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Literature review of material recovery technologies

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Abbreviations

AAM	Alkali-Activated Material
CA	Citric Acid
CRM	Critical Raw Material
DC	Direct Current
EC	European Commission
ED	Electrodialytic
EDS	Electrodialytic Separation
EDTA	Ethylenediaminetetraacetic Acid
EDTMP	Ethylenediamine Tetra Methylenephosphonic Acid
EU	European Union
GHG	Greenhouse Gas
LA	Lactic Acid
L/S	Liquid/Solid
MSW	Municipal Solid Waste
MSWI	Municipal Solid Waste Incineration
MSWI-BA	Municipal Solid Waste Incineration - Bottom Ash
MSWI-FA	Municipal Solid Waste Incineration - Fly Ash
OA	Oxalic Acid
PEE	Phosphorus Extraction Efficiency
REE	Rare Earth Element
RMIS	Raw Material Information System
SRM	Secondary Raw Material
SSA	Sewage Sludge Ash
TRL	Technology Readiness Level
WBA	Wood Biomass Ash
WBA-BA	Wood Biomass Ash - Bottom Ash
WBA-FA	Wood Biomass Ash - Fly Ash

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Objective

The main objective of this deliverable as defined in Task 3.1 of the AshCycle project is to provide an overview of different extraction methods of the valuable elemental resources, i.e. P, heavy metals and rare earth elements (REEs), from selected ashes on different technology readiness levels stated in TRL 2-9. The extraction technologies prioritized for further investigation in this project are those based on waste acids or excess renewable energy so as to have an optimum environmental profile and provide benefits in the form of local symbioses. The extraction methods include all scarce and valuable chemical elements with concentrations above certain limits from ashes considered within the present project, i.e. sewage sludge ash (SSA), municipal solid waste incineration bottom ash (MSWI-BA) and MSWI fly ash (MSWI-FA), wood biomass bottom ash (WBA-BA) and WBA fly ash (WBA-FA). Apart from the extraction of valuable components, an important part of this survey is defining potential applications for the mineral residues after the extraction process. Therefore an additional focus of this review is placed on screening available technologies for a recovery of Critical Raw Materials (CRMs) and base metals from selected ashes and their possible application in construction and other sectors so as to fully close the loop of the material circle.

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Introduction

The European Commission (EC) has recognized the necessity to recover CRMs not only from virgin materials, but as much as possible also from secondary sources. In 2015, the document ”Closing the loop - An EU action plan for the Circular Economy [1]” expressed the ECs aim to support the Raw Materials Information System (RMIS) which would provide data on Secondary Raw Materials (SRMs). SRMs can be sources used instead of primary raw materials. SRMs can also be a source for CRMs, which are of high economic importance due to the eventual scarcity on the EU market or supply disruption. The EU thus wishes to promote the recycling of these materials, especially the recovery of CRMs, as part of the move toward a circular economy [1].

One CRM is the element P, which is an irreplaceable resource and an essential nutrient for the growth of organisms [2, 3]. Most P is currently extracted from phosphate rocks, which are the primary and non-renewable sources occurring in a limited number of deposits worldwide [4]. 74% of the global phosphate rock deposits are located in Morocco, while the rest are found in the US, China, South Africa, and Jordan [5]. These global P resources will be depleted within a few decades, which is why the EC placed P and phosphate rocks on the list of 20 CRMs presented in Figure 1 in 2020 [6]. Solutions for a P recovery from various secondary resources such as ashes are hence required. In addition, optimized use of P in agriculture and soil stabilization to prevent erosion is encouraged. REEs are also a limited resource and due to the high demand for them in current technological and industrial production, the EC placed them in the CRMs list, where they are divided into light (LREEs) and heavy REEs (HREEs) [6, 7].

