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INCORPORATION OF CELLULOSE-BASED ADSORBENT ASH WITH POTENTIALLY TOXIC ELEMENTS INTO MORTAR: A SUSTAINABLE APPROACH

Doctoral (Ph.D.) Dissertation

By

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DEDICATION

This work is dedicated to:

The architects of my upbringing, the sculptors of my character, and the weavers of my dreams my radiant sun and gentle moon, my beloved father, Muneer Naser, and nurturing mother, Maysoon Abed El Khaleq - this tribute is a symphony of gratitude for your unwavering love.

My cherished family members.

My comrades and fellow travellers along this winding road, whose generosity in aiding me knows no bounds.

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LIST OF ACRONYMS AND CODES

AFm - aluminate-ferrite, monosubstituted hydration phase

AMA - Adsorbed mulch ash

APA - Adsorbed paper ash

ASTM C618 - Standard Specification for Coal Fly Ash and Raw or Calcined Natural Pozzolan for Use in Concrete

BSE - Back Scattered-Electron Imaging

CEN/TS 14429:2015 - Characterization of waste - Leaching behavior test - Influence of pH on leaching with initial acid/base addition

EU - European Union

ICP-OES - Inductively coupled plasma optical emission

IMAGE J - ImageJ is a Java-based image-processing program

LOI - loss of ignition

MEP - Multiple Extraction Procedure US EPA Method 1320

PTEs - Potentially toxic elements

SEM - Scanning electron microscope

SEM-EDS - Combined technique that uses a scanning electron microscope and energy-dispersive X-ray spectroscopy

SPLP -Synthetic Precipitation Leaching Procedure

TCLP -Toxicity Characteristic Leaching Procedure

WHO - World Health Organization

WTE - Waste to energy

XRF - X-ray fluoresc

1. INTRODUCTION OF THE WORK AND ITS AIMS

This chapter begins with the introduction of the research study, explaining the importance of this study and the research problem where the issue is identified with supporting data, followed by stating the aim of the study, and finally, the steps that were taken to achieve the aim are listed on the research objectives.

1.1. INTRODUCTION

Potentially toxic elements (PTEs) are ubiquitous environmental pollutants that significantly threaten human health because they tend to intoxicate, accumulate in, and remain in ecological media; these contaminants can be hazardous to organisms, including humans, even in low concentrations (Bahadir et al., 2006). Nguyen et al. (2020) classified potentially toxic elements as hazardous pollutants that contaminate different ecosystems due to the continuous discharge of pollutants from various sources, including industrial effluents, mining activities, agricultural runoff, and improper waste disposal. In water, these contaminants can directly harm living beings through oral intake as drinking water (Rehman et al. 2018) or indirectly when such contaminated water is utilized for agricultural crop irrigation, PTE deposition in agricultural areas, which accumulates in growing food crops and vegetables, contaminating the food chain and posing a concern to humans (Yang et al., 2019; Muhammad et al., 2019). PTE toxicity includes renal, stomach, heart, and muscular disorders and numerous types of cancer (Rehman et al., 2018).

Traditional methods to remove dissolved PTEs ions from wastewater include chemical precipitation, chemical oxidation and reduction, ion exchange filtration, electrochemical treatment, and evaporative recovery. However, as stated by Veglio & Beolchini (1997), these high-technology processes have significant drawbacks, such as incomplete metal removal requirements, expensive equipment and monitoring systems, high reagent energy, and the production of toxic sludge or other waste products that need to be disposed of. Therefore, the development of effective and eco-friendly adsorbents for the removal of PTEs is of paramount importance. Therefore, selecting, developing, and characterizing adsorbent materials are critical in designing an adsorption process for water treatment. Adsorbents for water treatment must have the following characteristics: low cost and availability, chemical stability, mechanical stability, good textural and physicochemical properties, high adsorption capacity, high efficiency, fast kinetics, and the ability to regenerate and reuse (Dotto and McKay, 2020). Many materials, including agriculture products,

red mud, clay minerals, fly ash, portland cement, and cellulose-based materials, have been tested as cheap and abundant adsorbents. (Mondal et al., 2019; Nag & Biswas, 2021).

Since the 1980s, living or dead microorganisms or biological materials have been used to remove contaminants in wastewater, such as PTEs, ions, and dyes (Çolak et al., 2009). Bio-based adsorbents are frequently suggested as a promising solution for removing toxins from polluted water (Hubbe, 2022). The biosorption process offers the advantages of low operating cost, the possibility of metal recovery regeneration of biosorbents, and the minimization of the volume of chemical and biological sludge to be disposed of (Beni and Esmaeili, 2020).

In recent years, cellulose-based adsorbents have attracted much interest as a potentially helpful material for removing PTEs from contaminated water. Cellulose is an attractive material for the adsorption of PTEs due to its unique physicochemical properties, such as its high surface area, biocompatibility, and low cost. The adsorption of PTEs onto cellulose-based adsorbents occurs through a combination of physical and chemical interactions, including ion exchange (Nata et al., 2022), electrostatic interactions, and complexation. Most cellulose-based adsorbents follow the pseudo-second-order kinetic model, which assumes that the rate-limiting step is chemisorption (Syeda and Yap. 2022). Other researchers modified cellulose-based adsorbents' surface chemistry and morphology through various methods such as chemical modification, physical treatment, and grafting of functional groups to enhance their adsorption capacity and selectivity (De Quadros et al., 2016). Many researchers have investigated the efficacy of different cellulose-based adsorbents in removing PTEs from polluted water but rarely discussed the fate of the cellulose-based adsorbents loaded with PTEs. Still, it is a significant concern due to the potential environmental impacts they may cause; although biosorption is an effective technique for removing potentially toxic elements (PTEs) from wastewater, it causes secondary pollution in the form of used adsorbents ending up in landfills, leading to PTE leaching into groundwater.

The final disposal methods of biosorbents are regeneration and reuse, incineration, landfilling, and other safe disposal methods (Huang et al. 2020). However, these methods, particularly landfilling, have limitations. According to the EU’s waste hierarchy, landfilling is the least preferable option and should be limited to the bare minimum quantity. In addition to the communication from the commission to the European parliament, council, the European Economic and social committee, and the committee of the regions toward a circular economy, a zero-waste program for Europe, COM No. 398 (CEN 2014), due to the limitations mentioned previously, other methods are

encouraged to be used to manage hazardous wastes. The simplest and oldest means of dealing with wastes is by producing energy from them, or what is known as "waste to energy" (WTE) (Menikpura et al. 2016). The main types of incineration waste are municipal wastes, non-hazardous industrial wastes, hazardous wastes, sewage sludge, and clinical wastes. Mostly, all non-recyclable non-hazardous wastes (MSW, waste from paper and wood industry) are co-incinerated and constitute the majority of the volume (Joseph et al. 2018), which are also considered low-cost adsorbents of PTEs (Sthiannopkao and Sreesai 2009) and (Jang et al. 2005). But incineration or combustion produces fly ash and bottom ash wastes, created as a by-product of wood-fired power plants, pulp and paper, and wood-burning industrial facilities. Most of the ash produced is disposed of in landfills, while small quantities are used as supplementary cementitious materials (Ayobami 2021). Most of the research work focuses on adding the wastepaper ash and wood biomass ash to ordinary portland cement or partially substituting cement in mortars or concrete in terms of the mechanical properties of the mix (Ahmad et al. 2013; Monosi et al. 2012; Zmamou et al. 2021; Ayobami 2021; Martínez-Lage et al. 2016; Wong et al. 2015; Frías et al. 2015; Carević et al. 2019; Castrillón and Gil 2020). Only a few researchers have studied the leaching of PTEs of mortar incorporated with fly ash and bottom ash of paper and wood biomass. (Ismail et al. 2019) used wastepaper sludge ash in the cement matrix to stabilize water treatment sludge, while (Carević et al. 2020) conducted leaching tests of wood biomass ash cement replacement by 15% of the cement composite. Also, insufficient research has been done on the final fate of cellulose-based adsorbents loaded with contaminants. Contaminated cellulose adsorbents typically include a lot of water and PTEs; therefore, they will not be suitable for landfilling, raising concerns about the leaching of pollutants into groundwater. In this study, the proposed solution to this problem was investigated through a procedure of compression and incineration of the contaminated cellulose adsorbent waste, followed by incorporating the resulting ash within a mortar matrix.

Mortar is a workable paste that is used as a binding agent for masonry work units such as stones, bricks, and concrete masonry work units to fill and seal uneven spaces between them as well as to add decorative colors or patterns to a masonry wall (Elangovan and Rajanandhini. 2018). It is generally produced by combining fine aggregate sand, water, and cementing materials like lime or cement. Recently, fly ash and bottom ash from industrial by-products have been widely used to replace Portland cement in cementitious composites like mortar (Sathonsaowaphak et al. 2009; Agrawal and Savoikar 2022; Omur et al. 2023). Other researchers studied the partial replacement of binder with waste wood ash and other cellulose-based materials ash. The results seem to indicate an auspicious pozzolanic material with no reduction in the mortar strength, enhanced durability of

the sample, and significantly contributing to the sustainability of the construction industry with added environmental value where waste wood ash is utilized and the amount of discarded ash and its adverse effects on the environment can be diminished significantly, this also contributes to reducing the amount of waste sent to landfills (Martínez-García, et al. 2022). Other studies have proved that adding calcium carbonate to a cementitious composite positively affects early-age strength, the hydration process, and durability (Cao et al. 2019). Another study result has shown that the binders of historic mortars composed of small grain–sized silica (SiO_2) and carbonated lime ($CaCO_3$) are considered the main part that imparts hydraulic character and high strength to the mortar (Sağın et al. 2012). Knowing that the main components of adsorbed paper ash (APA) and adsorbed mulch ash (AMA) are $CaCO_3$ and SiO_2 supports their suitability to be added to a mortar mix. To assess the feasibility of using APA and AMA mortar composites in engineering applications while ensuring environmental suitability by utilizing the leaching test. This research included the preparation and the use of two kinds of adsorption process waste ash – adsorbed paper ash (APA) and adsorbed mulch ash (AMA) – as an additive to mortar, which added an environmental and economic value through energy recovery during the ashing process following the ideas of the waste to energy and circular economy, in addition to immobilizing the PTEs into a cement matrix aiming to close the loop of pollution.

This study focused on investigating the leaching behavior of PTEs (Cd, Zn, Cu, and Pb) at five different artificial adsorption initial concentrations (0.5, 1, 5, 10, and 50 mg L^{-1}) and three different mixing weight proportions of ash with cement, as well as their pH-dependent behavior, to ensure environmental safety ensuring their suitability for use in constructions covering a wide range of acidic environments by using the CEN/TS 14429:2015 (BSI 2005) test method. To detect the release of PTEs from the mortar ash mixes, the ICP-OES technique was used then the concentrations were compared to the permissible leaching value limits for the waste categories according to EU directive 1999/31/EC. A literature review also showed that studies on cellulose-based materials ash in construction products are limited to mechanical strength, with only a few studies reporting on its microstructural characteristics. While no microstructural characterization study was reported on cellulose-based adsorbents ash loaded with PTEs, this research further investigated the elemental and microstructural characteristics of APA and AMA mortar composites by using an SEM device equipped with energy-dispersive X-ray spectroscopy (EDS) to ensure the development and improvement of materials, performance evaluations, and describe the immobilization and distribution of PTEs within a mortar composite.

1.2. Aim of the study

This research aimed to study the potential for paper and mulch waste previously used for the (PTEs) adsorption process and then ashed as an additive to mortar which added an environmental and economic value through energy recovery during the ashing process following the idea of the waste to energy, also reducing the quantity of waste landfilled, in addition to immobilizing the PTEs into a cement matrix aiming to close the loop of pollution to achieve environmental safety and sustainability in addition to the circular economy.

1.3. Study objectives

Based on the problem statement, the following objectives were identified:

1- Determine the leaching values of Cd, Pb, Zn, and Cu by using the ICP-OES technique and describe the behavior of each element in addition to studying the influence of the pH on the AMA and APA in mortar leaching concentrations.
2- Assess leaching as a component of environmental safety after comparing the measured leaching values with the standard permissible leaching limits value for the waste categories according to EU directive 1999/31/EC.
3- Determine the optimum weight ratio for APA or AMA addition to the mortar mix based on the compared leaching values.
4- Study the microstructural and elemental characteristics of (APA) and (AMA) mortar composites by using an SEM device equipped with energy-dispersive X-ray spectroscopy (EDS).
5- Compare the two used adsorbents' ash suitability as a construction material based on leaching experiments and the microstructural and elemental testing of the prepared mortar.

1.4. Research hypothesis

The research hypotheses aim to test various aspects of the research objectives, such as comparing the leaching behavior of different PTEs, assessing environmental safety and compliance with regulations, determining optimal mixing ratios, and comparing the suitability of APA and AMA as construction materials based on different parameters. The research will

use statistical analysis to ensure the feasibility and environmental suitability of using APA and AMA mortar composites in engineering applications.

The research hypotheses are as follows:

1-There will be a significant difference in the leaching behavior of different PTEs (Cd, Zn, Cu, and Pb) between the mortar composites containing APA and AMA at different initial concentration, eluent molarity, and ash percentage.

2-The leaching concentrations of PTEs (Cd, Zn, Cu, and Pb) from APA and AMA mortar composites will either meet or exceed the permissible leaching limits according to EU directive 1999/31/EC for waste categories.

3-There exists an optimal weight ratio of APA or AMA addition to the mortar mix that minimizes PTEs leaching while maintaining mechanical strength and ensuring environmental safety.

4-There will be a significant difference in the microstructural and elemental characteristics of the mortar composites containing APA and AMA.

2. LITERATURE OVERVIEW

2.1. Potential toxic elements

Due to their capacity to contaminate, build up, and remain in environmental media, potentially toxic elements (PTEs), also known as toxic metals, trace elements, trace metals, or heavy metals, are dangerous contaminants (Nguyen *et al*., 2020). Although other terms have been used to characterize these elements, "Potentially Toxic Elements" (PTEs) are thought to be more broad and acceptable from a chemical and environmental perspective than "toxic" or "heavy metals" (Hooda, 2010; Pourret & Hursthouse, 2019). Potentially toxic elements are separated into three main classes: the macronutrients, the micronutrients, and the toxic elements. The macronutrients are needed in large quantities by plants; the micronutrients are needed in small quantities; however, in large amounts, they can be toxic; and lastly, the toxic elements can indicate toxicity even in small quantities (Mbokazi 2021).

Human and animal exposure to toxic metals occurs via ingestion, dermal contact, and inhalation, highlighting the multiple routes by which these elements can enter organisms and accumulate (Bansal, 2020). The mobility of PTEs in soil may result in translocation and accumulation in plant tissues. Consumption of plant tissues by herbivores and omnivores typically results in the translocation and bioaccumulation of PTEs to higher trophic levels, resulting in the biomagnification of PTEs throughout a food web (Szynkowska *et al*., 2018). Human activities, such as mining, industrial operations, and agricultural practices, have considerably increased the environmental concentration of PTEs (Jehan *et al*., 2018). Potentially toxic elements in water and waste incineration ash pose significant health hazards to humans and the environment. For water treatment, many methodologies are used to remove these toxins, including the adsorption method, but unfortunately, this method will generate waste loaded with PTEs.

These wastes must be handled; for example, incineration is a step in solving the problem but will produce contaminated ash. In recent years, industrial by-products such as fly ash and bottom ash have been extensively used as replacements for Portland cement in cementitious composites like mortar (Sathonsaowaphak *et al*., 2009; Agrawal & Savoikar, 2022; Omur *et al*., 2023). Additionally, waste wood ash and other cellulose-based materials have been studied for partial binder replacement, showing promising results regarding mortar strength, enhanced durability, and environmental sustainability in the construction industry (Martínez-García *et al*., 2022). The chemical composition of waste wood ash includes macro-elements, micro-elements, and potentially toxic elements (Zając *et al*., 2018). Therefore, incorporating ash into construction

materials should be assessed. Because of their potential harm to the environment and organisms, further research is needed to assess the extent of their impact and develop sustainable solutions to minimize their adverse effects.

Exposure to potentially toxic elements (PTEs) such as cadmium (Cd), lead (Pb), zinc (Zn), and copper (Cu) can have various health effects. Cadmium exposure primarily occurs in humans through inhalation of tobacco smoke or particulate matter, ingesting contaminated food, and drinking contaminated water (Schaefer *et al*., 2020). Chronic inhalation of Cd can lead to kidney disease and cause bones to become weaker. Cd can cause various symptoms such as abdominal pain, vomiting, hepatic injury, renal failure, gastrointestinal erosion, and bone damage (Fatima *et al*., 2019), and has been associated with prostate and lung cancer (Moon et al., 2023) and hypertension (Aramjoo *et al*., 2022). Lead toxicity mainly affects organs such as the kidneys, liver, brain, heart, and nervous system (Collin *et al*.,2022). The nervous system is highly impacted by Pb toxicity, leading to symptoms such as poor attention, headaches, dullness, irritability, and memory loss. These symptoms can also worsen to paralysis, coma, or even death. Zinc toxicity has been linked to an increased risk of cardiovascular diseases, hypertension, nausea, muscular stiffness, stomach damage, and neurotoxic effects on human health (Obasi & Akudinobi, 2020). Excessive zinc intake can cause psychical dysfunctions and induce neural changes in the body, neuronal damage, and death associated with traumatic brain injury, stroke, seizures, and neurodegenerative diseases (Cheng *et al.*, 2021). Chronic copper toxicity is rare in humans and primarily associated with liver damage, while acute Cu intoxication leads to gastrointestinal effects such as abdominal pain, cramps, nausea, diarrhea, and vomiting. Understanding potentially toxic elements toxicity and potential routes of exposure is essential for instituting effective mitigation strategies and ensuring the safety of water resources and waste management practices.

2.2. Adsorption of Potentially Toxic Elements

The phenomenon of adsorption was first introduced by Heinrich Kayser in 1881 (Salih, 2018). Adsorption is defined as the process by which substances (atoms, ions, or molecules) are removed from a gas or liquid and adhere to the interphase of a solid material (Pillai, 2020). It has evolved into a critical technology for water treatment and has been extensively investigated by researchers in this domain. Distinguishing itself from absorption, which involves the incorporation of the absorbing substance within the volume of solid material, adsorption is a surface-based process that

involves the adhesion of adsorbates to the surface of the adsorbent material. Refer to Figure 1 for an illustration of the adsorption process.

Traditional techniques to eliminate dissolved PTE ions from wastewater include chemical precipitation, chemical oxidation and reduction, ion exchange filtration, electrochemical treatment, and evaporative recovery. However, these high-technology processes present significant drawbacks, as described by Veglio and Beolchini (1997), including incomplete removal of metals, high equipment, and monitoring system expenses, considerable reagent energy requirements, and the generation of toxic sludge or other waste products necessitating appropriate disposal. In contrast, the adsorption technique has garnered considerable attention due to its operational and design flexibility, the potential for adsorbate recovery and subsequent regeneration, and the cost-effectiveness of the process (Ahmadian *et al*., 2023). The regenerated adsorbent can be reused for multiple purposes, rendering adsorption an environmentally acceptable method (Kulkarni & Kaware, 2014; Carolin *et al*., 2017), although the number of regeneration cycles is limited and depends on the characteristics of the adsorbent material (Alsawy *et al*., 2022).

The success of adsorption primarily hinges on selecting, developing, and characterizing the adsorbent material, which is crucial in designing an efficient adsorption process for water treatment. An ideal adsorbent for water treatment should possess the following attributes: cost-effectiveness and availability, chemical and mechanical stability, favorable textural and physicochemical properties, high adsorption capacity, efficient adsorption kinetics, and the potential for regeneration and reuse (Dotto and McKay, 2020). Numerous materials, including agricultural products, red mud, clay minerals, fly ash, and Portland cement, have been tested as low-cost adsorbents (Mondal *et al*., 2019).

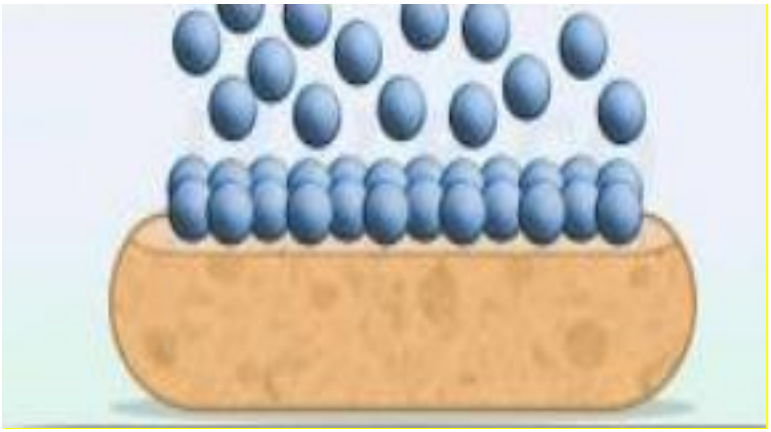

Figure 1. The adsorption process (Salih, 2017)

2.2.1. Cellulose-Based Adsorbents

Cellulose is nature's most prevalent biopolymer and is the primary component of plant fibers, which provide plant rigidity (Sharma *et al*., 2019). It is a long-chained linear polysaccharide composed of β-d-glucopyranose units linked by β-1.4 glycosidic linkages (Faruk *et al*., 2012; Henriksson & Berglund, 2007; O'Connell *et al*., 2008). Figure 2 shows the chemical structure of cellulose.

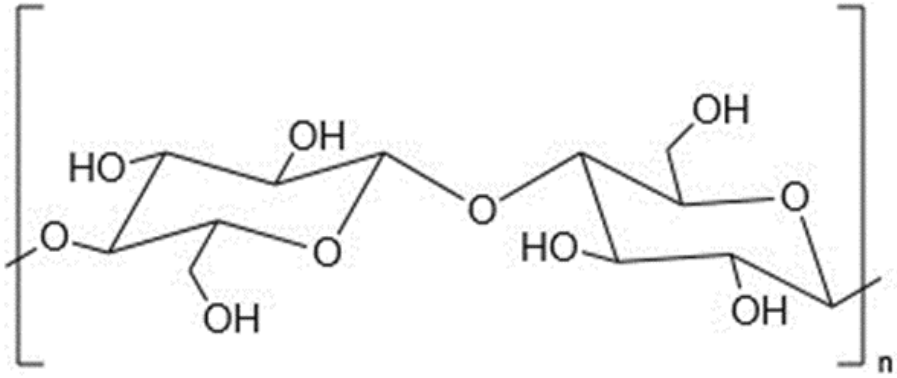

Figure 2. Chemical structure of cellulose

Cellulose-based materials have been used in various construction applications, primarily as intact wood. Furthermore, cellulose is abundant in commonly used materials such as cotton (90%), wood (50%), and dried hemp (57%). It has many applications in various fields but is most commonly used in producing paper, cardboard, and derivative products such as cellophane and rayon. It is also a significant component of cotton and linen textiles. In addition, powdered cellulose and microcrystalline cellulose are used in the pharmaceutical industry as inactive drug fillers. Also, cellulose is a versatile starting material for chemical conversions that produce artificial cellulose-based threads and films and several stable cellulose derivatives utilized in various industrial and consumer applications (Gupta *et al*., 2019).

The cellulose content is variable in agricultural and waste materials; some cellulose content in different materials is presented in Table 1.

Table 1.*The cellulose content in different materials*

Material	Cellulose %	Reference
Oakwood	41	Le Floch *et al* (2015)
Wastepaper	90-99	Sahin and Arslan (2008)
Cotton	95	Holtzapple (2003)
Leaf fiber	55 – 73	Hokkanen *et al*. (2016)
paulownia wood	43.93	Huo *et al* (2022)
Crop residues	30 –50	Koul *et al* (2022)
Wheat straw	30	Sundarraj& Ranganathan (2018)
Jute	51-78	Kalia *et al* (2011)
Maize Straw	28 - 44	Rehman *et al* (2014)
coconut waste	28.7	Rojas *et al* (2018)

Several natural cellulose-based materials have been tested by researchers and found promising results for PTEs removal, such as coconut shells (Tan *et al*., 1993; Low *et al*., 1995; Baes *et al*., 1996; Pino *et al*., 2006); Wood Sawdust (Sharma & Forster, 1994; Mohan &Singh, 2002; Dakiky *et al* ., 2002; Šćiban & Klašnja., 2003; Acar & Eren, 2006; Gupta & Babu, 2009; Putra *et al*.,2014), and paper waste as adsorbents for the removal of dyes and PTEs from wastewater (Fahad *et al*., 2018). These eco-friendly adsorbents show good affinity towards PTEs due to the cellulose content in these materials, which have good adsorption potential due to O-containing and hydroxyl functional groups (Jamshaid *et al*., 2017). Surface complexation, ion exchange, and electrostatic contact were all mentioned as adsorption methods by which these functional groups may efficiently coordinate with PTEs (Han *et al*., 2022). Binding affinity is greatly affected by whether PTE ions establish bonds with monodentate or bidentate functional groups (Zhang *et al*., 2020). In addition, PTEs' charge and valency affect the interface's electrical characteristics. Metal ions with positive charges, such as Cd (II), Pb (II), and Hg (II), have an electrostatic attraction towards functional groups with a negative charge. Arsenate (AsO_4^{3-}) ions, which are negatively charged, prefer to attach to positively charged sites. According to Han et al. (2022), the electrostatic

interactions between PTE ions and functional groups are enhanced for divalent and trivalent PTE ions compared to monovalent PTE ions due to the valency effect.

Most adsorption studies were conducted using untreated cellulosic materials, and only a few demonstrated good adsorption potential. Other researchers applied physical and chemical treatment, which significantly impacted the performance of these adsorbents. For example (Šoštarić *et al*., 2018) applied treatment with NaOH to apricot shells, increasing the adsorption capacity by 90%, 154%, and 61% for $Pb^{2+,}$ Cu^{2+}, and Zn^{2+}, respectively. (Wang *et al*., 2019) developed a carboxymethylated cellulose fiber for water purification. The prepared carboxymethylated cellulose fiber bio-adsorbent removed Cu (II) more effectively than the unmodified fibers, whose adsorption capacity increased 130-fold.

In another study, Huo et al. (2022) used a wood-based adsorbent modified by esterification with phosphoric acid. The ideal adsorption conditions were 318.15 K with pH= 6, and the phosphorylated wood maximum adsorption capacity was 130.2 mg g^{-1}, seven times higher than that of the alkaline extracted wood (18.5 mg g^{-1}).

The most recent studies are concentrated on synthesizing cellulose- and nano-cellulose-based adsorbents. For example, maleic acid-modified nano-cellulose has a greater maximum adsorption capacity for Pb^{2+} (115 mg g^{-1}) than maleic acid-modified macro (20 mg g^{-1}), according to Vadakkekara et al. (2019). Furthermore Hamad et al. (2020) prepared a hybrid nanofibers composite adsorbent membrane of modified cellulose nanofibers with modified hydroxyapatite for the removal of both lead (Pb^{2+}) and ferrous (Fe^{2+}) ions from simulated wastewater; the results demonstrated that this composite offers a higher adsorption capacity than either material alone. To achieve removal efficiencies of 99.7 and 95.47% for Pb (II) and Fe (III), respectively, the optimal conditions for the adsorption efficiency of Pb (II) and Fe (III) ions in wastewater were at the equilibrium time of 35 and 40 min., respectively, at pH = 6, room temperature, 0.1 g of adsorbent, V = 50 ml. Moreover, in a comparison study (Sirviö & Ivanka, 2020), the researchers compared the adsorption capacity of lignin-rich wood nanofibers and cellulose nanofibers produced from bleached cellulose fibers for lead and copper adsorption. Nanofibers of bleached cellulose pulp showed a lower adsorption capacity of copper and lead compared to lignin-rich nanofibers.

2.2.2. Potentially toxic elements adsorption by paper

The globe generates more than 300 million tons of paper annually, and demand is expected to double before 2030. Papers and cardboards account for 40% of municipal solid waste (Putro *et al*., 2019). Paper waste for PTE adsorption has gained significant attention recently due to its low cost, high availability, and eco-friendliness. Several types of paper waste have been investigated for their adsorption properties, including recycled paper, newsprint, cardboard, and office paper. The properties of the paper waste, such as its surface area, pore size distribution, and chemical composition, significantly impact its adsorption capacity.

Recent studies have demonstrated the potential of paper waste as an adsorbent for PTEs. For instance, waste printing paper exhibited a high adsorption efficiency of 90% and metal uptake of less than 25 mg g^{-1} for evaluated metal ions (Moyib *et al*., 2017). Extraction of micro-fibrillated cellulose from wastepaper using simple sulfonation resulted in a 250% increase in adsorption capacity for lead (Pb) ions compared to pristine micro-fibrillated cellulose (Sridhar & Park, 2020). In a study (Chakravarty *et al*., 2008), newspaper pulp was used as an adsorbent for removing copper from effluent. The study found that the newspaper pulp adsorbent removed copper effectively, with a maximal loading capacity of 30 mg g^{-1} at an initial Cu concentration of 20 mg L^{-1}. The study also found that the adsorption of Cu onto the newspaper is a physisorption spontaneous endothermic process. not only but also modified Newspaper pulp with citric acid was used as an adsorbent (Pitsari *et al*., 2013) investigated the modification by 0 .5 M and 1 M citric acid to improve lead adsorption, which increased adsorption capacity by 35% and 82%, respectively. Additionally, paper sludge waste, a by-product of the paper industry, has been explored for PTEs adsorption. Adsorbents derived from de-inking paper sludge showed higher removal of Cu^{2+} than virgin pulp mill sludge (Méndez *et al*., 2009).

Several researchers have investigated the use of modified paper residue for the adsorption of potentially toxic elements. Using paper functionalized with polyethyleneimine (Setyono &Valiyaveettil, 2016) successfully removed nanoparticles, $Ni^{2+,}$ $Cd^{2+,}$ and Cu^{2+} cations, and Cr (VI) anions from polluted water samples. Coated polyethyleneimine paper demonstrated significantly higher adsorption capacities for the pollutants examined in this study compared to untreated paper; these capacities were observed for PTEs Ni^{2+} (208 mg g^{-1}), Cd^{2+} (370 mg g^{-1}), and Cu^{2+} (435 mg g^{-1}) ions compared to nanoparticles (17-79 mg g^{-1}), and Cr (VI) (64 mg g^{-1}) anions. Finally, the factors that affect the adsorption of PTEs onto paper waste have been extensively investigated, including the pH of the solution (Fawzy & Gomaa, 2020), the initial concentration of the pollutant, the contact time (Dehghani et al., 2016), and the temperature.

Further research is needed to optimize paper waste's adsorption performance and explore its potential for large-scale applications.

2.2.3. Potentially toxic elements adsorption by wood mulch and sawdust

Mulch is a protective covering of material laid on top of the soil. Because of its low cost and simple availability, ground, shredded, or chipped wood is the most popular product in the mulch industry. In addition, mulching provides soil erosion prevention, moisture conservation, and weed control (Soleimanifar *et al*., 2019). The quantitative examination of oak wood macromolecule content yielded data from many authors; according to (Puech, 1978; Kollmann& Fengel, 1965; Herrera *et al*., 2014), the proportions of cellulose, hemicellulose, and lignin 41%, 26.35%, and 25.71%, respectively.

Sawdust is a powdery by-product of woodworking processes such as sawing and milling. It can sorb PTEs in stormwater (Deng, 2020). Wood mulch and sawdust are effective adsorbents for various pollutants, including potentially toxic elements. This review discusses some of the studies investigating the adsorption of PTEs by wood mulch and sawdust.

Different types of wood mulch have been investigated as adsorbents of PTEs from wastewater; in a study by (Jang *et al*., 2005), three types of mulch were used as sorbents to collect PTEs in urban runoff. The results revealed that hardwood bark mulch has the optimum physicochemical features for heavy metal ion adsorption. When the Hardwood mulch dosage was 4 g/L, and the pH was less than 6, more than 90% of the Pb (II) in the solution could be eliminated. Nevertheless, the required hardwood mulch dosage was more than 6 g/L, achieving better than 80% removal efficiency for Cu (II) and Zn (II). When the adsorption capabilities at pH 6.0 and 5.0 are compared, a slight variation indicates that the pH is unimportant.

In another study of hardwood wood adsorption of several contaminants by (Ray et al., 2006), the results showed that chromium (Cr(VI)), Copper (Cu^{2+}), cadmium (Cd^{2+}), zinc (Zn^{2+}), lead (Pb^{2+}), fluoranthene, naphthalene, butyl benzyl phthalate, 1,3dichlorobenzene, and benzo(a)pyrene were all sorbed by hardwood mulch from a spiked stormwater pollutant sorbed mass depending on pollutant species, contact time, and initial concentration. Further studies by Iqbal *et al*. (2020) proved that mulches are an excellent source for removing PTEs from soils solution, and the results showed that utilizing woodchips and compost in forest regions can create complexes with copper metal and transform it into a form that is not hazardous for crop plant growth.

Sawdust as an adsorbent of PTEs was studied by Šćiban *et al*. (2007); the study focused on the efficiency of potential toxic elements removal by using sawdust at quantities of 1.25, 2.5, 3.75, 5, 10 and 20 g L^{-1}, the most significant removal efficiencies for Cu(II), Zn(II), and Cd(II) were 76.2%, 37.5%, and 31.9%, respectively.

Recently modifications on wood mulch and sawdust adsorbents were applied by different researchers; for example, The biosorption ability of natural and modified poplar, cherry, spruce, and hornbeam sawdust in removing PTEs from acidic model solutions was investigated by (Kovacova *et al*., 2020) they studied the efficiency of alkaline modified sawdust for metal removal from model solutions at varying beginning concentrations of Cu(II) and Zn(II). Poplar treated by KOH had the maximum adsorption efficiency values for zinc (98.2% at pH 7.3) and copper (94.3% at pH 6.8). Sidhu *et al*. (2021) investigated other modifications, including using iron-based water treatment residuals coated wood mulch to reduce common contaminants in urban runoff. The results reveal that the unique adsorption media removed more Cu, Pb, Zn, and P than the typical uncoated mulch. Coated wood mulches were synthesized and evaluated in another study for removing PTEs and phosphorus (P) from synthetic urban stormwater (Soleimanifar *et al*., 2016). In batch experiments, the composite adsorption capacity was 97% lead (Pb), 76% zinc (Zn), 81% copper (Cu), and 97% phosphorus (P) for the tested synthetic stormwater containing (Pb = 100 g/L, Zn = 800 g/L, Cu = 100 g/L, P = 2.30 mg L^{-1}) at pH = 7.0.

Other reviewed modifications by (Meez *et al*., 2021) of sawdust as an adsorbent material to enhance its selectivity and capacity these pretreatment techniques involved various modifying agents, including (I) organic compounds (ethylene diamine, formaldehyde, epichlorohydrin, methanol, dyes; (ii) acid solutions (HCl, H_2SO_4, H_3PO_4, CH_3COOH, HNO_3, citric acid; (iii) mineral salts (NaCl, KCl, $NaHCO_3$),(iv) basic solutions (NaOH, $Ca(OH)_2$, KOH, Na_2CO_3); (v) phosphorylation treatment ($CO(NH_2)_2$ (urea) + H_3PO_4).

