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Effect of pH on the Leaching of Potentially Toxic Metals from Different Types of Used Cooking Pots

O. T. Fatunsin*, O. F. Adeyeye, K. O. Olayinka, T. O. Oluseyi

Department of Chemistry, University of Lagos, Akoka, Yaba, Lagos, Nigeria.

Abstract

Humans are exposed to Potentially Toxic Metals (PTMs) through many routes. Cooking foods in cookwares which are prone to material leaching can be an exposure route to PTMs. This study assessed the effect of pH on the leaching of some PTMs from used cooking pots into deionized water. Series of deionized waters were prepared from pH 3 to 7. Each water was brought to boil in clay, non-stick, stainless steel, cast aluminum, pressed aluminum and glass pots respectively. The PTMs leached from each sample pot were determined by Inductively Couple Plasma-Optical Emission Spectrophotometer (ICP-OES) (Agilent nu7m technologies 700 series). The deionized water from the aluminum cast pot and nonstick pot gave the highest concentration of aluminum ($2273 \mu g/L$) and Zinc ($24.39 \mu g/L$) respectively. While that from the clay pot gave the highest concentration of iron ($237 \mu g/L$) and that from the stainless-steel pot gave the highest concentration of iron ($237 \mu g/L$) and that from the glass pot. The results from this study showed more leaching of PTMs into deionized water occurred more at lower pHs (pH 3 to 5) than at neutral pH for almost all the pots. Thus, cooking of acidic foods in pots except when glass pots are used should be avoided. The results of this study therefore reveal the health implications associated with using metal pots for cooking slightly acidic foods as metals can be easily leached from the pots into the foods.

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Keywords: Leaching, potential toxic metals, aluminum pot, clay pot, non stick pot, aluminum cast pot, stainless steel pot, glass pot.

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1. Introduction

Potentially toxic metals (PTMs) are significant environmental pollutants which by ingestion, inhalation and dermal contacts they may enter the human body. Ingested metal are hazardous to humans because they tend to bio-accumulate and cause harm to internal organs [1]. Their concentrations increase in biological systems over time because they are slowly metabolized or excreted and are therefore stored in the system [2]. Metals, unlike organic molecules, do not require bio-activation or undergo enzymatic modification to produce reactive chemical species for detoxification process [3, 4] but use other mechanisms, such as long-term storage (for example cadmium and iron), biliary and/or urinary excretion. Though metal toxicity to a biological system depends on the amount ingested, exposure to certain metals, such as cadmium (Cd) and lead (Pb), may lead to severe toxic effects even at low concentrations [5, 6].

There are several sources/ routes of exposure to PTMs [7]. Leaching of PTMs into food and water from cookware

^{*}Corresponding author tel. no: +2348027178550.

Email address: ofatunsin@unilag.edu.ng (O. T. Fatunsin)

during cooking presents additional exposure to PTMs. Foods are usually cooked /processed in pots to make them edible, this process may leach metals into food [8]. Different types of pots are currently available for cooking. They vary from locally fabricated cast pot commonly called *ikoko irin* and clay pots, to industrially produced aluminum pots, non-stick pots, stainless steel pots and glass-pyrex pots. The fabricated cast pots in Nigeria are produced from Aluminum scraps (such as cans from food packaging, roofing sheets among others) by melting and molding them in sand moulds [9] while the clay pots are molded from wet clay and fired to make them strong. They are not glazed when they are to be used for cooking [10].