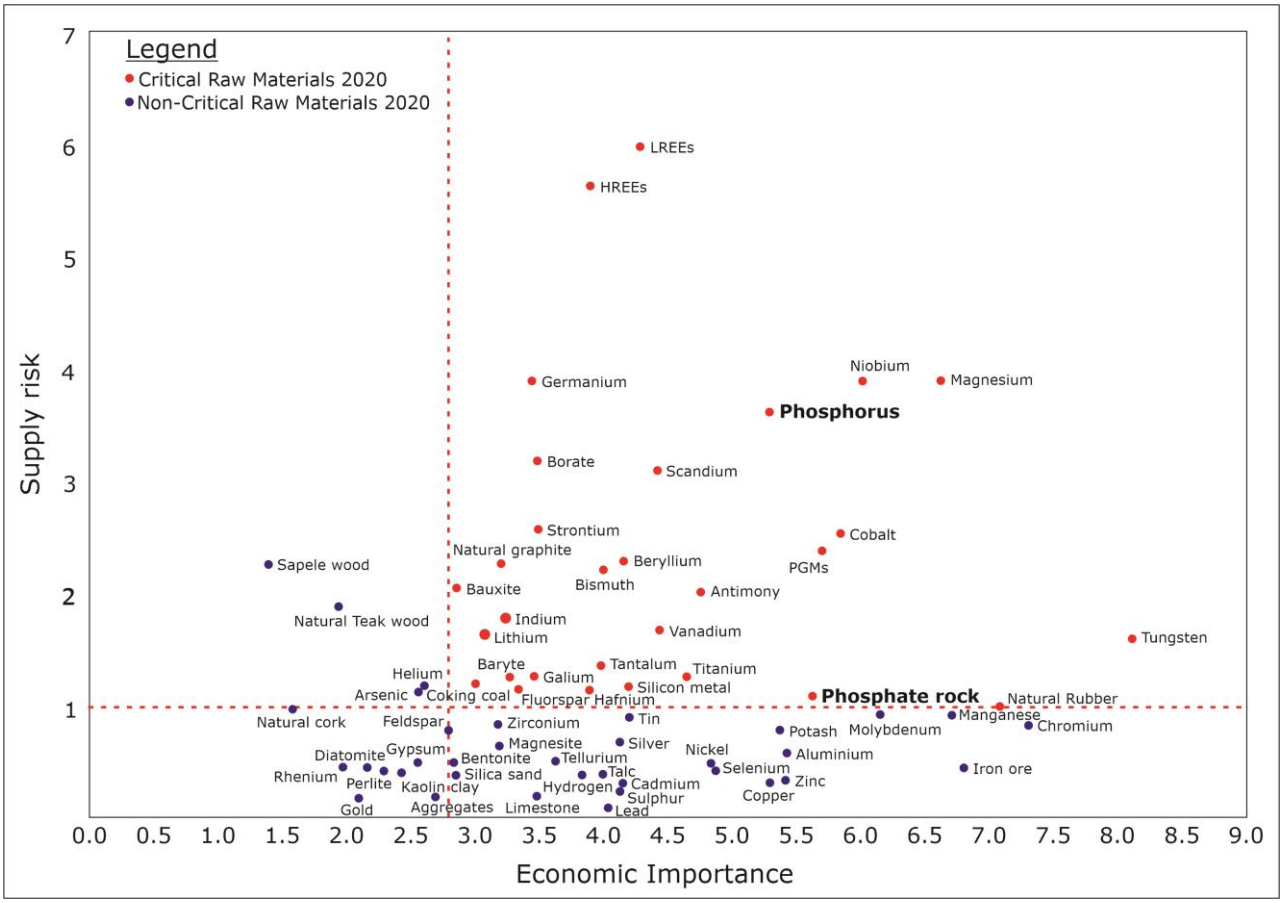

Figure 1: CRMs (list from 2020) in relation to the economic importance and supply risk, redrawn based on Ref. [6].

Apart from CRMs, there is a fundamental dependence on metals in different industries. The non-ferrous base metals Cu, Zn, and Pb are some of the most used metals in the world, only exceeded by Fe and Al. Projections predict an increase in the base demand for Cu by 140%, Zn by 81% and Pb by 46% compared to the 2010 demand until 2050 [8]. By the end of the 21st century, the projected demand is even higher for Cu and Zn (330% for Cu and 130% for Zn) [8]. At the same time, the expected depletion years (the year when the cumulative primary production exceeds the reserves) will decrease within the next 4-17 years without the peak year of primary production being reached for these elements [8]. Consequently, used ore grades become lower, leading to an increased energy demand for extraction, an effect typical for fossil fuels, and resulting in increased Greenhouse Gas (GHG) emissions [9, 10].

Table 1 summarizes concentrations of P and selected heavy metals reported in various ashes with typical ore concentrations when mined.

Table 1: Minimum and maximum content of P and heavy metals in treated SSA, MSWI-FA, MSWI-BA, WBA-FA and WBA-BA from existing literature compared to typical ore concentrations.

element	SSA [2, 4, 11–15]	MSWI-FA [16–18]	MSWI-BA [16]	WBA-FA [19–21]	WBA-BA [20]	Typical ore concentration [22, 23]
P (g/kg)	35-99	4-5	-	10-23	8-17	110-160
Zn (mg/kg)	895-2823	9000-70000	610-7800	446-2274	74-234	50000-150000
Pb (mg/kg)	70-460	5300-26000	100-13700	11-177	5-80	300000-400000
Cu (mg/kg)	423-839	600-3200	190-8200	89-161	65-111	5000-20000
Cr (mg/kg)	78-460	140-1100	23-3200	18-101	25-70	310000
Cd (mg/kg)	4-126	50-450	0.3-70	7-16	0.1-0.5	1000-10000

Legislation can be a powerful tool to promote recycling. Following the legislative developments in Switzerland and Germany, Austria has also adopted legislation for a mandatory P recovery from SSA. The Swiss Federal Council published an Ordinance on the Avoidance and the Disposal of Waste (SR 814.600), where P must be recovered and recycled from SSA (Section 3, Art.15, in force since Jan. 1st of 2019) and metals recovered from MSWI filter ash (Section 3, art. 32) [24]. There are also requirements and limit values for using waste as a raw material in this Ordinance (Annex 4, in force since Jan. 1st of 2022) [24]. Denmark has the Statutory Order no.1672/2016 on the use of residual products, where MSWI-BA is on the list to replace primary raw materials [16]. However, the ash must meet quantitative criteria regarding the content and leachability of certain inorganic substances [16]. The Statutory Order no. 732/2019 on the application of biomass ash in agriculture additionally determines the extent to which biomass ash can be requested as a replacement for commonly used fertilizers or soil improvers (i.e. which types of biomass ash are allowed, the limit values for the content of heavy metals, the maximum amount of allowed ash and its reactivity) [16]. The Finnish legislation on fertilizers has also been modernized, as a new Fertilizer Act (711/2022) came into force in July of 2022 [25].

1. RECOVERY FROM SELECTED ASHES

1.1. Introduction

The extraction of specific components from waste ashes is a wide field, as there are many different ashes and components to be extracted. The selected ashes of this project are rich in silica (SiO_2), the main constituent of soil, and macronutrients necessary for plant growth, such as Ca, Mg, P, S, as well as Al and Fe [26]. The content of potentially toxic elements such as Zn, Cu, Cr, Pb, Ni and Cd is also very important. The main elements of interest in this literature survey are P and the heavy metals Zn, Pb, Cd, Cr and Cu, which are usually extracted using various organic/inorganic acids, chelating agents or basic solvents, as well with electrochemical methods such as electrodialytic separation (EDS).

1.2. Overview of the technologies

Various techniques, such as wet extraction, thermochemical and electrochemical methods have been developed to extract or recover metals from different ashes [27, 28]. This overview also covers plants at TRL7 (see Subchapter *1.3. Methods applied on industrial scale*). Wet chemical extraction is the most widely used method for extracting P from various ashes due to its high P recovery rate, low cost and procedure simplicity. Choosing the right extractant is very important to achieve high extraction efficiency. Commonly used extractants are inorganic acids such as sulfuric acid (H_2SO_4) [2, 4, 11–14, 29–31], nitric acid (HNO_3) [4, 12, 14, 30, 31], hydrochloric acid (HCl) [4, 18, 32, 33], phosphoric acid (H_3PO_4) [28], organic acids and chelating agents such as citric acid (CA) [14, 29–31], oxalic acid (OA) [4, 13, 14, 29–31], lactic acid (LA) [30], EDTA [14, 29, 31], EDTMP [14], [29], [31], Cyanex 923 [34] and Cyanex 572 [17], bases such as NaOH [4] and/or salts such as $NaHCO_3$ [35].