2.2.4. The fate of used cellulose-based adsorbents

Cellulose-based adsorbents are widely used in various applications such as water treatment, environmental remediation, and removing heavy metals and dyes from wastewater. However, the fate of these adsorbents after their use has become a significant concern due to the potential environmental impacts they may cause. These semisolid forms are solid waste containing PTEs flowing through a transportation stream, as summarized in Figure 3 (Xiong et al., 2019).

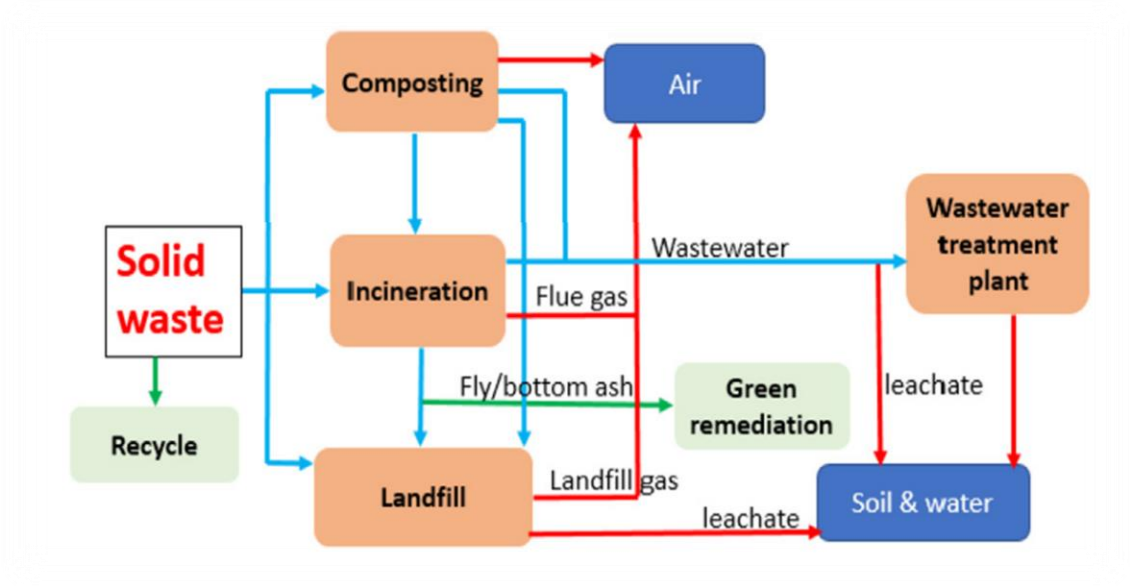

Figure 3. PTEs transportation in the solid waste stream. (Xiong et al., 2019)

Incineration is a frequent way of disposing of used cellulose-based adsorbents. Unfortunately, this process can release harmful air pollutants like volatile PTEs. Furthermore, the ash created by burning cellulose-based adsorbents may contain PTEs that are hazardous to human health and the environment. It causes secondary pollution through PTEs leaching into groundwater.

Another technique for getting rid of used cellulose-based adsorbents is landfilling. Unfortunately, the adsorbents can release hazardous chemicals into the soil and water nearby, endangering the environment. There may be issues with toxins seeping into groundwater since, in most situations, the contaminated cellulose-based adsorbent will contain high water content inappropriate for landfilling (Hubbe,2022). In addition, the degradation of cellulose-based adsorbents in landfills can contribute to the creation of greenhouse gases, mainly methane, significantly contributing to climate change.

Scientists have investigated several techniques for reusing and recycling cellulose-based adsorbents to address these challenges. The rejuvenation of the adsorbents using desorption techniques like heat or chemical treatment has been examined by several researchers (Liu *et al*., 2002), who carried out tests on the adsorption and desorption of Copper (II) using a spherical cellulose adsorbent. An aqueous solution of NaOH or HCl can be used to recover the Cu^{2+} ions that have been adsorbed on the adsorbent. The maximum recovery rate is nearly 100% using a 2.4 mol/L HCl solution. Moreover, the adsorption capacity was lowered after 30 cycles of adsorption/ desorption by only 7.2%. Therefore, the need for new adsorbents can be reduced, and waste can be reduced by reusing previously used adsorbents in various applications. For instance, the regeneration of adsorbents using the eutectic freeze crystallization process was explored by (Hubbe *et al*., 2018).

Another strategy is to repurpose used cellulose-based adsorbents as value-added products. Adsorbents, for example, can be pyrolyzed to form biochar, which can be utilized as a soil amendment or a renewable energy source and for further adsorption processes. Agarwal *et al*. (2015) investigated the removal of azo dye using biochar from pyrolysis of cellulose-based materials municipal solid waste. Table 2 summarizes some PTEs cellulose-based adsorbents and their treatment methods reported in the literature. Finally, the fate of used cellulose-based adsorbents is a critical issue that requires careful consideration. Further research is essential to develop more efficient and sustainable methods for managing used adsorbents.

Table 2. *Potentially toxic elements cellulose-based adsorbents and their treatment methods*

Potentially toxic elements	**Adsorbent**	**Treatment**	**Reference**
Ni, Zn, Cd	Residues of pine sawdust, sunflower seed hulls, and corn residues mix	Immobilization of used adsorbents in clay ceramics	Simón *et al.* (2022)
Pb, Ni	Paper sludge waste	Sulfur treatment with K_2S solution	Wajima (2013)
Cu	Grape bagasse	Pyrolysis	da Silva *et al.* (2022)
Zn, Ni, Cu, Co, Cd	Succinic anhydride-modified mercerized nanocellulose	Regeneration	Hokkanen *et al.* (2013)
Cr	Pine sawdust and oak wood ash	Regeneration	Núñez-Delgado *et al.* (2015)
Cd, Zn, Pb, Cu	Paper and Mulch adsorbents waste	Incineration and Immobilization of adsorbent ash into mortar	Naser *et al.* (2023)

2.3. Landfilling

The presence of diverse pollutants, encompassing potentially hazardous elements within landfill leachate, constitutes a matter of significant scientific apprehension. This phenomenon carries substantial ramifications for both public health and the ecotoxicological equilibrium of terrestrial and aquatic ecosystems, as corroborated by recent studies (Ahmad *et al*., 2021; Kolawole *et al*., 2023). Specifically, the contamination of water sources by Potentially Toxic Elements (PTEs) presents a direct peril to living organisms, manifesting through primary exposure mechanisms such as oral ingestion, exemplified by water consumption (Ur Rehman *et al*., 2018). Additionally, an indirect route of exposure emerges when tainted water is employed to irrigate agricultural crops. The consequence of this practice is the subsequent deposition of PTEs within agricultural fields, where these elements accumulate within the matrices of growing food crops and vegetables, thus engendering a pernicious cycle of food chain contamination with concomitant threats to human health (Yang *et al.,* 2015).

Within the terrestrial ecosystem, soil is pivotal, serving as the ultimate reservoir for PTEs and a conduit for their dispersion into water bodies, organisms, and the atmosphere. PTEs exhibit persistence and gradual accrual within soil matrices, augmenting the overall soil toxicity through interactions with inorganic and organic constituents. Notably, these elements may adsorb to clay particles, sulfides, and various organic compounds, alongside potential associations with hydrated iron (Fe) and manganese (Mn) oxides (Li *et al*., 2022). Nevertheless, the transient sequestration of these PTEs within soil particles ultimately culminates in their release into the surrounding aquatic environments (Jadoon *et al*., 2019).

The modes of human exposure to PTEs encompass dermal contact absorption, direct ingestion, and inhalation, with drinking water and inhaling soil particles identified as principal pathways. These toxic metals, upon entering the human body, can precipitate adverse health effects, including but not limited to renal, gastric, cardiac, and muscular ailments and various forms of malignancy. Sources of PTEs and their transfer to humans are illustrated in Figure (4).

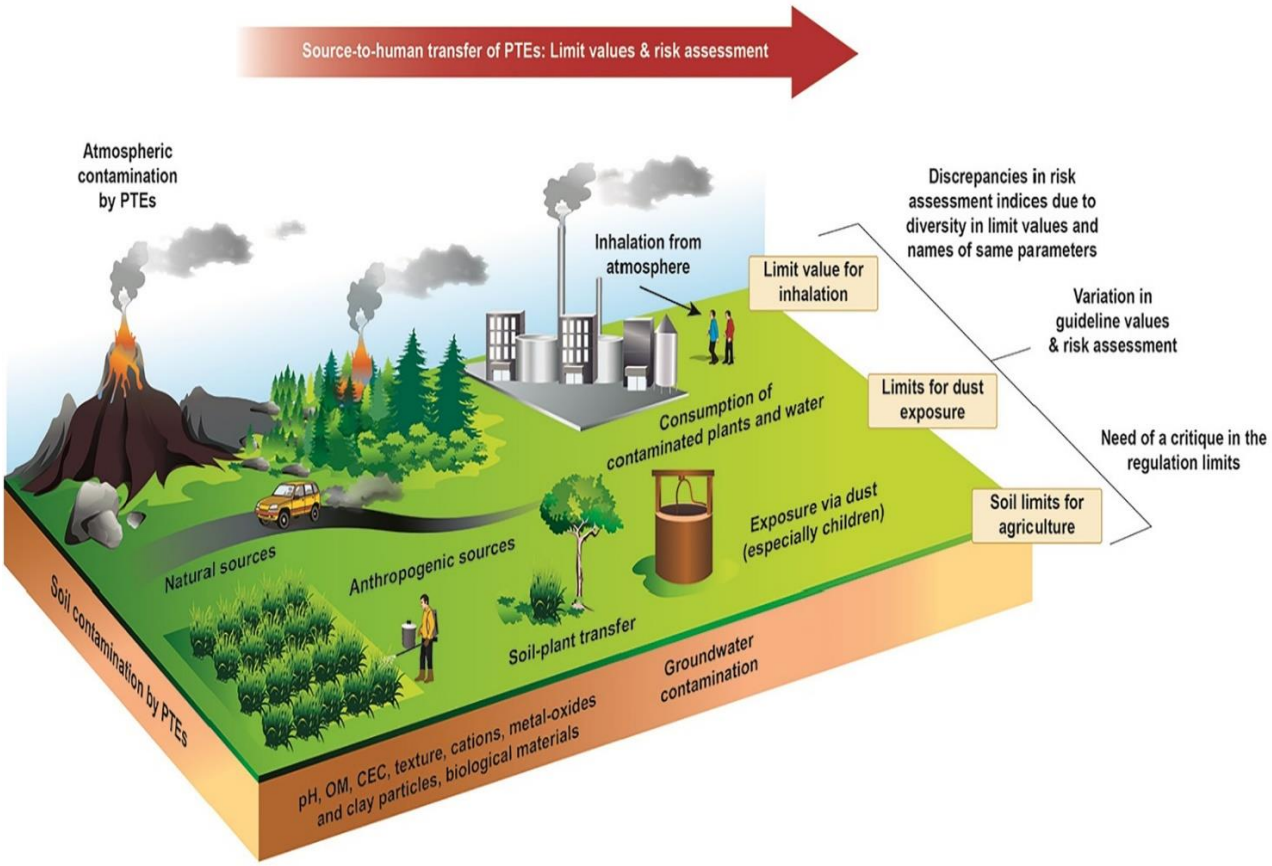

Figure 4. Sources of PTEs and their transfer to humans (Antoniadis et al., 2019)

Waste incineration facilities are prolific generators of ash materials, many of which contain Potentially Toxic Elements (PTEs). The environmental impact of ash landfilling has been rigorously investigated, as exemplified by the study conducted by Twaróg *et al*. (2020) in Poland. This investigation comprised a comprehensive case study assessing the influence of metallurgical ash landfilling on both the soil environment and the atmosphere. The study encompassed the analysis of pseudo-total soil concentrations for a suite of elements, including Cd, Cr, Cu, Fe, Mn, Ni, Pb, Ti, Zn, Li, Sr, and V. Furthermore, the study involved the measurement of CH_4 and CO_2 emissions at various sampling locations. The findings unveiled notably heightened concentrations of Cu, Cr, Pb, and Zn on the surface of the ash dump and along its periphery. The maximal concentrations recorded were 82 mg kg^{-1} for Cu, 23 mg kg^{-1} for Cr, 1144 mg kg^{-1} for Pb, and 8349 mg kg^{-1} for Zn, exceeding both natural background levels and typical values observed in regional soils. Additionally, the study revealed the surpassing of natural background values for Fe, Mn, Ni, Li, Sr, and V. Notably, no methane emissions were detected along the sampling line, while carbon dioxide emissions ranged from 7 to 42 g m^{-2} day-1. The disposal of ash materials thus exhibited a

significant degree of soil contamination with PTEs. This contamination, coupled with alterations in the soil environment, may engender the migration of pollutants into adjacent regions, thereby posing a substantial threat to the environment and living organisms.

The generation of ash from various sources, including wood biomass and paper waste, has garnered substantial attention in recent years due to its environmental and economic implications. This literature review aims to provide an overview of the quantities of ash produced from these sources, highlighting regional disparities and trends. Additionally, it explores the challenges associated with managing this burgeoning ash production and the environmental consequences of current disposal practices. Wood biomass and paper waste production rates were studied by various scholars and summarized in Table 3.

Wood ashes' chemical composition and engineering properties exhibit considerable variability, contingent upon multiple influencing factors. Approximately 70% of wood ash undergoes direct landfill disposal, while approximately 20% serves as a soil amendment, and the remaining 10% fulfills various alternative applications (Wang & Haller, 2022).

Within the European Union (EU), the year 2015 witnessed an estimated production of approximately 7.3 million tons of wood biomass ash (WBA), mirroring the ascendant utilization of wood biomass as a renewable energy source (Obernberger & Supancic, 2009). Croatia, as an example, records an annual consumption of 37,900 tons of wood biomass by a single biomass plant, contributing to WBA production at a rate of 3.1% relative to the initial biomass input (Štirmer *et al*., 2018). Hungary similarly registers an annual mass of 30,000 tons of wood ash, principally managed as waste, with prevalent disposal methods encompassing the filling of former mineshafts and conventional landfilling (Tóth *et al*., 2011).

Globally, the paper industry engenders an annual production exceeding 450 million tons of paper, with projections anticipating a rise to 500 million tons by the culmination of 2020 (Ali *et al*., 2013). Notably, Canada, a noteworthy contributor to paper waste, has an annual output exceeding 1 million tons of wood ash, originating from pulp and paper mills and wood/forest biomass ashes. Nevertheless, this is likely to be a dwindling trend due to the ascent of disposal costs, stringent regulatory measures, and the restriction of landfill capacity, prompting the diminishment or obsolescence of landfilling as a viable disposal avenue (Cherian & Siddiqua, 2019; Lessard *et al*., 2017). Notably, the disposal of Wood biomass ash incurs significant financial obligations, exemplified by annual expenses exceeding 1.7 million € in Austria solely for landfilling (Bohrn & Stampfer, 2014). Eu reports disposal costs per tonne ranging between 100 and 500 EUR (Štirmer&

Carević, 2022). As challenges mount in securing novel landfill sites and conforming to stringent EU landfill directives, it is anticipated that the economic and ecological burdens entailed in WBA disposal will further escalate (James *et al*., 2012).

In the context of paper waste, the annual production rates of fly ash and bottom ash from Canadian paper factories alone assume substantial proportions, approximating 60 and 25 tons, respectively. These materials are frequently subjected to stockpiling or conventional landfilling practices, thereby bestowing environmental challenges (Lessard *et al*., 2017). In summation, the substantial quantities of ash stemming from wood biomass and paper waste underscore the imperative for adopting sustainable management approaches. In light of the burgeoning interest in wood biomass as a renewable energy source and the burgeoning demand for paper, it becomes imperative to confront the challenges associated with ash disposal. Sustainable practices, including ash recycling and alternative utilization avenues, assume pivotal significance in ameliorating financial and ecological burdens while concurrently championing the sustainable deployment of these resources. It becomes discernible that further research endeavors and policy initiatives are necessary to address these issues comprehensively, subsequently catalyzing a transition toward more ecologically favourable ash management practices.

Table 3 .*Waste production rates of wood biomass and paper studied by various scholars*

Ash type and region	Annual Ash Quantity (tons)	Reference
Wood Biomass (EU)	7.3 x 10^7	Obernberger and Supancic, (2009)
Wood Biomass (Croatia)	37,900	Štirmer et al. (2018)
Wood Biomass (Hungary)	30,000	Tóth *et al.* (2011)
Paper Waste (Canada)	>1,000,000	(Cherian & Siddiqua, 2019)
Paper Waste (Canada, One Factory)	60 (fly ash) - 25 (bottom ash)	Lessard *et al*. (2017)
Forest residue (America)	231 million	Siddique et al. (2019)

2.4. Leaching of Potentially Toxic Elements

Leaching, as defined by Van der Sloot et al. (2018), refers to the process of releasing constituents from a solid material into the aqueous phase upon contact with water. The extent of release into the aqueous phase can be determined through constituent liquid-solid partitioning. Non-managed waste incineration ash may harm the environment and cause the release of many potentially toxic elements into the surrounding environment (Udoeyo *et al*., 2006). Testing leaching is mandatory for waste incineration ash to characterize its ecotoxicity (Stiernström *et al*., 2014); many test methods are used worldwide for this purpose, like TCLP (Toxicity Characteristic Leaching Procedure), SPLP (Synthetic Precipitation Leaching Procedure), ASTM-D3987 (American Society for Testing and Materials), MEP (Multiple Extraction Procedure US EPA Method 1320), and CEN/TS 14429:2015 Characterization of waste - Leaching behavior test - Influence of pH on leaching with initial acid/base addition (Kadir *et al*., 2016; Xu *et al*., 2019; Masud *et al*., 2021; Darama *et al* ., 2021).

Numerous studies have investigated the leaching risk of incorporating wood and paper ash into cementitious composites. Ismail *et al.* (2019) conducted a study in which wastepaper bottom ash and fly ash derived from paper recycling plants were utilized for soil stabilization. The environmental impact of these materials was evaluated following the guidelines outlined in the EN 12457-2 standard. The findings from the leaching test emphasized the importance of considering the concentration of barium when working with these ash materials. Notably, adding wastepaper ash as a binding agent significantly reduced barium content. Comparing the results to the European directive (Directive, 1999/31/EC), the soil stabilized with wastepaper ash was classified as an inert material.

In a separate study, Carević *et al.* (2020) conducted leaching tests on cement composites containing 15% wood biomass ash as a replacement for cement. The investigation encompassed both monolithic and crushed cement composites, with the elements Cd, Cr, Cu, Ni, Pb, and Zn being studied. The results indicated that the leachates from the monolithic composites failed to meet the criteria for categorization as inert waste. However, for the monolithic samples, they satisfied the criteria outlined in the "Soil Quality Degree" standard.

Drljača *et al*. (2022) examined the leaching behavior of wood biomass ash obtained from two distinct sources. The first source involved the combustion of hardwood types (spruce and fir) in a heating plant, resulting in ash formation. The second source consisted of wood bark combustion. Leaching tests were performed using distilled water as an extraction agent. To evaluate the

potential release of toxic elements, the same cement composite was subjected to leaching tests using a fresh solution of deionized water, targeting Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Ba, and Pb. The leaching tests conducted on composites prepared from wood ash in combination with cement demonstrated minimal leaching of ash, with all heavy metals being bound within the cement matrix. This indicates the potential suitability of wood ash for construction purposes.

The aforementioned studies underscore the significance of considering the specific ash material and its composition when evaluating leaching behavior and environmental impact in cementitious composites. Incorporating distinct ash materials, such as wastepaper ash or wood biomass ash, can yield different outcomes regarding the leaching of contaminants. These differences can arise from variations in the percentage of ash added to the cementitious composites, the initial concentration of contaminants present in the ashes, and the pH of the studied leachates. Acquiring a comprehensive understanding of the behavior of these materials is essential for assessing their appropriateness for construction applications and ensuring adherence to environmental regulations.

2.5. Factors affecting PTEs leaching from adsorbent ash mortar composites

2.5.1. The pH

In the realm of environmental science, a comprehensive understanding of the crucial role that pH plays in the solubility of PTEs is imperative (Keshavarzi *et al*., 2015). Despite the essential nature of many elements in minimal quantities for living organisms, certain PTEs can surpass naturally occurring background concentrations, raising concerns about their potential toxicity. It is essential to possess in-depth knowledge of PTEs to implement defensive measures against excessive exposure (Jaishankar *et al*., 2014).

The significance of PTEs in soil is categorized into macronutrients, micronutrients, and toxic elements, underscoring their multifaceted roles (Khumalo *et al*., 2020). Plants, vital for their life cycle, rely on essential macronutrients (e.g., nitrogen, phosphorus, potassium, calcium, sulfur, and magnesium) and micronutrients (such as iron, zinc, manganese, nickel, and copper) from the soil (Kabata-pendias and Pendias, 2001). Micronutrients, crucial in low concentrations, can become highly toxic in excess, while macronutrients are required abundantly for optimal plant growth (Stanojković Sebić *et al*., 2017). In contrast, nonessential elements like mercury, lead, cadmium, and arsenic are recognized for their harmful effects, leading to significant health problems (Shiva Kumar and Srikantaswamy, 2014). Among these, cadmium, due to its toxicity, propensity to

accumulate in soils, and bioaccumulation in plants and animals, emerges as particularly concerning (Al-hwaiti and Ranville, 2010).

The intricate adsorption/desorption behavior of cadmium is significantly governed by pH, with a secondary influence from the solution concentration of cadmium and the presence of competing cations or complexing ligands. An Eh-pH diagram for cadmium shown in Figure 5 reveals that Cd^{2+} is the soluble form at pH < 5 and under moderately to highly oxidizing redox potentials. Notably, in near neutral to moderately alkaline pH (6 to about 12), cadmium carbonate remains stable, while cadmium hydroxide is stable at pH > 12.5. Furthermore, in sulfidic environments, cadmium sulfide retains stability across a broad pH range (Nickel, 2007). This underscores the critical influence of pH on the speciation and mobility of cadmium in diverse environmental conditions.

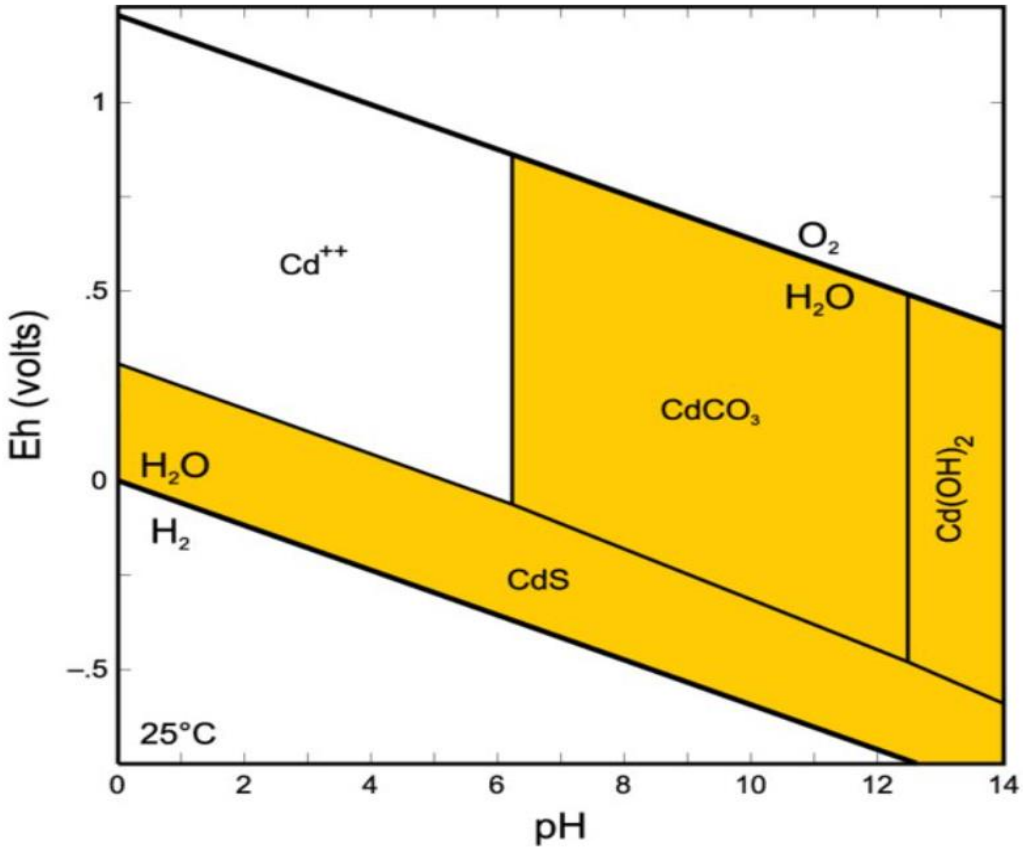

Figure 5. Eh-pH diagram for cadmium (total Cd = 10^{-5} molal, total C = 10^{-3} molal, total S = 10^{-3} molal; all organic cadmium complexes are suppressed; activity coefficients for all species are set equal to 1). (Nickel, 2007).

The leachability of Potentially Toxic Elements is known to be predominantly governed by pH, which is considered a critical chemical parameter. The extent of PTEs released from binder materials is highly dependent on the pH of the surrounding environment (Tiwari *et al*., 2015; Berra *et al*., 2019; Masud *et al*., 2021; van der Sloot & van Zomeren, 2012; Król, 2011; Dijkstra *et al*., 2004; van der Sloot & Kosson, 2010; Saveyn *et al*., 2014).

Table 4 summarizes research studies on leaching test methods, pH values, and materials studied. These research studies have employed different leaching test methods to assess the leaching behavior of PTEs under varying pH conditions. For instance, Xu et al. (2019) compared leaching tests commonly used in China, such as HJ/T 299-200 and HJ/T 300-2007, with internationally recognized leaching tests, including 1311 TCLP (USEPA 1992a), 1312 Synthetic Precipitation Leaching Procedure of USEPA SW-846 (USEPA 1992b), and CEN/TS 14429 Leaching behavior tests. Their findings indicated that the CEN/TS 14429 test method was more suitable for examining the leaching behavior of PTEs in various materials or products under diverse environmental pH conditions, especially when evaluating recycled raw materials or products derived from hazardous industrial wastes.

Table 4. *Summary of research studies on leaching test methods, pH values*

Reference	Leaching test method	pH value	Materials studied
Kalembkiewicz & Sitarz-Palczak, 2015	USEPA [USEPA] TCLP [TCLP 1311] ASTM D 3987-85	Nitric acid: 0.6 ± 0.1 Acetic acid: 4.0 ± 0.1 Distilled water: 6.3 ± 0.1	N/A (Test conditions provided)
Berra *et al*., 2019	NEN 7345 leaching TEST	pH 6 (acidic environment)	Hardened cement pastes prepared with a blended cement (70% Portland cement – 30% washed fly ash)
Masud *et al*., 2021	N/A	2.5 and 3.0	High-Performance Mortar (HPM) containing high-volume fly ash as cement replacement
Xu *et al*., 2019	HJ/T 299-2007 HJ/T 300-2007 US EPA SW-846 Methods 1311 US EPA SW-846 Methods 1312 CEN/TS 14429	3.20 ± 0.05 2.64 ± 0.05 2.88 ± 0.05 4.20 ± 0.05 The pH of the reagent varies depending on the sample	Two types of municipal solid waste incineration fly ash (grate firing fly ash - GFFA, fluidized bed fly ash - FBFA) used as AAMs brick raw materials

2.5.2. Ash type

In recent years, researchers have extensively studied cellulose-based biomass ash and found it to be highly efficient for various applications in civil engineering. These applications include its use in bricks, panels, geopolymers, alkali-activated materials, road construction, soil stabilization, concretes, mortars, and other eco-friendly building materials (Raheem & Adenuga, 2013; Da Luz & Sousa, 2013; Emeh & Igwe, 2016; Yliniemi *et al*., 2016; Eliche-Quesada *et al*., 2017; Akinyemi & Dai, 2020; Dimter *et al*., 2021; Wang & Haller, 2022).

However, before utilizing wood ash in such applications, it is crucial to evaluate its specific characteristics, as they can significantly influence the properties of cementitious matrices. Steenari *et al*. (1999a) studied the elemental composition and mineralogical characteristics of diverse ash types derived from different biomass sources. They subjected these ash samples to leaching tests to assess potentially toxic elements released. The results revealed that the leaching behavior of PTEs was influenced by various factors, such as the elemental composition, mineralogical properties of the ashes, and the specific fuel sources used for their production. The origin of the ash, which is determined by the type of fuel utilized (biomass, gas, oil, wood waste, peat, or board production residues), plays a crucial role in defining its unique composition. This diversity of inorganic species in the fuel used for ash generation leads to variations in the properties of the resulting ash, significantly affecting the leaching potential of PTEs under different environmental conditions and scenarios.

Zając *et al*. (2018) stated that the differences in ash composition can be attributed to various factors, such as the type of biomass, the species of origin, the growth conditions and age of the plant, fertilization, the use of pesticides, time and techniques of harvesting, transport conditions, wood storage, as well as technology and burning conditions. This ash variability poses complexity in its application, as its properties depend on multiple factors, with the chemical composition being the main determinant.

As the concentrations of constituents, elements, and morphology vary depending on the type of ash used in mortars and concrete, it is impossible to predict how using a particular ash will impact concrete durability (Fava *et al*., 2018). it becomes necessary to assess the final product through leaching and solubilization tests to determine the potential encapsulation of heavy metals in the C-S-H matrix, if present, and to avoid any undesirable reactions with new products. Therefore, new

materials must undergo thorough technical and environmental analysis before becoming viable for application.

According to a recent review by Nascimento *et al*. (2023), wood bottom ash exhibits different chemical and physical characteristics depending on the species of origin, the parts of the plant that generated the ashes, and the burning conditions. However, in general, wood bottom ash contains a higher amount of Ca, which may be surpassed by Si in some cases. The chemical variation can affect the pozzolanic activity of wood bottom ash. In conclusion, cellulose-based biomass ash, particularly wood ash, shows immense promise as an eco-friendly construction material in civil engineering. However, due to the diversity of factors affecting ash characteristics, careful evaluation is necessary before its application to ensure its suitability and performance in various construction scenarios.

2.6. Immobilization of PTEs

The immobilization efficiency of potentially toxic elements (PTEs) refers to their ability to be securely trapped and resist leaching. Several factors can influence immobilization efficiencies, such as the properties of the raw materials, the characteristics of PTEs, their forms, and the pH conditions during the immobilization process (Malviya & Chaudhary, 2006; Vu & Gowripalan, 2018). The efficiency is quantified using the following equation:

$$\text{Immobilization efficiency (\%)} = (Cr - Ci)/Cr \times 100 \quad (1)$$

Cr represents the amount of PTEs in the contaminated ash, and *Ci* represents the amount of PTEs leached out from mortar specimens.

Strength development serves as a reliable indicator of solidification, while the leaching test stands out as a paramount measure for assessing the extent of immobilization of Potentially Toxic Elements (PTEs). Numerous leaching methods have been widely employed to gather essential data for informed environmental decision-making. Consequently, selecting an appropriate leaching test method becomes crucial in evaluating the efficiency of PTE immobilization within cementitious composites. The process of immobilization treatment effectively mitigates the potential hazards posed by PTEs-contaminated solid waste to the environment. This technology reduces the potential migration of PTEs by modifying the physical and chemical properties of the waste materials. Over the past few decades, various immobilization methods have been developed, including cement-based solidification/stabilization (S/S) (Ucaroglu & Talinli, 2012), geopolymer-based S/S (Guo *et al*., 2017), vitrification (Coruh & Ergun, 2006), carbonation (Li *et al*., 2007), Colloidal silica medium to obtain safe inert (Benassi *et al*., 2016), and others. Figure 6 illustrates the classification of these diverse immobilization techniques. Notably, cementitious materials and thermal treatments emerge as particularly attractive methods because they encapsulate PTEs in durable and compact waste forms. Over the past few decades, the solidification-stabilization method using cement-based S/S has been widely used for treating solid waste contaminated with Persistent Toxic Elements (PTEs) (Batchelor, 2006).

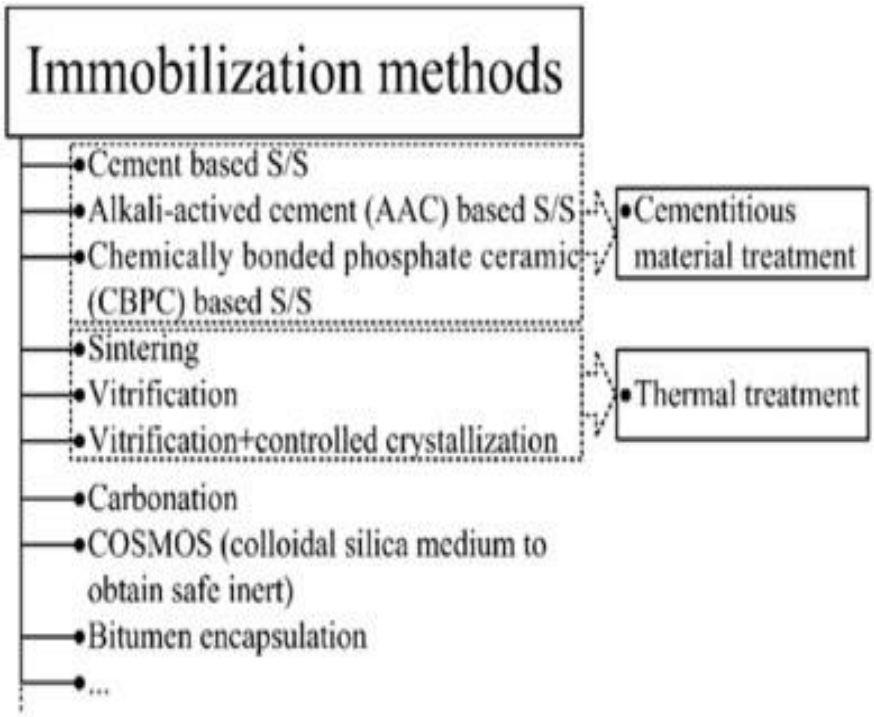

Figure 6. PTEs immobilization methods (Guo et al., 2017)

The primary binder in this technique is Portland cement (Gougar *et al*., 1996; Shi & Spence, 2004). It was also one of the first materials used in nuclear waste S/S applications (Conner & Hoeffner, 1998). In the context of immobilizing cellulose-based biomass ash, a study conducted by Pavlíková *et al*. (2018) involved the incorporation of wood chip ash into a mortar, and then subjecting the resulting composites to leaching tests. The research outcomes showcased the successful immobilization of chlorides in the ash within the silicate matrix. Similarly, in another investigation conducted by Chen *et al*. (2018), Portland cement was utilized to solidify and stabilize rice husk and sewage sludge co-combustion ashes containing various PTEs (Cu, Cd, Pb, Zn, Fe, Mn, Cr, Ni, Ba). The three-step TCLP leaching method was employed to assess the concentrations of leached metals, and the results indicated that the addition of cement effectively immobilized the PTEs. Limited research has been conducted on the leaching of PTEs from mortar containing fly ash and bottom ash of wastepaper. Ismail et al. (2019) explored the use of wastepaper sludge ash in the cement matrix to stabilize water treatment sludge containing Cd, Pb, Ni, and Cr. The leaching test results further validated the efficacy of immobilization within cementitious composites.