Street et al., [11] in South Africa investigated the risk of metal exposure from the use of artisanal cookware. These cook wares were produced from metal scraps and e-waste. Using XRF and inductive coupled plasma mass spectroscopy, total and leached silver (Ag), asernic (As), baron (Ba), Cd, colbalt (Co), chromium (Cr), copper (Cu), Iron (Fe), mecury (Hg), manganese (Mn), molybedium (Mo), nickel (Ni), Pb, antimony (Sb), selenium (Se), tin (Sn), vanadium (V), Al and zinc (Zn) were evaluated. The mean total Al was 509 mg/ L and was over 100 times the EU maximum permissible level allowed for cookware. Pb was detected in the leachates of the pot samples with some concentration higher than the maximum EU permissible level (10 μ g/L) for 1st, 2nd and 3rd migrations respectively of Pb. Cd and Hg were also detected in the leachates from the pots. Chagas et al., [12] investigated the leaching of Pb from clay pot of Brazil origin using inductively coupled Plasma Optical Emission Spectroscopy (ICP-OES). The concentration of Pb leached was found to be higher than 2.0 mg/kg, the value regulated by the Brazilian Health Regulatory Agency and the concentration increased with increase in contact time of food with the pot. In another study by Odularu et al., [13] Al concentration of rice cooked in old and new aluminum, stainless steel, steel and clay pot respectively showed that Al was leached from all their pot samples. The Al concentrations varied between 186.83 ± 75.18 and $350 \pm 130 \,\mu\text{g/g}$. In 2016, Ojezele *et al.*, [14] also analyzed the level of metals (Fe, Zn, Cd, Ni, Mn, Cr, Co, Pb, Cu and Al) in rice cooked in iron, stainless steel, Aluminum and clay pots. They also observed increased levels of metals in rice cooked in pots compared to the all glass pot used as control. Kamerud et al., [15] in 2013 also found stainless steel pots leached metals. Nickel and chromium increased by 24 and 35 fold respectively in tomato sauce cooked in stainless steel pots, however the amount leached reduced with subsequent use and was dependent on factors like grade of pot, length of cooking and cookware usage. Lar et al., [16] found that varying amount of metal (Al, Ca, Fe, Mg and Na) leached from clay pots and cast pots made in different parts of Nigeria (clay-Ife, Ilorin, Minna, Sokoto, Makurdi, Lokoja, Plateau and Calabar; Cast pots- Plateau, Nassarawa, Anambra , Kaduna) when distilled water is boiled in them.

Previous researches conducted on the leaching of metals from different cook wares used specific food in their studies

such as tomatoes sauce [15, 17], fish stew [12], 3% acetic acid solution [11], rice [13], vegetables [18], fruit juices [19], beans [20], except Lar et al., [16], Alabi et al., [21] Alabi and Adeoluwa, [22] and Noemie et al [17] used water. With food substances, parameters such as pH cannot be easily monitored or varied. Though, Lar et al., [16] used water in their study they only determined metals leached from cast and clay pots while Alabi et al., [21], Alabi and Adeoluwa, [22] only studied Aluminum pots. Noemie et al [17] studied the effect of salty water and tomato juice on leaching from cast aluminum pots. So far no study has been carried out on the amount of PTMs leached nor from nonstick pots and at varying pH's in Nigeria despite the presence of these pots in virtually every home. Ingestion is the main route of toxic metal exposure to the human body [23] thus there is a need to study the leaching of metals to foods from cooking pots. Therefore the aim of this study is to determine the effect of pH on the amount of PTMs leached from different cookware pots into food during cooking.

2. Materials and Methods

2.1. Sampling

Samples of used pressed aluminum, clay, stainless steel, non-stick and casted aluminum cooking pots of similar sizes (1 liter) were randomly collected from different households in Lagos state, Nigeria (Plate 1). The pots were properly washed with soap, rinsed with tap water followed by distilled water, allowed to air dry. An all glass pyrex glass pot was used as the control for this study.

2.2. Sample preparation

One liter of distilled, de-ionized water were boiled in each pot (clay, non-stick, stainless steel, aluminum cast pot (*koko-Irin*), aluminum and glass pots) and aliquots stored in cleaned plastics storage bottles (plastics that have been soaked in 0.1 M nitric acid overnight) and analyzed within 24 hours by an ICP-OES (Agilent nu7m technologies 700 series) for aluminum (Al), chromium (Cr), Iron (Fe), Nickel (Ni), lead (Pb), and zinc (Zn).

pH values of water samples were adjusted using a 1 Molar sodium hydroxide and 1 Molar hydrochloric acid respectively with a calibrated pH meter. The water samples were adjusted to pH values of 3, 4, 5, 6, and 7 respectively. These pH values were chosen to stimulate pH of food usually eaten since there is no standardized test for the study of metals leached for pot thus to simulate the cooking condition, one liter of water at pH of 3, 4, 5, 6, and 7 were brought to boil at 100°C for 30 minutes in the different pots.