The most commonly used inorganic acid and the cheapest extractant on the market is H_2SO_4 [12]. Its main advantages are easy transportation due to its low volatility, the possibility of concentrating up to 98% and ensuring less co-dissolution of heavy metals, especially Pb [2, 4]. Other inorganic acids, such as HNO_3 [4, 12, 14, 30, 31], HCl [4, 18, 32, 33] and H_3PO_4 [5] have also been used. HCl may facilitate the occurrence of unwanted complexation reactions [2], and H_3PO_4 is comparably expensive [5]. Organic acids are usually chosen in research for their reduction properties (especially OA, which is the strongest naturally occurring organic acid) and for their environmentally friendly production (CA and LA) [4, 13, 14, 29–31]. Organic acids also dissolve much more metals than H_2SO_4 due their chelating capacity and significant amounts of P, which is unfavorable for P recovery. OA is the most efficient extractant among organic acids as it combines a high P extraction efficiency with relatively low co-extraction of heavy metals; however, H_2SO_4 has an economic advantage over OA due to the lower costs for optimal P extraction [4, 14].

Chelating agents have a marginal effect on the morphology and particle size distribution, however compared to inorganic and organic acid extractants, they are

less effective for P and Zn recovery [27, 31]. EDTA performed better than EDTMP for trace elements, such as Zn, Pb and Cd, so EDTA could be used before the P extraction process to remove significant quantities of metals without leaching P [29]. Fang et. al. [36] also published a study where a combination of EDTA and H_2SO_4 was used, which was not effective for P extraction. The use of Cyanex, as a highly stable P-based chelating extractant, has been shown to be effective in the extraction of heavy metals (Zn, Pb and Cd) [17, 34]. Alkali bases such as NaOH dissolve almost no heavy metals, mainly due to the high pH of around 13 at the end of extraction procedure. They are also ineffective for P extraction, because Ca-phosphates show a poor solubility in alkaline environments (especially when the molar ratio of P/Ca is lower than 1), while Al- and Fe-phosphates are highly soluble in such media [4, 11].

In order to achieve optimal extraction conditions, the requirements for the highest P extraction efficiency (PEE), the lowest possible co-extraction of heavy metals and the lowest possible operating costs in the process need to be satisfied. In addition, variables such as the type of extractant and its concentration, contact time (optimally two hours [2, 11, 13, 29, 31]), liquid/solid (L/S) ratio (optimally 20:1 [2, 11, 12, 29]), extraction temperature and ash composition should be considered as they significantly affect the PEE [2, 11, 13]. Longer extraction times (e.g., one week compared to two hours) result in a reduced PEE and a greater leaching of heavy metals [12]. It is also necessary to consider the variability of certain elemental contents, e.g. the P content in SSA can be partially attributed to differences in wastewater treatment systems and incineration conditions [37]. The sampling period also appears to influence its content, as e.g. a lower P content was measured in the summer months while higher contents were measured in February and March [38]. This can be explained by different food habits and leisure activities of people in different seasons [38]. The recovery of P by wet chemical extraction can be effectively applied to different types of SSA, as they contain higher amounts of P [2, 4, 11–13, 29–31]. Ashes from wood biomass and municipal solid waste are not as rich in P as SSA, but they are rich in Zn, which is also important to recover [32–34, 39, 40].

A literature review of the extractants used to extract P, Zn, Pb, Cu, Cr, and/or Cd from the selected ashes is presented in Table 1 along with the respectively achieved extraction rates. More detailed tables are available in the Appendix. In the case of Zn, the highest extraction rate of 99.3% was achieved using Cyanex 923 [34], while a 100% extraction rate was reported for P [12, 13, 30]. Extraction rates of 76% for Cr [28], 81% for Pb [34], 97% for Cd [33] and 100% for Cu [34] were also reported. Although chemical extraction achieves high extraction efficiencies, it requires further purification and the treatment of insoluble acid residues [13, 14, 27, 29, 41, 42]. It also often requires undesirably large amounts of acids, which has encouraged researchers to develop alternative methods [28].
One of the alternatives for a high removal of Zn (around 90%), Pb and Cd from ash is the thermochemical method, although there are concerns regarding this process due to operating costs, a high energy input and equipment lifetimes [41–43]. Potassium chloride (KCl), magnesium chloride ($MgCl_2$) and calcium chloride ($CaCl_2$) are often added in a thermochemical process where a high concentration of chlorine compounds

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can be extremely corrosive [41, 44, 45]. Increasing the treatment temperature led to higher Pb and Zn removal rates [45]. Above 1400 °C the thermal removal of heavy metals also enables the separation of Fe, which increases the bioavailability of P in the ash [41]. The most promising one-step extraction method is EDS, whose main advantage over other techniques (e.g. acid/base extraction) is the ability to separate P from the remaining waste material and remove heavy metals from ash [3, 46–49]. Here electromigration is responsible for transporting P towards the anode and heavy metals mainly towards the cathode, which is very important in the extraction of P from mixed component wastes [46]. After the treatment, P is recovered from the anolyte by filtration to separate the liquid from the remaining solids, and the heavy metals are solubilized in the catholyte [47]. However, the EDS process is time consuming (3-28 days [46, 48–50]) and the operating costs are relatively high [42, 46, 51]. Electrochemical and thermochemical methods are promising, but it is always necessary to determine the optimal experimental conditions and find ways to reduce the energy consumption of the process [27].