2.7. Immobilization mechanism

The chemical stabilization mechanisms of cement-based S/S are well discussed in the literature. The possible immobilization mechanisms of PTEs in concrete and mortar could be (1) sorption, (2) chemical incorporation (surface complexation, precipitation, co-precipitation, and (3) micro-encapsulation or macro-encapsulation (Trussell & Spence, 1994; Glasser, 1997). Table 5 summarizes some PTEs immobilization mechanisms and efficiency reported in the literature.

Table 5. *Potentially toxic elements immobilization mechanisms and efficiency within cementitious composites*

Immobilized element	Mechanism of immobilization	Immobilization efficiency	Reference
Lead (Pb)	precipitation and chemisorbed by Si chains	Pb precipitates as Pb hydroxides, The hydrates sorb 21.35 %; the rest (5.96 %) cannot be immobilized	Wang& Wang, 2022
Lead (Pb)	Precipitation as insoluble lead silicates, hydroxides, and carbonate	N/A	Liu *et al*. 2023
Zinc (Zn)	Chemosorption Zn can occupy different crystallographic positions in the tobermorite structure depending on the presence or absence of co-absorbing elements.	N/A	Vespa *et al*. 2014
Copper (Cu) + Zinc (Zn)	chemical complexation and ionic substitution within a magnesium sulfate cement composite	immobilization efficiency (up to 99%) for Cu and Zn	Tan *et al*. 2023
Cadmium(Cd) + Lead (Pb)	Precipitation as cadmium hydroxides Chemical adsorption, physical encapsulation, and physical adsorption.	Cd was 99.97% Pb and 94.67% at 28 days	Wang *et al*. 2022

2.8. Instrumental analytical methods

In the realm of environmental analysis, the presence of potentially toxic elements (PTEs) is a concern across diverse ecosystems. Therefore, various instrumental analytical techniques are employed to assess the concentration levels of PTEs in different samples (Helaluddin *et al*., 2016). Determinative methods utilized for PTE analysis include flame atomic absorption spectrometry (FAAS) (Ademola *et al*., 2015), atomic fluorescence spectroscopy (AFS), anodic stripping voltammetry, inductively coupled plasma optical emission spectrometry (ICP-OES), and inductively coupled plasma mass spectrometry (ICP-MS) among others, which have been extensively employed for determining PTE contents in marine environments (Okoro *et al*., 2012). These techniques, such as atomic absorption spectrometry (AAS), atomic emission/fluorescence spectrometry (AES/AFS), ICP-MS, ICP-OES, neutron activation analysis (NAA), X-ray fluorescence (XRF), and anodic stripping voltammetry (AVS), have been recognized as the most prevalent analytical approaches (Helaluddin *et al*., 2016). According to Feist et al. (2008), instrumental techniques like ICP-OES or AAS enable simultaneous or sequential determination of multiple elements. Sereshti *et al*. (2012) highlight that ICP-OES allows rapid multi-element analysis, making it suitable for various complex and organic matrices applications. Moreover, ICP-OES exhibits low interference effects even with highly resistant elements due to the inert and high-temperature medium employed during sample atomization (Sereshti *et al.,* 2012). Similarly, Fassel and Kniseley (1974) emphasize that ICP-OES has emerged as a valuable methodology for assessing metals in diverse sample matrices. The technique utilizes different nebulizers or sample introduction methods to introduce fluid samples into an argon plasma induced by radiofrequency (RF). ICP-OES techniques are widely adopted in research and environmental control laboratories. Successful implementation of these techniques relies on favorable detection limits, linear calibration curves, compatibility with acid dissolution, and low sensitivity to matrix effects (Bettinelli *et al*., 2000).

Heltai *et al.* (2019) conducted a scientific investigation on the utilization of flexible multi-elemental inductively coupled plasma optical emission spectrometry (ICP-OES) detection for fractionating the content of potentially toxic elements (PTEs) in solid environmental samples using a sequential extraction procedure. The primary objective of their study was to elucidate the application of the multi-elemental ICP-OES method as a versatile and dependable approach for assessing the mobility of PTEs in soils and sediments through the BCR (European Community Bureau of Reference) fractionation technique. The authors concluded that the ICP-OES method was effectively employed for monitoring the remediation efforts of PTE-contaminated lake

systems, such as the Gödöllő-lake system, and could also be successfully applied for characterizing the environmental mobility of PTEs and other micro- and macro-elements in soils.

Concerning our study of mortar composites for leaching detection Similarly, Maherzi *et al*. (2023) investigated the leaching of soluble PTEs and anions from mortar composites, wherein waterway sediments were partially replaced with waste sediment. They aimed to identify the mechanisms of releasing chemical substances from monolith samples into water. The concentrations of PTEs such as (As, Ba, Cd, Cr, Cu, Co, Mo, Ni, Pb, Sb, Se, Zn, Ca, Fe, K, and Mg) and anions (including sulfates, chlorides, and fluorides) in the leachate were successfully measured using ICP-OES and ionic chromatography techniques. Taken together, the aforementioned studies highlight that ICP-OES demonstrates sufficient detection capability for studying PTEs in environmental investigations, particularly when a simultaneous multi-elemental analysis is required.

2.9. Characterization Methods

2.9.1. X-Ray Diffraction (XRD)

Wood bottom ash (WBA) is a residual material generated as a by-product during the incineration or combustion processes of wood-fired power plants, pulp and paper mills, and wood-based industrial facilities (Ayobami, 2021).

Extensive scientific investigations have focused on characterizing wood ash and wastepaper ash using X-ray diffraction (XRD) analysis to elucidate their crystalline phases, thus providing valuable insights into their composition and structural properties.

In a study by Carrasco *et al*. (2014), XRD analysis was conducted on wood bottom ash (WBA) incorporated into concrete, revealing quartz as the prevailing crystalline phase. Additionally, other crystalline materials, including calcite, ankerite, langbeinite, and calcium silicate, were identified within the ash. Conversely, Chowdhury *et al.* (2015) observed higher amorphous phases, particularly SiO_2, in the wood bottom ash XRD pattern, alongside discernible crystalline peaks. The amorphous nature of silica in wood bottom ash, known for its pozzolanic characteristics, makes it suitable for cementitious applications. Sklivaniti *et al.* (2017) investigated the XRD profile of wood bottom ash derived from the combustion of olive plant trimmings, indicating that calcite ($CaCO_3$) dominated approximately 70% of the major phases. Additional notable phases included fairchildite, quartz, and grossular. Furthermore, a chemical bonding between potassium and sulfur resulted in the formation of arcanite. These compositions were found to be significantly influenced by various biomass growing conditions, such as fertilizer type and soil characteristics.

In the case of wastepaper ash, Baloochi *et al.* (2022) utilized XRD analysis to identify crystalline phases, detecting the presence of calcite, lime, quartz, larnite, aluminum, and halite. Moreover, minor amounts of portlandite, likely attributed to environmental moisture, were observed. Some similarities were noted between these ashes and previous studies, such as the presence of lime and calcite (Bai *et al*., 2003; Kinuthia *et al*., 2001; Segui *et al*., 2013; and Spathi *et al.,* 2015).

In the context of mortar ash composites, Sağın *et al*. (2012) investigated the composition of historical mortars. The results of their study revealed that the binders in these mortars primarily consisted of fine-grained silica and calcium carbonate, which played a crucial role in imparting hydraulic properties and high strength to the mortar. Similarly, Cao *et al.* (2019) demonstrated that adding calcium carbonate to cementitious composites positively impacts early-age strength, the hydration process, and overall durability. Considering that the main constituents of adsorbed paper ash (APA) and adsorbed mulch ash (AMA) are $CaCO_3$ and SiO_2, their inclusion in mortar mixes is suitable and can be justified based on their compatibility with the desired properties.

2.9.2. Scanning Electron Microscopy (SEM)

In the microscale range below 1mm within the cementitious paste matrix, sand or fine aggregate particles embed with a specific microstructural shape resembling the arrangement of mortar constituents. This arrangement is influenced by various factors, including the chemical and mineralogical composition of mortar constituents, the quantity of aggregates and other additives, the water-to-cement ratio, the mixing process, and the type of cement employed (Gao *et al.,* 2018; Gominho, 2016). Predicting the behavior of such materials poses significant challenges due to the complexity of their microstructure, morphology, and composition. Microscopy techniques have been utilized to investigate materials, enabling the development and enhancement of materials, quality assurance, reverse engineering, and performance evaluation by analyzing their structural properties (Stefanidou & Pavlidou, 2018).

The advancement of energy-dispersive X-ray spectrometry (EDS) systems technology, as highlighted by Thompson (2017), has facilitated the acquisition of qualitative and quantitative information for materials analysts. For example, identifying phases containing calcium hydroxide (CH) crystals, which exhibit various shapes and sizes, is relatively straightforward with EDX analysis. However, other phases, like calcium sulfate hydrates, have a more continuous morphology. Two phases in cement hydration products are of particular interest. The first is the AFt phase, denoting the presence of calcium aluminate ferrite trisubstituted or calcium aluminate trisubstituted phases in the hydrated (or hardened) cement paste. The second phase is the AFm

phase, which refers to an "alumina, ferric oxide, monosubstituted" phase or aluminate ferrite monosubstituted, Al_2O_3, Fe_2O_3 mono. AFm phases are significant hydration products in Portland cement and hydraulic cement hydration. According to Celik (2015), AFt forms needle-like crystals in the early stages of hydration, while AFm appears as hexagonal platy crystals. Early formed AFm tends to crystallize in clusters or rosettes of irregular plates, whereas those formed later develop into well-developed yet very thin hexagonal plates.

Regarding the SEM analysis of mortar wood ash composites, Awolusi *et al*. (2017) investigated the microstructure features of conventional Portland cement mortar incorporating waste wood ash from sawdust. The SEM image revealed larger interspatial distances between sawdust ash particles compared to cement particles, indicating a more compact arrangement of the latter. Gabrijel *et al.* (2021) conducted an SEM image study. They found that adding wood ash to concrete increased particle sphericity, which improved packing and enhanced workability when combined with smaller particle sizes.

The microstructure investigation of incorporating paper waste ash into mortar composites has received limited attention in the existing literature. However, Wong *et al*. (2022) conducted a study in which waste newspaper ash was employed as a partial replacement for cement in treated concrete cylinders. The study utilized SEM analysis to examine the microstructure of the newspaper ash concrete composite sample compared to a reference sample. The SEM analysis revealed a dense surface with a reduced number of voids in the micrograph of the newspaper ash concrete composite sample after a 28-day curing period, indicating improved compactness and reduced porosity compared to the reference sample. Overall, the relation between the literature findings is that incorporating wood ash or paper waste ash into cementitious composites can positively impact the microstructure. Adding these waste materials can lead to a more compact arrangement of particles, improved particle sphericity, enhanced packing, and reduced voids, potentially improving cementitious materials' density, workability, and mechanical properties.

2.10. Regulatory measures

The process of industrialization and urbanization has led to an augmented release of Potentially Toxic Elements (PTEs) into the natural environment, encompassing soil, lakes, rivers, groundwater, and oceans. The hazardous nature of these toxic elements is determined by their persistence, toxicity, and ability to accumulate in organisms. Unfortunately, PTEs can be mobilized and transported through water or wind to various compartments of the environment, such as air, water, sediment, and soil (Achour *et al.,* 2022).

Numerous physical, chemical, and biological remediation approaches have addressed PTE contamination in soil and water. Among these techniques, adsorption is widely employed (Azeem *et al.,* 2022). While adsorption effectively removes PTEs from wastewater, it results in secondary pollution when the used adsorbents are disposed of in landfills, leading to the leaching of PTEs into groundwater (Naser *et al.,* 2023). In accordance with Directive 1999/31 of the European Union's waste hierarchy, landfilling is the least desirable option and should be minimized to the utmost extent necessary. To facilitate the transition to a circular economy, the Landfill Directive imposes restrictions on landfilling waste that can be recycled or recovered for material or energy purposes. By 2030, the directive limits the landfilling of municipal waste to 10%, aiming to further reduce it to that level by 2035. Landfills are categorized into hazardous waste, non-hazardous waste, and inert waste.

Various governing bodies implement standard limits and regulations to ensure public health and environmental safety concerning potential toxic elements. These standards provide guidelines for acceptable levels of toxic elements in different environmental media, including air, water, soil, and food. Examples of such standards and regulations include the Environmental Protection Agency (EPA) Standards, European Union Regulations, World Health Organization (WHO) International Standards, and National Standards.

In Hungary, for instance, Joint Decree No. 10/2000 (VI.2.) KöMEüM-FVM-KHVM establishes limit values to safeguard groundwater and the geological medium, while Governmental Decree No. 219/2004 (VII21.) Korm. Provides background concentration and pollution limit values for the geological medium and groundwater. Table 6 shows the standard limits of PTEs in different media and regulatory bodies. It is essential to acknowledge that standard limits and regulations can vary between countries and regions. These limits are subject to periodic review and updating based on scientific research and emerging knowledge regarding the toxicity of different elements. Compliance with these standards plays a crucial role in safeguarding human health, protecting the environment, and ensuring the overall well-being of communities.

***Table 6**. Standard limits of potentially toxic elements in different media and regulatory bodies*

Regulatory	Average concentration in Hungarian soil (µg/g)	Hungarian (Threshold limit) (µg/L)	WHO (ppm)	WHO (ppm)	Leaching limit Values based on wase acceptance crieteri,a (mg/Kg),L/S=10		
Media	Soil concentration	Groundwater	Drinking water	Sediments	Inert waste landfill	Non-Hazardous waste land fill	Hazardous waste land fill
Cd	0.01 - 2	1	0.001 - 0.005	0.1	0.04	1	5
Zn	50 - 100	200	5	≤1	4	50	200
Cu	10 - 15	30	3	0.05 - 0.15	2	50	100
Pb	15 - 30	10	0.1	5	0.5	10	50
Refrence	(Ezejiofor et al., 2013)	Joint Decree No. 10/2000 (VI. 2.)	(Olayinka et al., 2021)		EU Directive 1999/31		

3. MATERIALS AND METHOD

This chapter explains the steps taken in detail to achieve the results. At the beginning of this chapter, a summarized methodology is shown in a flowchart illustrated in Figure 7, followed by a detailed description of the raw materials, the adsorption process, the preparation of the adsorbent ash, and the preparation of incorporated mortar ash composites. This chapter also presents the analysis of raw materials and the chemical properties of the prepared mortar ash composites that might affect the immobilization of PTEs within a mortar composite. Lastly, the quantitative and qualitative Methods used to determine PTEs are mentioned in detail.

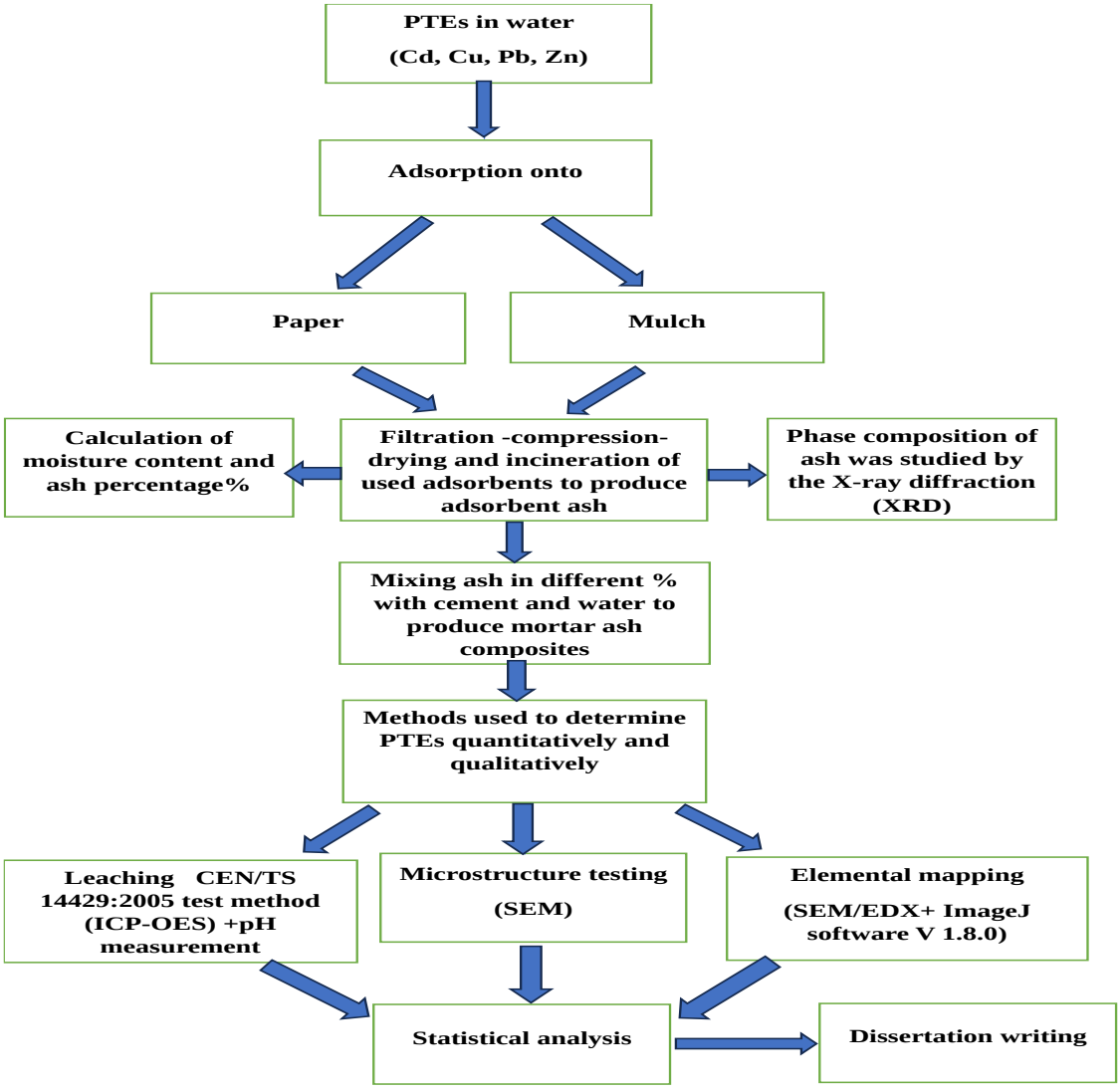

Figure 7. Methodology flowchart

3.1. Batch adsorption method

Two cellulose-based adsorbents, depicted in Figure 8, were employed in this study. The first adsorbent is a wastepaper including cardboard and receipts obtained from a local store in Gödöllő, Hungary. The sample was washed several times with distilled water, dried, ground then kept in sterilized plastic bottles. The second adsorbent is mulch obtained from Oak trees. The sample was washed several times with distilled water, dried, ground then kept in sterilized plastic bottles.

Chemical grade salts [$Pb(NO_3)_2$, $CdCl_2 \cdot 2.5H_2O$, $Cu(NO_3)_2 \cdot 3H_2O$, $Zn(CH_3CO_2)_2 \cdot 2H_2O$] were used to prepare the concentrated stock solution 1000 mg L^{-1} of each of (Pb(II), Cu(II), Zn(II), Cd(II)) ions by dissolving 2.03 g of cadmium dichloride, 1.60 g of lead nitrate, 3.80 of Copper (II) nitrate and 3.36 of Zinc acetate in the same solution. This stock solution was subsequently diluted to five different concentrations (0.5, 1, 5, 10, 50) mg L^{-1}. Each adsorbent was applied at a concentration of 1 g in 100 ml. The samples were shaken for four hours. The type and operating conditions of the shaker used: (Multi Rotator /PTR-60 Grant-bio, speed =24 rpm, vibration mode = 1°, reciprocal mode=2°). Triplicate repetition of the adsorption process was conducted at five different concentrations (0.5, 1, 5, 10, 50) mg L^{-1}. After adsorption, samples were filtered using ashless MN 640m.Ø90mm filter paper. The total elemental content of PTEs filtrate was determined by a Horiba Jobin Yvon Activa M Inductively Coupled Plasma–Optical Emission Spectrometer (ICP-OES). The obtained data were used to calculate the adsorption capacity of PTEs in mg g^{-1} according to the equation (1):

$$q = [(C_o - C_e) /m] *V \qquad (1)$$

Where q is the uptake (mg g^{-1}), C_0 and C_e are the liquid phase concentrations of PTEs at initial and equilibrium (mg L^{-1}), V is the volume (L), and m is the amount of adsorbent (g).

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Figure 8. Raw materials mulch and paper

3.2. Adsorbent ash preparation

In this study, two kinds of PTEs-contaminated ash were produced in the laboratory after conducting the adsorption experiments. The remaining adsorbents on filter paper were compressed to separate the adsorbed fraction as in Figure 9; this procedure was followed by oven drying at 105°C. Finally, samples were burned in the furnace at 580°C for four hours. Ash of mulch and paper are shown in Figure 10.

Figure 9. Compression of used adsorbents

Figure 10. Ash of mulch and paper after (adsorption-compression- oven drying-burning)

3.3. Determination of adsorbents' moisture content (%)

The moisture content of 30 samples utilizing mulch and paper as adsorbents was assessed in accordance with (ISO 287:2017), This procedure was done in triplicate for each adsorption initial concentration sample. The following procedure was performed for each sample:

1- Container weight determination; Weigh the empty container in grams.
2- Initial Mass Determination: Weigh the sample+ Container accurately to determine its initial mass.
3- Drying: Place the sample in an oven preheated to a temperature of 105°C. The drying Period allows the sample to dry in the oven until it reaches a constant mass. This is typically achieved when the difference between two successive drying and weighing sessions, separated by a time interval of at least half the initial drying period, does not exceed 0.1% of the initial mass.
4- The calculation of the moisture content was done by using the formula (2) below:

$$\% \text{ Moisture Content} = [(W_i - W_f) / (W_i - W_c)] \times 100 \qquad (2)$$

Where: W_i = initial weight; W_f = oven-dry weight, and W_c = sample container weight; all in grams. Note that W_i and W_f include the container weight.

3.4. Determination of the ash percentage (%)

The determination of the percentage of ash of mulch and paper adsorbents was performed on 30 samples according to the standard test method ASTM D1102-84 (2013) as follows:

1- Ignited the empty crucible and cover it over a burner or in the muffle at 600°C, cool it in a desiccator, and weigh it to the nearest 0.1 mg.
2- Placed the test specimen in the crucible, determined the weight of the crucible plus specimen, and placed it in the drying oven at 100 to 105°C with the crucible cover removed. After 1 h, replaced the cover on the crucible, cooled in a desiccator, and weighed. Repeated the drying and weighing until the weight is constant to within 0.1 mg.
3- Placed the crucible and contents, with the cover removed, in the muffle furnace and ignite until all the carbon is eliminated. The temperature of ignition was 580 °C.
4- Removed the crucible with its contents to a desiccator, cooled, and weighed accurately.

5- Calculated the percentage (%) of ash, based on the weight of the moisture-free sample, as follows in formula 3:

$$\text{Ash percentage (\%)} = (W1 / W2) * 100 \qquad (3)$$

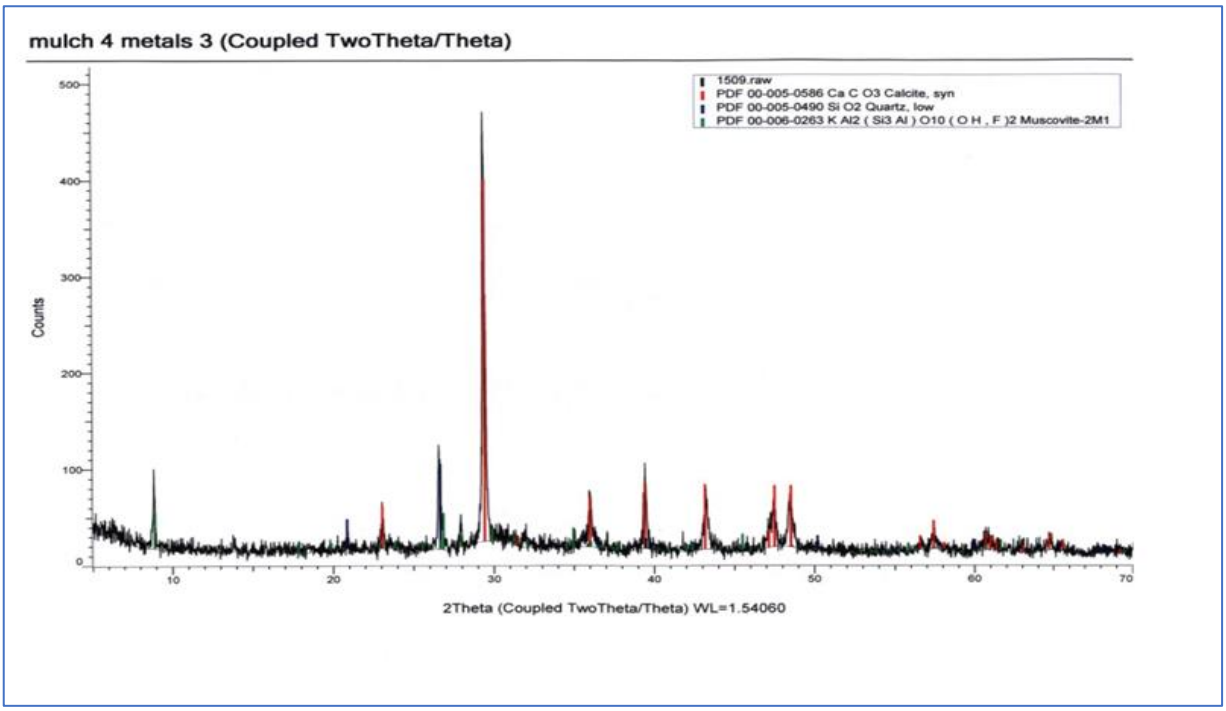
W1 = weight of ash, and

W2 = weight of the oven-dry sample

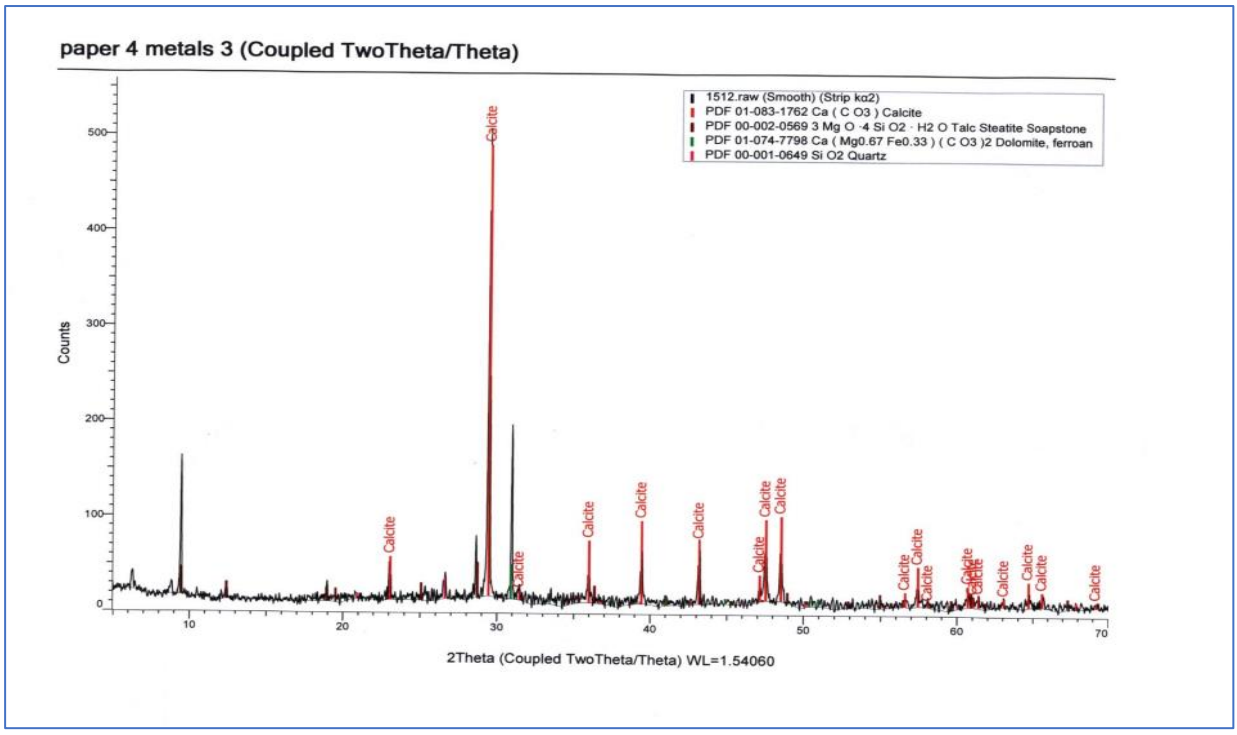
This procedure was applied in triplicate repetitions for each adsorbent, covering different initial PTEs adsorption process concentrations (0.5, 1, 5, 10, 50) mg L^{-1}.

3.5. X-Ray Diffraction (XRD) of adsorbents ash

The phase composition was studied by the X-ray diffraction (XRD) method (D4 Bruker, Germany). The obtained data confirmed that the predominant crystalline phase in the paper-contaminated ash samples was calcite $CaCO_3$, accompanied by smaller quantities of Quartz SiO_2, Talc $H_2Mg_3O_{12}Si_4$, and Dolomite $CaMg(CO_3)_2$. In the mulch-contaminated ash, the predominant crystalline phase was calcite $CaCO_3$, accompanied by smaller quantities of quartz SiO_2 and Muscovite $KAl_2(Si_3Al)O_{10}(OH,F)_2$. The phase composition of the paper and mulch ashes is given in Figure 11.



a)



b)

Figure 11. XRD pattern of (a) PTE-contaminated mulch ash (AMA),(b) PTE-contaminated paper ash (APA)

3.6. Mortar adsorbent ash mixture preparation

Ash mortar mixture preparation involved weighing the various constituents of the mixture. Mix proportions are listed in Table 7. The preparation steps are summarized as follows: Sand was added to AMA or APA ash and mixed to improve the homogenization of the mortar. Cement was added with additional dry mixing. Finally, water addition had a fixed ratio of water/binder (w/b). The specimens in Figure 12 were left for 28 days under temperature-controlled conditions in the laboratory at 20°C±2°C. The procedure also included the preparation of blank mortar samples. For each analyzed parameter, three repetitions were made to ensure the accuracy of the work.

Table 7. *Mix proportions of mortar ash specimen constituents based on weight. XP (1 % paper ash), XM (1 % Mulch ash), YP (2% paper ash), YM (2% mulch ash), ZP (3% paper ash), ZM (3% mulch ash), B0 (Blank)*

Set	Cement	Sand	w/b	Ash
PX	1	3	0.5	0.01
PY	1	3	0.5	0.02
PZ	1	3	0.5	0.03
MX	1	3	0.5	0.01
MY	1	3	0.5	0.02
MZ	1	3	0.5	0.03
BO	1	3	0.5	0

Figure 12. Mortar-incorporated adsorbed ash composites

3.7. Leaching experimental testing

Experiments for the leaching of PTEs from the APA mortar composite and AMA mortar composite were carried out in accordance with the CEN/TS 14429:2015 test method. The mortar samples were crushed and sieved through a 1 mm sieve. The HNO_3 reagent was prepared in two concentrations (4 M HNO_3 and 0.43 M HNO_3). The crushed sample was weighted, and the reagent was added at a weight/volume ratio of 1/10 in a PE bottle. The sealed PE bottles were placed on a rotary agitator. The nitric acid mixture sample was rotated at 24 rpm for four h. After mixing, the elute was filtered using an MN 640 m ashless filter paper. The removed fluid's pH was measured using the JENWAY 3510 pH meter and acidified with concentrated nitric acid to pH < 2 before testing.

3.8. Instrumental analysis

The total elemental content of PTEs leachate solutions was determined by a HORIBA Jobin Yvon ACTIVA M Inductively Coupled Plasma – Optical Emission Spectrometer (ICP-OES) using the operation parameters proposed by the manufacturer and yttrium internal standard. Table 8 indicates the ICP-OES operational parameters used to determine the following elements (with ICP-OES emission lines): Cd (228.802 nm), Cu (324.754 nm), Pb (220.353 nm), Zn (213.857 nm).

Table 8. *ICP-OES operational parameters for trace elemental analysis*

Operational parameters	Settings
Incident RF-power	1200 W
Outer gas flow (Argon)	16 L/min
Sheath gas flow (Argon)	0.3 L/min
Auxiliary gas flow (Argon) 0.6 L/min	0.6 L/min
Nebulizer gas pressure (Argon) 2.86 bar	2.86 bar
Nebulizer solution uptake (Meinhardt-cyclonic spray	0.85 mL/min

3.9. Scanning electron microscopy (SEM)

The microstructure and elemental mapping study of the mortar ash composite samples were performed using scanning electron microscopy (SEM). SEM analysis investigated the structural differences between mortar before and after incorporating (APA) and (AMA) into mortar composites. SEM images provide a better analysis of the morphology and the porosity and identify the core-shell structure of the samples. The instrument used for SEM analysis was a Hitachi S-4700 field emission scanning electron microscope combined with a Bruker (former Röntec) QX2 energy dispersive X-ray fluorescence spectrometer before analysis to enable the user to gather high-quality information from SEM. An extra step involved coating the sample with an additional thin layer (~10 nm) of a conductive material; the sputter coater used to coat the samples were gold. The elemental mapping of the studied elements (Cu, Pb, Zn, Cd, Si, Ca, Al) was carried out at 20 kV acceleration voltage and 9 µA beam current. The acquisition time of the individual samples was at least 1 hour.

3.10. Image Analysis Procedure

The elemental mapping covered the adsorbed elements (Cd, Zn, Cu, Pb) and others (Si, Ca, and Al). The working method of image analysis of the elemental maps of each of the seven elements in AMA, APA, and blank mortar composites, was carried out using ImageJ software V 1.8.0. ImageJ is one of the first tools for analyzing digital images. The National Institutes of Health produced the initial version in 1997. This program's open-source code, made accessible under a BSD-2 license, is a significant benefit. It can be used for image analysis and other purposes (Boruczkowski *et al*., 2022). Several steps were performed to conduct an in-depth analysis of the elemental maps. Each image was cropped, filtered, adjusted, and transformed to 8-bit using Image J software V 1.8.0 (Zohar & Haruzi, 2021). The analyzed elemental maps provided information on the estimated area coverage percentage (%) for the adsorbed PTEs and other major elemental content in the studied mortar composites.

