing the best available techniques under directive 2010/75/EU, the upper limit of suspended particles is 5 mg/m^3 at the outflow of the chimney. In Figure 3, it is indicated that during the study period the measurements from the outflow concentration were below the upper limit of 5 mg/m^3.

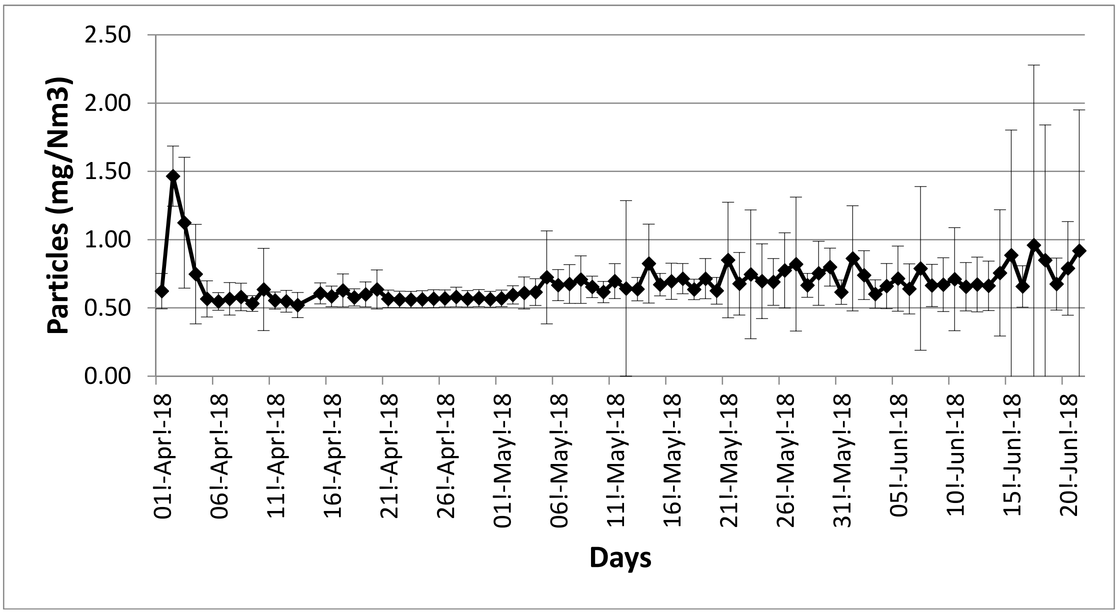


Figure 3. Particle concentration emitted from the stack during the period 1 April 2018 to 21 June 2018.

3.3. Heavy Metals Associated with the Particles

As already stated, the scope of this work was to define the heavy metals retained from the air-pollution control system. As a result, analysis of heavy-metal concentrations in the bag houses, in the dust selected from the tanks of the two gravity settlers, and the two bag houses was performed by using a Wavelength Dispersive X-Ray Fluorescence System (WDXRF, Rigaku, ZSX Primus II) in the laboratory. The Quant Application took place using four national institute of standards and technology NIST standards (NIST 1646a, NIST1468a, NIST 2584, NIST 2710a). Twelve filters were collected from the bag houses: six from the PK720 (furnace bag house), three collected from the first bag in the bag house row, and three from the last bag in the row. From every bag, three samples with a diameter of 40 mm were cut. The first sample was from the upper side of the bag, the second from the middle, and the third one from the bottom. The same procedure was carried out for bag filters from PK820 (sanitary bag house). The dust collected from the tanks of the two gravity settlers and the bag houses was sieved to 60 µm and pressed to a pellet at 1520 kPa. The mean values of the three samples of each bag house filter are shown in Table 1. The results from the dust analysis are shown in Table 2. There are also Supplementary Tables S1 and S2 where LOQ and blanks from bag filters analysis, and LOQ from heavy metal concentrations in the dust from Gravity Settlers and from Bag houses are indiacted, respectively.

Table 1. Heavy metal concentrations in bag houses PK720 (bag house furnace) and PK820 (sanitary bag house) in gr/kg. **N.D.** stands for nondetected. **STDEV** Standard Deviation.

Bag House Filters	**PK720 (Bag House Furnace)**				**PK820 (Sanitary Bag House)**			
	Input		**Output**		**Input**		**Output**	
Elements	Average (gr/Kg)	STDEV	Average (gr/Kg)	STDEV	Average (gr/Kg)	STDEV	Average (gr/Kg)	STDEV
Ag	N.D.		N.D.		N.D.		0.05	
Al	0.04	0.00	0.06	0.01	1.55	0.22	0.95	0.09
Bi	N.D.		N.D.		N.D.		N.D.	
Ca	N.D.		0.03	0.02	1.07	0.17	4.68	0.23
Co	N.D.		N.D.		N.D.		N.D.	
Cr	N.D.		N.D.		N.D.		0.06	
Cu	0.17	0.03	0.29	0.09	8.44	2.03	1.23	0.24
Fe	0.02	0.02	0.32	0.17	6.46	0.36	2.28	1.08
Ga	0.68	0.16	0.87	0.11	1.58	0.07	0.43	0.09
K	0.50	0.07	0.60	0.11	N.D.		N.D.	
Mg	N.D.		N.D.		N.D.		N.D.	
Mn	N.D.		N.D.		N.D.		N.D.	
Na	16.90	0.37	22.55	0.82	17.29	0.73	3.29	0.58
Ni	N.D.		N.D.		0.62	0.05	0.13	0.02
Pb	3.80	0.42	4.74	0.15	9.03	0.13	3.53	0.17
Sr	N.D.		N.D.		N.D.		N.D.	
Tl	10.43		N.D.		N.D.		N.D.	
Zn	N.D.		N.D.		N.D.		N.D.	