Table 2: Summary of chemicals used to extract P, Zn, Pb, Cu, Cr, and/or Cd from selected ashes in the respective references. The corresponding maximum extraction rates achieved are stated below.

		SSA	MSWI	WBA
Inorganic acids	**H_2SO_4** **HNO_3** **HCl** **HCl + H_2O_2** **HCl + H_3PO_4**	[2, 4, 11–14, 29–31] [4, 12, 14, 30, 31] [4] - -	- - [18, 32, 33] [33] [28]	- - [18] - -
Organic acids	**Citric acid (CA)** **Oxalic acid (OA)** **Lactic acid (LA)**	[14, 29–31] [4, 13, 14, 29–31] [30]	- - -	- - -
Chelating agents	**EDTA** **EDTMP** **Cyanex 572** **Cynex 923** **LIX860N-I**	[14, 29, 31] [14, 29, 31] - - -	- - [17] [34] [17, 34]	- - - - -
Base	**NaOH**	[4]	-	-
Salt	**$NaHCO_3$**	-	-	[35]
Maximum extraction rate	**P (%)** **Zn (%)** **Pb (%)** **Cu (%)** **Cr (%)** **Cd (%)**	100 71 56 66 58 50	- 99 81 100 76 97	>40 - - - - -

1.3. Methods applied on an industrial scale

There are prototype plants (i.e., at TRL7) for the extraction of P from SSA (EasyMining, S), the extraction of salts and Zn from MSWI (StenaRecycling, DK), and the full-scale commercially available process FLUWA/FLUREC operating in Switzerland for the recovery of metals. These extraction processes at the high TRL level are based

on using waste acid from nearby industry or from the wet scrubber of the incineration plant itself.

A wet extraction method using HCl and lime for recovering commercial P, Fe and Al products called Ash2Phos was developed by Easymining, Sweden. The process has recovery rates of 90-95% for P, 60-80% for Al and 10-20% for Fe from SSA [52]. Simultaneously, the heavy metal content in connection to P is reduced by at least 96%, making it a very pure and clean fertilizer product [52]. SSA is dissolved in HCl at 40 °C and the P, Fe and Al are separated as pure calcium phosphate ($Ca_3(PO_4)_2$), ferric chloride ($FeCl_3$) and sodium aluminate ($NaAlO_2$) [52]. The separation process is based on chemical precipitation steps in a unique combination. The solution is thereafter neutralized to remove heavy metals. Lime (CaO) is used during the precipitation steps and for neutralization. The produced phosphorus-calcium rich product ($Ca_5(PO_4)_3OH$, CAS no 12167-74-7) contains a minimum of 16.5% P and 35% Ca and can be used as raw material for feeds or fertilizer applications [52]. The Fe and Al products can be reused in wastewater treatment plants [52]. After the treatment, the non-dissolved SSA is filtrated, washed and neutralized. This product is also called ”silicate sand” (48.3% SiO_2, 22.9% Fe_2O_3, 7.2% Al_2O_3) and has the potential to be used as a partial cement replacement in mortars after milling [53]. A full scale plant able to annually treat 30,000 tons SSA is under construction in Sweden and plants in Germany are also under development [52].

A recovery of metals from MSWI-FA is achieved by the FLUWA/FLUREC processes developed in Switzerland by the AIK Technik AG, as well as by the HaloSep process in Denmark developed by Stena Recycling. In 2018, >60% of MSWI-FA was treated with the FLUWA process in Switzerland [33]. The FLUWA process is based on wet extraction by adding acidic (HCl) and neutral (NaCl) waste scrub water to MSWI-FA where 60-80% Zn, 80-95% Cd, 50-85% Pb, 50-85% Cu can be extracted [33, 54]. The metal enriched filtrate after FLUWA needs to be further processed to recover the metals which is either done by leading the filtrate to a wastewater treatment plant for the precipitation of a metal hydroxide sludge that can be recovered in smelting plants or the FLUREC process. The FLUREC process is a recovery process for high-purity Zn from the filtrate after FLUWA. Cd, Pb and Cu are separated by reductive separation (cementation) using Zn powder as a reducing agent [54]. This cement phase contains 50-70 % Pb, which can be recovered in a smelter [54]. Zn is removed from the remaining liquid by solvent extraction, followed by electrowinning to recover high-grade Zn (>99.99% Zn) which can be sold [55]. The remaining fly ash particles (filter cake) are currently landfilled.

The HaloSep process is also a wet extraction method using HCl scrubber liquid with MSWI-FA that produces brine and a neutralized washed FA. The resulting residues from the process are a stabilized FA, a metal fraction and a brine solution. The metals are precipitated from the brine into a filter cake containing up to 38-40% Zn that can be recovered at smelters. The remaining brine contains salt products ($CaCl_2$, NaCl, KCl) which are useful for industrial applications. The treated FA complies with the European leaching limits for acceptance in landfills [54], but can also be used in

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construction [56]. A full-scale HaloSep plant is operating at the incineration plant Vestforbrænding in Copenhagen, Denmark and plants in other countries are under exploration [56].

There are additional plants or factories using the process of P recovery from SSA into H_3PO_4 [42]. One of them is a sewage sludge incineration plant in Werdohl, Germany that uses the Remondis TetraPhas process. It consists of leaching P from SSA by H_3PO_4 and a purification of the P concentrated acid leachate allowing an 80% P extraction. The product is called RePacid, mainly contains H_3PO_4 and can be directly used by the industry [42]. Another solvent-extraction process called Phos4life was designed in the Canton of Zürich, Switzerland, where technical H_3PO_4 (74%) is the main product. Here P from SSA is extracted by H_2SO_4 and more than 95% of it can be recovered from SSA in the form of H_3PO_4 [42]. Another well-known P production company is Nippon Phosphoric Acid Co. Ltd (NPA) in Achi, Japan, where the H_3PO_4 is also obtained through a wet extraction process followed by filtration and purification. Gypsum ($CaSO_4 \cdot 2H_2O$) with possible applications in cement, plasterboard or soil improvement is a by-product of this process [42].