3.11. Statistical analysis

To achieve the stated objectives, we employed a range of statistical methodologies for data analysis. Data collected from various experimental trials were meticulously recorded and stored within Microsoft Excel spreadsheets. First, the outlier and missing values were handled using the

appropriate techniques. The interquartile range method was used to remove the outlier values (Vinutha *et al*., 2018). The subsequent statistical analysis was carried out using the GenStat 12th edition software. In the next step, data distribution was evaluated (normal or not normal), where possible, the data was normalized otherwise, the statistical tests were implied based on the data distribution. The research approach employed was factorial design, which permits the investigation of primary and interaction effects involving two or more independent variables in relation to one or more dependent variables (Nordstokke & Colp, 2014). For comparing means among more than two variables and when dealing with a higher number of factors, we employed appropriate forms of Analysis of Variance (ANOVA), including one-way, two-way, or multifactor ANOVA (Kim, 2014). The investigated hypothesis aimed to study the factorial effects of four independent variables on the leaching concentrations of PTEs. These independent variables are:

- Initial Concentration of PTEs: Measured in milligrams per liter mg L^{-1}.
- Incorporated Ash Percentage %: This variable represents the percentage of incorporated ash in the sample.
- Molarity of HNO_3 eluant used: This variable represents the molarity of nitric acid HNO_3 used as the eluant in the leaching process, measured in moles per liter M.
- Adsorbent type: A categorical variable representing different types of adsorbents used in the leaching process. In this study, the effect of AMA and APA, when incorporated into the mortar, on the leaching concentration of PTEs was investigated.
- The dependent variable is the leaching concentration of PTEs, which is also measured in milligrams per kilogram mg kg^{-1}.

Null Hypothesis (H0): There is no significant factorial effect of the initial concentration of PTEs, the incorporated ash percentage %, the molarity of the used HNO_3, and the adsorbent type on the leaching concentrations of PTEs. In other words, the factors do not have a statistically significant impact on the leaching concentrations of PTEs.

Alternative Hypothesis (Ha): There is a significant factorial effect of at least one of the independent variables on the leaching concentrations of PTEs. In other words, the factors have a statistically significant impact on the leaching concentrations of PTEs.

To test this hypothesis, a factorial experimental design with appropriate statistical analysis (ANOVA) will be conducted to determine whether the combination of these independent variables has a statistically significant effect on the leaching concentrations of PTEs. If the p-value obtained from the analysis is less than the chosen significance level ($\alpha = 0.05$), we will reject the null hypothesis and conclude that there is a significant factorial effect on the leaching concentrations of PTEs.

4. RESULTS AND DISCUSSION

4.1. Adsorption

The study examined the adsorption capacities of potentially toxic elements (PTEs), specifically lead (Pb), copper (Cu), cadmium (Cd), and zinc (Zn), within mulch and paper substrates. These findings are graphically represented in Figure 13(a, b). The raw data for the adsorption capacity is presented in (Appendix A2 and A3). The correlation analysis reveals a strong positive correlation between the initial concentration of all studied PTEs and the corresponding adsorption capacity of both adsorbent materials, this indicates that as the initial concentration of PTEs increases, the adsorption capacity also increases, suggesting a direct relationship between these two variables in the studied system.

In mulch samples, the competitive adsorption capacities among these metal cations exhibited a slightly descending order: Pb ≥ Cu ≥ Cd > Zn, with respective capacities of 0.496, 0.495, 0.491, and 0.482 mg g^{-1}. This trend is in accordance with prior findings reported by Chirenje *et al*. (2006), which suggested that wood ash effectively immobilized these four metals. The quantities of metals retained by the wood ash also followed the Pb > Cu > Cd > Zn sequence. Similarly, modified coated wood mulches follow the same order for pb, Cu, and Zn (Soleimanifar *et al*., 2016; Sidhu *et al*., 2021). Furthermore, an intriguing consistency emerged between adsorption affinity and electronegativity, suggesting that electronegativity provided a compelling explanation for our experimental observations. Electronegativity values, as presented in Table 9, for the four PTEs are ranked in decreasing order: Pb^{2+} (2.33) > Cu^{2+} (1.90) > Cd^{2+} (1.69) > Zn^{2+} (1.65). Electronegativity plays a significant role in PTE adsorption, as metals with higher electronegativity tend to form stronger covalent bonds with oxygen atoms on the adsorbent surface. Consequently, Pb^{2+}, possessing the highest electronegativity value, exhibited the most substantial biosorption capacity, whereas Zn^{2+}, characterized by the lowest electronegativity value, displayed the least biosorption capacity, as supported by research conducted by Pham *et al*. (2021) and Han *et al*. (2022).

In contrast, when examining the adsorption capacities of these same metal cations within the paper, the competitive adsorption capacities followed a descending order of Pb > Cu > Zn > Cd, with respective capacities of 1.48, 1.26, 0.81, and 0.76 mg g^{-1}. This outcome is consistent with the study by Ding *et al*. (2018), which looked at the adsorption capacity of carbonized paper packaging boxes for aqueous Pb, Zn, and Cd. Their study revealed high aqueous Pb, Zn, and Cd sorption capacities, with Langmuir maximum sorption capacities of 458, 146, and 10.7 mg g^{-1}, respectively.

Notably, cadmium exhibited the lowest adsorption among these elements. Furthermore, it was observed that the paper showed a superior adsorption capacity compared to mulch for all the investigated PTEs. The varying affinities of mulch and paper adsorbents for PTE adsorption could be attributed to differences in the pH levels of the solutions. After the adsorption process, the average pH for mulch was measured at 4.69, while for paper, it registered at 5.58. Lower pH levels were associated with reduced adsorption of metal ions due to competition with H^+ ions for active adsorption sites (Farghali *et al*., 2013; Arshadi *et al*., 2014; Akpomie *et al*., 2015). Conversely, at higher pH levels, the presence of H^+ ions decreased on the adsorbent surface, resulting in increased adsorption of metal ions, as supported by Kushwaha *et al*. (2017). These results can also be explained by the findings of Cholico-Gonzalez *et al*. (2020) investigated the adsorption behavior of Pb(II), Cd(II), and Zn(II) onto Agave bagasse a lignocellulosic surface charge as it is affected by pH. The point of zero charge pH PZC of 4.7 indicates that at pH < 4.7, the ΔpH values (and the biosorbent surface) are positive, whereas at pH > 4.7, the surface is negative.

Table 9. *Basic physical and chemical properties of potentially toxic elements (Xiuling, 2021)*

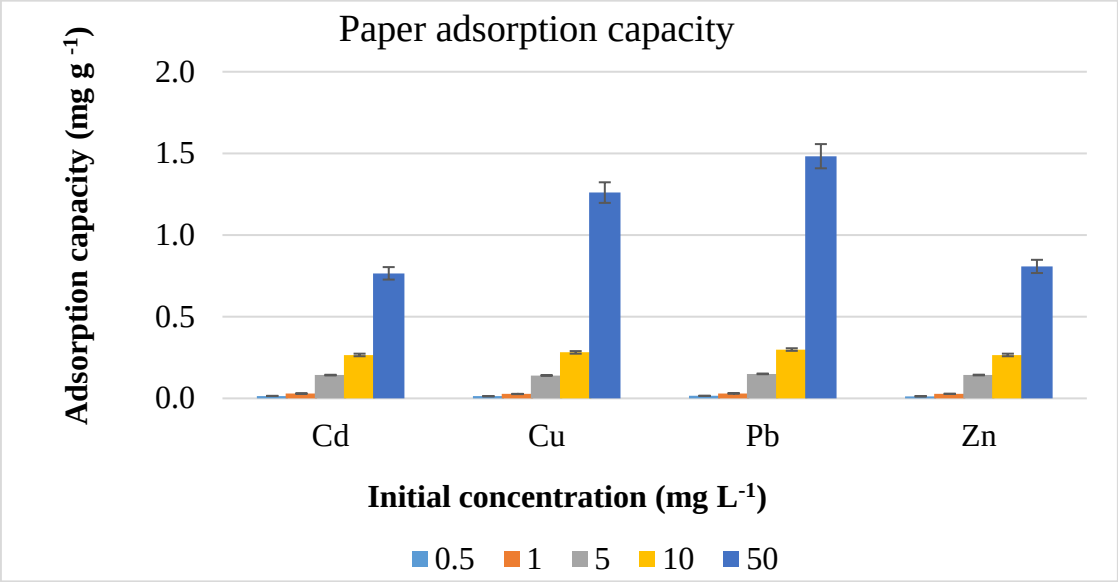
Ion species	Ion radius [pm]	Electronegativity	Hydrolysis constant	Water and ion radius [nm]	Charge-to-diameter ratio
Pb^{2+}	119	2.33	7.80	0.40	1.68
Cu^{2+}	73	1.90	7.34	0.42	2.74
Zn^{2+}	74	1.65	8.96	0.43	2.10
Cd^{2+}	95	1.69	9.20	0.43	2.70

These cheap adsorbents show good affinity towards dyes and PTEs due to cellulose in these materials, which have good adsorption potential due to O-containing and hydroxyl functional groups (Jamshaid *et al*., 2017).

In addition (Han *et al*., 2022) also discussed that these surface functional groups could efficiently coordinate with PTEs via various adsorption pathways, including surface complexation, ion exchange, and electrostatic interaction. Such observation could be explained by Pearson HSAB theory (Pearson, 1963), as Cu (II) and Pb (II) were considered harder than Cd(II) and preferred to bind to hard bases such as COOH and OH because O-containing functional groups are well-known as hard bases while PTE ions in aqueous solutions, can act as a Lewis acid (an electron acceptor) and react with the surface functional group as Lewis base (an electron donator) to form Lewis salt-

type compounds. Based on the Pearson hard-soft-acid-base (HSAB) theory, hard acids prefer to coordinate with hard bases and soft acids with soft bases. According to this theory, the Lewis acid and bases can be further divided into hard or soft or borderline types for the studied elements Ca(II), Zn(II), Al(III), Fe(III), Si(IV) ions belong to hard acids (Li *et al*., 2020); Pb(II), Cu(II), and Fe(II) ions are borderline acids (Esrafili et al., 2018) Cd(II), and Cu(I), ions are soft acids (Tang *et al*., 2018; Xia *et al*., 2017).

a)



b)

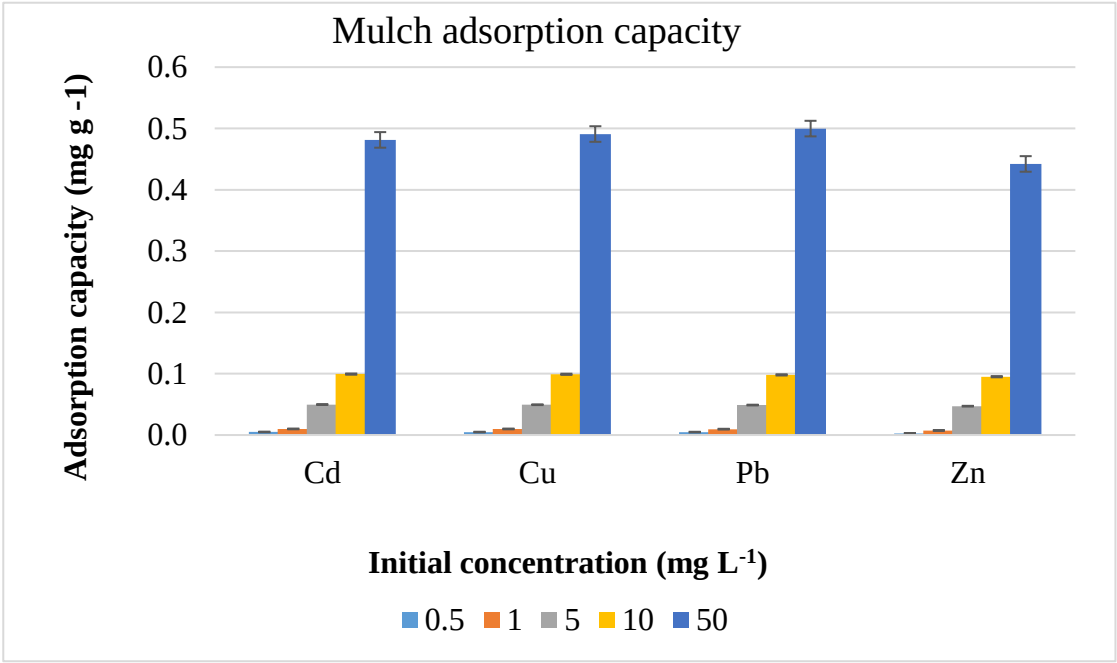


Figure 13 a) adsorption capacity of paper b) adsorption capacity of mulch

4.2. Moisture content%

Moisture is a fundamental attribute of wood, albeit not an inherent one, yet it holds significant importance due to its influential role in the behavior of this material during various phases of industrial processing and application, as observed by Tsuchikawa and Schwanninger in 2013. Industries reliant on wood chips as a primary raw material consider moisture content as a pivotal quality parameter. Beyond ensuring the final product's quality, it also minimizes losses and reduces reagent expenses, as Fardim *et al*. indicated in 2005. This importance extends to pulp and paper industries, where precise assessments of moisture content in wood chips aid in defining cooking parameters, optimizing industrial processes, and efficiently managing raw materials and reagent consumption, as highlighted in the research by Amaral *et al*. in 2020.

In the context of our investigation, we have quantified the increase in water content resulting from the adsorption process of Potentially Toxic Elements (PTEs); The calculations relied on wet-based moisture content. The moisture content of mulch and paper adsorbent samples is indicated in Tables 10 (a) and 10 (b) respectively. Specifically, the moisture content within Mulch adsorbents exhibited a range between 78.3 ± 0.27 (%) and 81.6 ± 0.21 (%), while paper-based adsorbents demonstrated moisture levels ranging from 51.3 ± 0.28 (%) to 56.4 ± 0.03 (%). These findings align with the observations made by Chen *et al.*(2014), where the moisture content of paper pulp ranged from 50% to 66.7% upon saturation with water adsorption. Additionally, a study conducted by Khazaei in 2008 exploring various wood sample species, indicated that the moisture reached an excess of 60% for most wood species.

4.3. Ash percentage%

Ashing is an essential initial step in the proximate or specific mineral analysis process, involving the determination of inorganic mineral residues following combustion. To ensure the integrity of this analysis, plant materials are typically subjected to oven-drying before grinding and ashing, with the primary aim being the removal of moisture, as elucidated by Harris and Marshall in 2017. This study investigates the ash content percentages of mulch and paper adsorbents and their characteristics. The results indicate variations in ash content, particularly in paper adsorbents, influenced by incineration temperature. The findings emphasize the importance of controlling incineration temperature and selecting appropriate feedstock to achieve desired ash yields in paper ash production. Additionally, the study discusses the significance of Loss on Ignition (LOI) in fly ash and its effects on concrete properties.

Table 10 (a) presents the ash content percentages of mulch, while table 10 (b) presents the ash content percentages of paper adsorbent samples. Mulch adsorbents exhibited ash percentages ranging from 6.10 ± 0.33 (%) to 7.88 ± 0.99 (%). These findings align with previous studies, indicating a wood ash percentage of 6.2% at 550°C (Steenari *et al*., 1999b; Al-Mefarrej *et al*., 2013).

Paper adsorbent ash content ranged from 13.8 ± 0.26 (%) to 14.8 ± 0.25 (%). Notably, Wielgosiński et al. (2021) reported an average ash percentage of 12% for paper/cardboard, attributing the higher percentage in this study to incineration temperature variations. Hui et al. (2020) demonstrated that increasing incineration temperature resulted in decreased ash content in paper sludge, affecting particle characteristics and chemical composition.

Mohebbi *et al*. (2015). observed that Loss on ignition results may overestimate the amount of organic carbon as the ignition mass loss is not only due to burning of organic carbon, but also due to other possible reactions such as calcination of inorganic carbonates, desorption of physically and chemically bound water (e.g., dehydration of portlandite), and oxidation of sulfur and iron minerals.

The LOI of fly ash has significant implications for concrete properties. Ngo *et al*. (2018) emphasized the increasing trend in specifying lower LOI limits (ranging from 3% to 6%) in many countries for quality assurance. However, it was noted that high LOI (12%) fly ash can enhance concrete strength due to carbon particles acting as air-reducing agents.

In the study by Ngo *et al*. (2018), the ash used had an LOI of 15.8%, exceeding the ASTM C618 limitation of 6%. Eight concrete mixtures with varying fly ash replacements were designed. Results showed improved workability and reduced unit weight with increasing fly ash content. Concrete mixtures with 10% and 20% fly ash exhibited higher compressive strength, especially in the long term, emphasizing the potential benefits of higher LOI fly ash. Despite exceeding traditional LOI limits, the study highlighted the significance of LOI in ash and its potential benefits in concrete applications.

Table 10. *a) Moisture content and ash percentage for mulch (M) adsorbents, b) Moisture content and ash percentage for paper(P) adsorbents*

Table 10*.a)*

Mulch sample No	Moisture content %	Ash percentage %	Standard deviation of moisture content	Standard deviation of ash percentage	Average moisture content %	Average ash percentage %
M1 (0.5 mg L^{-1})	81.9	6.35	0.21	0.33	81.6	6.10
M2(0.5mg L^{-1})	81.5	6.23				
M3(0.5 mg L^{-1})	81.5	5.73				
M1 (1mg L^{-1})	80.4	7.91	0.13	0.99	80.5	7.88
M2(1mg L^{-1})	80.6	6.87				
M3(1mg L^{-1})	80.5	8.86				
M1 (5mg L^{-1})	78.7	6.37	0.40	0.18	79.1	6.30
M2(5mg L^{-1})	79.5	6.44				
M3(5mg L^{-1})	79.1	6.10				
M1 (10mg L^{-1})	80.6	6.45	0.05	0.51	80.5	6.45
M2(10mg L^{-1})	80.6	6.96				
M3(10mg L^{-1})	80.5	5.93				
M1 (50mg L^{-1})	78.3	7.04	0.27	0.41	78.3	6.85
M2(50mg L^{-1})	78.0	6.38				
M3(50mg L^{-1})	78.6	7.13				

Table 10. *b)*

Paper Sample No	Moisture content %	Ash percentage %	Standard deviation of moisture content	Standard deviation of ash percentage	Average moisture content %	Average ash percentage %
P1 (0.5mg L^{-1})	54.0	14.1	0.11	0.26	54.1	13.8
P2(0.5mg L^{-1})	54.3	13.9				
P3(0.5mg L^{-1})	54.2	13.5				
P1 (1mg L^{-1})	56.1	15.2	0.22	0.49	56.3	14.7
P2(1mg L^{-1})	56.5	14.3				
P3(1mg L^{-1})	56.4	14.5				
P1 (5mg L^{-1})	51.0	15.0	0.28	0.25	51.3	14.8
P2(5mg L^{-1})	51.3	14.9				
P3(5mg L^{-1})	51.5	14.5				
P1 (10mg L^{-1})	54.3	14.5	0.25	0.34	54.5	14.4
P2(10mg L^{-1})	54.8	14.8				
P3(10mg L^{-1})	54.5	14.1				
P1 (50mg L^{-1})	56.5	14.0	0.03	0.21	56.4	14.0
P2(50mg L^{-1})	56.4	13.8				
P3(50mg L^{-1})	56.4	14.2				

4.4. Leaching of PTEs

Leaching test assessment on the crushed APA and AMA mortar samples was done by comparing the values of the tested samples with the leaching value limits for the waste categories according to EU directive 1999/31/EC shown in Table 6). With reference to Figure 14, concentrations of Zn leached from the tested crushed mortar samples at the 0.43M HNO_3 trial come under the inert waste category.

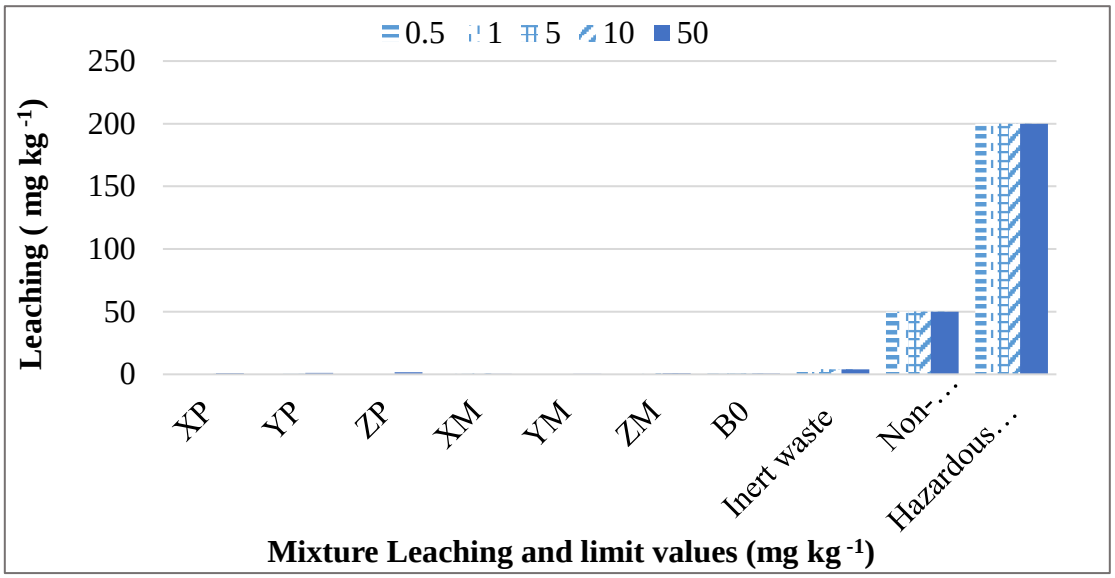


Figure 14. Zn leaching concentrations at 0.43M HNO_3 compared to limit values for the individual waste categories. XP (1 % paper ash), XM (1 % Mulch ash), YP (2% paper ash), YM (2% mulch ash), ZP (3% paper ash), ZM (3% mulch ash), B0 (Blank) mortar composites. five initial concentrations of PTEs (0.5, 1, 5, 10, 50) mg kg $^{-1}$.

The leaching results of Cu at 0.43M HNO_3 illustrated in Figure 15 also lie under the inert waste category in all composites.

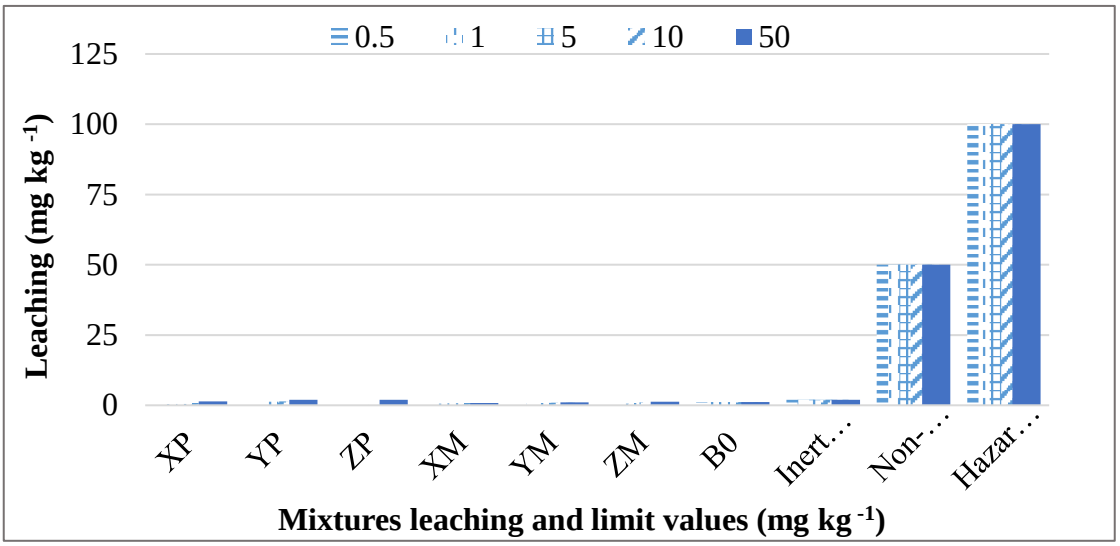


Figure 15. Cu leaching concentrations at 0.43M HNO_3 compared to limit values for the individual waste categories.

Pb concentration is slightly higher than that of the inert waste limit category in the case of APA mortar samples but inert in the AMA mortar samples according to Figure 16. The explanation for the different Pb leaching values is apparent when comparing the results of the Pb adsorption capacity in the mulch samples, which was 0.496 mg g^{-1} at 50 mg L^{-1} initial concentration of adsorption solution. In contrast, in the paper samples, the adsorption capacity reached 1.483 mg g^{-1} at the 50 mg L^{-1} initial concentration of Pb, in which paper showed more affinity to adsorb Pb.

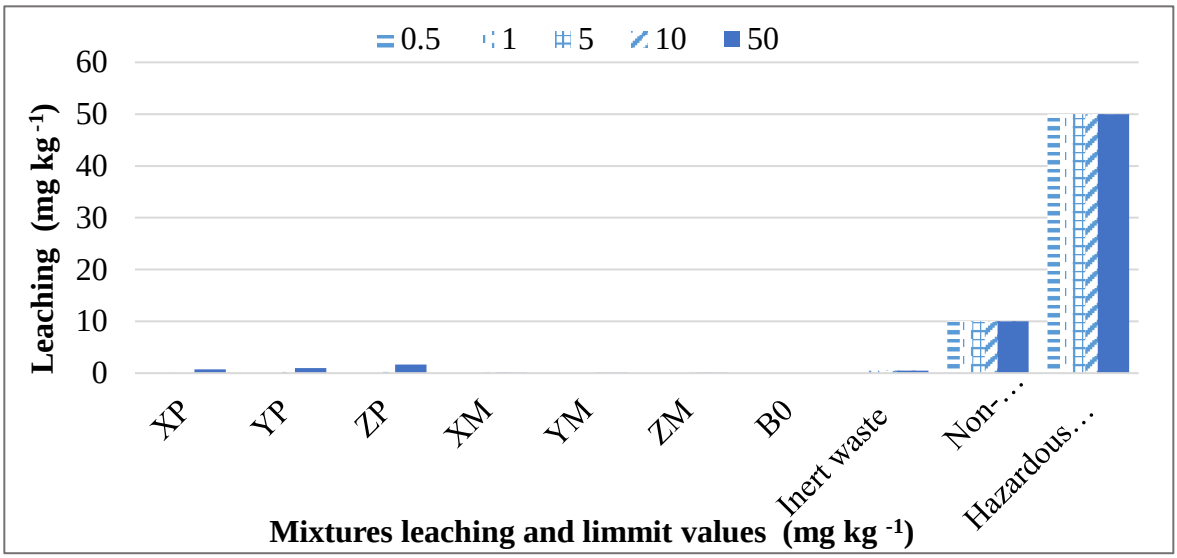


Figure 16. Pb leaching concentrations at 0.43M HNO_3 compared to limit values for the individual waste categories.

The Cd leached concentration at 0.43 M HNO_3 in Figure 17 was lower than the non-hazardous waste limit value.

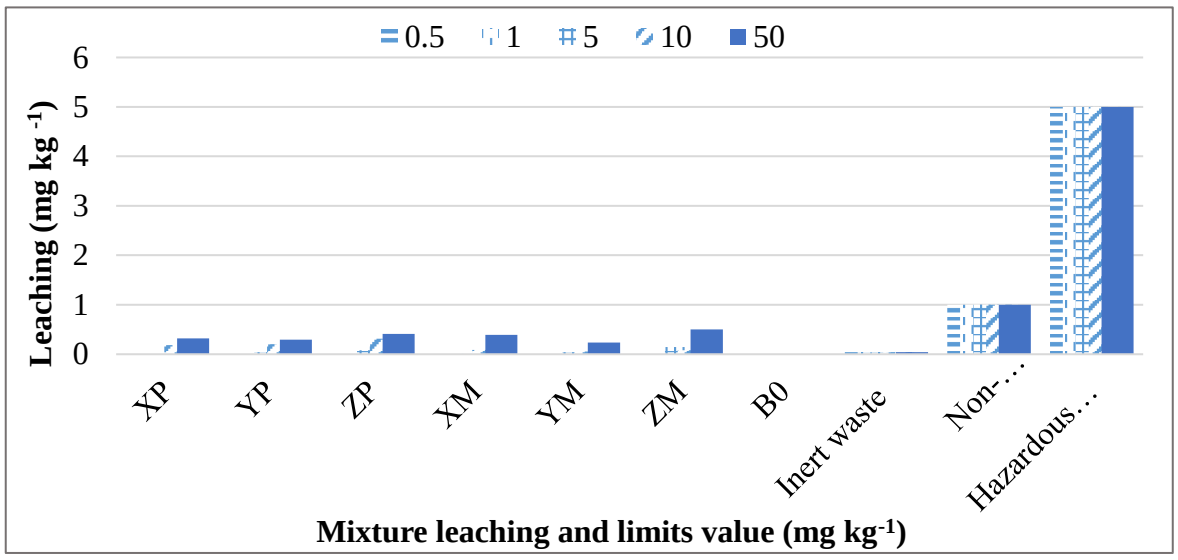


Figure 17. Cd leaching concentrations at 0.43M HNO_3 compared to limit values for the individual waste categories

Referring to Figure 18, the concentrations of Zn leached from the tested crushed mortar samples at the 4 M HNO_3 trial were within the safe limits under the non-hazardous waste category with higher leaching concentrations in the case of AMA mortar composites compared to APA leaching concentrations.

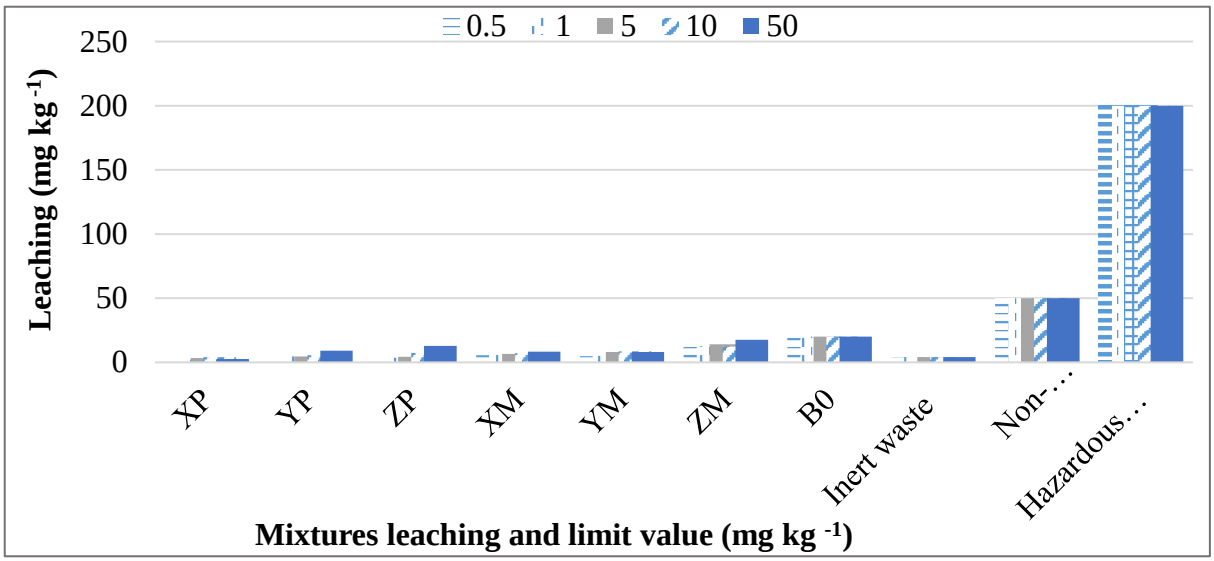


Figure 18. Zn leaching concentrations at 4 M HNO_3 compared to limit values for the individual waste categories.

The concentrations of Cu leached from the tested crushed mortar samples at the 4 M HNO_3 trial illustrated in (Figure 19), the results were within the safe limits under the non-hazardous waste category with higher leaching concentrations in the case of APA mortar composites compared to AMA leaching concentrations.

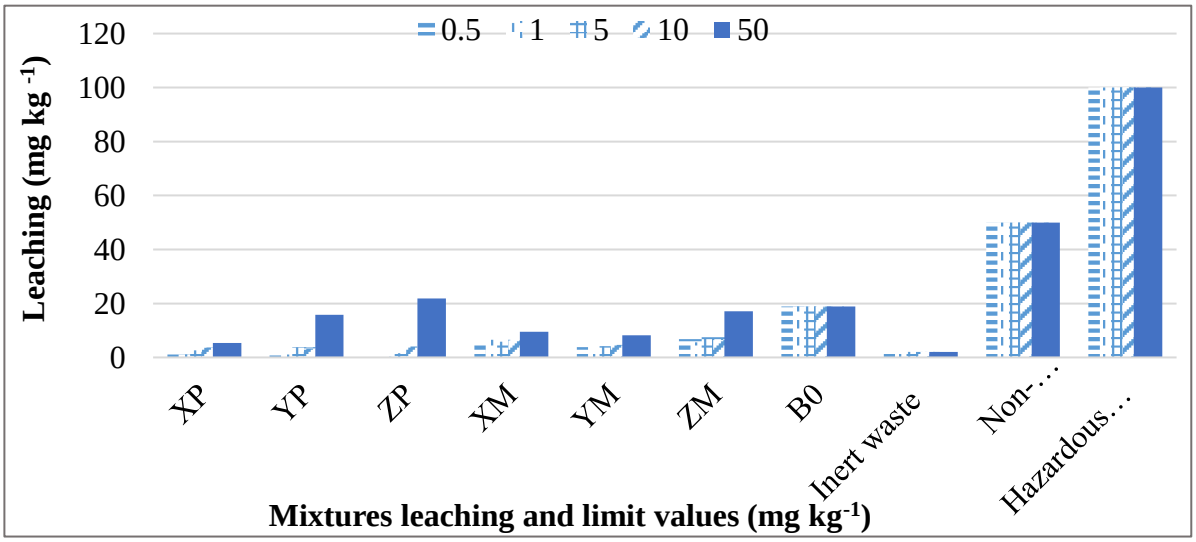


Figure 19. Cu leaching concentrations at 4 M HNO_3 compared to limit values for the individual waste categories.

Pb leached from the tested crushed mortar samples at the 4 M HNO_3 trial were within the safe limits under the non-hazardous waste category with higher leaching concentrations from APA mortar composites compared to AMA mortar composites (Figure 20).