Table 2. Heavy metal concentrations in the dust from gravity settlers and from bag houses. **N.D.** stands for non-detected.

Elements	**Gravity Settlers**		**Bag Houses**	
	MC720 (ppm)	**MC820(ppm)**	**PK720 (ppm)**	**PK820 (ppm)**
Al	N.D.	N.D.	N.D.	N.D.
Ca	N.D.	N.D.	N.D.	N.D.
Fe	990	820	1580	1670
Mg	N.D.	N.D.	N.D.	N.D.
P	90	6680	120	7340
K	N.D.	N.D.	N.D.	N.D.
Si	N.D.	N.D.	N.D.	N.D.
Na	25950	17410	28470	16880
S	N.D.	N.D.	N.D.	N.D.
Ti	N.D.	N.D.	N.D.	N.D.
AS	48	333	172	325
Cd	2501	120	1791	116
Cr	N.D.	N.D.	N.D.	N.D.
Cu	75	395	167	455
Pb	19774	2032	21096	21965
Mn	N.D.	N.D.	N.D.	N.D.
V	N.D.	N.D.	N.D.	N.D.
Zn	172	262	597	222
Ba	229	169	271	302
Ce	N.D.	N.D.	N.D.	N.D.
Co	N.D.	N.D.	N.D.	N.D.
Ga	116	89	105	87
La	N.D.	N.D.	N.D.	N.D.
Mo	4	5	5	4
Nd	63	50	58	48
Ni	14	62	34	75
Rb	20	34	27	35
Sc	N.D.	N.D.	N.D.	N.D.
Sr	42	45	N.D.	48
Th	54	45	51	45
U	N.D.	N.D.	N.D.	N.D.

Table 2. *Cont.*

Elements	Gravity Settlers		Bag Houses	
	MC720 (ppm)	MC820(ppm)	PK720 (ppm)	PK820 (ppm)
Sb	820	3671	1889	3962
Hg	8	7	7	7
Br	421	N.D.	240	N.D.
Cs	N.D.	N.D.	N.D.	N.D.
Bi	26	N.D.	25	21
Sm	N.D.	N.D.	N.D.	N.D.
W	N.D.	N.D.	N.D.	N.D.
Zr	N.D.	N.D.	N.D.	N.D.
Cl	836	641	1142	447

The detailed chemical composition from the filters in Table 1 shows that the highest concentrations in PK720 were for Na, Tl, and Pb. In PK820, large amounts of Cu, Fe, Ca, Na, and Pb were found. PbO and PbO_2 were extracted from the lead-battery scrap, and they reacted with the additive C in the melting furnace to give Pb. The high concentrations of Na came from soda ash (Na_2CO_3), which is used as a flux agent to liquidize the slag. Fe and Cu are additives in the furnace. Fe is used in order to grab sulfur compounds, while Cu is a metal that is used in antimonial alloys for battery construction and is also found in battery poles.

Moreover, there were elements found in PK820 that were absent or in low concentrations in PK720. For example, Ni and Ag was found only in the sanitary bag house and also Ca, Cu, Fe, and Al were in much higher concentrations in PK820 than in PK720. This is because the sanitary air comes from refinery kettles where all these elements are emitted from the refinery process. During the refinery process, the metal impurities (Cu, Zn, Fe, Al, Ni, As, Sn, Sb, Ag, Bi) included in the raw lead must be removed in order to obtain the final lead products: soft lead (which has a high purity, higher than 99.985%), hard lead, and various lead alloys. These metals are removed by performing a series of chemical reactions on the molten lead. A chemical oxidation process is used to remove all residual elements. For example, a mixture of sodium nitrate and sodium hydroxide is added to the molten lead. Alternatively, air enriched with oxygen is added. The mixed oxides produced are then skimmed from the molten lead surface and stored to be fed back to the smelting furnace.

K and Tl come only from the furnace and its surrounding air, and Na is found in much higher concentrations in PK720 filters (furnace) than in PK820 (sanitary air).

There are perceptible differences in heavy-metal concentrations between the first-row filter and the last in every bag house. The concentrations in PK720 were higher in the last-row filter compared to the first for all elements. The opposite was found for PK820. There was no regulation in the way the filter gathers the elements. This probably depends on other parameters, such as the size of the particles trapped in the filter or the way dust removal takes place.