Methods for extracting resources from ashes at locations where waste acid is not available in the close vicinity are also important. Here electrochemical technologies are beneficial because their major input is an electrical direct current (DC) which can be directly gained from renewable sources and the extraction can be performed in periods with excess grid energy. The extraction technologies prioritized for further investigation in this project will be those based on waste acids or excess renewable energy so as to have the best environmental profile and provide local symbioses.

2. POTENTIAL USE OF TREATED ASHES

Waste incineration is steadily increasing in Europe, but there are environmental concerns about the solid residues requiring pre-treatment and commonly disposed of in landfills [57]. Possibilities for using pre-treated ashes exist in agriculture, soil stabilization, the building sector (i.e., supplementary cementitious materials, precursors for alkali-activated materials or as artificial fillers or fine aggregates). Many researchers have been attracted towards the utilization of waste ashes in construction materials in recent years [57]. However, a good understanding of their chemical, physical and microstructural characteristics is necessary to utilize different waste ashes in full scale [58]. Especially the question how to maintain the characteristics of ash constant when heterogeneous materials (such as sewage sludge, MSW or varying types of biomasses) are incinerated is highly important for large-scale utilization.

2.1. Potential use of SSA

Sewage sludge is the most common and continuously generated by-product of waste water treatment, containing the second highest amount of P (after bone meal) [3, 5, 37]. It has a great potential for P-recovery with an appropriate thermal treatment [2,

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4, 11–14, 29–31, 41, 42]. Sewage sludge has been directly used as agriculture fertilizer for decades, but its limitations are increasing all over the world due to high contents of heavy metals and organic pollutants. Accordingly, a thermal treatment of sewage sludge is considered to be the best way for its disposal [41]. The incineration of sewage sludge at about 850 °C is widely used in the EU and currently the most efficient method, reducing the volume by 90% (the mass by 70%) and removing organic pollutants and pathogens [37, 42, 47].

Nevertheless, heat treatment produces SSA, containing 6-12 wt% of P [16], usually in the form of $AlPO_4$ and $Ca_3(PO_4)_2$, which are poorly bioavailable. SSA also contains Fe and toxic trace elements, such as Zn, Pb, Ni, Cr and Cd and is mostly landfilled [5, 12, 38]. Pre-treatment is required to prevent a loss of this potential P source and its use as a fertilizer. The aim of pre-treatment is to increase the bioavailability of P and remove heavy metals, which often exceed the legal limits for fertilizer production, see Table 3 [11, 38, 59, 60]. With, for example, innovative EDS, 80-90% of the P can be recovered while also achieving a low content of heavy metals [47–49]. A high concentration of CaO and SiO_2 in the SSA after the P extraction is the main reason for using SSA as a building material component [5].

Cement production is among the human activities that contribute significantly to CO_2 emissions, as cement production requires a large amount of raw material and energy. Every 600 kg of cement causes about 400 kg of CO_2 to be released into the atmosphere [61–65]. Cement is used as a binder for concrete, the second most used material in the world. Sustainable development of the building industry thus requires three approaches; production using renewable energy, using recycled products, and replacing cement [59]. SSA is a material comparable to lightweight sand and is less dense than Portland cement [37]. It consists of porous particles of irregular shapes, which is not ideal for a classification as a potential cementitious material [37]. For further use in construction materials, it should be considered that the extraction of P with H_2SO_4 produces $CaSO_4$, which negatively affects the cement properties [4]. Using OA as an extractant produces Ca-oxalate, which does not have this negative effect [4]. SSA also typically contains an elevated amount of about 14% Al_2O_3 compared to Portland cement, which contains about 5%, indicating a natural suitability of SSA for use in aerated concrete [37]. The high Al_2O_3 content in SSA may also benefit the chloride attack resistance in concrete applications due to the chloride binding capacity of amorphous Al_2O_3 [37]. SSA can be used as a possible cement replacement material, but it requires a pre-treatment due to the undesirable effects of the contained heavy metals and P recovery. It has a potential as cement replacement in mortar [12, 66] or as a partial replacement for clay in clay bricks [47]. Due to the small grain size, SSA is also suitable for potential use as a filler or fine aggregate component in mortar and concrete, where the effects on strength performance are manageable for SSA contents up to 15 wt% [37, 40, 67]. Research on the reuse of SSA as an aluminosilicate precursor material for alkali-activation/geopolymerization has also begun recently [68, 69].

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Table 3: Legal limits for trace elements in EU fertilising products (in mg/kg), adapted from Regulation (EU) 2019/1009 [60].

Element	Organic fertiliser	Organo-mineral fertiliser	Inorganic fertiliser
As	40	40	40
Cd	1.5	3	3
Cr	2	2	2
Cu	300	600	600
Hg	1	1	1
Ni	50	50	100
Pb	120	120	120
Zn	800	1500	1500

2.2. Potential use of MSWI

Municipal solid waste (MSW) contains wood waste but also paper, plastic, glass, wood and textile scrap material that cannot be degraded naturally. In the last few decades, the total mass of MSW has increased drastically due to rapid urbanization and an increased world population. This has encouraged many countries to properly dispose of this waste [57, 70, 71]. The so-called ”green economy” has begun, encouraging the reduction of waste, reuse of materials through recycling or recovery and supports sustainability [70, 72]. Several different treatments of MSW outlined in Figure 2 have been developed, among which incineration is one of the effective and popular methods due to the volume reduction of MSW by 90%, and heat/energy recovery. Two main residues are produced by incineration; around 80% MSWI-BA while the rest is MSWI-FA [17, 32, 70, 71, 73, 74].