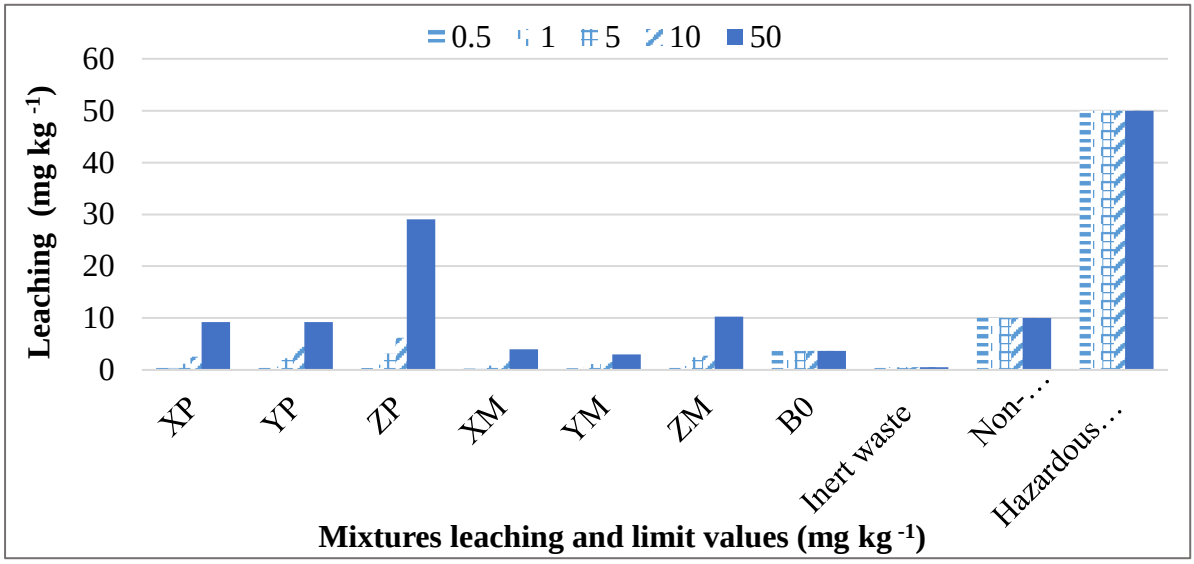


Figure 20. Pb leaching concentrations at 4 M HNO_3 compared to limit values for the individual waste categories.

Only Cd leached above the regulatory limit (5 mg kg^{-1}) (Figure 21). This result corresponds to the findings of Yuan (2018); the author used the tank test, which showed that long-term exposure to cement concrete containing municipal solid waste incineration fly ash in a water environment might lead to Cd pollution. However, the Cd leached concentration was lower than the regulatory limit in the study conducted by Carević et al. (2020), in which crushed mortar samples of wood ash were incorporated into the cement mortar; this difference can be explained by the low Cd concentration in the wood ash used in their experiment.

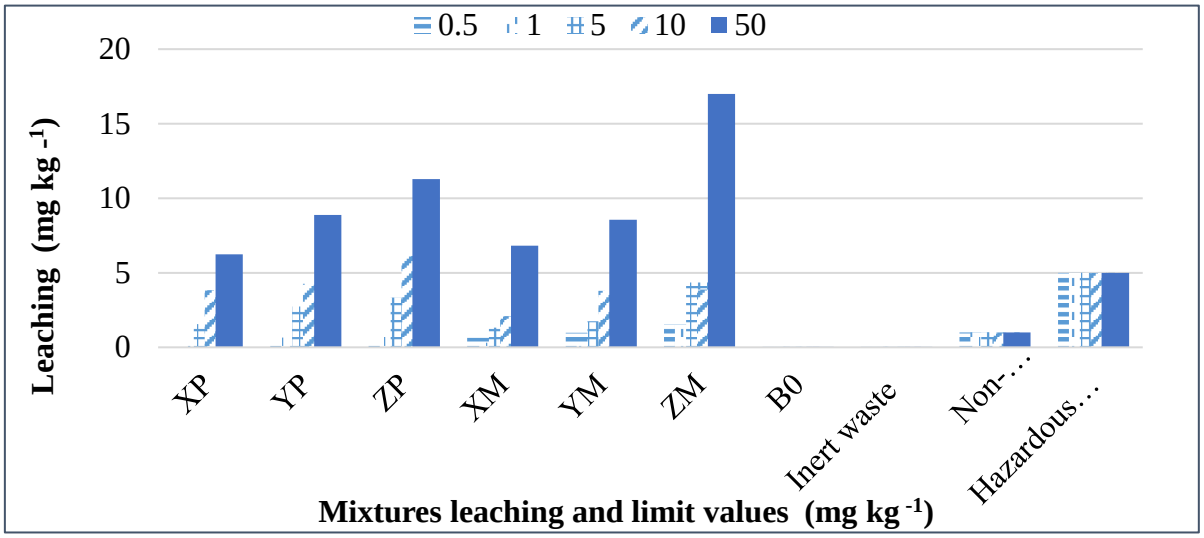


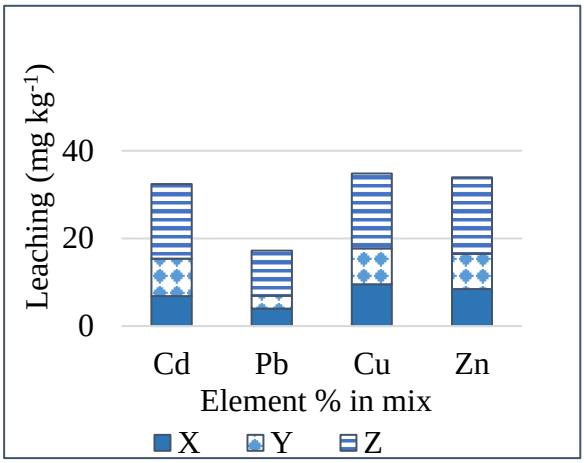
Figure 21. Cd leaching concentrations at 4 M HNO_3 compared to limit values for the individual waste categories.

4.5. The pH effect on PTEs leaching

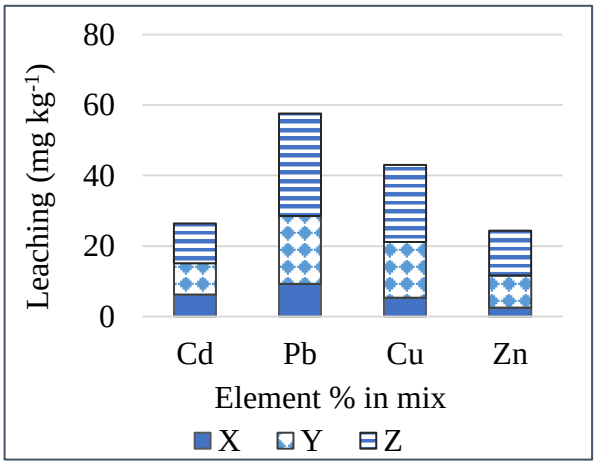
The amount of PTEs released from its binders is strongly pH-dependent on the surrounding environment. (Tiwari *et al*., 2015; Berra *et al*., 2019; Masud *et al*., 2021; van der Sloot and van Zomeren, 2012; Król, 2011; Dijkstra *et al*., 2004; van der Sloot and Kosson, 2010; Saveyn *et al*., 2014).

Many leaching tests study the effect of pH on the leaching of PTEs. (Xu *et al*., 2019) compared the leaching tests commonly used in China: HJ/T 299-2007and HJ/T 300-2007, with leaching tests widely used in the world: 1311 TCLP and 1312 Synthetic Precipitation Leaching Procedure of USEPA SW-846, and CEN/TS 14429 Leaching behavior tests. They concluded that the CEN/TS14429 test method is more proper for examining the PTEs leaching behavior of the test materials or products under diverse environmental pH conditions when evaluating recycled raw materials or products manufactured from hazardous industrial wastes.

In this study, two trial tests with nitric acid concentrations of 0.43 M and 4 M were conducted on the crushed samples with a final leachate average pH of - 0.25 for (AMA) mortar - 0.07 for (APA)mortar samples at the 4M HNO_3 trial, and pH of 4.69 for (AMA) mortar 5.58 for (APA) mortar samples at 0.43 M HNO_3 trial. The results show that (AMA) mortar is more acidic than (APA) mortar, and the pH was lower than the simulated acid rains with pH of 3.0 and 2.5 (Kanazu *et al*., 2001). For (AMA) mortar mix samples, Figure 22(a-c) and (APA) Figure 22(b-d) show that the leached concentration of Pb, Cd, Cu, and Zn increased as the pH values of the solution decreased, which corresponds with (Udoeyo *et al*., 2006) findings. At the 0.43 M HNO_3 leaching test of (APA) samples at (Z=3%) of added ash , the highest leaching amount of Cu, Zn, Cd, and Pb was 1.96 mg kg^{-1}, 1.81 mg kg^{-1}, 0.41 mg kg^{-1}, and 1.69 mg kg^{-1} respectively, while at 4M leaching test the highest leaching amount of Cu, Zn, Cd, and Pb was 21.87 mg kg^{-1}, 12.72 mg kg^{-1}, 11.29 mg kg^{-1}, and 29.07 mg kg^{-1} respectively for (Z= 3%) of added ash samples. Based on the findings of this study, future research involving more extensive analysis will aid in ensuring better use of this waste material; the future research work suggested must include the Study of leaching of non-crushed mortar samples for long-term exposure. Further studies should consider the mechanical strength and the durability of adsorbed paper ash mortar composite and the adsorbed mulch ash mortar composite to ensure that PTEs do not affect the properties of the mortar.



(a)



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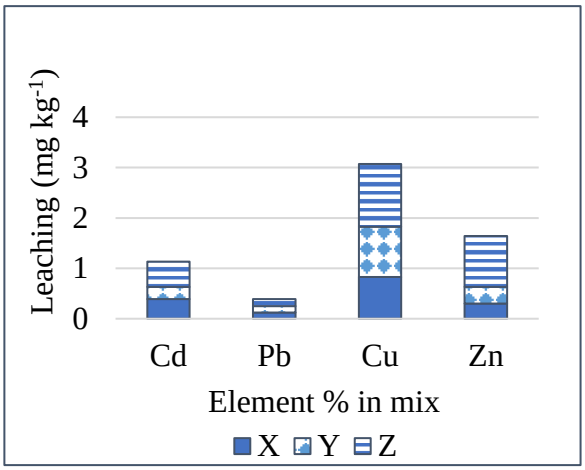
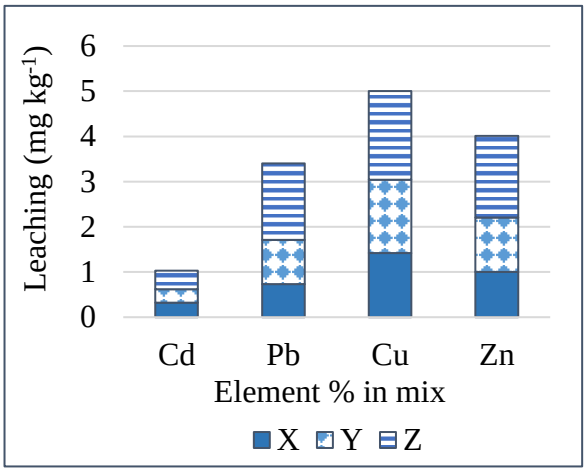


(c)



(d)

Figure 22. (a) maximum leaching in mulch ash concrete mix (4M HNO_3), (b) maximum leaching in paper ash concrete mix (4M HNO_3), (c) maximum leaching in mulch 0.43M HNO_3, (d) maximum leaching in paper 0.43 M HNO_3.All results are based on average values of 3 replicates of 50 mgL^{-1} initial concentration of PTEs.

4.6. Adsorbent type combined effect on PTEs leaching and adsorption

Ash's elemental composition and structural characteristics can vary significantly depending on its source. When incorporating ash into mortars and concrete, it becomes challenging to predict how using a specific type of ash will impact the concrete's durability, as pointed out by Fava *et al.* (2018). Consequently, it is essential to conduct assessments on the final product using leaching tests. Hence, any new material must undergo a thorough technical and environmental analysis before it can be deemed suitable for practical applications.

The leaching results from the AMA mortar composite followed the order Cu >Zn > Cd>Pb, with respective values of 1.25, 1.01, 0.50, and 0.14 mg kg^{-1} at 0.43 M HNO_3 shown in Figure 22 (c) at (Z =3%) mulch ash composite. This observation could be attributed inversely to the ionic radius of PTEs, as indicated in Table 9. While APA leaching test results followed the order Cu >Zn>Pb> Cd with leaching amounts of 1.96, 1.81, 1.69, and 0.41 mg kg^{-1}, respectively shown in Figure 22 (d) at (Z=3%) of paper ash composite. Notably, cadmium was the metal with the least leached from APA mortar composites. These results highlight how adsorption affinity also played a pivotal role in determining the resulting leachate concentrations of PTEs. For instance, cadmium exhibited consistently low adsorption capacity when paper was used as the adsorbent.

Additionally, pH, as noted by Hoang *et al.* (2022), is another influencing factor in the adsorption and leaching behavior of lignocellulosic biomass-activated carbon. After the leaching process with 0.43 M HNO_3, the measured pH values were 5.25 for (AMA) mortar and 7.22 for (APA) mortar at 50 mg L^{-1} initial concentration of PTE and 3% of ash content added in mortar. These results align with the idea that variations in pH contribute to the differing affinities for PTE adsorption, with (AMA) mortar being more acidic than (APA) mortar, which aligns with their respective adsorption preferences due to the pivotal role of acidity in this phenomenon. In this study the mulch and paper wastes were used as adsorbents showing different affinities to PTEs adsorption, which also influences the resultant APA and AMA mortar composites leachate concentration of PTEs.

4.7. PTEs immobilization efficiency of incorporated adsorbent ash

The immobilization efficiency (%) in APA mortar is higher than in AMA mortar, according to Table 11. The possible immobilization mechanisms of PTEs in concrete and mortar could be (1) sorption, (2) chemical incorporation (surface complexation, precipitation, co-precipitation, diodochy), and (3) micro-encapsulation or macro-encapsulation (Trussell and Spence 1994; Glasser, 1997). With reference to Figure 22 (a-b), PTEs show different leaching behaviors at 4M HNO_3 ; for example, Cd and Zn are more leached in the AMA mortar mix samples. Pb and Cu are more leached in the APA mortar mix. Furthermore, as the weight proportion of the contaminated ash added to the mortar increases, so does the content of PETs in the leachate. This result holds true for the AMA and APA mortar composites, as evidenced by the weight percentage addition of X =1%, Y =2%, and Z = 3% ash to the mortar. The leaching amount of the four PTEs at Z=3% of ash from the AMA mortar samples at 0.43 M HNO_3 follows the order Cu > Zn > Cd > Pb (Figure 22-c), and at 4 M HNO_3, the leaching order is Zn≈ Cd≈ Cu > Pb (Figure 22-a).

The order of leaching in the APA mortar samples at (Z=3% of ash 0.43 M HNO3) is Cu > Zn >Pb > Cd (Figure 22-d); at 4 M HNO_3, the order is Pb > Cu > Zn≈ Cd (Figure 22-b). These leaching results show that incorporating the APA and AMA into the mortar matrix confirms that the PTEs (Pb, Cu, Zn, and Cd) are successfully immobilized within the mortar matrix. Based on the obtained values, environmental safety can be ensured.

***Table 11**. Immobilization efficiency % of PTEs encapsulated within the mortar. SD standard deviation.*

Element	AMA (Z)%	SD	APA (Z)%	SD
Cd	96.1	0.002	97.7	0.002
Pb	97.9	0.001	97.7	0.001
Cu	93.6	0.002	98.6	0.002
Zn	87.7	0.001	97.7	0.001

4.8. Microstructural characterization of incorporated adsorbent ash into the mortar

Scanning Electron Microscopy (SEM) has emerged as a powerful tool for the characterization and analysis of materials, particularly in the field of construction materials. In the context of mortar composites, SEM analysis allows for the investigation of microstructure features, providing valuable insights into the arrangement and distribution of different components within the composite material.

The SEM examination in this study revealed differences in the microstructure of APA, AMA, and blank samples, referring to Figure 23 of the SEM images. The results showed that, Unlike APA mortar, blank mortar has the most microcracks and porosity, while AMA mortar has the least. The microstructure of AMA mortar is significantly denser than that of APA and blank mortar. According to SEM examination, the inclusion of AMA and APA enhanced the microstructure of the mortar due to its micro-filling capabilities. The hydration phase was aluminate-ferrite, monosubstituted (AFm), a well-developed yet very thin hexagonal plate, indicating late formation for all composites. Another apparent color difference appeared in SEM images where both the adsorbed paper ash and adsorbed mulch ash, when incorporated in the mortar, led to a darker color compared to blank mortar this finding is consistent with the results of (Lessard *et al*., 2017) when using fly and bottom ash of paper waste in a concrete composite.

The results of this study suggest that further studies should examine the mechanical strength and durability of the PTEs adsorbed paper ash mortar composite and the adsorbed mulch ash mortar composites to ensure that PTEs do not affect these properties. It is also recommended to investigate the usage of AMA and APA in other applications in civil engineering, including use in bricks, panels, geopolymers, and other alkali-activated materials, concrete, and other green building materials. Finally, a long-term study for aged samples of APA and AMA mortar composites is recommended to see the effect of age on microstructure; further research involving more extensive analysis will help to ensure better use of this waste material.

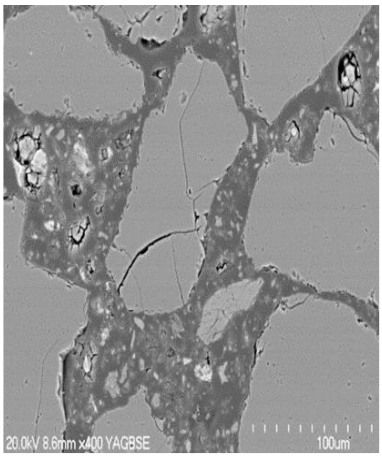
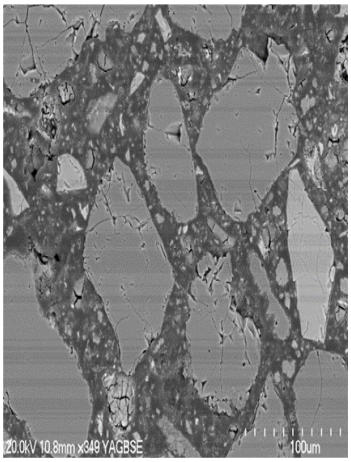
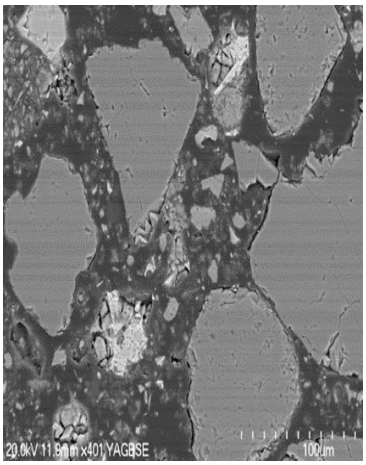
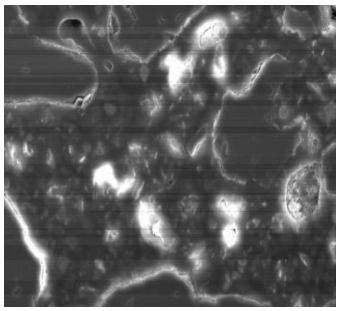
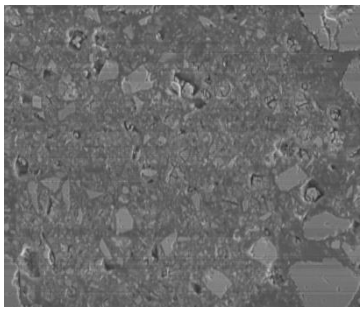
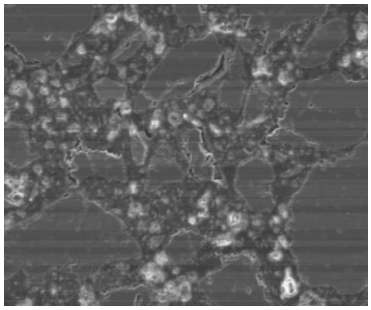
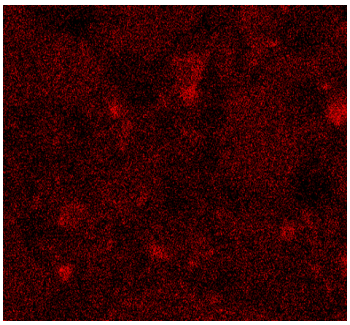
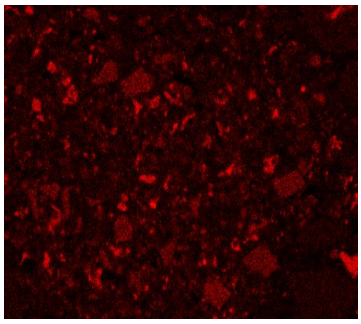
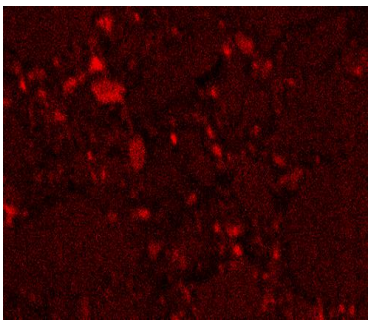
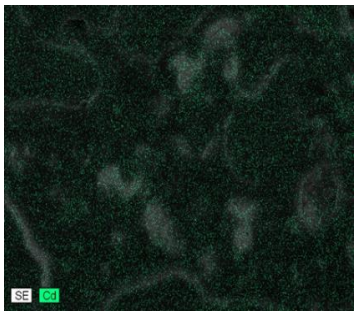
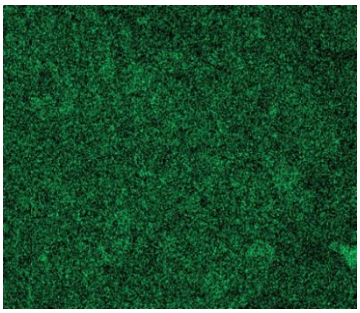
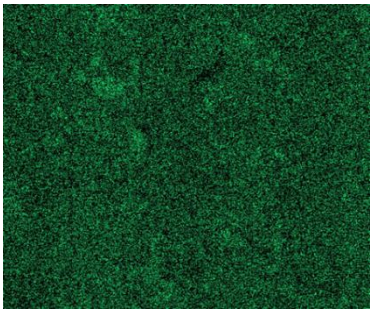
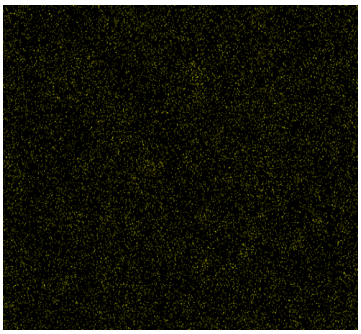
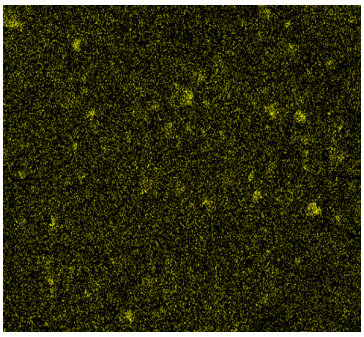
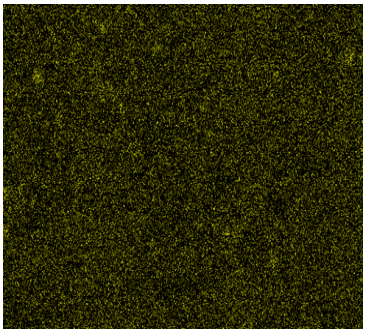
Blank mortar microstructure	APA mortar microstructure	AMA mortar microstructure
		

Figure 23. The microstructure of Blank, APA, and AMA mortar composites

4.9. Elemental mapping

Research findings on elemental mapping are presented in Figure 24. The elemental distribution of Al, Cd, Zn, Pb, Cu, Si, and Ca in the AMA, APA, and blank mortar composites was examined using SEM-EDS. The analysis revealed that the adsorbed potentially toxic elements - Cd, Zn, Cu, and Pb- exhibited a uniform distribution throughout the APA and AMA mortar matrix structure. However, their distribution intensity was higher than the blank mortar sample, indicating evidence of PTE immobilization.

Several steps were performed to conduct an in-depth analysis of the elemental maps. Each image was cropped, filtered, adjusted, and transformed to 8-bit using Image J software V 1.8.0 (Zohar & Haruzi, 2021).

Element	Blank mortar	Mulch mortar mix	paper mortar mix
BSE			
Al-K			
Cd			
Zn			

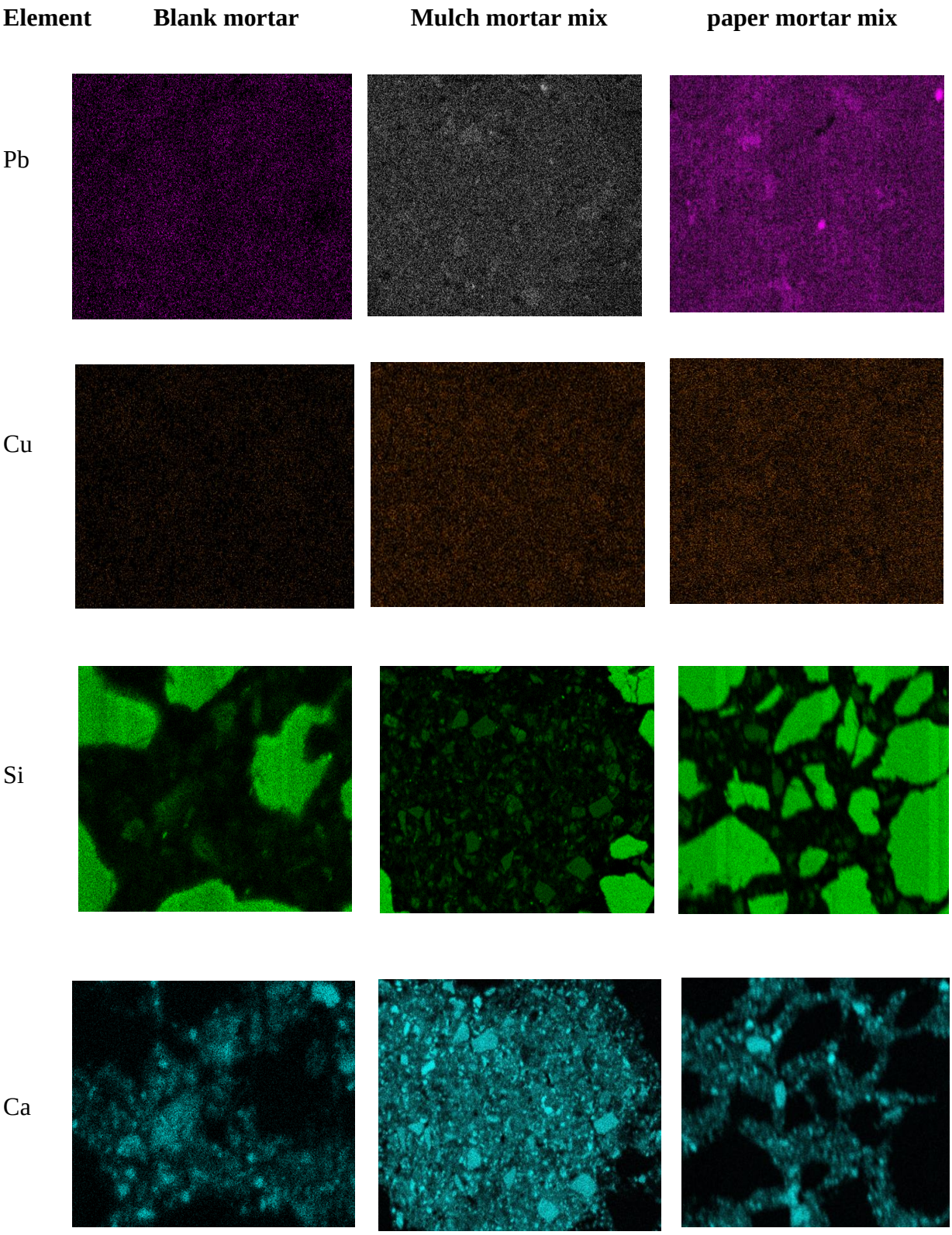


Figure 24. Elemental mapping of APA, AMA, and blank mortar samples

The analyzed elemental maps provided information on the estimated area coverage percentage (%) for the adsorbed PTEs and other major elemental content in the studied mortar composites. Figure 25 displays the estimated percentage of the area coverage for each element of the adsorbed PTEs within AMA, APA, and blank mortar composites. The percentage of area coverage of PTEs within AMA mortar composites followed the following order: Pb >Cd>Zn>Cu, accounting for 32.1, 28.6, 13.8, and 12.4%, respectively. This order was also followed by the blank mortar composite, accounting for 12.5, 8.6, 4.5, and 4.2%, respectively.

In the APA mortar composite, the percentage of area coverage followed the following order: Cd>Pb>Zn>Cu, accounting for 27.7%, 26.6%, 14.5%, and 14.1%, respectively. The highest percentage of area coverage was observed for Pb, accounting for 32.1%, followed by Cd at 28.6% within the AMA mortar composite. In contrast, the APA mortar composite exhibited slightly lower percentages of area coverage for Pb and Cd, with values of 26.6% and 27.7%, respectively. The Cd behavior in APA mortar composites could be attributed to the results of pH, which shows that APA composites tend to shift towards a more basic environment than AMA composites. This behavior aligns with the findings of Singh *et al*. (2023), which demonstrate that Cd has a higher adsorption affinity than Pb when the media is basic. In all the tested mortar composites, the area coverage percentage for Zn was consistently observed to be slightly higher than that for Cu.

An inverse correlation between the estimated percentage of area coverage for adsorbed PTEs and the leaching results from the mortar composites was observed, suggesting valuable insights into the immobilization mechanisms for these PTEs. Specifically, the immobilization mechanisms for Pb, Cd, and Zn in cementitious materials can be explained as follows:

Lead (Pb): The predominant immobilization mechanism for Pb in cementitious materials involves precipitation as insoluble lead silicates (e.g., $PbSiO_3$, Pb_2SiO_4, Pb_3SiO_5, and Pb_4SiO_6) (Grubb *et al*.,2009; Liu *et al*., 2023). Cadmium (Cd): In cement hydration systems, Cd is primarily immobilized through the precipitation of hydroxides. These insoluble phases are subsequently adsorbed onto the C-S-H surface or fill the pore structure of cement pastes (Wang *et al*., 2018; Wang *et al*., 2022). Zinc (Zn): precipitation as hydroxides or carbonates may be the major mechanism for immobilizing Zn in solidification/stabilization systems (Liu *et al*., 2020). Zinc, being amphoteric, is soluble in a highly alkaline environment provided by cement hydration and exists as hydroxyl complexes ($[Zn(OH)_4]^{2-}$ and $[Zn(OH)_3]^-$). These hydroxyl complexes are less prone to adsorption onto the surface of C-S-H due to their negative charge (Liu *et al*., 2023). Zn immobilized less than Pb and Cd because of its mechanism of immobilization. It is an amphoteric metal that is soluble in highly alkaline environments and hardly adsorbs onto C-S-H, while Pb and

Cd precipitate as hydroxides, filling the pore structure of cement pastes. This comprehensive understanding of the immobilization mechanisms helps explain the observed order of PTEs and provides insights into how these mechanisms influence leaching behavior in mortar composites.

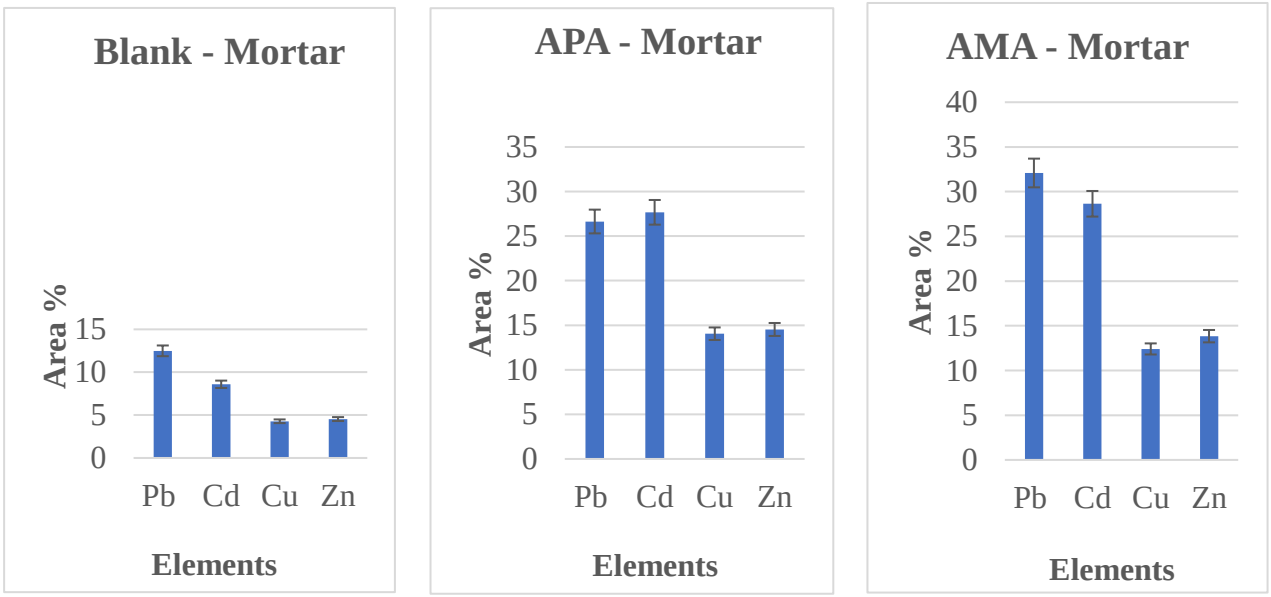


Figure 25. Area coverage percentage (%) for adsorbed elements estimated using ImageJ software V 1.8.0 for AMA, APA, and blank mortar composites.

The percentage of area coverage of the major elements within AMA, APA, and blank mortar composites compared are presented in Figure 26. The results showed that the percentage coverage of the major elements within the AMA mortar composite followed the following descending order: Pb>Ca>Cd>Al>Fe>Si>Zn>Cu.

In contrast, the order in APA mortar composite was Si>Pb>Cd>Al>Ca>Fe>Zn>Cu. Silicon in APA mortar samples has the highest area coverage percentage because calcium silicate is usually used as paper filler to improve the paper bulk, one of the most critical properties in papermaking (Qiu *et al*., 2020). In AMA mortar composite, the Calcium area coverage percentage was higher than APA mortar composite; this result is consistent with the findings of the researchers Zając *et al*. (2018), who studied the chemical composition of oak mulch ash and other wood ashes and found that calcium was the element with the highest elemental content in these ashes.