2.3. ICP -OES Analysis

Extracts from the study were kept at 4°C for metallic element analysis using the Inductively Couple Plasma-Optical Emission Spectrophotometer (ICP-OES) (Agilent nu7m technologies 700 series). All operating parameters for the ICP-OES were optimized for the sample solutions which base on



Plate 1: Pictures of the pots used in this study

de-ionized water. Plasma observation axial, nebulizer Licht emodified, spray chamber cyclonic, torch injector quartz diameter, plasma power, coolant gas, auxiliary gas, nebulizer gas were all optimized. The uptake rate for solutions were set at 2.0 mL/ min, replicate read time was 45s, pre-flush time was set at 60s [24]. The analytical method was validated by using instrumental detection limit (IDL), limit of detection (LOD), limit of quantification (LOQ), precision, and accuracy studies. In this study, the IDL for each metal was calculated from the analysis of seven replicates of the blank concentration. IDL $= 3 \times \text{Sbl}$, where Sbl is the standard deviation of the seven calibrated blank solutions, LOD for each metal was determined using seven replicates of method blank which were digested using the same procedures as the samples. $LOD = 3 \times Sbl$, where Sbl is the standard deviation of the seven method blank solution and the LOQ is the lowest concentration of an analyte in the sample which can be quantitatively determined with some levels of uncertainty. This can be achieved in triplicate of seven method blanks which are digested as the actual samples. LOQ = $10 \times \text{Sbl}$), where Sbl is the standard deviation of the seven method blank solution [25].

2.4. Quality Control

Appropriate measures were taken to prevent contamination and ensure reliability of data. They include actions such as the use of glass pot as control. Deionized water that was initially distilled was used throughout the experiment as blank. All glass wares were soaked overnight and rinsed with 0.1 mole HNO₃ and allowed to dry. They were also rinsed with the solutions to be measure in them prior to use. Recovery studies were carried out using standards to spike de-ionized water for analyses and values between 75 and 100 % were obtained.

3. RESULTS AND DISCUSSION

The glass, aluminum, cast, stainless steel, clay and nonstick pots were used to boil distilled water at pH 3, 4 5, 6 and 7. Foods are usually within the pH of 3 to 7. The amounts of PTMs leached were quantified as described in the methodology and the results are as shown in Table 1 and Figures 1 to 5 (including Tables 1S to 5S; supplementary data).

3.1. Effect of pH on the amount of PTMs leached from glass pot

Glass pot was used as control to compare with other pots which were all made of metals and the results are shown in Table 1. No metal was found to leach from the glass pots (as the leached metals from the glass pot were all below the limit of detection for each metal). This may be due to the type of glass used in this study. The glass material was made of Pyrex-type sodium borosilicate known to have improved thermal shock resistance, excellent weathering resistance, very low solubility, resistant to chemicals (inert) and excellent chemical durability against most leaching solution. They are commonly used for cookware, chemical laboratory ware, flat panel devices and fluorescent lamps and are considered as one of the best glasses. They are usually made of 80.6% SiO₂, 12.6% B₂O₃, 4.2% Na₂O, 2.2% Al₂O₃, 0.1% CaO, 0.1% Cl, 0.05% MgO, and 0.04% Fe₂O₃ [26].

Table 1: Concentration of leached PTMs from glass pot (μ g/L).

Metals	pH 3	pH 4	pH 5	pH 6	pH 7
Al	≤ 0.03	≤ 0.03	≤ 0.03	≤ 0.03	≤ 0.03
Cr	≤ 0.02				
Fe	≤ 0.02				
Ni	≤ 0.05				
Pb	≤ 0.10				
Zn	≤ 0.01				

3.2. Effect of pH on the amount of PTMs leached from aluminum pot

Aluminum pot sometimes referred to as pressed aluminum pots was used to boil distilled water of pHs 3 to 7 and the results of analyses of the water samples for leached metals are as shown in Table 1S (Supplementary data) and Figure 1. At pH 3 and 4, Zn was found to leach but from pH 5 to 7, no Zn leached. Other elements monitored did not leach from pH 3 to pH 7. Aluminum may not have been found in the leachate because of the quality of aluminum pot used and the remediating effect of hot water on aluminum pot. Karbouj et al., [27] discovered in their study on aluminum cook ware that boiling water in the cookware prior to cooking decreases the amount aluminum thus remediated aluminum leaching. In this study, the leaching of aluminum pot and other pots at the different pHs were done using the same pot with deionised water after each other. This process must have inhibited the leaching of aluminum. However, Alabi et al., [21] found that when water was boiled in new aluminum pots for 1 hour it leached 0.023 mg/L of aluminum. While three year old pressed aluminum pots leached 0.029 mg/L and 6 years old aluminum pots leached 0.048 mg/L of aluminum. Lomolinon et al., [24] also found that the amount of aluminum leached from aluminum cooking materials depends on quality of aluminum material it is made of.