Finally, we calculated the total mass of the Pb emitted in the atmosphere. Provided that we calculated the mean Pb concentration in the bag house filters in gr/kg and used the mean concentration of the dust in the stack, we could find the amount of Pb emitted in the atmosphere. The Pb concentration in the stack was found to be 0.0036 mg Pb/m^3 which is below the upper limit of 0.2 mg/m^3. According to the European Commission, implementing EU Decision 2016/1032, establishing the best available techniques under Directive 2010/75/EU, the limit of Pb in the outflow of the stack should be 0.2 mg/$m^{3.}$ Calculating this amount with the total air flow passed through the stack during a year, we calculated the total Pb amount emitted in the atmosphere to be almost 3.7 kg.

In Table 2, the element concentrations (ppm) from the dust in the two gravity settler tanks (MC720 and MC820) and from the two bag house tanks (PK720 and PK820) are listed. As expected, chemical analysis highlighted variable and high Na and Pb metal content in the dust obtained from all tanks. According to these results, the dust from the sanitary air mainly contained P, Fe, Na, Pb, and Sb. For the furnace air, the main metals found were Fe, Na, Cd, Pb, Sb, and Cl. Another interesting issue is that a high concentration of Sb was found in the dust from sanitary air. Furthermore, metals including

As and Cu were found mainly in the dust from the sanitary air, and Zn mainly in the dust from the furnace air. Finally, other impurities were found in the samples were Nd, Ni, Rb, Sr, Th, Hg, and Bi.

Ettler et al. [21] conducted a survey and studied the mineralogy and solubility of air pollution control residues from a secondary lead smelter. Comparing the results from our study to this survey we found that the values of heavy metals in bag filters from furnaces are comparable for Fe and Cu. Pb, Ca, Al, Na, and K concentrations in our study were lower, and only Ca showed a higher value. This is probably due to the fact that these two industries follow different methods in production/recovery lines.

There have been several studies conducted to investigate the influence of heavy metals on the environment and also on human-health [25–31]. Early life exposure to heavy metals, such as Cobalt (Co), Copper (Cu), Thalium (Tl), and Selenium (Se) has negative effects to human development [32]. High concentrations of Cadmium (Cd), Copper (Cu), Lead (Pb), and Arsenic (As) increase the health risks to children in contaminated areas [33]. High Pb concentrations have caused regional contaminations around the world [27,34–36]. Health effects from exposure to Cd include problems to kidneys [37] and bones [38]. According to Lanphera et al. [39], even low-level lead exposure increases the risk factor for death, and particularly for cardiovascular-disease death. This is also supported by Obeng-Gyasi et al. [40]. Furthermore, lead could harm the function of kidneys [41,42]. High lead exposure in the work-place has been associated with adverse hepatobiliary clinical makers [43] and with problems in liver function [44].

4. Conclusions

Particle removal efficiency of the whole system was calculated to be 0.9991. It was found that Pb and Na had the highest concentrations in both the PK720 and PK820 filters. Furthermore, Ag and Ni were found only in sanitary bag house filters, with Ca, Cu, Fe, and Al in higher concentrations than in the furnace bag house filters. This is because the particles in the sanitary bag house filters come from chemical reactions performed in metallurgical kettles for secondary lead refinement. Particles from furnace fumes include elements not found in sanitary bag house filters, such as Tl and K, or found in much higher concentrations, such as Na. There are also differences in heavy metals between the first row filters and the last-row ones in every bag house. Finally, it was calculated that the Pb emitted annually in the atmosphere is ca. 4 kg.

Concerning the dust from different areas of the air-pollution control system, chemical analysis highlighted the high metal content of Na and Pb in all cases. However, the highest Na concentrations were found in the dust from MC720 and PK720 (furnace) and, for Pb, the concentration was much lower in the dust from sanitary gravity settler MC820. The dust from the sanitary fumes was mainly enriched with P, Fe, Na, Sb, and Pb, and from the furnace mainly with Fe, Na, Cd, Pb, Sb, and Cl. In all cases, impurities of Nd, Ni, Rb, Sr, Th, Hg, and Bi were found.

This study determined a total range of unintentional heavy-metal emissions from a lead battery recycling process. This information should be useful for understanding the concentrations of heavy metals produced by this process, and in developing a heavy-metal emission inventory. More investigations need to be performed to obtain further information.

The obtained results indicate that the air-pollution control system trapped high amounts of particles containing toxic metals. Measurements for heavy metals, indoors and outdoors, should be performed in the future to confirm that the air-pollution control system could contribute to reducing the potential environmental and health impacts in the area.

Supplementary Materials: The following are available online at http://www.mdpi.com/1996-1073/11/12/3465/s1, Table S1: LOQ and blanks from bag filters analysis values in μg/cm^2, Table S2: LOQ in ppm from heavy metal concentrations in the dust from Gravity Settlers and from Bag houses.

Author Contributions: Conceptualization, K.K.; Methodology, G.L.; Validation, G.L.; Formal Analysis K.K., D.K. and A.S.; Investigation, K.K., D.K., and A.S.; Resources A.S.; Data curation, K.K., D.K. and A.S.; Writing-Original Draft Preparation, K.K.; Writing-Review & Editing, K.K.; Visualization, K.K. and A.S.; Supervision, G.L.

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Conflicts of Interest: The authors declare no conflicts of interest.

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