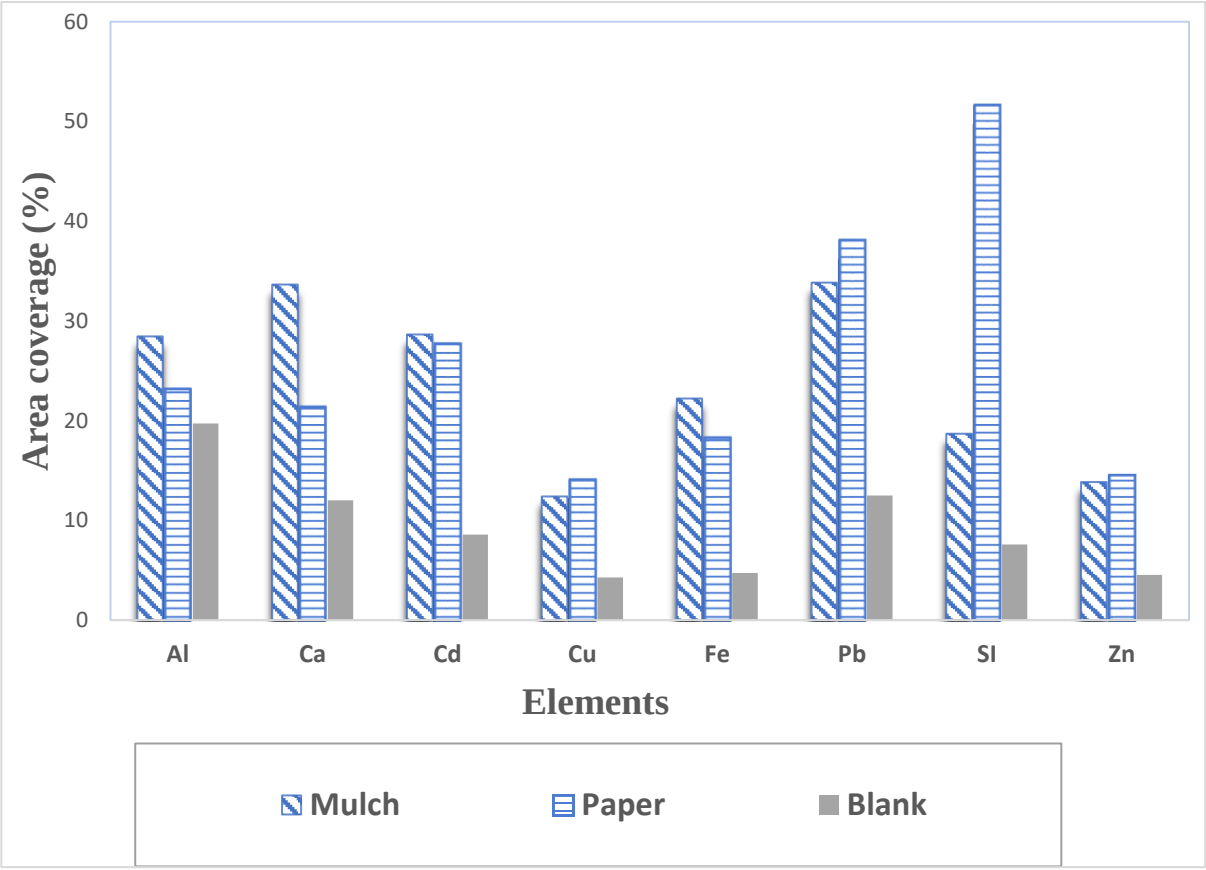


Figure 26. Compared Area coverage percentage (%) for major elements estimated using ImageJ software V 1.8.0

4.10. Statistical analysis

This study investigated the effect of incorporating AMA and APA into the mortar on the leaching concentration of PTEs. The dependent variable is the leaching concentration of PTEs, also measured in mg kg^{-1}. Tables presenting the statistical data results are provided in Appendix 4. The research hypotheses for this study are as follows:

Null Hypothesis (H0): There is no significant factorial effect of the initial concentration of PTEs, the incorporated ash percentage %, the molarity of the used HNO_3, and the adsorbent type on the leaching concentrations of PTEs.

Alternative Hypothesis (Ha): There is a significant factorial effect of at least one of the independent variables on the leaching concentrations of PTEs. In other words, the factors have a statistically significant impact on the leaching concentrations of PTEs.

4.10.1. Molarity factor

This study presents an analysis of the leached concentrations of four potentially toxic elements (Cadmium, Copper, Lead, and Zinc) across two different molarity conditions—0.43 M and 4 M. The primary goal of this analysis is to determine if there are statistically significant differences in PTE leaching concentrations associated with varying eluant molarity levels. The data underwent statistical analysis, including mean calculation, determination of p-values, and computation of the least significant difference (LSD) for each element. A p-value of less than or equal to 0.05 indicates statistical significance, suggesting that molarity substantially influences PTE concentrations.

As illustrated in Figure 27, which shows the AMA samples' statistical analysis results, the findings for Cadmium reveal that it leached at 0.43 M HNO_3, yielding a mean concentration of 1.25· Conversely, at 4 M, the mean Cadmium concentration substantially increases to 4.40 mg kg^{-1}. The p-value associated with Cadmium is less than or equal to 0.05, signifying a statistically significant disparity in Cadmium concentrations between the two molarity levels. Notably, the LSD value for Cadmium is determined to be 0.12. Since the absolute difference (3.15) is significantly larger than the LSD (0.12), this indicates that the difference in Cd concentration between the two molarity levels (0.43 M and 4 M) is indeed statistically significant. This finding confirms the influence of molarity on Cd concentration, as discussed earlier.

As depicted in Figure 27, the Copper (Cu) concentration analysis reveals that at 0.43 M, the mean Cu concentration measures 0.69 mg kg^{-1}. In contrast, at 4 M, the mean Cu concentration experiences a significant increase, reaching 7.74 mg kg^{-1}. The associated p-value for Copper (Cu) is less than or equal to 0.05, indicating a statistically significant disparity in Cu concentrations attributed to varying molarity levels. Notably, the Least Significant Difference (LSD) calculated for Copper (Cu) is 0.12. A direct comparison between the absolute difference in Cu concentrations 7.04 mg kg^{-1} and the LSD (0.12) further validates the statistical significance of the difference between the molarity levels, affirming the influence of molarity on Cu concentrations.

Within the context of Lead (Pb) concentrations, the data displayed in Figure 27 indicates that at 0.43 M, the mean Pb concentration is 0.065 mg kg^{-1}. However, at 4 M, the mean Pb concentration notably increases to 2.48 mg kg^{-1}. The accompanying p-value for Lead (Pb) is less than or equal to 0.05, signifying a statistically significant distinction in Pb concentrations due to variations in molarity levels. The computed Least Significant Difference (LSD) for Lead (Pb) is measured at 0.08. The comparison of the absolute difference in Pb concentrations (2.41) with the LSD (0.08)

underscores the statistical significance of the variation between the two molarity conditions, reinforcing the influential role of molarity on Pb concentrations.

As depicted in Figure 27, the results related to Zinc (Zn) concentrations portray a distinct pattern. At 0.43 M, the mean Zn concentration is 1.70 mg kg^{-1}; at 4 M, the mean Zn concentration experiences a notable increase, amounting to 10.5 mg kg^{-1}. The p-value associated with Zinc (Zn) is less than or equal to 0.05, signifying a statistically significant discrepancy in Zn concentrations as influenced by molarity levels. Moreover, the calculated Least Significant Difference (LSD) for Zinc (Zn) is quantified at 0.19. A direct comparison between the absolute difference in Zn concentrations (8.84) and the LSD (0.19) underscores the statistical significance of the difference between the molarity levels. This reaffirms the pivotal role of molarity in influencing Zinc (Zn) concentrations, echoing the trends observed in the other elements.

The results unequivocally establish that molarity significantly influences the concentrations of Cd, Cu, Pb, and Zn. In all cases, elevating the molarity from 0.43 M to 4 M substantially increases PTE concentrations. These findings have important implications for understanding the behavior of these elements within different chemical environments.

This statistical analysis confirms that molarity substantially influences the leaching concentrations of Cadmium, Copper, Lead, and Zinc. The marked increases in PTE concentrations at higher molarity levels underscore the necessity of considering molarity when studying the behavior of these elements. This insight is valuable for further research and environmental assessments, ensuring accurate characterizations of PTE concentrations.

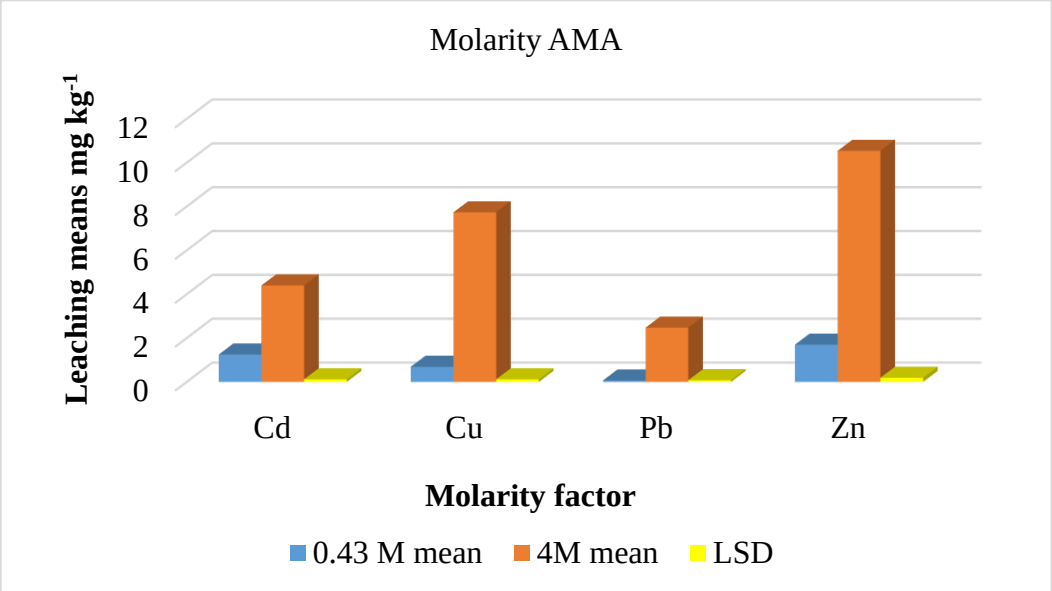


Figure 27. AMA molarity factor statistical analysis

In the case of the APA Samples, the statistical analysis illustrated in Figure 28 revealed that the p-values for all four elements (Cd, Cu, Pb, Zn) are less than 0.05. This means there is strong evidence to reject the null hypothesis (H0) for each element. The LSD values for each element represent the smallest difference between means that is statistically significant. The LSD values are smaller than the differences between the means for each element at the two molarity levels (0.43 M and 4 M). Therefore, we can conclude that the molarity of the eluant (M) has a significant factorial effect on the leaching concentrations of Cd, Cu, Pb, and Zn. The alternative hypothesis (Ha) is supported. In summary, the data suggests that the molarity of the eluant (M) has a statistically significant impact on the leaching concentrations of PTEs (Cd, Cu, Pb, Zn). The null hypothesis is rejected in favor of the alternative hypothesis

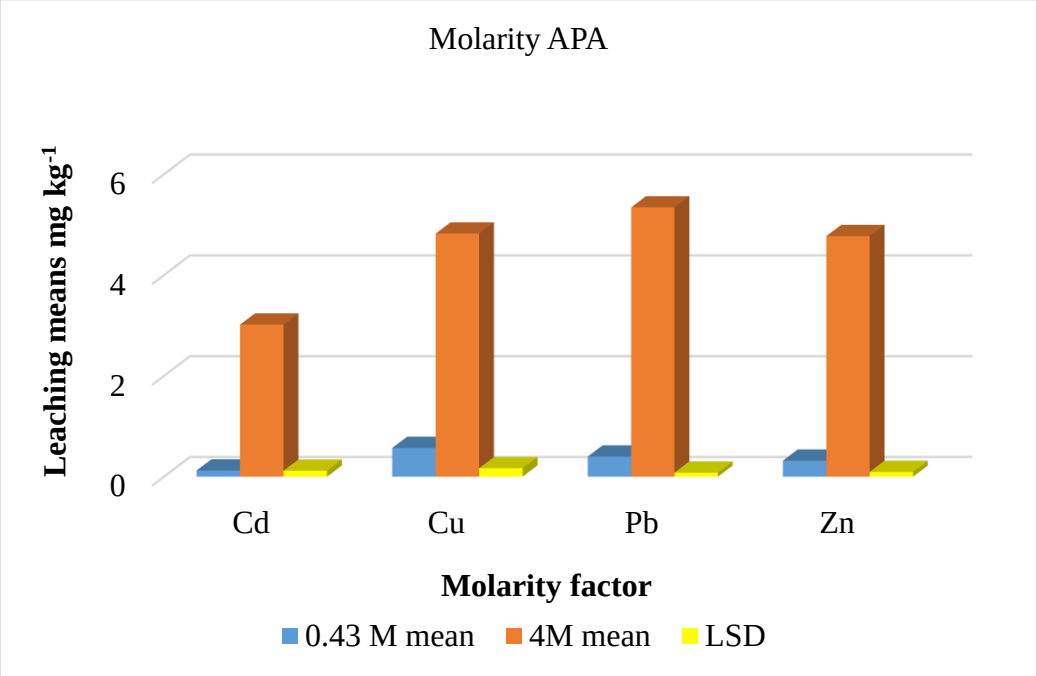


Figure 28. APA molarity factor statistical analysis

4.10.2. Percentage % factor

This study examines the impact of varying ash percentages (1% and 3%) on the mean leaching concentrations of four potentially toxic elements: Cu, Cd, Pb, and Zn. Statistical analysis is performed to assess whether statistically significant differences between the two ash percentage levels exist. As presented in Figure 29 for AMA samples, the results consistently reveal that the absolute differences in leaching concentrations at these two ash percentage levels significantly

surpass the corresponding Least Significant Difference (LSD) values. This unambiguously affirms the substantial role of ash percentage in shaping the leaching concentrations of these potentially Toxic elements.

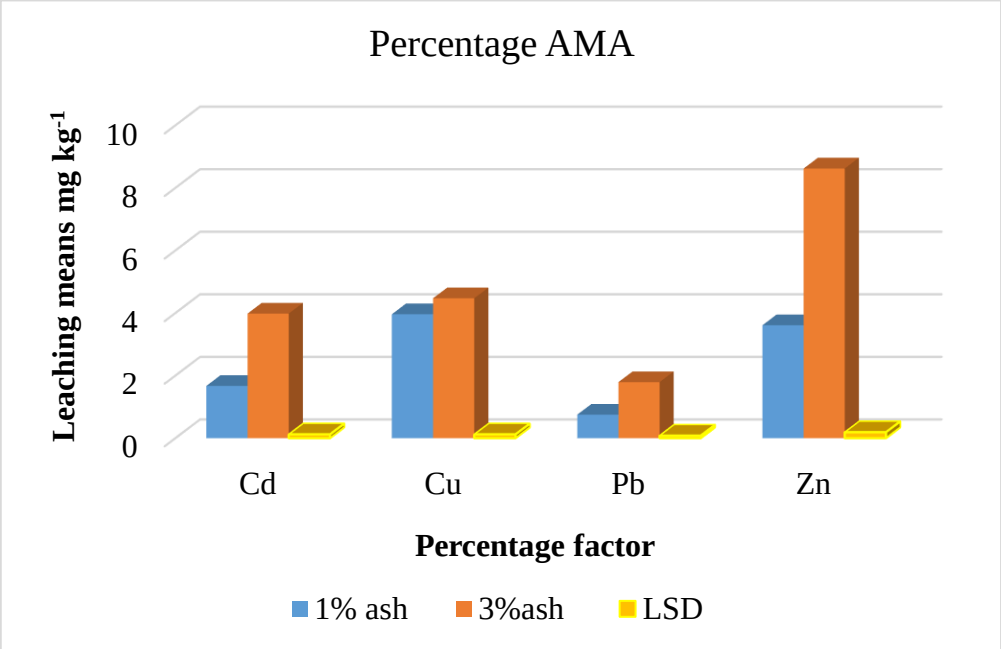


Figure 29. AMA ash percentage factor statistical analysis

The statistical data for APA samples is illustrated in Figure 30. The p-values for all four elements (Cd, Cu, Pb, Zn) are less than 0.05. This means there is strong evidence to reject the null hypothesis (H0) for each element. The LSD values for each element represent the smallest difference between means that is statistically significant. The LSD values are smaller than the differences between the means for each element at the two percentage levels (1% and 3%). Therefore, we can conclude that the percentage has a significant factorial effect on the leaching concentrations of Cd, Cu, Pb, and Zn. The alternative hypothesis (Ha) is supported.

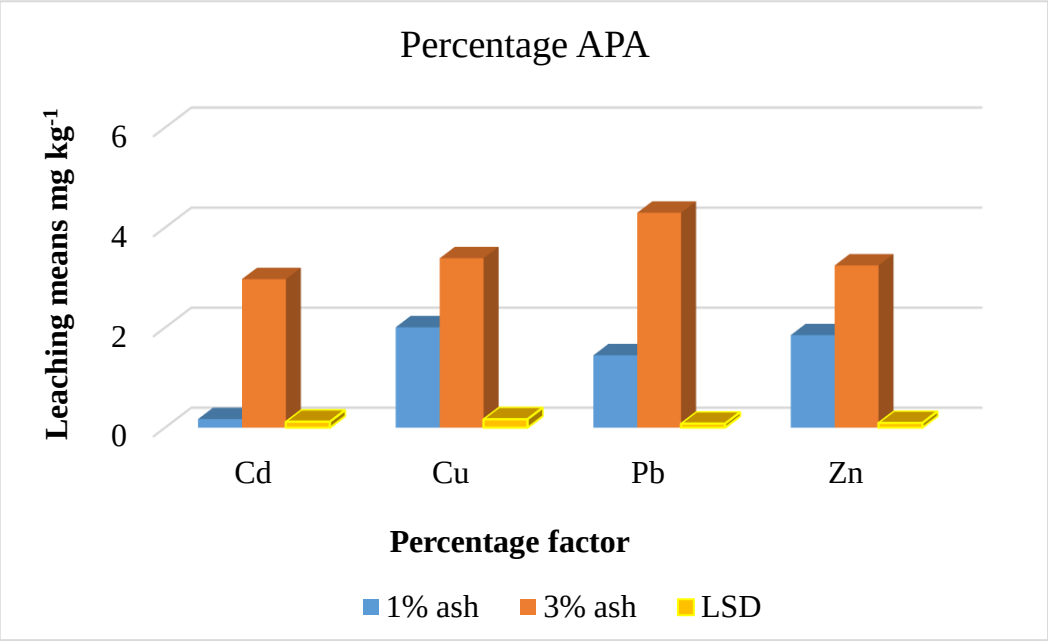


Figure 30. APA ash percentage factor statistical analysis

4.10.3. Initial concentration factor

The initial concentration of Potentially Toxic Elements plays a pivotal role in leaching. As depicted in Figure 31, the AMA dataset offers a comprehensive view of the average leaching concentrations of Cadmium, Copper, Lead, and Zinc across a range of initial concentrations (0.5, 1, 5, 10, and 50) mg L^{-1}. The statistical analysis confirms that for most elements (Copper, Lead, and Zinc), there are statistically significant differences in leaching concentrations across the various initial concentrations, as indicated by p-values less than 0.001. However, it is noteworthy that the concentrations of Cadmium and Lead deviate from this trend. The calculations reveal that significant differences in Cadmium and Lead concentrations only became evident at the 5 mg L^{-1} initial concentration, as the absolute differences surpassed the respective LSD values. This suggests that, unlike the other elements, Cadmium and Lead required a higher initial concentration (5 mg L^{-1}) to exhibit statistically significant differences in leaching behavior. The initial concentration statistical data for APA samples is illustrated in Figure 32. The p-values for all four elements (Cd, Cu, Pb, Zn) are less than 0.05. This suggests strong evidence to reject each element's null hypothesis (H0). The LSD values for each element represent the smallest difference between means that is statistically significant. The LSD values are smaller than the differences between the means for each element at the various initial concentrations. Consequently, we can conclude that the initial concentrations of Potentially Toxic Elements have a statistically significant impact on Cd, Cu, Pb, and Zn leaching concentrations.

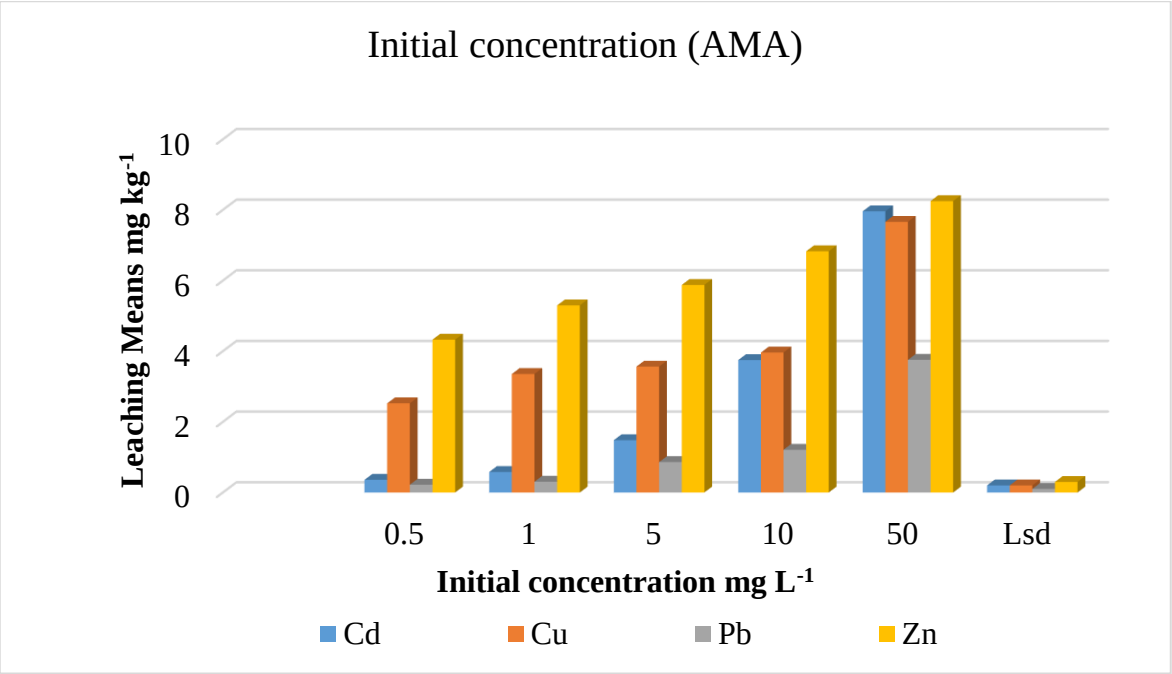


Figure 31. AMA initial concentration factor statistical analysis

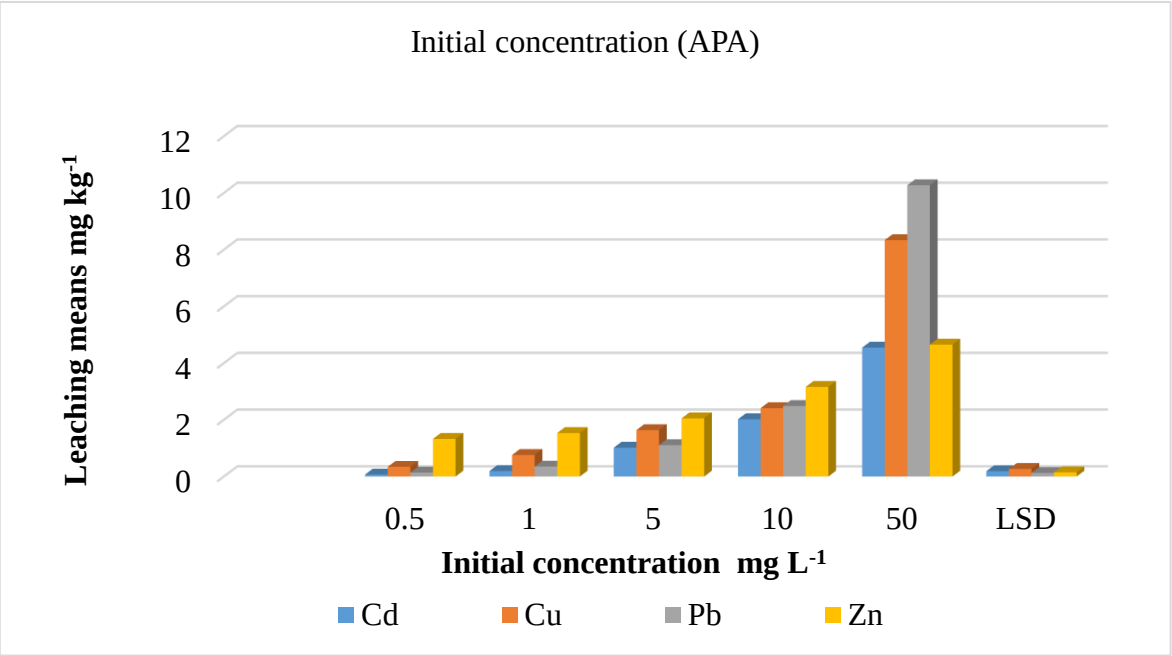


Figure 32. APA initial concentration factor statistical analysis

5. CONCLUSION AND RECOMMENDATIONS

This research addresses the utilization of cellulose-based adsorbent ashes, specifically adsorbed paper ash (APA) and mulch ash (AMA), in mortar composites. The study investigates their affinity for potentially toxic elements (PTEs), their immobilization mechanisms, leaching behavior, and microstructure. This research contributes significantly to the development of sustainable construction materials by demonstrating the effectiveness of cellulose-based adsorbent ashes in mortar composites. By reducing waste and environmental impact while enhancing material performance, these findings align with the principles of waste-to-energy and the circular economy, offering a promising avenue for eco-friendly construction practices. Further research in these areas will lead to more widespread adoption and greater environmental benefits. In conclusion, this study highlights the potential of incorporating waste adsorbents into mortar mixes as a promising solution for addressing the environmental challenges associated with the adsorption of potentially toxic elements (PTEs). Our findings demonstrate the following key points:

-Effective immobilization of PTEs: the inclusion of APA and AMA in mortar matrices successfully immobilizes PTEs (Pb, Cu, Zn, and Cd) in accordance with EU directives, effectively preventing their leaching into the environment.

-Environmental safety: leaching tests indicate that PTE concentrations leached from the tested crushed mortar samples, even in aggressive conditions (0.43 M HNO_3), fall within the inert waste category. Although APA mortar samples slightly exceed the inert waste limit for Pb, both APA and AMA composites remain well below hazardous waste limits, ensuring environmental safety.

-Variable leaching behaviors: our research highlights different leaching behaviors for PTEs. Cd and Zn exhibit higher leaching tendencies in AMA mortar samples, while Pb and Cu are more prone to leaching in APA mortar samples. Therefore, careful consideration of the specific PTEs of concern is essential when selecting the appropriate waste adsorbent.

-Optimal weight ratio: the optimum weight ratio for the addition of APA or AMA to mortar mixes is determined to be 3%. Exceeding this ratio could potentially lead to leaching that exceeds permissible limits, posing a threat to environmental safety.

-Elemental mapping and microstructure: According to Scanning Electron Microscopy (SEM) analysis, the AMA mortar has a denser microstructure than APA mortar and blank mortar, lending credence to the idea that the addition of AMA and APA improved the mortar's microstructure thanks to their micro-filling capabilities. Elemental mapping results show that adsorbed Cd, Zn, Cu, and Pb are distributed reasonably uniformly throughout the APA and AMA mortar matrices,

with a greater intensity of dispersion than the blank mortar sample, indicating evidence of immobilization of the PTEs.

Based on these findings, we recommend the following for future research:

-Extensive Analysis: Future studies should conduct more comprehensive analyses to further explore the utilization of APA and AMA in construction materials, with a focus on their long-term performance and stability. To assess the effects of aging and prolonged exposure, research should include studies on non-crushed mortar samples over extended periods.

-Mechanical Strength and Durability: Investigate the impact of APA and AMA on the mechanical strength and durability of mortar composites to ensure that these waste materials do not compromise the properties of the mortar.

-Civil Engineering Applications: Explore the use of APA and AMA mortar composites in various civil engineering applications, assessing their suitability and performance in real-world construction scenarios.

In addition to the previously mentioned recommendations, a proposed new avenue for future research is the study of adsorbent incorporation into white cement composites. While our current research focuses on conventional mortar mixes, exploring the application of adsorbed paper ash (APA) and adsorbed mulch ash (AMA) in white cement composites presents a promising direction. Future studies should investigate the following aspects:

Adsorption Efficiency, Aesthetic Impact, Environmental Performance, and the compatibility of APA and AMA with white cement, examining their effects on the mechanical properties and long-term durability of the composites. Exploring the integration of waste adsorbents into white cement composites aligns with the broader objective of sustainable construction materials. This research will not only contribute to reducing waste and environmental impact but also offer architects and builders eco-friendly alternatives without compromising the visual appeal and performance of white cement-based architectural elements.

6. NEW SCIENTIFIC RESULTS

1. The findings of this study indicate different affinities and capacities of mulch and paper in adsorbing PTEs. Notably, the paper-based adsorbent exhibited a superior capacity for adsorbing the investigated PTEs. Additionally, the study revealed that Cadmium, with a critical leaching limit of 5 mg kg^{-1}, demonstrated better immobilization within the APA mortar composite, despite the paper adsorbing twice the quantity of mulch.

2. In the investigation of leaching concentrations of PTEs, including Cadmium, Zinc, Copper, and Lead, within both AMA and APA mortar composites, it has been discerned that these concentrations fall below the permissible leaching limits as outlined by the EU directive 1999/31/EC for waste categories. Furthermore, the study has unveiled a crucial revelation: an optimal weight ratio has been determined for the inclusion of APA or AMA in the mortar mixture, and this ratio is found to be 3%. It is of paramount significance that this ratio is not exceeded, as exceeding this threshold significantly increases the risk of leaching surpassing permissible limits, consequently posing a substantial threat to environmental safety.

3. In this study, scanning electron microscopy (SEM) was employed to analyze the microstructures of APA, AMA, and the reference blank samples. The results reveal a significant distinction: the blank mortar displays a higher prevalence of microcracks, and porosity compared to the APA mortar, which exhibits fewer such features. Notably, the AMA mortar demonstrates the densest and most compact microstructure among the three specimens. The SEM analysis indicates that the incorporation of AMA and APA in the mortar enhances its microstructure, primarily due to their exceptional micro-filling properties.

4. The study utilized elemental maps to assess the surface area coverage percentage of adsorbed PTEs and other major elements in mortar composites. An intriguing inverse correlation was observed between the surface area coverage of adsorbed PTEs and the leaching behavior of mortar composites, shedding light on the mechanisms behind PTE immobilization. The observed lower immobilization of Zn was demonstrated due to its unique characteristics, solubility in highly alkaline environments, and limited adsorption onto Calcium-Silicate-Hydrate (C-S-H). In contrast, Pb and Cd precipitated as hydroxides, occupying the cement paste's pore structure These findings offer valuable insights into the sequence of PTE behavior and the crucial role played by immobilization mechanisms in determining leaching patterns in mortar composites.

7. SUMMARY

This study delves into the innovative use of waste ash derived from biosorption processes, specifically adsorbed paper ash (APA) and adsorbed mulch ash (AMA), as valuable additives in mortar. The primary objectives of this research are twofold: to tackle pressing environmental issues associated with the disposal of used adsorbents in landfills and to effectively immobilize potentially toxic elements (PTEs) like Cd, Zn, Cu, and Pb within a cement matrix, thus preventing their seepage into groundwater. This approach not only addresses environmental concerns but also offers the dual advantages of reducing landfill waste and harnessing energy during the ashing process while simultaneously confining PTEs within the cement matrices.

To comprehensively assess the impact of these ash additives and ensure their compatibility with environmental safety standards, this study employs a multifaceted approach. Firstly, the study investigates the leaching behavior of mortar composites at various artificial adsorption initial concentrations (ranging from 0.5 to 50 mg L^{-1}) and explores different mixing weight ratios of ash with cement. To quantify the total elemental content of PTEs in leachate solutions, the research employs an inductively coupled plasma-optical emission spectrometer (ICP-OES).

The study's findings are promising, with results indicating that the ash from used adsorbents can be considered environmentally acceptable as a mortar additive. This is primarily due to its demonstrated ability to stabilize PTEs, with leaching values for Pb, Zn, and Cu falling comfortably below the permissible limits set for hazardous waste. The sole exception was Cd, which exceeded regulatory limits only in cases of extreme acidity. This research explores a sustainable and environmentally friendly approach to addressing the secondary pollution associated with biosorption processes by repurposing waste ash as an additive in mortar. The results suggest that this approach can effectively immobilize most PTEs within the cement matrix, ensuring environmental safety and compliance with regulatory limits. However, there may be a need for further measures or treatments to control the leaching of Cd under specific conditions. In addition to these techniques, the research also utilizes a scanning electron microscope (SEM) equipped with energy-dispersive X-ray spectroscopy (EDS) to scrutinize the elemental and microstructural characteristics of the mortar composites. Elemental maps generated through this analysis are further processed using ImageJ software, offering insights into the microstructural improvements achieved by incorporating the ash additives. Elemental mapping reveals a consistent distribution of PTEs within the mortar matrix structures, indicating successful immobilization. The study finds that the order of percentage area coverage for immobilized elements correlates with the order of

leaching, demonstrating the effectiveness of the immobilization process. This research embodies a sustainable and ecologically friendly approach to mitigating the secondary pollution stemming from biosorption processes by repurposing waste ash in mortar applications. The results underscore the effectiveness of this approach in immobilizing the majority of PTEs within the cement matrix at the optimum addition of 3% of ash to cement, thereby ensuring compliance with environmental safety standards and regulatory thresholds. Nonetheless, it is worth considering additional measures or treatments to address Cd leaching in specific conditions. It also promotes sustainability by repurposing waste ash, reducing pollution, and improving the microstructure of mortar composites, contributing to environmentally sound construction practices and sustainable waste management.

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9. APPENDICES

A1: Bibliography

Acar, F. N., & Eren, Z. (2006). Removal of Cu (II) ions by activated poplar sawdust (Samsun Clone) from aqueous solutions. Journal of hazardous materials, 137(2), 909-914.

Achour, Y., Souissi, R., Tlil, H., Souissi, F., & Motelica-Heino, M. (2022). Mobility of Potentially Toxic Elements (Pb, Zn, Cd, As, Sb) in Agricultural Carbonated Soils Contaminated by Mine Tailings (Northern Tunisia): A New Kinetic Leaching Approach with Organic Acids. Water, 14(20), 3337.

Ademola, A.K., Olaoye, M.A. and Abodunrin, P.O. (2015), "Radiological safety assessment and determination of heavy metals in soil samples from some waste dumpsites in Lagos and Ogun state, south-western, Nigeria," Journal of Radiation Research and Applied Sciences, Elsevier Ltd, Vol. 8 No. 1, pp. 148–153.

Agarwal, M., Tardio, J., & Mohan, S. V. (2015). Pyrolysis biochar from cellulosic municipal solid waste as adsorbent for azo dye removal: equilibrium isotherms and kinetics analysis. International Journal of Environmental Science and Development, 6(1), 67.

Agrawal, V. M., &Savoikar, P. P. (2022). Sustainable use of normal and ultra-fine fly ash in mortar as partial replacement to ordinary Portland cement in ternary combinations. Materials Today: Proceedings, 51, 1593-1597. https://doi.org/10.1016/j.matpr.2021.10.409.