MSWI-BA is classified as non-hazardous waste and mainly consists of amorphous SiO_2, Al_2O_3 and CaO, however its exact composition varies from country to country [70]. MSWI-BA can be reused in the construction field as an alternative light aggregates [70] or as an alternative material for cement production [75]. It is commonly utilized in road construction [76]. MSWI-FA is classified as a hazardous waste as it contains soluble salts, dioxins and a significant amount of heavy metals. It is usually landfilled, which is not only harmful to the environment and human health due to the presence of potentially leachable contaminants, but also means a loss of the resources in the ash [17, 28, 32, 39, 50, 73, 74, 77]. MSWI-FA has a large potential for extracting metals such as Zn, Pb and Cd [33]. Sekito et. al. [78] reported a 2-fold higher content of Zn and Pb in MSWI-FA compared to MSWI-BA, while the content of Cd was even 13-fold higher. Therefore, MSWI-FA must be pre-treated before further use, so much research has focused on how to extract and recover various metals from it. In MSWI-FA, the pH has a significant effect on the removal efficiency for heavy metals [39]. Many metals have a high solubility at low pH levels, so using strong acids as the extractant is necessary. As MSWI-FA is alkaline, alternative methods are desired to avoid the consumption of large acid volumes. A new microwave-assisted acid extraction method has recently been developed [28]. Significant advantages of this method are lower costs, shorter processing times and a better efficiency of metal extraction compared to conventional heating [28, 79–81].

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Electrodialytic (ED) treatment is another innovative method also used for contaminated SSA [47]. It reduces the content of heavy metals and salts and increases the reactivity of Si and the Si/Al ratio [50, 82]. Such a pre-treatment method can make MSWI-FA into a potential precursor in geopolymers based on alkali-activated material (AAM) that can naturally trap heavy metals inside its matrix. As MSWI-FA is alkaline (having a pH of around 11), ED treatment results in an acidic pH, similar to the common natural precursor in geopolymers, i.e. metakaolin. The combination of MSWI-FA pre-treatment and inertization of up to 20 wt% of MSWI-FA in geopolymers achieves the lowest metal leaching and high compressive strength, making it a potential construction material [50, 82]. Previous studies have shown that the use of raw MSWI-FA is also possible but made the geopolymer matrix less stable. Liu et. al. [74] reported that adding 10% of metakaolin to MSWI-FA significantly improved the compressive strengths, while Łach et al. [83] immobilized 70 mass% of raw MSWI-FA using a metakaolin-based geopolymerization process.

MSWI-FA has also been studied as a potential replacement of cementitious materials, but adding it to cement-based products means that technical and environmental requirements such as sufficient strength, durability and leaching limits of heavy metals from the products must be met [84]. The main problem with using MSWI-FA as a cement substitute is the presence of leachable toxic heavy metals and a high salt content. It is beneficial to use water washing and mechanochemical [84] or ED pre-treatments [85] which can improve the performance of MSWI-FA before it is used in mortar, concrete or bricks. The mechanochemical processes can stabilize the heavy metals and activate the MSWI-FA, allowing it to partially replace Portland cement in building materials [84], while an ED treatment can remove heavy metals and soluble salts from the MSWI-FA suspension which is thus decontaminated [86, 87].

As an ultrafine material, MSWI-FA is also a potential substitute for clay in bricks, which should stabilize the heavy metals, reduce raw material imports, and at the same time conserve primary clay resources [86]. Studies have shown that fired bricks with an addition of 2.5-5 wt% [87–89] treated MSWI-FA may be feasible in the clay brick industry.

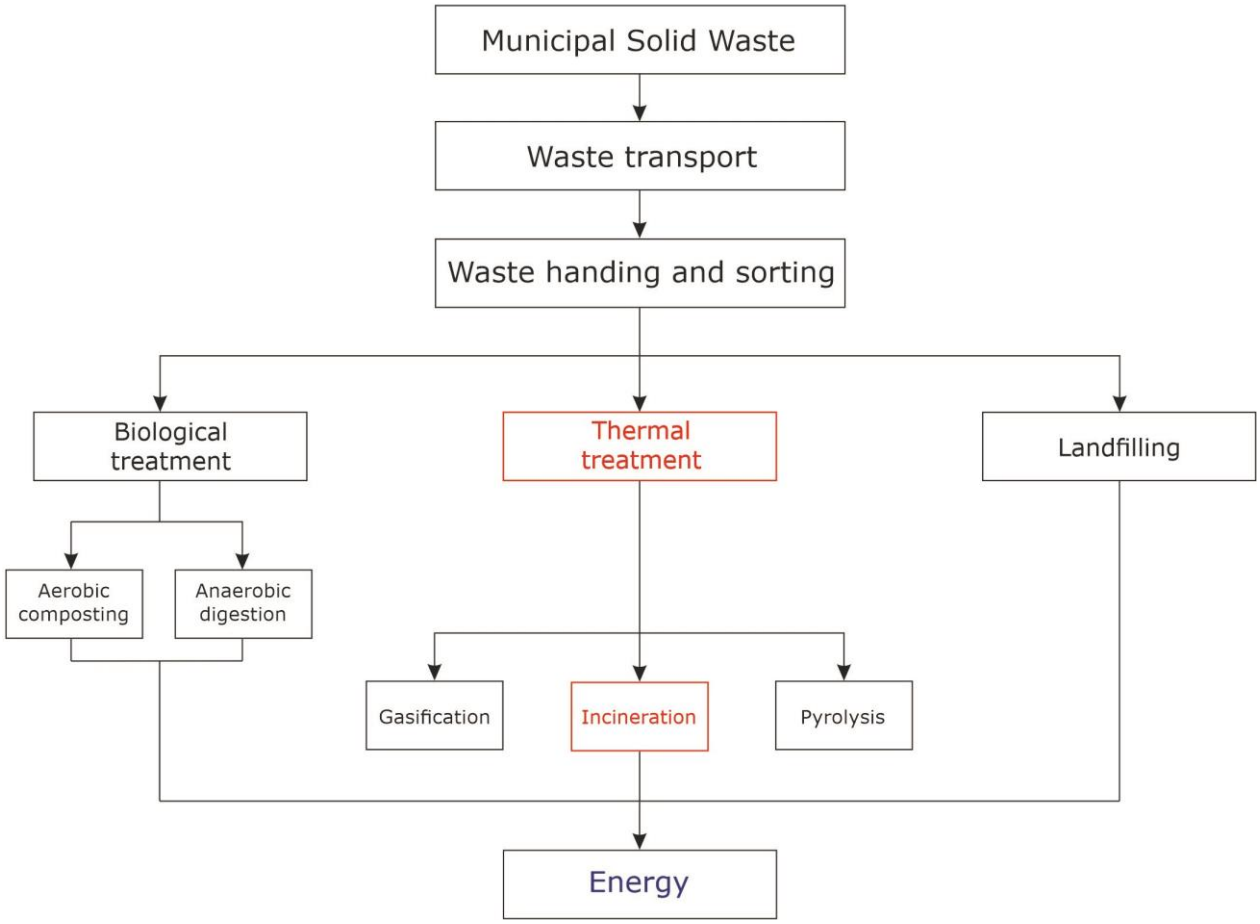

Figure 2: Different treatments for MSW management, redrawn based on Ref. [70].

