



Use of industrial residues for heavy metals immobilization in contaminated site remediation: a brief review

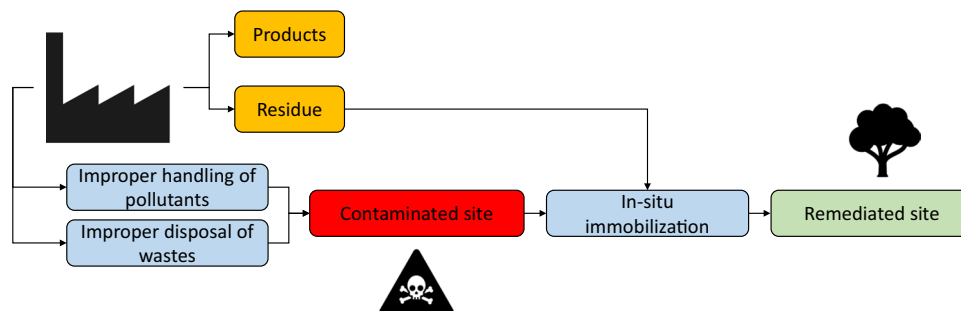
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Abstract

The increasing use of industrial residues for the remediation of landscapes contaminated with heavy metals diminishes the negative environmental impact of the contamination itself, reduces the demand for primary raw materials and minimizes the costs for the disposal of the residues. On the other hand, industrial residues often contain heavy metals themselves, which make their application for contaminated site remediation controversial. This study assembles and compares results of different investigations, such as laboratory tests, greenhouse tests and full-scale field tests, concerning heavy metals immobilization in soils all over the world. This review begins with an overview of the principles of immobilization and then focusses on two major groups of industrial residues: (i) residues from metallurgy (slags and red mud) and (ii) residues from thermal processes, i.e. incineration and pyrolysis. The feasibility of industrial residue applications in contaminated site remediation is presented exemplarily for the immobilization of arsenic, cadmium, cobalt, chromium, copper, manganese, nickel, lead and zinc. Red mud and steel slag additives show a high removal efficiency for specific heavy metals at contaminated field sites, whereas fly ash and biochar applications exhibit a high performance for various heavy metals uptake at laboratory scale, bearing a high potential for the extension to full-industrial scale. The latter materials may increase the soil pH, which favours the sorption of cationic heavy metals, but may decrease the sorption of hazardous oxyanions.

Graphical abstract



Highlights

- Red mud and steel slag are successfully used for the in situ immobilization of heavy metals.
- Red mud application may increase the mobility of arsenic and copper in alkaline media.
- Fly ash and biochar are highly promising according to laboratory-scale studies.
- Alkaline residues (red mud, steel slag, fly ash) are efficient for remediating cationic metals (e.g. Pb, Zn).

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Introduction

Along with industrialization and technological progress, the anthropogenic impact on the environment has increased significantly over the last two centuries. Human activities have caused an enduring level of contamination in particular in soils, surface-near sediments and the aquatic environments due to, e.g. mining (Concas et al. 2006), fossil fuel combustion (Kapička et al. 1999), traffic and transportation (Ma et al. 2016), agricultural chemicals (Perkovich et al. 1996), households and industrial waste disposal (Querol et al. 2006) or industry (Sedlazeck et al. 2017). The increasing contamination of the terrestrial and aquatic environment with persistent heavy metals is one of the most severe problems in recent decades, arising from their high toxicity, fast accumulation, non-biodegradability and endurance (Nagajyoti et al. 2010). The partially toxic or cancerogenic, but generally health-damaging, heavy metal ions, such as arsenic ($\text{As}^{3+/5+}$), cadmium (Cd^{2+}), cobalt (Co^{2+}), chromium ($\text{Cr}^{3+/6+}$), copper (Cu^{2+}), manganese (Mn^{2+}), nickel (Ni^{2+}), lead (Pb^{2+}) and zinc (Zn^{2+}), can react with bioparticles in the human body and other life forms, which can cause numerous diseases and disorders even at low concentration levels (Femina Carolin et al. 2017; Ma et al. 2018).