Ahmad, S., M. I. Malik, M. B. Wani, and R. Ahmad. 2013. "Study of concrete involving use of waste paper sludge ash as partial replacement of cement." IOSR J. Eng. 3 (11): 6–15. https://doi.org/10.9790/3021 -031130615.

Ahmad, W., Alharthy, R. D., Zubair, M., Ahmed, M., Hameed, A., & Rafique, S. (2021). Toxic and heavy metals contamination assessment in soil and water to evaluate human health risk. Scientific reports, 11(1), 1-12.

Ahmadian, M., Derakhshankhah, H., & Jaymand, M. (2023). Recent advances in adsorption of environmental pollutants using metal–organic frameworks-based hydrogels. International journal of biological macromolecules, 123333.

Akinyemi BA, Dai C (2020) Development of banana fibers and wood bottom ash modified cement mortars. Constr Build Mater 241 https://doi.org/10.1016/j.conbuildmat.2020.118041/

Akpomie, K. G., & Dawodu, F. A. (2015). Potential of a low-cost bentonite for heavy metal abstraction from binary component system. Beni-suef university journal of basic and applied sciences, 4(1), 1-13.

Al-Hwaiti, M. S., & Ranville, J. F. (2010). Distribution of potentially toxic metal and radionuclide contamination in soils related to phosphogypsum waste stockpiling in the Eshidiya Mine, Jordan.

Ali, A., Hashmi, H. N., & Baig, N. (2013). Treatment of the paper mill effluent – A review. Annals of Faculty Engineering Hunedoara - International Journal of Engineering, 11(3), 337–11.

Al-Mefarrej, H. A., Abdel-Aal, M. A., & Nasser, R. A. (2013). Chemical evaluation of some lignocellulosic residues for pulp and paper production. American-Eurasian J Agric Environ Sci, 13, 498-504.

Alsawy, T., Rashad, E., El-Qelish, M., & Mohammed, R. H. (2022). A comprehensive review on the chemical regeneration of biochar adsorbent for sustainable wastewater treatment. Npj Clean Water, 5(1), 29.

Amaral, E. A., Santos, L. M., Costa, E. V., Trugilho, P. F., & Hein, P. R. (2020). Estimation of moisture in wood chips by Near Infrared Spectroscopy. *Maderas. Ciencia y tecnología*, *22*(3), 291-302.

American Society for Testing and Materials. (2012). ASTM C 618-Standard specification for coal fly ash and raw or calcined natural pozzolan for use in concrete. West Conshohocken: ASTM.

Antoniadis, V., Shaheen, S. M., Levizou, E., Shahid, M., Niazi, N. K., Vithanage, M., ... & Rinklebe, J. (2019). A critical prospective analysis of the potential toxicity of trace element regulation limits in soils worldwide: Are they protective concerning health risk assessment?-A review. Environment international, 127, 819-847.

Aramjoo, H., Arab-Zozani, M., Feyzi, A., Naghizadeh, A., Aschner, M., Naimabadi, A., ... & Samarghandian, S. (2022). The association between environmental cadmium exposure, blood pressure, and hypertension: a systematic review and meta-analysis. Environmental Science and Pollution Research, 29(24), 35682-35706.

Arshadi, M., Amiri, M. J., & Mousavi, S. (2014). Kinetic, equilibrium and thermodynamic investigations of Ni (II), Cd (II), Cu (II) and Co (II) adsorption on barley straw ash. Water Resources and Industry, 6, 1-17.

Awolusi, T. F., Sojobi, A. O., & Afolayan, J. O. (2017). SDA and laterite applications in concrete: Prospects and effects of elevated temperature. Cogent Engineering, 4(1), 1387954.

Ayobami, A. B. 2021. "Performance of wood bottom ash in cement-based applications and comparison with other selected ashes: Overview." Resour. Conserv. Recycl. 166: 105351. https://doi.org/10.1016/j .resconrec.2020.105351.

Azeem, M., Shaheen, S. M., Ali, A., Jeyasundar, P. G., Latif, A., Abdelrahman, H., ... & Zhang, Z. (2022). Removal of potentially toxic elements from contaminated soil and water using bone char compared to plant-and bone-derived biochars: A review. Journal of Hazardous Materials, 427, 128131.

Baes, A. U., Umali, S. J. P., & Mercado, R. L. (1996). Ion exchange and adsorption of some heavy metals in a modified coconut coir cation exchanger. Water Science and Technology, 34(11), 193-200.

Bahadir, T., Bakan, G., Altas, L., Buyukgungor, H. (2006) The investigation of lead removal by biosorption: An application at storage battery industry wastewaters. Enzyme and Microbial Technology. 41, 98– 102.

Bai, J., Chaipanich, A., Kinuthia, J. M., O'farrell, M., Sabir, B. B., Wild, S., & Lewis, M. H. (2003). Compressive strength and hydration of wastepaper sludge ash–ground granulated blastfurnace slag blended pastes. Cement and Concrete Research, 33(8), 1189-1202.

Baloochi, H., Aponte, D., & Barra, M. (2022). Waste paper ash as a hydraulic road binder: Hydration, mechanical and leaching considerations. Journal of Environmental Management, 314, 115042.

Bansal, O. P. (2020). Health risks of potentially toxic metals contaminated water. Heavy metal toxicity in public health, 63.

Batchelor, B. (2006). Overview of waste stabilization with cement. Waste management, 26(7), 689-698.

Benassi, L., Pasquali, M., Zanoletti, A., Dalipi, R., Borgese, L., Depero, L. E., ... & Bontempi, E. (2016). Chemical stabilization of municipal solid waste incineration fly ash without any

commercial chemicals: first pilot-plant scaling up. ACS Sustainable Chemistry & Engineering, 4(10), 5561-5569.

Beni, A. A., & Esmaeili, A. (2020). Biosorption, an efficient method for removing heavy metals from industrial effluents: a review. Environmental Technology & Innovation, 17, 100503.

Berra, M., N. M. Ippolito, T. Mangialardi, A. E. Paolini, and L. Piga. (2019). "Leaching test procedure for assessing the compliance of the chemical and environmental requirements of hardened woody biomass fly ash cement mixtures." Waste Manage. (Oxford) 90: 10–16. https://doi.org/10 .1016/j.wasman.2019.04.038.

Bettinelli, M., Beone, G.M., Spezia, S. and Baffi, C. (2000), "Determination of heavy metals in soils and sediments by microwave-assisted digestion and inductively coupled plasma optical emission spectrometry analysis", Vol. 424, pp. 289–296.

Bohrn, G., & Stampfer, K. (2014). Untreated wood ash as a structural stabilizing material in forest roads. Croatian Journal of Forest Engineering: Journal for Theory and Application of Forestry Engineering, 35(1), 81-89.

Boruczkowski, T., Boruczkowska, H., Drożdż, W., Miszczak, M., & Leszczyński, W. (2022). Use of ImageJ software for assessment of mechanical damage to starch granules. Processes, 10(4), 630. https://doi.org/10.3390/pr10040630.

Cao, M., X. Ming, K. He, L. Li, and S. Shen. 2019. "Effect of macro-, micro- and nano-calcium carbonate on properties of cementitious composites—A Review " Materials 12 (5): 781. https://doi.org/org/10.3390 /ma12050781.

Carević , I., M. Serdar, N. Štirmer, and N. Ukrainczyk. 2019. "Preliminary screening of wood biomass ashes for partial resources replacements in cementitious materials." J. Cleaner Prod. 229: 1045–1064. https://doi .org/org/10.1016/j.jclepro.2019.04.321.

Carević , I., N. Štirmer, M. Trkmić , and K. KostanićJurić . 2020. "Leaching characteristics of wood biomass fly ash cement composites." Appl. Sci. 10 (23): 8704. https://doi.org/org/10.3390/app10238704.

Carolin, C. F., Kumar, P. S., Saravanan, A., Joshiba, G. J., & Naushad, M. (2017). Efficient techniques for the removal of toxic heavy metals from aquatic environment: A review. Journal of environmental chemical engineering, 5(3), 2782-2799.

Carrasco, B., Cruz, N., Terrados, J., Corpas, F. A., & Pérez, L. (2014). An evaluation of bottom ash from plant biomass as a replacement for cement in building blocks. Fuel, 118, 272-280.

Castrillón, J. J., and H. Gil. 2020. "Mechanical properties of mortars modified with wood waste ash." J. Indian Acad. Wood Sci. 17 (1): 90–99. https://doi.org/org/10.1007/s13196-020-00258-w.

Celik, K. (2015). Development and characterization of sustainable self-consolidating concrete containing high volume of limestone powder and natural or calcined pozzolanic materials. University of California, Berkeley.

CEN (European Committee for Standardization). 2014. Towards a circular economy: A zero waste program for Europe. Communication No. 398, Brussels, Belgium: CEN.

CEN/TS 14429. (2005). Characterization of Waste–Leaching Behaviour Tests–Influence of pH on Leaching with Initial Acid/base Addition.

Chakravarty, S., Pimple, S., Chaturvedi, H. T., Singh, S., & Gupta, K. K. (2008). Removal of copper from aqueous solution using newspaper pulp as an adsorbent. Journal of hazardous materials, 159(2-3), 396-403.

Chen, C., Jin, Y., & Chi, Y. (2014). Effects of moisture content and CaO on municipal solid waste pyrolysis in a fixed bed reactor. *Journal of analytical and applied pyrolysis*, *110*, 108-112.

Chen, Y., Wang, T., Zhou, M., Hou, H., Xue, Y., & Wang, H. (2018). Rice husk and sewage sludge co-combustion ash: Leaching behavior analysis and cementitious property. Construction and Building Materials, 163, 63-72.

Cheng, H., Yang, B., Ke, T., Li, S., Yang, X., Aschner, M., & Chen, P. (2021). Mechanisms of metal-induced mitochondrial dysfunction in neurological disorders. Toxics, 9(6), 142.

Cherian, C., & Siddiqua, S. (2019). Pulp and paper mill fly ash: A review. Sustainability, 11(16), 4394.

Chirenje, T., Ma, L. Q., & Lu, L. (2006). Retention of Cd, Cu, Pb and Zn by wood ash, lime and fume dust. Water, Air, & Soil Pollution, 171, 301-314.

Cholico-Gonzalez, D., Ortiz Lara, N., Fernandez Macedo, A. M., & Chavez Salas, J. (2020). Adsorption behavior of Pb (II), Cd (II), and Zn (II) onto agave bagasse, characterization, and mechanism. ACS omega, 5(7), 3302-3314.

Chowdhury, S., Mishra, M., & Suganya, O. M. (2015). The incorporation of wood waste ash as a partial cement replacement material for making structural grade concrete: An overview. Ain Shams Engineering Journal, 6(2), 429-437.

Çolak, F., Atar, N., & Olgun, A. (2009). Biosorption of acidic dyes from aqueous solution by Paenibacillus macerans: Kinetic, thermodynamic and equilibrium studies. Chemical engineering journal, 150(1), 122-130.

Collin, M. S., Venkatraman, S. K., Vijayakumar, N., Kanimozhi, V., Arbaaz, S. M., Stacey, R. S., ... & Swamiappan, S. (2022). Bioaccumulation of lead (Pb) and its effects on human: A review. Journal of Hazardous Materials Advances, 7, 100094.

Conner, J. R., & Hoeffner, S. L. (1998). A critical review of stabilization/solidification technology. Critical Reviews in Environmental Science and Technology, 28(4), 397-462.

Çoruh, S., & Ergun, O. N. (2006). Leaching characteristics of copper flotation waste before and after vitrification. Journal of environmental management, 81(4), 333-338.

Da Luz Garcia, M., & Sousa-Coutinho, J. (2013). Strength and durability of cement with forest waste bottom ash. Construction and building Materials, 41, 897-910.

Da Silva, C. M., da Boit Martinello, K., Lütke, S. F., Godinho, M., Perondi, D., Silva, L. F., and Dotto, G. L. (2022). Pyrolysis of grape bagasse to produce char for Cu (II) adsorption: a circular economy perspective. Biomass Conversion and Borefinery, 1-18.

Dakiky, M., Khamis, M., Manassra, A., & Mer'Eb, M. (2002). Selective adsorption of chromium (VI) in industrial wastewater using low-cost abundantly available adsorbents. Advances in environmental research, 6(4), 533-540.

Darama, S. E., Gürkan, E. H., Terzi, Ö., & Çoruh, S. (2021). Leaching performance and zinc ions removal from industrial slag leachate using natural and biochar walnut shell. Environmental Management, 67, 498-505.

De Quadros Melo, D., de Oliveira Sousa Neto, V., de Freitas Barros, F. C., Raulino, G. S. C., Vidal, C. B., & do Nascimento, R. F. (2016). Chemical modifications of lignocellulosic

materials and their application for removal of cations and anions from aqueous solutions. Journal of Applied Polymer Science, 133(15).

Dehghani, M. H., Sanaei, D., Ali, I., & Bhatnagar, A. (2016). Removal of chromium (VI) from aqueous solution using treated waste newspaper as a low-cost adsorbent: kinetic modeling and isotherm studies. Journal of molecular liquids, 215, 671-679.

Deng, Y. (2020). Low-cost adsorbents for urban stormwater pollution control. Frontiers of Environmental Science & Engineering, 14, 1-8.

Dijkstra, J. J., J. C. Meeussen, and R. N. Comans. (2004). "Leaching of heavy metals from contaminated soils: An experimental and modeling study." Environ. Sci. Technol. 38 (16): 4390–4395. https://doi.org/10 .1021/es049885v.

Dimter S, Simun M, Zagvozda M, Rukavina T (2021) Laboratory evaluation of the properties of asphalt mixture with wood ash filler. Mater (Basel) 14(3). https://doi.org/10.3390/ma14030575.

Ding, Z., Xu, X., Phan, T., & Hu, X. (2018). Carbonized Waste Corrugated Paper Packaging Boxes as Low-Cost Adsorbent for Removing Aqueous Pb (II), Cd (II), Zn (II), and Methylene Blue. *Polish Journal of Environmental Studies*, *27*(6).

Directive of 6/2009. (IV.14.) KvVM-EüM-FVM about of the permissible limits of contaminants in soils, in sediments and below-ground waters and its assessments (Együttes rendelet a földtani közeg és a felszín alatti víz szennyezéssel szembeni védelméhez szükséges határértékekről és a szennyezések méréséről).

Directive, L. (1999). Council Directive 1999/31/EC of 26 April 1999 on the landfill of waste. *Official Journal of the European Communities*, *182*(1), 26-04.

Dotto, G., McKay, G. (2020) Current scenario and challenges in adsorption for water treatment Journal of Environmental Chemical Engineering, 8 (4) , art. no. 103988.

Drljača, D., Vukić, L., Dragić, D., Borković, A., Botić, T., Dugić, P., ... & Savković, J. (2022). Leaching of heavy metals from wood biomass ash, before and after binding in cement composite. Journal of the Serbian Chemical Society, 87(9), 1091-1108.

Elangovan, G., &Rajanandhini, M. V. (2018). EXPERIMENTAL STUDY AND SEM ANALYSIS ON MORTAR CUBE WITH WOOD ASH FOR PARTIAL REPLACEMENT OF CEMENT. Management Research, 5(3), 263-269. DOI: 10.5281/zenodo.1218517.

Eliche-Quesada D, Felipe-Sese MA, Lopez-Perez JA, Infantes-Molina A (2017) Characterization and evaluation of rice husk ash and wood ash in sustainable clay matrix bricks. Ceram Int 43(1):463–475. https://doi.org/10.1016/j.ceramint.2016.09.181

Emeh C, Igwe O (2016) The combined effect of wood ash and lime on the engineering properties of expansive soils. Int J Geotech Eng 10(3):246–256. https://doi.org/10.1080/19386362.2015.1125412.

Esrafili, L., Safarifard, V., Tahmasebi, E., Esrafili, M. D., & Morsali, A. (2018). Functional group effect of isoreticular metal–organic frameworks on heavy metal ion adsorption. New Journal of Chemistry, 42(11), 8864-8873. https://doi.org/10.1039/C8NJ01150H.

Fahad, B. M., Ali, N. S., & Hameed, T. T. (2018). Using paper waste as adsorbent for methyl violet dye removal from wastewater. Journal of Engineering and Sustainable Development, 22(01).

Fardim, P., Ferreira, M. M., & Durán, N. (2005). Determination of mechanical and optical properties of Eucalyptus kraft pulp by NIR spectrometry and multivariate calibration. Journal of wood chemistry and technology, 25(4), 267-279.

Farghali, A. A., Bahgat, M., Allah, A. E., & Khedr, M. H. (2013). Adsorption of Pb (II) ions from aqueous solutions using copper oxide nanostructures. Beni-Suef University Journal of Basic and Applied Sciences, 2(2), 61-71.

Faruk, O., Bledzki, A, K., Fink, H.-P., and Sain, M. (2012). "Biocomposites reinforced with natural fibers: 2000-2010," Progress in Polymer Science 37 (11), 1552-1596. DOI: 10.1016/j.progpolymsci.2012.04.003.

Fassel, V. A., & Kniseley, R. N. (1974). Inductively coupled plasma. Optical emission spectroscopy. Analytical Chemistry, 46(13), 1110A-1120a.

Fatima, G., Raza, A. M., Hadi, N., Nigam, N., & Mahdi, A. A. (2019). Cadmium in human diseases: It's more than just a mere metal. Indian Journal of Clinical Biochemistry, 34, 371-378.

Fava, G., Naik, T. R., & Pierpaoli, M. (2018). Compressive strength and leaching behavior of mortars with biomass ash. Recycling, 3(3), 46.

Fawzy, M. A., & Gomaa, M. (2020). Use of algal biorefinery waste and waste office paper in the development of xerogels: A low cost and eco-friendly biosorbent for the effective removal of congo red and Fe (II) from aqueous solutions. Journal of environmental management, 262, 110380.

Frías, M., O. Rodríguez, and M. S. De Rojas. 2015. "Paper sludge, an environmentally sound alternative source of MK-based cementitious materials. A review." Constr. Build. Mater. 74: 37–48. https://doi.org/10 .1016/j.conbuildmat.2014.10.007.

Gabrijel, I., JelčićRukavina, M., &Štirmer, N. (2021). Influence of Wood Fly Ash on Concrete Properties through Filling Effect Mechanism. Materials, 14(23), 7164. https://doi.org/10.3390/ma14237164.

Gao, Y., Hu, C., Zhang, Y., Li, Z., & Pan, J. (2018). Investigation on microstructure and microstructural elastic properties of mortar incorporating fly ash. Cement and Concrete Composites, 86, 315-321.

Glasser, F. P. (1997). "Fundamental aspects of cement solidification and sta bilisation." J. Hazard. Mater. 52 (2–3): 151–170.

Gominho, M.S. (2016). Microstructural Characterization of Mortars.Civil Engineering Department, Instituto Superior Técnico, Universidade Técnica de Lisboa, Portugal.

Gougar, M. L. D., Scheetz, B. E., & Roy, D. M. (1996). Ettringite and C S H Portland cement phases for waste ion immobilization: A review. *Waste management*, *16*(4), 295-303.

Grubb, D. G., Moon, D. H., Reilly, T., Chrysochoou, M., & Dermatas, D. (2009). Stabilization/solidification (S/S) of Pb and W contaminated soils using type I/II Portland cement, silica fume cement and cement kiln dust. Glob. Nest. J, 11, 267-282.

Guo, B., Liu, B., Yang, J., & Zhang, S. (2017). The mechanisms of heavy metal immobilization by cementitious material treatments and thermal treatments: A review. Journal of environmental management, 193, 410-422.

Gupta, P. K. , Raghunath, S. S. , Prasanna, D. V. , Venkat, P., Shree, V., Chithananthan, C., Choudhary, S., Surender, K., & Geetha, K. (2019). An Update on Overview of Cellulose, Its

Structure and Applications. In A. R. Pascual, & M. E. E. Martín (Eds.), Cellulose. IntechOpen. https://doi.org/10.5772/intechopen.84727.

Gupta, S., & Babu, B. V. (2009). Removal of toxic metal Cr (VI) from aqueous solutions using sawdust as adsorbent: Equilibrium, kinetics and regeneration studies. Chemical Engineering Journal, 150(2-3), 352-365.

Hamad, A. A., Hassouna, M. S., Shalaby, T. I., Elkady, M. F., Abd Elkawi, M. A., & Hamad, H. A. (2020). Electrospun cellulose acetate nanofiber incorporated with hydroxyapatite for removal of heavy metals. International journal of biological macromolecules, 151, 1299-1313.

Han, B., Weatherley, A. J., Mumford, K., Bolan, N., He, J. Z., Stevens, G. W., & Chen, D. (2022). Modification of naturally abundant resources for remediation of potentially toxic elements: A review. Journal of Hazardous Materials, 421, 126755.

Harris, G. K., & Marshall, M. R. (2017). Ash analysis. Food analysis, 287-297.

Helaluddin, A., Khalid, R.S., Alaama, M. and Abbas, S.A. (2016), "Main Analytical Techniques Used for Elemental Analysis in Various Matrices", Pharm Res Tropical Journal of Pharmaceutical Research Journal Citation ReportsScience Edition, Vol. 15 No. 152, pp. 427–427.

Heltai, G., Győri, Z., Fekete, I., Halász, G., Kovács, K., Takács, A., Khumalo, L., et al. (2019), "Application of flexible multi-elemental ICP-OES detection in fractionation of potentially toxic element content of solid environmental samples by a sequential extraction procedure", Microchemical Journal, Elsevier, Vol. 149 No. January, p. 104029.

Henriksson, M., and Berglund, L. A. (2007). "Structure and properties of cellulose nanocomposite films containing melamine formaldehyde," Journal of Applied Polymer Science 106 (4), 2817-2824. DOI: 10.1002/app.26946.

Herrera, R., Erdocia, X., Llano-Ponte, R., & Labidi, J. (2014). Characterization of hydrothermally treated wood in relation to changes on its chemical composition and physical properties. Journal of analytical and Applied Pyrolysis, 107, 256-266.

Hoang, A. T., Kumar, S., Lichtfouse, E., Cheng, C. K., Varma, R. S., Senthilkumar, N., ... & Nguyen, X. P. (2022). Remediation of heavy metal polluted waters using activated carbon from lignocellulosic biomass: An update of recent trends. Chemosphere, 302, 134825.

Hokkanen, S., Bhatnagar, A., & Sillanpää, M. (2016). A review on modification methods to cellulose-based adsorbents to improve adsorption capacity. Water research, 91, 156-173.

Hokkanen, S., Repo, E., and Sillanpää, M. (2013). Removal of heavy metals from aqueous solutions by succinic anhydride modified mercerized nanocellulose. Chemical Engineering Journal, 223, 40-47.

Holtzapple,M.T.(2003).CELLULOSE.ElsevierEBooks,998–1007. https://doi.org/10.1016/b0-12-227055-x/00185-1.

Hooda, P.S. (2010), Trace Elements in Soils, edited by Hooda, P.S., 1st ed., John Wiley & Sons, Ltd, Chichester, West Sussex, United Kingdom, available at: https://doi.org/10.13140/RG.2.1.3377.1123.

Huang, D., Li, B., Ou, J., Xue, W., Li, J., Li, Z., ... & Guo, X. (2020). Megamerger of biosorbents and catalytic technologies for the removal of heavy metals from wastewater: Preparation, final disposal, mechanism and influencing factors. Journal of environmental management, 261, 109879.

Hubbe, M. A.(2022). “What to do with toxic, contaminated cellulose-based adsorbents.” BioResources 17 (1): 3–6. https://doi.org/10.15376/biores.17.1.3-6.

Hubbe, M. A., Becheleni, E. M. A., Lewis, A. E., Peters, E. M., Gan, W., Nong, G., Mandal, S., and Shi, S. Q. (2018). "Recovery of inorganic compounds from spent alkaline pulping liquor by eutectic freeze crystallization and supporting unit operations: A Review," BioRes. 13(4), 9180-9219.

Hui, L. Y., Min, J. H., Kim, S. H., & Lee, J. Y. (2020). Fundamental study of the production of paper ash made from paper sludge I: Evaluation of the characteristics of paper ash made from paper sludge in a specialty paper mill. Journal of Korea TAPPI, 52(3), 113-119.

Huo, H., Yu, Y., Zhang, X., Tang, M., Chen, C., Wang, S., & Min, D. (2022). Phosphorylated wood designed as a biosorbent for effectively removing Ni2+ from wastewater. Industrial Crops and Products, 188, 115727.

Iqbal, R., Raza, M. A. S., Valipour, M., Saleem, M. F., Zaheer, M. S., Ahmad, S., ... & Nazar, M. A. (2020). Potential agricultural and environmental benefits of mulches—a review. Bulletin of the National Research Centre, 44(1), 1-16.

Ismail, N., M. F. Arshad, H. M. Saman, and C. W. Kang. (2019). "Water treatment sludge stabilizer binder by waste paper sludge ash for solidification/stabilisation technique." Int. J. Integr. Eng. 11 (1): 113–119. https://doi.org/10.30880/ijie.2019.11.01.014.

ISO 287:2017(en) Paper and board — Determination of moisture content of a lot — Oven-drying method.

Jadoon, S., Muhammad, S., Hilal, Z., Ali, M., Khan, S., & Khattak, N. U. (2019b). Spatial distribution of potentially toxic elements in urban soils of Abbottabad city, (N Pakistan): Evaluation for potential risk. Microchemical Journal. https://doi.org/10.1016/j.microc.2019.104489.

Jaishankar, M., Tseten, T., Anbalagan, N., Mathew, B. B., & Beeregowda, K. N. (2014). Toxicity, mechanism and health effects of some heavy metals. Interdisciplinary toxicology, 7(2), 60.

James AK, Thring RW, Helle S, Ghuman HS (2012) Ash management review-applications of biomass bottom ash. Energies5(10):3856–3873. https://doi.org/10.3390/en5103856.

Jamshaid, A., Hamid, A., Muhammad, N., Naseer, A., Ghauri, M., Iqbal, J., ... & Shah, N. S. (2017). Cellulose-based Materials for the Removal of Heavy Metals from Wastewater–An Overview. ChemBioEng Reviews, 4(4), 240-256. https://doi.org/10.1002/cben.201700002.

Jang, A., Y. Seo, and P. L. Bishop. 2005. "The removal of heavy metals in urban runoff by sorption on mulch." Environ. Pollut. 133 (1): 117–127. https://doi.org/10.1016/j.envpol.2004.05.020.

Jehan, S., Khattak, S. A., Muhammad, S., Ahmad, R., Farooq, M., Khan, S., et al. (2018). Ecological and health risk assessment of heavy metals in the Hattar industrial estate, Pakistan AU—Jehan, Shah. Toxin Reviews. https://doi.org/10.1080/15569543.2018.1478858.

Joint Decree No. 10/2000 (VI. 2.) KöM-EüM-FVM-KHVM of the Minister of Environment, Minister of Public Health, Minister of Agriculture and Regional Development and the Minister of Transport, Communication and Water Management on the limit values required to the quality protection of groundwater and the geological media.

Joseph, A. M., R. Snellings, P. Van den Heede, S. Matthys, and N. De Belie. 2018. “The use of municipal solid waste incineration ash in various building materials: A Belgian point of view.” Materials 11 (1): 141. https://doi.org/10.3390/ma11010141.

Kabata-Pendias, A. (2010). Trace elements in soils and plants: Trace Elements in Soils and Plants, 1–520.

Kadir, A. A., Hassan, M. I. H., & Abdullah, M. M. A. B. (2016, June). Investigation on leaching behaviour of fly ash and bottom ash replacement in self-compacting concrete. In IOP Conference Series: Materials Science and Engineering (Vol. 133, No. 1, p. 012036). IOP Publishing.

Kalembkiewicz, J., & Sitarz-Palczak, E. (2015). Efficiency of leaching tests in the context of the influence of the fly ash on the environment. Journal of Ecological Engineering, 16(1).

Kalia S, Kaur BSKI. Cellulose Fibers: Bio- and Nano-Polymer Composites [Internet]. Kalia S, Kaith BS, Kaur I, editors. Berlin, Heidelberg: Springer Berlin Heidelberg; 2011. 743 p. Available from: http://link.springer.com/10.1007/978-3-642-17370-7.

Kanazu, T., Matsumura, T., Nishiuchi, T., & Yamamoto, T. (2001). Effect of simulated acid rain on deterioration of concrete. In Acid rain 2000: Proceedings from the 6 th International Conference on Acidic Deposition: Looking back to the past and thinking of the future Tsukuba, Japan, 10–16 December 2000 Volume III/III Conference Statement Plenary and Keynote Papers (pp. 1481-1486). Springer Netherlands.

Keshavarzi, B., Moore, F., Ansari, M., Rastegari Mehr, M., Kaabi, H., & Kermani, M. (2015). Macronutrients and trace metals in soil and food crops of Isfahan Province, Iran. Environmental monitoring and assessment, 187, 1-22.

Khazaei, J. (2008) Water Absorption Characteristics of Three Wood Varieties. Cercetari Agronomicein Moldova, XLI, 134-145.

Khumalo, L., Heltai, G., & Horváth, M. (2020). The migration of potentially toxic elements during the recultivation of the uranium mining deposit in Mecsek. Acta Hydrol Slovaca, 20, 210-7.

Kim, H. Y. (2014). Analysis of variance (ANOVA) comparing means of more than two groups. Restorative dentistry & endodontics, 39(1), 74-77.

Kinuthia, J. M., O'Farrell, M., Sabir, B. B., & Wild, S. (2001). A preliminary study of the cementitious properties of wastepaper sludge ash ground granulated blast-furnace slag (WSA-GGBS) blends. In Recovery and Recycling of Paper (pp. 93-104). Thomas Telford Publishing.

Kolawole, T. O., Iyiola, O., Ibrahim, H., & Isibor, R. A. (2023). Contamination, ecological and health risk assessments of potentially toxic elements in soil around a municipal solid waste disposal facility in Southwestern Nigeria. Journal of Trace Elements and Minerals, 5, 100083.

Kollmann, F., & Fengel, D. (1965). Changes in the chemical composition of wood by thermal treatment. Holz als Roh-und Werkstoff, 23, 461-468.

Koul, B., Yakoob, M., & Shah, M. P. (2022). Agricultural waste management strategies for environmental sustainability. Environmental Research, 206, 112285.

Kovacova, Z., Demcak, S., Balintova, M., Pla, C., & Zinicovscaia, I. (2020). Influence of wooden sawdust treatments on Cu (II) and Zn (II) removal from water. Materials, 13(16), 3575.

Kratochvil, D., & Volesky, B. (1998). Advances in the biosorption of heavy metals. Trends in biotechnology, 16(7), 291-300.

Król, A. (201)1. "Problems of assessment of heavy metals leaching from construction materials to the environment." Archit. Civ. Eng. Environ. 3: 71–76.

Kulkarni, S., & Kaware, J. (2014). Regeneration and recovery in adsorption-a review. Int. J. Innov. Sci. Eng. Technol, 1(8), 61-64.

Kushwaha, A. K., Gupta, N., & Chattopadhyaya, M. C. (2017). Adsorption behavior of lead onto a new class of functionalized silica gel. Arabian Journal of Chemistry, 10, S81-S89.

Le Floch, A., Jourdes, M., & Teissedre, P. L. (2015). Polysaccharides and lignin from oak wood used in cooperage: Composition, interest, assays: A review. Carbohydrate research, 417, 94-102.

Lessard, J. M., Omran, A., Tagnit-Hamou, A., & Gagne, R. (2017). Feasibility of using biomass fly and bottom ashes in dry-cast concrete production. Construction and Building Materials, 132, 565-577.

Li, X., Ma, W., Li, H., Zhang, Q., & Liu, H. (2020). Sulfur-functionalized metal-organic frameworks: Synthesis and applications as advanced adsorbents. Coordination Chemistry Reviews, 408, 213191. https://doi.org/10.1016/j.ccr.2020.213191.

Li, Q., Wang, Y., Li, Y., Li, L., Tang, M., Hu, W., ... & Ai, S. (2022). Speciation of heavy metals in soils and their immobilization at micro-scale interfaces among diverse soil components. Science of the Total Environment, 825, 153862.

Li, X., Bertos, M. F., Hills, C. D., Carey, P. J., & Simon, S. (2007). Accelerated carbonation of municipal solid waste incineration fly ashes. Waste management, 27(9), 1200-1206.

Liu, J., Wu, D., Tan, X., Yu, P., & Xu, L. (2023). Review of the Interactions between Conventional Cementitious Materials and Heavy Metal Ions in Stabilization/Solidification Processing. Materials, 16(9), 3444.

Liu, J., Zha, F., Xu, L., Kang, B., Yang, C., Zhang, W., ... & Liu, Z. (2020). Zinc leachability in contaminated soil stabilized/solidified by cement-soda residue under freeze-thaw cycles. Applied Clay Science, 186, 105474.

Liu, M., Deng, Y., Zhan, H., & Zhang, X. (2002). Adsorption and desorption of copper (II) from solutions on new spherical cellulose adsorbent. Journal of Applied Polymer Science, 84(3), 478-485.

Low, K. S., Lee, C. K., & Wong, S. L. (1995). Effect of dye modification on the sorption of copper by coconut husk. Environmental technology, 16(9), 877-883.

Maherzi, W., Ennahal, I., Bouaich, F. Z., Benzerzour, M., Rais, Z., Mamindy-Pajany, Y., & Abriak, N. E. (2023). Assessment of Dynamic Surface Leaching of Monolithic Polymer Mortars Comprised of Wastes. Materials, 16(6), 2150.

Malviya, R., and R. Chaudhary. 2006. “Factors affecting hazardous waste solidification/stabilization: A review.” J. Hazard. Mater. 137 (1): 267–276.https://doi.org/10.1016/j.jhazmat.2006.01.065.

Martínez-García, R., Jagadesh, P., Zaid, O., Șerbănoiu, A. A., Fraile-Fernández, F. J., de Prado-Gil, J., ... &Grădinaru, C. M. (2022). The present state of the use of waste wood ash as an eco-efficient construction material: A review. Materials, 15(15), 5349. https://doi.org/10.3390/ma15155349.

Martínez-Lage, I., M. Velay-Lizancos, P. Vázquez-Burgo, M. Rivas-Fernández, C. Vázquez-Herrero, A. Ramírez-Rodríguez, and M. Martín-Cano.(2016). “Concretes and mortars with waste paper industry: Biomass ash and dregs.” J. Environ. Manage. 181: 863–873. https:// doi.org/10.1016/j.jenvman.2016.06.052.

Masud, F., R. S. Edwin, and I. S. Jaliki. (2021). “Leaching assessment of high-performance mortar containing high volume fly ash as cement replacement (effect of pH level and particle size).” IOP Conf. Seri.: Earth Environ. Sci. 871 (1): 012014. https://doi.org/10.1088/1755-1315/871/1 /012014.

Mbokazi L. (2021). Determination of radionuclides and potentially toxic elements associated with the spoil bank remediation on the environment at the abandoned uranium mining area [Unpublished Ph. D. dissertation]. Hungarian University of Agricultural and Life Sciences.