2.3. Potential use of WBA

Wood biomass ash (WBA) is produced during wood biomass combustion, where two types of ash are generated: WBA-BA, collected from the bottom of a combustion chamber and WBA-FA, which is subdivided into fine fly ash (size of the particles <1 µm), collected from electrostatic filters or bag house filters and coarse fly ash (size of the particles >1 µm), collected from the cyclone or boilers [62, 90, 91]. In general, CaO and SiO_2 are the major chemical components in WBA, while other compounds such as Al_2O_3, Fe_2O_3, K_2O, Na_2O, MgO, P_2O_5, and SO_3 are present in lower amounts [18, 58]. Minor contents of As, Cd, Cr, Cu, Pb, and Ni were also detected [18, 58]. Depending on the ash type, significant differences in the content of volatile heavy metals can be detected; they are the main concern when using WBA. A higher concentration of heavy metals was measured in WBA-FA samples compared to WBA-BA, as heavy metals are more concentrated in smaller particle size fractions (<75 µm) [21, 90–92]. The particles of fine FA fraction are also lighter and smaller, which makes them easy to inhale and thus pose a health risk as e.g. Cd accumulates in the kidneys and affects bone density [93].

The chemical composition of WBA-FA also differs from coal FA, as WBA-FA usually contains more alkali elements and less Al [91]. Nutrients, such as P and Mg are primarily found in the WBA-BA and coarse WBA-FA. WBA-FA shows significant lower

Cd concentrations compared to MSWI-FA [18, 20, 21, 94]. The chemical and physical properties of WBA depend on the combustion technology, heat treatment temperature, tree species and the geographic location where the WBA was collected, however, other factors, such as soil conditions, climate characteristics and storage methods also influence its properties [18, 21, 62, 64, 91–93, 95, 96].

As wood biomass is considered to be a CO_2-neutral source of energy, it is environmentally desirable to use WBA in the construction industry [90]. This would not only reduce rising disposal costs, as 70% of WBA still ends up in landfills, but also preserve natural resources and reduce GHG emissions [90]. WBA has the potential to be used in various construction areas: as a partial replacement of aggregates or mineral admixtures in concrete production [91], as a partial replacement of raw materials for clinker production [90, 91], as filler/partial sand replacement material in cement-based materials [20, 95], in brick production [95], road construction [95] and others. Proper storage and transport conditions are important for WBA use in cementitious composites, as carbonation and hydration can occur suddenly during these procedures in wet circumstances and thus strongly determine the quantity of CaO and other carbonate elements [95]. It is also very important to find the optimal cement/ash ratio so that required strength of the cement composites does not decrease [95, 97]. Replacing up to 45% of cement with WBA-FA is suitable for construction purposes, however WBA-FA has more potential as a filler material than as a cement replacement material in construction [64, 95]. Most studies report that the optimal content of WBA-FA and WBA-BA to replace part of the cement in mortars is 10 wt% [63, 65, 95, 98].

WBA-FA can also be used as a complete or partial replacement material in the preparation of geopolymer mortars, which could reduce the cost of geopolymer source materials and the cost of WBA disposal in landfills [99, 100]. The use of WBA as a forest fertilizer also has potential, however future research should focus on the effect of trace element solubility on the leaching process that occurs naturally in the forest soil [21, 101]. Another important factor, preventing the use of WBA on certain soil typologies is its alkaline pH (usually higher than 12) [102]. Accordingly, Pasqali et. al. [102] proposed a technology to stabilize heavy metals in WBA and lower its pH, based on the use of other by-products (coal FA, rice husk ash and MSWI-FA). MSWI-FA has a similar pH as WBA and is a source of leachable heavy metals, while its concentration of P is low. Ca-rich coal FA was used in the stabilization procedure, while rice husk ash was chosen as a heavy metal stabilizer due to its amorphous silica content. Wolffers et. al. [18] recently reported the recovery of heavy metals from WBA-FA based on acid leaching, a process also applied to MSWI-FA. In Switzerland, the disposal of WBA-FA in landfills will be prohibited in 2023 due to the elevated concentrations of very toxic Cr(VI) and other heavy metals [18]. In Switzerland, the FLUWA process has already been developed for the recovery of Zn, Pb, Cu and Cd from the similarly generated MSWI-FA, which must be treated before disposal, so this process also represents a promising method for WBA-FA and for the co-processing of both types of ash [18].

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3. SUMMARY AND CONCLUSIONS

This literature review focused on the technologies which are available for the recovery of valuable elements, such as P and metals, such as Cu, Zn and Pb, from MSWI, SSA and WBA. The most common extraction techniques for P and Zn recovery are wet extraction, thermochemical, and electrochemical (ED) methods. Based on this review and the results from ash characterization and categorization (Task 2.1 – Deliverable D 2.1 Characterization and categorization of the ashes) the next step in the project is an extraction and potential evaluation of specific ashes. Further actions are undertaken to verify the efficiency through different methods of extraction (Task 3.2: Development of electrodialytic, electrocoagulation, or ligand-based extraction as robust method for extraction of valuable resources).

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Appendix

Data presented supplement of Table 2, introduced in Subchapter 1.2.Overview of the technologies.