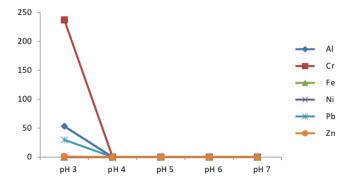


Figure 1: Concentration of leached PTMs from aluminum pot (μ g/L)

3.3. Effect of pH on the amount of PTMs leached from Cast Aluminum pot

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Aluminum cast pots are produced by the informal sector (artisanal sector) in many African Countries including Nigeria from recycling of scrap metal and e-waste. The cast pot is neither as smooth nor compacted in appearance as pressed aluminum pot usually industrially manufactured. Cast pots can be cracked if smatched unlike the pressed pot and it also flakes when water is stored in it for a while. Probably due to the temperature of smelting, type of mould and the purity or type of the starting material. Temperature employed in the informal sector is less, sand moulds are used and starting materials are aluminum scraps. Cast aluminum pot (Koko Irin), was used to boil de-ionised water of pHs 3 to 7 and the results of analyses of the water samples for leached metals are as shown in Figure 2 (Table 2S of Supplementary data). Aluminum leached increased from pH 3-5 and then reduced at pH 6 before increasing slightly at pH 7 which was still lower than at pH 3. Thus the pH 5 leached the highest amount of Al (2273 μ g/L). In a study by Verissimo et al., [8] when food samples were cooked with different acidic additives (lemon juice, wine vinegar and cider apple vinegar) and low pH in cast aluminum pot, increased leaching of Al was observed. Similar trend was observed in this study. Amount of aluminum leached at pH 3 to 5 were higher than amounts leached at pH 6 to 7. Cr was only leached at pH 7 while other metals were not leached at all pH tested. In the study by Street et al., [11] the average concentration of Al leached from cast pot was 509,000 μ g/L which was above the EU maximum permissible limit of 5000 μ g/kg for cook ware and Pb was detected in all their samples with some instances at concentrations higher than the 10 ug/L. Weidenhamer et al., [28] also detected lead in the extract from leached casted aluminum pots made from scraps in Cameroun. However, in this study the concentration of Al leached from cast pots were between 1147 and 2273 μ g/L which are lower than the values reported in Street et al., [11] and lower than the EU permissible value for pots.

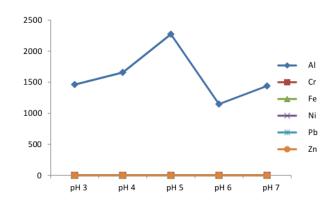


Figure 2: Concentration of leached PTMs from cast pot (Koko Irin) (µg/L)

3.4. Effect of pH on the amount of PTMs leached from clay pot

Deposits of clay exist in many parts of Nigeria and are usually used in pottery [29]. In the clay pot, Pb was not detected

at all the pH tested. The concentration of Al gradually reduced from pH of 3 to pH 4 and increased till pH 7 as shown in Figure 3 (Table 3S Supplementary data). Clay is known to be of a source of aluminum [30]. The concentration of Fe gradually increased from pH 3 to 5 and at pH 6 it decreased and increased at pH 7. Boisa et al., [31] observed that the concentration of Fe leached upon exposure of Ara-Ekiti clay pot to water of Acidic pH, Neutral pH, alkaline pH were (1.15 mg/L 0.16 mg/L, and 0.08 mg/L respectively). Aleksanyan et al., [32] reported the tendency of Fe to leach at near neutral to neutral pH conditions. The Concentration of Zn was higher at pH 5 than at pH 7, this varying trend in concentration of Zn leached at the different pH conditions was also recorded by Boisa et al. [31]. Pb was not detected at all pH conditions. However, Chagas et al., [12] in their study of Pb leached from clay pot found high levels higher than 2.0 mg/kg which they attributed to the substance used for glazing the pots. The pot used in this study was not glazed but was an oven baked hardened pot. The high level of Fe recorded in this study may be attributed to the availability of the metal in the earth's crust. Soils from Nigeria are known to be rich in iron [31, 33] and the pot used was made in Nigeria.