Meez, E., Rahdar, A., & Kyzas, G. Z. (2021). Sawdust for the removal of heavy metals from water: a review. Molecules, 26(14), 4318.

Méndez, A., Barriga, S., Fidalgo, J. M., & Gascó, G. (2009). Adsorbent materials from paper industry waste materials and their use in Cu (II) removal from water. Journal of Hazardous Materials, 165(1-3), 736-743.

Menikpura, S. N. M., J. Sang-Arun, and M. Bengtsson. 2016. “Assessment of environmental and economic performance of waste-to-energy facilities in Thai cities.” Renewable Energy 86: 576–584. https://doi.org/10 .1016/j.renene.2015.08.054.

Mohan, D., & Singh, K. P. (2002). Single-and multi-component adsorption of cadmium and zinc using activated carbon derived from bagasse—an agricultural waste. Water research, 36(9), 2304-2318.

Mohebbi, M., Rajabipour, F., & Scheetz, B. E. (2015, May). Reliability of loss on ignition (LOI) test for determining the unburned carbon content in fly ash. In World of Coal Ash Conference (WOCA), Nashville, TN, May (pp. 5-7).

Mondal, M., Manoli, K., Ray, A .(2019) Removal of arsenic(III) from aqueous solution by concrete-based adsorbents. Volume 98, January2020 Can. J. Chem. Eng. 1–7, 2019.

Monosi, S., D. Sani, and M. Letizia Ruello. 2012. “Reuse of paper mill ash in plaster blends.” Open Waste Manage. J. 5 (1): 5–18. https://doi.org /10.2174/1876400201205010005.

Moon, S., Lee, J., Yu, J. M., Choi, H., Choi, S., Park, J., ... & Park, Y. J. (2023). Association between environmental cadmium exposure and increased mortality in the US National Health and Nutrition Examination Survey (1999–2018). Journal of Exposure Science & Environmental Epidemiology, 1-9.

Moyib, O. K., Ayedun, M. A., Awokoya, O. J., & Omotola, O. E. (2017). Waste printing paper as analogous adsorbents for PTEs in aqueous solution. Nigerian Journal of Chemical Research, 22(1), 29-38.

Muhammad, S., Ullah, R., & Jadoon, I. A. (2019). Heavy metals contamination in soil and food and their evaluation for risk assessment in the Zhob and Loralai valleys, Baluchistan province, Pakistan. Microchemical Journal, 149, 103971.

Nag, S., Biswas, S. (2021). Cellulose-Based Adsorbents for Heavy Metal Removal. In: Inamuddin, Ahamed, M., Lichtfouse, E., Asiri, A. (eds) Green Adsorbents to Remove Metals, Dyes and Boron from Polluted Water. Environmental Chemistry for a Sustainable World, vol 49. Springer, Cham. https://doi.org/10.1007/978-3-030-47400-3_5.

Nascimento, L. C., Junior, G. B., de Castro Xavier, G., Monteiro, S. N., Vieira, C. M. F., de Azevedo, A. R. G., & Alexandre, J. (2023). Use of wood bottom ash in cementitious materials: a review. Journal of Materials Research and Technology.

Naser, H., Horváth, M., & Czinkota, I. (2023). Incorporation of Adsorbent Ash with Potentially Toxic Elements into Mortar: A Sustainable Approach. Journal of Hazardous, Toxic, and Radioactive Waste, 27(1), 04022037.

Nata, I. F., Irawan, C., Putra, M. D., Wijayanti, H., Dewi, Y. S., & Meliana, Y. (2022). A Sustainable Amine Magnetic Biocomposite Based on Rice Husk–Sugarcane Bagasse Fiber for Lead and Contaminant Adsorption in Aqueous Solution. Magnetochemistry, 8(12), 183.

Nickel, N. (2007). Monitored Natural Attenuation of Inorganic Contaminants in Ground Water.

Ngo, S. H., Huynh, T. P., Thi Le, T. T., & Thi Mai, N. H. (2018, June). Effect of high loss on ignition-fly ash on properties of concrete fully immersed in sulfate solution. In IOP conference series: materials science and engineering (Vol. 371, p. 012007). IOP Publishing.ement of ASTM C 618 .

Nguyen, B. T., Do, D. D., Nguyen, T. X., Nguyen, V. N., Nguyen, D. T. P., Nguyen, M. H., ... & Bach, Q. V. (2020). Seasonal, spatial variation, and pollution sources of heavy metals in the sediment of the Saigon River, Vietnam. Environmental Pollution, 256, 113412.

Nordstokke, D., Colp, S.M. (2014). Factorial Design. In: Michalos, A.C. (eds) Encyclopedia of Quality of Life and Well-Being Research. Springer, Dordrecht. https://doi.org/10.1007/978-94-007-0753-5_982

Núñez-Delgado, A., Fernández-Sanjurjo, M. J., Álvarez-Rodríguez, E., Cutillas-Barreiro, L., Nóvoa-Muñoz, J., and Arias-Estévez, M. (2015). Cr (VI) sorption/desorption on pine sawdust and oak wood ash. International Journal of Environmental Research and Public Health, 12(8), 8849-8860.

O'Connell, D. W., Birkinshaw, C., and O'Dwyer T., F. (2008). "Heavy metal adsorbents prepared from the modification of cellulose: A review," Bioresource Technology 99, 6709-6724. DOI: 10.1016/j.biortech.2008.01.036.

Obasi, P. N., & Akudinobi, B. B. (2020). Potential health risk and levels of heavy metals in water resources of lead–zinc mining communities of Abakaliki, southeast Nigeria. Applied Water Science, 10(7), 1-23.

Obernberger I, Supancic K (2009) Possibilities of ash utilisation from biomass combustion plants. Proceedings of the 17th European biomass conference & exhibition.

Okoro, H.K., Fatoki, O.S., Adekola, F.A., Ximba, Bhekumusa, J. and Snyman, Re.G. (2012), "A Review of Sequential Extraction Procedures for Heavy Metals Speciation in Soil and Sediments", Journal of Environmental & Analytical Toxicology, Vol. 01 No. S1, pp. 1–9.

Omur, T., Miyan, N., Kabay, N., Birol, B., &Oktay, D. (2023). Characterization of ferrochrome ash and blast furnace slag based alkali-activated paste and mortar. Construction and Building Materials, 363, 129805. https://doi.org/10.1016/j.conbuildmat.2022.129805.

Pavlíková, M., Zemanová, L., Pokorný, J., Záleská, M., Jankovský, O., Lojka, M., ... & Pavlík, Z. (2018). Valorization of wood chips ash as an eco-friendly mineral admixture in mortar mix design. Waste Management, 80, 89-100.

Pearson, R. G. (1963). Hard and soft acids and bases. Journal of the American Chemical society, 85(22), 3533-3539.

Pham, B. N., Kang, J. K., Lee, C. G., & Park, S. J. (2021). Removal of heavy metals (Cd2+, Cu2+, Ni2+, Pb2+) from aqueous solution using Hizikia fusiformis as an algae-based bioadsorbent. Applied Sciences, 11(18), 8604.

Pillai, S. B. (2020). Adsorption in water and used water purification. Handbook of Water and Used Water Purification, 1-22.

Pino, G. H., De Mesquita, L. M. S., Torem, M. L., & Pinto, G. A. S. (2006). Biosorption of cadmium by green coconut shell powder. Minerals engineering, 19(5), 380-387.

Pitsari, S., Tsoufakis, E., & Loizidou, M. (2013). Enhanced lead adsorption by unbleached newspaper pulp modified with citric acid. Chemical Engineering Journal, 223, 18-30.

Pourret O, Hursthouse A. It's Time to Replace the Term "Heavy Metals" with "Potentially Toxic Elements" When Reporting Environmental Research. International Journal of Environmental Research and Public Health. 2019; 16(22):4446. https://doi.org/10.3390/ijerph16224446.

Puech, J. L. (1978). Substances Phenoliques Des Eaux-De-vie d'armagnac. III. Sur La Presence Des Ethoxyles Dans La Lignine D'extraction Et Les Produits De SA Degradation.

Putra, W. P., Kamari, A., Yusoff, S. N. M., Ishak, C. F., Mohamed, A., Hashim, N., & Isa, I. M. (2014). Biosorption of Cu (II), Pb (II) and Zn (II) ions from aqueous solutions using selected waste materials: Adsorption and characterisation studies. Journal of Encapsulation and Adsorption Sciences, 2014.

Putro, J. N., Santoso, S. P., Soetaredjo, F. E., Ismadji, S., & Ju, Y. H. (2019). Nanocrystalline cellulose from waste paper: Adsorbent for azo dyes removal. Environmental Nanotechnology, Monitoring & Management, 12, 100260.

Qiu, Y., Cao, S., Chen, F., You, S., & Zhang, Y. (2020). Synthesis of calcium silicate as paper filler with desirable particle size from desilication solution of silicon-containing waste residues. Powder technology, 368, 137-148.

Raheem AA, Adenuga OA (2013) Wood ash from bread bakery as partial replacement for cement in concrete. Int J Sustain Constr 4(1):75–81.

Ray, A. B., Selvakumar, A., & Tafuri, A. N. (2006). Removal of selected pollutants from aqueous media by hardwood mulch. Journal of hazardous materials, 136(2), 213-218.

Rehman, K., Fatima, F., Waheed, I., & Akash, M. S. H. (2018). Prevalence of exposure of heavy metals and their impact on health consequences. Journal of cellular biochemistry, 119(1), 157-184.

Rehman, N., de Miranda, M. I. G., Rosa, S. M., Pimentel, D. M., Nachtigall, S. M., & Bica, C. I. (2014). Cellulose and nanocellulose from maize straw: an insight on the crystal properties. Journal of Polymers and the Environment, 22, 252-259.

Rojas-Valencia, M. N., Galeana-Olvera, E., Fernández-Rojas, D. Y., Mendoza-Buenrostro, C., & Alejandro, H. (2018). Isolation of cellulose nanofibrils from coconut waste for the production of sewing thread. Adv Mater Sci, 3(1), 1-3.

Sağın, E. U., H. Böke, N. Aras, and Ş Yalçın. 2012. "Determination of $CaCO_3$ and SiO_2 content in the binders of historic lime mortars." Mater. Struct. 45 (6): 841–849. https://doi.org/10.1617/s11527-011-9802-1.

Sahin, H. T., & Arslan, M. B. (2008). A study on physical and chemical properties of cellulose paper immersed in various solvent mixtures. International Journal of Molecular Sciences, 9(1), 78-88.

Salih, A. M. (2018). The purification of industrial wastewater to remove heavy metals and investigation into the use of zeolite as a remediation tool.

Sathonsaowaphak, A., Chindaprasirt, P., &Pimraksa, K. (2009). Workability and strength of lignite bottom ash geopolymer mortar. Journal of Hazardous Materials, 168(1), 44-50. https://doi.org/10.1016/j.jhazmat.2009.01.120.

Saveyn, H., P. Eder, E. Garbarino, L. Muchova, O. Hjelmar, H. A. van der Sloot, and A. Oberender.(2014). Study on methodological aspects regarding limit values for pollutants in aggregates in the context of the possible development of end-of-waste criteria under the EU Waste Framework Directive. JRC Technical Rep. Luxembourg: Publications Office of the European Union.

Schaefer, H. R., Dennis, S., & Fitzpatrick, S. (2020). Cadmium: Mitigation strategies to reduce dietary exposure. Journal of food science, 85(2), 260-267.

Šćiban, M., & Klašnja, M. (2003). Optimization on of usage of wood sawdust adsorbent of heavy metal ions from water. ISIRR 3:51–56.

Šćiban, M., Radetić, B., Kevrešan, Ž., & Klašnja, M. (2007). Adsorption of heavy metals from electroplating wastewater by wood sawdust. Bioresource technology, 98(2), 402-409.

Segui, P., Aubert, J. E., Husson, B., & Measson, M. (2013). Valorization of wastepaper sludge ash as main component of hydraulic road binder. Waste and Biomass Valorization, 4, 297-307.

Sereshti, H., Heravi, Y. E., & Samadi, S. (2012). Optimized ultrasound-assisted emulsification microextraction for simultaneous trace multielement determination of heavy metals in real water samples by ICP-OES. Talanta, 97, 235-241.

Setyono, D., & Valiyaveettil, S. (2016). Functionalized paper—A readily accessible adsorbent for removal of dissolved heavy metal salts and nanoparticles from water. Journal of hazardous materials, 302, 120-128.

Sharma, A., Thakur, M., Bhattacharya, M., Mandal, T., & Goswami, S. (2019). Commercial application of cellulose nano-composites–A review. Biotechnology Reports, 21, e00316.

Sharma, D. C., & Forster, C. F. (1994). A preliminary examination into the adsorption of hexavalent chromium using low-cost adsorbents. Bioresource Technology, 47(3), 257-264.

Shi, C., & Spence, R. (2004). Designing of cement-based formula for solidification/stabilization of hazardous, radioactive, and mixed wastes. *Critical Reviews in Environmental Science and Technology*, *34*(4), 391-417.

Shiva Kumar, D., & Srikantaswamy, S. (2014). Factors affecting on mobility of heavy metals in soil environment. International Journal for Scientific Research & Development, 2(3), 201-203.

Siddique, R., Singh, M., & Singhal, A. K. (2019). Use of unprocessed wood ash as partial replacement of sand in concrete. ACI Materials Journal, 116(6), 77-86.

Sidhu, V., Barrett, K., Park, D. Y., Deng, Y., Datta, R., & Sarkar, D. (2021). Wood mulch coated with iron-based water treatment residuals for the abatement of metals and phosphorus in simulated stormwater runoff. Environmental Technology & Innovation, 21, 101214.

Singh, V., Pant, N., Sharma, R. K., Padalia, D., Rawat, P. S., Goswami, R., ... & Deifalla, A. M. (2023). Adsorption Studies of Pb (II) and Cd (II) Heavy Metal Ions from Aqueous Solutions Using a Magnetic Biochar Composite Material. Separations, 10(7), 389.

Sirviö, J. A., & Visanko, M. (2020). Lignin-rich sulfated wood nanofibers as high-performing adsorbents for the removal of lead and copper from water. Journal of hazardous materials, 383, 121174.

Simón, D., Palet, C., Costas, A., and Cristóbal, A. (2022). Agro-industrial waste as potential heavy metal adsorbents and subsequent safe disposal of spent adsorbents. Water, 14(20), 3298.

Sklivaniti, V., Tsakiridis, P. E., Katsiotis, N. S., Velissariou, D., Pistofidis, N., Papageorgiou, D., & Beazi, M. (2017). Valorisation of woody biomass bottom ash in Portland cement: A characterization and hydration study. Journal of environmental chemical engineering, 5(1), 205-213.

Soleimanifar, H., Deng, Y., Barrett, K., Feng, H., Li, X., & Sarkar, D. (2019). Water treatment residual-coated wood mulch for addressing urban stormwater pollution. Water Environment Research, 91(6), 523-535.

Soleimanifar, H., Deng, Y., Wu, L., & Sarkar, D. (2016). Water treatment residual (WTR)-coated wood mulch for alleviation of toxic metals and phosphorus from polluted urban stormwater runoff. Chemosphere, 154, 289-292.

Šoštarić, T. D., Petrović, M. S., Pastor, F. T., Lončarević, D. R., Petrović, J. T., Milojković, J. V., & Stojanović, M. D. (2018). Study of heavy metals biosorption on native and alkali-treated apricot shells and its application in wastewater treatment. Journal of Molecular Liquids, 259, 340-349.

Spathi, C., Young, N., Heng, J. Y., Vandeperre, L. J., & Cheeseman, C. R. (2015). A simple method for preparing super-hydrophobic powder from paper sludge ash. Materials Letters, 142, 80-83. Spectroscopy", Analytical Chemistry, Vol. 46 No. 13, pp. 1110A-1120A.

Sridhar, V., & Park, H. (2020). Extraction of microfibrillar cellulose from wastepaper by NaOH/urethane aqueous system and its utility in removal of lead from contaminated water. Materials, 13(12), 2850.

Stanojković-Sebić, A., Maksimović, J., Dinić, Z., Poštić, D., Iličić, R., Stanojković, A., & Pivić, R. (2017). Microelements and heavy metals content in frequently utilized medicinal plants collected from the power plant area. Natural Product Communications, 12(2), 1934578X1701200211.

Steenari, B. M., Karlsson, L. G., & Lindqvist, O. (1999a). Evaluation of the leaching characteristics of wood ash and the influence of ash agglomeration. Biomass and Bioenergy, 16(2), 119-136.

Steenari, B. M., Schelander, S., & Lindqvist, O. (1999b). Chemical and leaching characteristics of ash from combustion of coal, peat and wood in a 12 MW CFB–a comparativ study. Fuel, 78(2), 249-258.

Stefanidou, M., &Pavlidou, E. (2018). Scanning mortars to understand the past and plan the future for the maintenance of monuments. Scanning, 2018.

Sthiannopkao, S., and S. Sreesai. 2009. "Utilization of pulp and paper industrial wastes to remove heavy metals from metal finishing wastewater." J. Environ. Manage. 90 (11): 3283–3289. https://doi.org/10.1016/j .jenvman.2009.05.006.

Stiernström, S., Enell, A., Wik, O., Hemström, K., & Breitholtz, M. (2014). Influence of leaching conditions for ecotoxicological classification of ash. Waste management, 34(2), 421-429.

Štirmer, N., Carević, I., Milovanović, B., & Baricevic, A. (2018). Market Potential of Wood Biomass Ash Utilisation in Cement Composites-Croatian Case Study. Proceedings of the SynerCrete, 18.

Štirmer, N., & Carević, I. (2022). Utilization of Wood Biomass Ash in Concrete Industry. Biomass, Biorefineries and Bioeconomy.

Sundarraj, A. A., & Ranganathan, T. V. (2018). A review on cellulose and its utilization from agro-industrial waste. Drug Invent. Today, 10(1), 89-94.

Syeda, H. I., & Yap, P. S. (2022). A review on three-dimensional cellulose-based aerogels for the removal of heavy metals from water. Science of The Total Environment, 807, 150606.

Szynkowska, M. I., Pawlaczyk, A., & Maćkiewicz, E. (2018). Bioaccumulation and biomagnification of trace elements in the environment. Recent advances in trace elements, 251-276.

Tan, W. T., Ooi, S. T., & Lee, C. K. (1993). Removal of chromium (VI) from solution by coconut husk and palm pressed fibres. Environmental technology, 14(3), 277-282.

Tan, Y., Zhang, Z., Yang, D., Dong, J., Cheng, X., & Yu, H. (2023). Immobilization of Zn (II) and Cu (II) in basic magnesium-sulfate-cementitious material system: Properties and mechanism. Journal of Hazardous Materials, 446, 130720.

Tang, N., Niu, C. G., Li, X. T., Liang, C., Guo, H., Lin, L. S., ... & Zeng, G. M. (2018). Efficient removal of Cd2+ and Pb2+ from aqueous solution with amino-and thiol-functionalized activated carbon: Isotherm and kinetics modeling. Science of the Total Environment, 635, 1331-1344.

Thompson, K. (2017). Large-Area Quantitative Phase Mapping in the Scanning Electron Microscope. Microscopy Today, 25(2), 36-45.

Tiwari, M. K., S. Bajpai, U. K. Dewangan, and R. K. Tamrakar. (2015). "Suitability of leaching test methods for fly ash and slag: A review." J. Radiat. Res. Appl. Sci. 8 (4): 523–537. https://doi.org/10.1016/j .jrras.2015.06.003.

Tóth, P. – Bulla, M. – Nagy, G. (2011): Energetika. [Energetics.] Pannon Egyetem, Veszprém. 204 p. (in Hungarian).

Trussell, S., & Spence, R. D. (1994). A review of solidification/stabilization interferences. Waste management, 14(6), 507-519.

Tsuchikawa, S., & Schwanninger, M. (2013). A review of recent near-infrared research for wood and paper (Part 2). Applied Spectroscopy Reviews, 48(7), 560-587.

Twaróg, A., Mamak, M., Sechman, H., Rusiniak, P., Kasprzak, E., & Stanek, K. (2020). Impact of the landfill of ashes from the smelter on the soil environment: case study from the South Poland, Europe. Environmental geochemistry and health, 42(5), 1453-1467.

Ucaroglu, S., & Talinli, I. (2012). Recovery and safer disposal of phosphate coating sludge by solidification/stabilization. Journal of environmental management, 105, 131-137.

Udoeyo, F. F., Inyang, H., Young, D. T., & Oparadu, E. E. (2006). Potential of wood waste ash as an additive in concrete. Journal of materials in civil engineering, 18(4), 605-611.

Ur Rehman, I., Ishaq, M., Ali, L., Khan, S., Ahmad, I., Din, I. U., & Ullah, H. (2018). Enrichment, spatial distribution of potential ecological and human health risk assessment via toxic metals in soil and surface water ingestion in the vicinity of Sewakht mines, district Chitral, Northern Pakistan. *Ecotoxicology and environmental safety*, *154*, 127-136.

USEPA (United States Environmental Protection Agency). (1992a). Toxicity characteristic leaching procedure (TCLP) (Accessed July 1992). Method No. 1311. US Environmental Protection Agency (USEPA): Washington, DC, USA.

USEPA (United States Environmental Protection Agency). (1992b). Synthetic Precipitation Leaching Procedure SW-846. Method No. 1312. US Environmental Protection Agency (USEPA): Washington, DC, USA.

Vadakkekara, G. J., Thomas, S., & Nair, C. R. (2019). Maleic acid modified cellulose for scavenging lead from water. International journal of biological macromolecules, 129, 293-304.

Van der Sloot, H. A., & Kosson, D. S. (2010). Leaching assessment methodologies for disposal and use of bauxite residues. Hans van der Sloot Consultancy.

van der Sloot, H. A., and A. van Zomeren. (2012). "Characterisation leaching tests and associated geochemical speciation modelling to assess long term release behaviour from extractive wastes." Mine Water Environ31 (2): 92–103. https://doi.org/10.1007/s10230-012-0182-8.

van der Sloot, H. A., Kosson, D. S., & van Zomeren, A. (2018). Landfilling of different kinds of waste: Leaching behavior. Solid Waste Landfilling: Concepts, Processes, Technologies. Amsterdã: Elsevier, cap, 20, 1077-1093.

Veglio, F., Beolchini, F. (1997) Removal of metals by biosorption: a review. Hydrometallurgy. 44, 301 -316.

Vespa, M., Dähn, R., & Wieland, E. (2014). Competition behaviour of metal uptake in cementitious systems: An XRD and EXAFS investigation of Nd-and Zn-loaded 11 Å tobermorite. Physics and Chemistry of the Earth, Parts A/B/C, 70, 32-38.

Vinutha, H. P., Poornima, B., & Sagar, B. M. (2018). Detection of outliers using interquartile range technique from intrusion dataset. In Information and decision sciences (pp. 511-518). Springer, Singapore.

Vu, T. H., & Gowripalan, N. (2018). Mechanisms of heavy metal immobilisation using geopolymerisation techniques–A review. Journal of Advanced Concrete Technology, 16(3), 124-135.

Wajima, T. (2013). Conversion of waste paper sludge into heavy metal adsorbent using sulfur impregnation. Advanced Materials Research, 699, 759-764.

Wang, D., & Wang, Q. (2022). Clarifying and quantifying the immobilization capacity of cement pastes on heavy metals. Cement and Concrete Research, 161, 106945.

Wang, H., Zhu, Z., Pu, S., & Song, W. (2022). Solidification/Stabilization of Pb2+ and Cd2+ contaminated soil using fly ash and GGBS based geopolymer. Arabian Journal for Science and Engineering, 47(4), 4385-4400.

Wang, J., Liu, M., Duan, C., Sun, J., & Xu, Y. (2019). Preparation and characterization of cellulose-based adsorbent and its application in heavy metal ions removal. Carbohydrate polymers, 206, 837-843.

Wang, R., & Haller, P. (2022). Applications of wood ash as a construction material in civil engineering: a review. Biomass Conversion and Biorefinery, 1-21.

Wang, Y., Han, F., & Mu, J. (2018). Solidification/stabilization mechanism of Pb (II), Cd (II), Mn (II) and Cr (III) in fly ash based geopolymers. Construction and Building Materials, 160, 818-827.

Wielgosiński, G., Czerwińska, J., & Szufa, S. (2021). Municipal solid waste mass balance as a tool for calculation of the possibility of implementing the circular economy concept. Energies, 14(7), 1811.

Wong, H. S., R. Barakat, A. Alhilali, M. Saleh, and C. R. Cheeseman. (2015). "Hydrophobic concrete using waste paper sludge ash." Cem. Concr. Res. 70: 9–20. https://doi.org/10.1016/j.cemconres.2015.01.005.

Wong, L. S., Chandran, S. N., Rajasekar, R. R., & Kong, S. Y. (2022). Pozzolanic characterization of waste newspaper ash as a supplementary cementing material of concrete cylinders. Case Studies in Construction Materials, 17, e01342.

Xia, Z., Baird, L., Zimmerman, N., & Yeager, M. (2017). Heavy metal ion removal by thiol functionalized aluminum oxide hydroxide nanowhiskers. Applied Surface Science, 416, 565-573. https://doi.org/10.1016/j.apsusc.2017.04.095.

Xiong, X., Liu, X., Iris, K. M., Wang, L., Zhou, J., Sun, X., ... & Tsang, D. C. (2019). Potentially toxic elements in solid waste streams: Fate and management approaches. Environmental pollution, 253, 680-707.

Xiuling, L. (2021, May). Preparation of Graphene Oxide-Molecular Sieve Composite Adsorbent and its Adsorption Performance for Heavy Metals in Water. In IOP Conference Series: Earth and Environmental Science (Vol. 784, No. 1, p. 012022). IOP Publishing.

Xu, P., Q. Zhao, W. Qiu, and Y. Xue. (2019). "The evaluation of the heavy metal leaching behavior of MSWI-FA added alkali-activated materials bricks by using different leaching test methods." Int. J. Environ. Res. Public Health 16 (7): 1151. https://doi.org/10.3390/ijerph16071151.

Yang, J., Cao, J., Xing, G., & Yuan, H. (2015). Lipid production combined with biosorption and bioaccumulation of cadmium, copper, manganese and zinc by oleaginous microalgae Chlorella minutissima UTEX2341. Bioresource technology, 175, 537-544.

Yang, L., Liu, G., Di, L., Wu, X., You, W., & Huang, B. (2019). Occurrence, speciation, and risks of trace metals in soils of greenhouse vegetable production from the vicinity of industrial areas in the Yangtze River Delta, China. Environmental Science and Pollution Research, 26(9), 8696–8708.

Yliniemi J, Nugteren H, Illikainen M, Tiainen M, Weststrate R, Niinimaki J (2016) Lightweight aggregates produced by granulation of peat-wood fly ash with alkali activator. Int J Miner Process 149:42–49. https://doi.org/10.1016/j.minpro.2016.02.

Zając, G., Szyszlak-Bargłowicz, J., Gołębiowski, W., & Szczepanik, M. (2018). Chemical characteristics of biomass ashes. Energies, 11(11), 2885.

Zhang, H., Xu, F., Xue, J., Chen, S., Wang, J., & Yang, Y. (2020). Enhanced removal of heavy metal ions from aqueous solution using manganese dioxide-loaded biochar: Behaviour and mechanism. Scientific reports, 10(1), 1-13.

Zmamou, H., N. Leblanc, D. Levacher, and J. Kubiak. (2021). "Recycling of high quantities of wastepaper sludge ash for production of blended cements and alternative materials." Environ. Technol. Innovation 23: 101524. https://doi.org/10.1016/j.eti.2021.101524.

Zohar, I., & Haruzi, P. (2021). Image Analysis for Spectroscopic Elemental Dot Maps: P, Al, and Ca Associations in Water Treatment Residuals as a Case Study. Frontiers in Environmental Chemistry, 2, 719300. https://doi.org/10.3389/fenvc.2021.719300.

A2. Tables presenting adsorption capacity of mulch samples at (0.5,1,5,10,50) mg L^{-1} initial concentrations.

Cadmium/Mulch

Initial Concentration (mg L^{-1})	Adsorption Capacity Cd (mg g^{-1})	Average (mg g^{-1})	SD
0.5	0.005		
0.5	0.005		
0.5	0.005	0.005	0
1	0.010		
1	0.010		
1	0.010	0.010	0
5	0.050		
5	0.050		
5	0.050	0.050	0
10	0.099		
10	0.099		
10	0.099	0.099	0
50	0.491		
50	0.492		
50	0.491	0.491	0

Copper/Mulch

Initial Concentration (mg L^{-1})	Adsorption Capacity Cu (mg g^{-1})	Average (mg g^{-1})	SD
0.5	0.005		
0.5	0.010		
0.5	0.005	0.007	0.003
1	0.010		
1	0.010		
1	0.010	0.010	0
5	0.049		
5	0.049		
5	0.049	0.049	0
10	0.099		
10	0.099		
10	0.099	0.099	0
50	0.495		
50	0.496		
50	0.494	0.495	0.001

Lead/Mulch

Initial Concentration (mg L^{-1})	Adsorption Capacity Pb (mg g^{-1})	Average (mg g^{-1})	SD
0.5	0.005		
0.5	0.005		
0.5	0.005	0.005	0
1	0.009		
1	0.010		
1	0.010	0.010	0
5	0.049		
5	0.049		
5	0.049	0.049	0
10	0.098		
10	0.098		
10	0.098	0.098	0
50	0.496		
50	0.496		
50	0.496	0.496	0

Zinc/Mulch

Initial Concentration (mg L^{-1})	adsorption Capacity Zn (mg g^{-1})	Average (mg g^{-1})	SD
0.5	0.003		
0.5	0.003		
0.5	0.003	0.003	0
1	0.006		
1	0.008		
1	0.008	0.007	0.001
5	0.047		
5	0.047		
5	0.047	0.047	0
10	0.095		
10	0.095		
10	0.095	0.095	0
50	0.482		
50	0.483		
50	0.482	0.482	0

A3. Tables presenting adsorption capacity of paper samples at (0.5,1,5,10,50) mg L^{-1} initial concentrations.

Cadmium /Paper

Initial Concentration (mg L^{-1})	Adsorption Capacity Cd (mg g^{-1})	Average (mg g^{-1})	SD
0.5	0.015		
0.5	0.015		
0.5	0.015	0.015	0
1	0.029		
1	0.030		
1	0.030	0.030	0
5	0.143		
5	0.142		
5	0.142	0.142	0.001
10	0.265		
10	0.267		
10	0.266	0.266	0.001
50	0.769		
50	0.762		
50	0.762	0.764	0.004

Copper /Paper

Initial Concentration (mg $^{L-1}$)	adsorption Capacity Cu (mg g^{-1})	Average (mg g^{-1})	SD
0.5	0.013		
0.5	0.013		
0.5	0.013	0.013	0
1	0.027		
1	0.027		
1	0.027	0.027	0
5	0.141		
5	0.140		
5	0.140	0.140	0
10	0.281		
10	0.283		
10	0.282	0.282	0.001
50	1.259		
50	1.261		
50	1.261	1.260	0.001

Lead/Paper

Initial Concentration (mg L^{-1})	adsorption Capacity Pb (mg g^{-1})	Average (mg g^{-1})	SD
0.5	0.016		
0.5	0.016		
0.5	0.016	0.016	0
1	0.030		
1	0.030		
1	0.030	0.030	0
5	0.150		
5	0.150		
5	0.150	0.150	0
10	0.299		
10	0.299		
10	0.299	0.299	0
50	1.484		
50	1.482		
50	1.482	1.483	0.001

Zinc/Paper

Initial Concentration (mg L^{-1})	adsorption Capacity Zn (mg g^{-1})	Average (mg g^{-1})	SD
0.5	0.013		
0.5	0.013		
0.5	0.012	0.013	0.001
1	0.027		
1	0.028		
1	0.028	0.028	0.001
5	0.144		
5	0.144		
5	0.141	0.143	0.002
10	0.266		
10	0.267		
10	0.264	0.266	0.002
50	0.814		
50	0.802		
50	0.802	0.806	0.006

A4. Tables presenting the statistical data results showing the effect of the initial concentration of PTEs in mg L^{-1}, the incorporated ash percentage %, the molarity of the used HNO_3, and the adsorbent type on the leaching concentrations of PTEs in mg kg^{-1}.

Initial concentration (AMA)

Initial concentration (mg L^{-1})	Cd (mg kg^{-1})	Cu (mg kg^{-1})	Pb (mg kg^{-1})	Zn (mg kg^{-1})
0.5	0.360	2.525	0.221	4.327
1	0.580	3.354	0.304	5.299
5	1.480	3.564	0.861	5.876
10	3.750	3.962	1.210	6.829
50	7.960	7.663	3.755	8.250
$p \leq 0.05$	<0.001	<0.001	<0.001	<0.001
LSD	0.200	0.180	0.130	0.300

Molarity (AMA)

Molarity (M)	Cd (mg kg^{-1})	Cu (mg kg^{-1})	Pb (mg kg^{-1})	Zn (mg kg^{-1})
0.43 M mean	1.250	0.692	0.065	1.700
4M mean	4.400	7.736	2.476	10.54
$p \leq 0.05$	<0.001	<0.001	<0.001	<0.001
LSD	0.120	0.120	0.080	0.190

Percentage (AMA)

Percentage	Cd (mg kg^{-1})	Cu (mg kg^{-1})	Pb (mg kg^{-1})	Zn (mg kg^{-1})
1% mean	1.670	3.960	0.750	3.610
3% mean	3.980	4.470	1.790	8.620
$p \leq 0.05$	<0.001	<0.001	<0.001	<0.001
LSD	0.120	0.120	0.080	0.190

Initial concentration (APA)

Initial concentration ($mg\ L^{-1}$)	Cd ($mg\ kg^{-1}$)	Cu ($mg\ kg^{-1}$)	Pb ($mg\ kg^{-1}$)	Zn ($mg\ kg^{-1}$)
0.5	0.071	0.342	0.140	1.326
1	0.194	0.760	0.351	1.535
5	1.018	1.637	1.106	2.052
10	2.021	2.410	2.488	3.161
50	4.545	8.335	10.271	4.653
LSD	0.188	0.267	0.127	0.149
$p \leq 0.05$	<0.001	<0.001	<0.001	<0.001

Molarity (APA)

Molarity (M)	Cd ($mg\ kg^{-1}$)	Cu ($mg\ kg^{-1}$)	Pb ($mg\ kg^{-1}$)	Zn ($mg\ kg^{-1}$)
0.43 M mean	0.123	0.570	0.400	0.318
4M mean	3.017	4.823	5.342	4.773
$p \leq 0.05$	<0.001	<0.001	<0.001	<0.001
LSD	0.119	0.169	0.080	0.094

Percentage (APA)

Percentage	Cd ($mg\ kg^{-1}$)	Cu ($mg\ kg^{-1}$)	Pb ($mg\ kg^{-1}$)	Zn ($mg\ kg^{-1}$)
1% mean	0.171	2.007	1.446	1.849
3% mean	2.969	3.386	4.296	3.242
$p \leq 0.05$	<0.001	<0.001	<0.001	<0.001
LSD	0.119	0.169	0.080	0.094