There is no consistent definition of heavy metals in the scientific literature, but Hawkes (1997) defines them as “a block of all the metals in groups 3 to 16 that are in periods 5 and greater”. In contrast, Ali and Khan (2018) define them as “naturally occurring metals having atomic numbers (Z) greater than 20 and an elemental density greater than 5 g cm^{-3} ”. The toxicity level of most of these heavy metals depends mainly on the concentration, speciation and bio-availability, with the latter being predetermined by ligand complexation and oxidation state of the specific chemical component (Jaishankar et al. 2014). Heavy metals are soluble in certain pH ranges, which strongly affect their persistency versus mobility in natural and also technical surroundings. The solubility of most heavy metals depends on the type of the chemical bonding (mineral, (ad)sorptive, complexed, etc.), but is generally highest in the acidic pH range, although others are also soluble in the circum-neutral to alkaline range (Brümmer 1986). Many metals such as Zn, Cd and Pb show a higher mobility at lower pH (Hermann and Neumann-Mahlkau 1985), whereas others such as Mo show a maximal adsorption in this range (Goldberg et al. 1996). Furthermore, most of the heavy metal ions show specific oxidation–reduction (redox) features, as well as distinct aquo-speciation and ligand complex formation

characteristics, which define their mobility, chemical reactivity and toxicity among others in the environment (Femina Carolin et al. 2017; Friesl-Hanl and Horak 2011).

The high number of contaminated sites worldwide and the distinct (site-specific) contamination require fast, efficient, economic and safe methods for the remediation of hazardous heavy metal ions. In recent decades, immobilization of heavy metals has become one of the most widely used techniques for environmental clean-up and protection, as it reduces the mobility and bioavailability of the heavy metal ions of concern. The immobilization methods developed so far aim to improve the quality of soils, sediments and (ground)water, besides ensuring safe agriculture products and minimizing risks for human beings and the environment, e.g. by reducing the phytotoxicity or leaching into the groundwater (Friesl-Hanl and Horak 2011; Ma et al. 2018). Immobilization of heavy metals in contaminated site remediation can be conducted *ex situ* (Xia et al. 2019) and *in situ* (Czupryna et al. 1989). State-of-the-art in *in situ* immobilization is the use of cementitious or clay-supported suspensions (Dörrie and Längert-Mühlegger 2010; Baldermann et al. 2021a). This method leads to the formation of hydrated binder phases, which incorporate the heavy metals in their structure, but also clog the pores in the soil, thereby decreasing the permeability (Paria and Yuet 2006; Baldermann et al. 2021b). However, cement production does not only consume limestone and clay raw materials, it is also responsible for 5% of the global carbon dioxide (CO_2) emissions (Worrell et al. 2001). Therefore, the use of wastes and by-products as alternatives for *in situ* immobilization is highly interesting.

The use of waste materials, such as from metallurgy, incineration and pyrolysis industry, for the immobilization of heavy metal ions has gained increasing attraction over the last decades, improving both economic and ecologic aspects of current environmental remediation strategies (Lwin et al. 2018). The recycling and/or re-use of such waste materials significantly reduces the estimated disposal costs of the waste and creates an environmentally friendly, durable and sustainable alternative to the use of other primary raw materials. Moreover, there is an increasing demand for an efficient treatment and handling of industrial waste materials as the disposal costs increased significantly in the last decades due to legal restrictions and decreasing space in existing landfills worldwide (Chowdhury et al. 2014). Furthermore, the remediation of landscapes contaminated by heavy metals with primary raw materials is very often expensive due to high production, processing, transportation and construction costs compared to recycling of waste materials, although the

risk of new contamination needs to be barred in every case (Friesl-Hanl and Horak 2011).