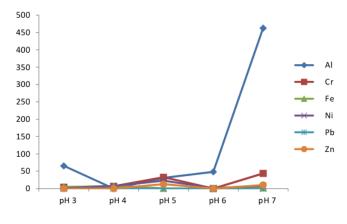


Figure 3: Concentration of leached PTMs from clay pot (μ g/L)

3.5. Effect of pH on the amount of PTMs leached from non-stick pot

The concentration of Al, Fe, Cr, Ni and Pb leached were all below the limit of detection, meanwhile the Zn (24.39 μ g/L) was only detected at pH 3 as shown in Figure 4 (Table 4S). The Zn may have been from the exposed part of the handle inside the pot that had contact with water. Most part of non-stick pot are coated with Polytetrafluoroethylene (PTFE, Teflon) or its other substitutes which are usually organic in nature [34].

3.6. Effect of pH on the amount of PTMs leached from stainless steel pot

Stainless steel cookware is made from metal alloy primarily made of mostly iron and chromium along with differing percentages of molybdenum, nickel, titanium, copper and vanadium. Stainless steel is known to exhibit good thermal conductivity and good corrosion resistance due to its ability to readily form an iron/chromium-enriched passive layer [35].

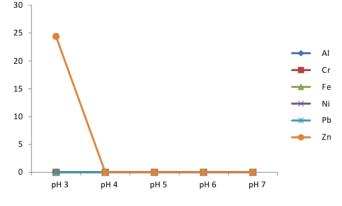


Figure 4: Concentration of leached PTMs from non- stick pot (µg/L)

The result obtained in this study shows that stainless steel pot leached all the metals of interest at pH 3 only (Fe 237, Al 53.18, Pb 29.87 and Zn 1.09 ug/L) except Ni and Cr which were below the detection limit as shown in Figure 5 (Table 5S Supplementary data). Okazaki et al., [36] conducted a seven day immersion test using several solutions on stainless steel and it was found that the quantities of Fe and Ni released gradually decreased with increasing pH from 2 to 7.5. Hedberg et al., [37] stated that acidic pH causes changes n the ionic strength, affects corrosion and dissolution processes, affects ligand conformation and their adsorption behavior, changes the surface hydroxide which all results into increased corrosion of stainless steel. Dan and Ebong, [18] also found that stainless steel pot leached more Fe than those cooked with aluminum pot, this tallied with the result obtained from this study. They noted that the Fe content in cooked food was higher than in uncooked food, and attributed it to the over fifty percent iron content of stainless steel. In the study by Herting, [38] Cr and Pb leached into acetic acid solutions which are also acidic solutions from stainless steel surface. In this study Pb was also detected at an acidic pH of 3 for solutions in stainless steel pot. Lead is a very dangerous heavy metal poison that can accumulate in the body over time to cause neurological and behavioral, renal, cardiovascular dysfunction. Thus, it is important that its levels are well monitored and are way below toxic levels in foods at all times.

The concentration of Zn $(1.09 \ \mu g/L)$ suggests that the stainless steel may not directly cause or be involved in the toxic effects of zinc in the body as the recommended intake values for zinc range zinc is between 7 and 16 mg/day for adults, pregnant and lactating women, depending on sex and dietary phytate intake [39] while for infants and children its is from 2.9 to 14.2 mg/day [40].

The aluminum cast pot - *koko irin* (2273 μ g/L) leached the highest concentration of aluminum while the clay pot leached the highest concentration of Cr and Ni, (7.27 and 22.63 μ g/L). Nonstick pot leached the highest concentration of Zn (24.39 μ g/L) and stainless steel pot leached the highest concentration of Fe (237 μ g/L) and Pb (24.39 μ g/L). No PTM was leached from the glass pot.