Table A1: Summary of the inorganic acids used to extract P, Zn, Pb, Cu, Cr, and/or Cr from selected ashes with their measured element content (in g/kg or mg/kg) and corresponding maximum extraction rates (in %).

Extractant	Conc. (mol/L)	P (g/kg)	P (%)	Zn (mg/kg)	Zn (%)	Pb (mg/kg)	Pb (%)	Cu (mg/kg)	Cu (%)	Cr (mg/kg)	Cr (%)	Cd (mg/kg)	Cd (%)	SSA	MSWI	WBA	Ref.
H_2SO_4	0.05 0.1 0.19 0.19 0.2 0.2 0.25 0.4 0.5 0.5	55.0 35.0 69.9 99.0 86.0 39.9 78.0 93.1 53.5 40.4	>95 88.3 100 ~88 92 94 93 96.4 >70 74	895 2198 1700 2000 2024 2198 2823 n.d. 2198 2198	 ~36 ~42 ~42	460 93 85 112 84 97 267 n.d. 70 70	 ~1 38.4	423 839 540 690 783 839 753 n.d. 621 621	 ~38 ~40 ~40	460 n.d. n.d. n.d. 78 n.d. 142 n.d. 137 137	 ~5 57.7	126 n.d. n.d. n.d. n.d. n.d. 9.6 n.d. 4.5 4.5	 ~28 50	x x x x x x x x x x			[13] [11] [12] [12] [2] [29] [4] [30] [31] [14]
HNO_3	0.3 0.4 0.5 0.5 1.5 1.5	78.0 93.1 53.5 40.4 69.9 99.0	89 100 >70 ~71 ~80 100	2823 n.d. 2198 2198 1700 2000	~32 ~36 ~36 16 71	267 n.d. 70 70 85 112	~24 40 56 47	753 n.d. 621 540 690	~36 ~38 	142 n.d. 137 n.d. n.d.	~5 ~52 	9.6 n.d. 4.5 4.5 n.d. n.d.	~27 ~6 	x x x x x x			[4] [30] [31] [14] [12] [12]
HCl	0.3 1.0 1.0 1.0 1.0	78.0 n.d. 4.5 4.8 3.8	98.8	2823 13000 65420 44607 21550	~32 80 75 ~58	267 3100 11920 8143 21015	~30 1 ~1	753 800 2512 2131 1131	40	142 n.d. 468 360 1221	~5	9.6 100 370 277 71	~28 71 40	x	 x x x	 x	[4] [32] [33] [18] [18]
$HCl + H_2O_2$	1.0 + 9.8	4.5		65420	68	11920	62	2512		468		370	97		x		[33]
$HCl + H_3PO_4$	2.0 + 3.0						30.2		71		75.7		56.2		x		[28]

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Table A2: Summary of the organic acids, chelating agents, alkali bases, and salts used to extract P, Zn, Pb, Cu, Cr, and/or Cr from selected ashes with their measured element content (in g/kg or mg/kg) and corresponding maximum extraction rates (in %).

Extractant	Conc. (mol/L)	P (g/kg)	P (%)	Zn (mg/kg)	Zn (%)	Pb (mg/kg)	Pb (%)	Cu (mg/kg)	Cu (%)	Cr (mg/kg)	Cr (%)	Cd (mg/kg)	Cd (%)	SSA	MSWI	WBA	Ref.
Citric acid ($C_6H_8O_7$)	0.2 0.4 0.5 0.5	39.9 93.1 53.5 40.4	~80 59.3 >70 72	2198 n.d. 2198 2198	 ~23 ~23	97 n.d. 70 70	 13.3	839 n.d. 621 621	 ~16 ~16	n.d. n.d. n.d. n.d.	 ~25	n.d. n.d. 4.5. 4.5	 ~7	x x x x			[29] [30] [31] [14]
Oxalic acid ($C_2H_2O_4$)	0.05 0.2 0.55 0.4 0.5 0.5	55.0 39.9 78.0 93.1 53.5 40.4	100 >95 95.4 100 >70 74	895 2198 2823 n.d. 2198 2198	 37 56.9 56.9	460 97 267 n.d. 70 70	 ~1 4	423 839 753 n.d. 621 621	 37 65.8 65.8	460 n.d. 142 n.d. 137 137	 ~8 ~53	126 n.d. 9.6 n.d. 4.5 4.5	 ~13 ~13	x x x x x x			[13] [29] [4] [30] [31] [14]
Lactic acid ($C_3H_6O_3$)	0.4	93.1	28.4	n.d.		n.d.		n.d.		n.d.		n.d.		x			[30]
EDTA ($C_{10}H_{16}N_2O_8$)	0.02 0.05 0.05 0.05	39.9 53.5 40.4 39.9	~20 <30 ~24 ~40	2198 2198 2198 2198	 ~14 ~14 	97 70 70 97	 37 	839 621 621 839	 ~5 ~5 	n.d. 137 137 n.d.	 ~42 	n.d. 4.5 4.5 n.d.	 ~6 	x x x x			[29] [31] [14] [29]
EDTMP ($C_6H_{20}N_2O_{12}P_4$)	0.05 0.05 0.05	53.5 40.4 39.9	<30 ~13 ~25	2198 2198 2198	~10 10 	70 70 97	~22 	621 621 839	~9 ~9 	137 137 n.d.	~26 	4.5 4.5 n.d.	~6 	x x x			[31] [14] [29]
Cyanex 923 **Cyanex 572** **LIX860N-I**	1.0 1.0 1.0	n.d. 4.0 n.d.		5800 5800 5800	99.3 91 	5700 5700 5700	~81 	5400 5400 5400	 100	190 190 190		90 90 90	~88 		x x x		[34] [17] [34]
NaOH	0.5	78.0	40	2823	~3	267	~3	753	~2	142	~2	9.6	~4	x			[4]
$NaHCO_3$	0.5	17.0	>40	n.d.		n.d.		n.d.		n.d.		n.d.				x	[35]

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