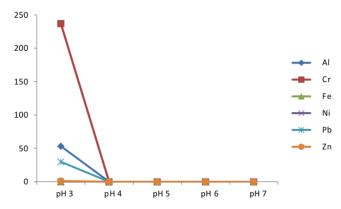


Figure 5: Concentration of leached PTMs from stainless steel pot (μ g/L)

The type of food with respect to the pH was found to be an important factor during the study of metal leachability from cooking pots. Semwal et al., [41] also found that migration of metals into the food was higher with acidic foods than with alkaline foods. They found that more metals leached when food of low (pH of 4.25) was cooked in aluminum cooking pot and that the concentration of Al metal increased by 20.3 mg/kg [8] and also reported that red cabbage samples cooked with different acidic additives (lemon juice, wine vinegar and cider apple vinegar) showed increased leaching of aluminum. Similarly, this study showed that leaching PTMs were considerably higher at more acidic pHs than at neutral pH for most of the pots. Thus cooking of acidic foods should generally be avoided. Longterm exposure to PTMs can lead to gradually progressing physical, muscular, and neurological degenerative processes that imitate diseases such as multiple sclerosis, Parkinson's disease, Alzheimer's disease and muscular dystrophy [42]. Repeated long-term exposure of some metals and their compounds may even cause cancer [43].

4. Conclusion

The results obtained from this study shows that the pots (aluminum, cast Aluminum, clay, nonstick, stainless steel) used for the analysis leached considerable concentrations of PTMs into the distilled water on varying pH especially at acidic pHs while no PTM was leached from the glass pot. More leaching were observed between pH 3 to 5 than between pH 6 to 7 for most the pot types. Since PTMs are known to bioaccumulate, there are health implications associated with the amount leached from the pots. Thus the public should be enlightened about exposure to metals from foods. It should be a public health priority

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Supplementary Data

Table 1S: Concentration of leached PTMs from aluminum pot (μ g/L).

Metals	pH 3	pH 4	pH 5	pH 6	pH 7
Al	≤ 0.03				
Cr	≤ 0.02				
Fe	≤ 0.02				
Ni	≤ 0.05				
Pb	≤ 0.10				
Zn	11.66	1.75	≤ 0.01	≤ 0.01	≤ 0.01

Table 2S: Concentration of leached PTMs from cast pot (Koko Irin) (µg/L).

Metals	pH 3	pH 4	pH 5	pH 6	pH 7
Al	1462	1657	2273	1147	1440
Cr	≤ 0.02	≤ 0.02	≤ 0.02	≤ 0.02	1.99
Fe	≤ 0.02				
Ni	≤ 0.05				
Pb	≤ 0.10				
Zn	≤ 0.01				

Table 4S: Concentration of leached PTMs from non- stick pot (μ g/L).

pH 3	pH 4	pH 5	pH 6	pH 7
≤ 0.03	≤ 0.03	≤ 0.03	≤ 0.03	≤ 0.03
≤ 0.02	≤ 0.02	≤ 0.02	≤ 0.02	≤ 0.02
≤ 0.02	≤ 0.02	≤ 0.02	≤ 0.02	≤ 0.02
≤ 0.05	≤ 0.05	≤ 0.05	≤ 0.05	≤ 0.05
≤ 0.10	≤ 0.10	≤ 0.10	≤ 0.10	≤ 0.10
24.39	≤ 0.01	≤ 0.01	≤ 0.01	≤ 0.01
	$ \begin{array}{l} \leq 0.03 \\ \leq 0.02 \\ \leq 0.02 \\ \leq 0.05 \\ \leq 0.10 \end{array} $	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	$\begin{array}{r rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$

Table 5S: Concentration of leached PTMs from stainless steel pot (μ g/L).

Metals	pH 3	pH 4	pH 5	pH 6	pH 7
Al	65.43	≤ 0.03	30.84	48.25	462.69
Fe	4.03	6.61	32.55	≤ 0.02	43.68
Cr	4.42	7.27	0.62	≤ 0.02	1.88
Ni	0.80	7.42	22.63	1.35	7.81
Pb	≤ 0.10				
Zn	≤ 0.01	≤ 0.01	12.73	≤ 0.01	10.36

Metals	pH 3	pH 4	pH 5	pH 6	pH 7
Al	53.18	≤ 0.03	≤ 0.03	≤ 0.03	≤ 0.03
Fe	237	≤ 0.02	≤ 0.02	≤ 0.02	≤ 0.02
Cr	≤ 0.02				
Ni	≤ 0.05				
Pb	29.87	≤ 0.10	≤ 0.10	≤ 0.10	≤ 0.10

Table 3S: Concentration of leached PTMs from cast pot (Koko Irin) (μ g/L).