Besides the economic aspects of environmental protection, the conservation and sustainable use of natural resources are a main concern of many nations. The use of industrial waste materials for remediation efforts bears a high potential for the massive reduction in the environmental footprint. Moreover, pioneer studies have demonstrated that positive side effects can arise from the use of waste materials: treated soils may attain improved stress–strain properties, which lead to the possibility of construction applications, such as backfill or road subgrade (Dermatas and Meng 2003). As a consequence, the recycling of industrial residues in contaminated site remediation is a matter of increasing interest in many regions worldwide, such as in Asia, Europe, Africa and North America.

Many waste streams have been examined for the treatment of heavy metals contamination in the past. Consequently, several reviews exist on in situ immobilization of heavy metals: Wang et al. (2009) give a broad overview about immobilization agents and mechanisms, focusing on primary raw materials. Gougar et al. (1996) provide a review on ettringite and calcium silicate hydrate phases, which may also occur in industrial residues, giving focus on the utilization of Portland cement in environmental remediation. Miretzky and Fernandez-Cirelli (2008) describe the use of phosphates for Pb immobilization, but they do not consider wastes or by-products as immobilization agents. Mahar et al. (2015) present a review on the immobilization of Pb and Cd, which also covers the utilization of wastes in terms of animal manure, but do not consider true industrial residues. Bolan and Duraisamy (2003) also include manure in their study, but only briefly mention other wastes, such as fly ashes. Kumpiene et al. (2008) mention fly ashes from biomass energy industry, sewage and paper mill sludges, as well as gypsum and lime-rich industrial residues for in situ immobilization of heavy metals, but they do not focus on these secondary resources. Kumpiene et al. (2019) describe the use of permeable reactive barriers for the upscaling of in situ immobilization approaches from the laboratory scale to the field scale and also consider wastes, such as biochar and coal fly ashes, but their review concentrates on the used amendments. Industrial residues, such as fly ash, slag, bauxite residue and gravel sludge, are also considered by Guo et al. (2006) as additives for in situ remediation, but the study focuses on economic and ecological assessment methods and does not compare individual case studies. The oblivion of the common industrial residues from metallurgy, incineration and pyrolysis for in situ immobilization of heavy metals calls for a review on this topic.

Apart from the technical feasibility of using industrial residues for remediation works, the legal framework in

each country needs to be reviewed to enable the implementation of both economic and ecological methods for contaminated site remediation. Particularly, the respective countries need to evaluate the possibility of secondary contamination with heavy metals due to the desired insertion of waste materials under varying site-specific environmental conditions. For example, one of the relevant criteria to take into consideration in the EU for the substitution of primary raw materials is the “End-of-waste status”. This concept ensures that the use of waste materials for immobilization is not classified as illegal landfill. Furthermore, guideline values for critical pollutants in waste streams are defined in the concept of the End-of-Waste as well as the need to prevent from any adverse environmental effects, which may be caused by the use of the waste materials itself (Waste Framework Directive 2008). However, to assess the country-specific legislation regarding in situ immobilization is beyond the scope of this review. Instead, we describe the principles of heavy metals immobilization in inorganic matrices and discuss the potential effects of using metallurgical residues and residues from incineration and pyrolysis in environmental remediation.

This literature review was carried out between October 2019 and February 2020 in Leoben, Austria.

Materials and methods

Published approaches utilizing industrial waste materials for heavy metals immobilization and environmental remediation have been assembled and systematically evaluated. The considered studies include either laboratory tests, greenhouse tests or full-scale field tests and have been conducted within different institutions across Asia, Europe, Africa and North America. The reviewed literature focuses primarily on the used waste material, test scale, investigated heavy metals, feasibility of immobilization and the respective country. The source and characteristics of the used waste material, methodology and performance of heavy metals immobilization were also compared. In addition to these parameters, relevant and recent publications concerning the overall importance and the principles of heavy metals immobilization in inorganic matrices were assembled and evaluated. The conclusions and novel implications of this literature review will be presented in this work. Firstly, the main principles of heavy metals immobilization in inorganic matrices will be presented to provide a deeper understanding of relevant aspects and influencing factors. Secondly and based on these principles, the feasibility of using industrial residues,



such as metallurgical residues and residues of incineration and pyrolysis, for the immobilization of heavy metals at contaminated sites will be presented. The following heavy metals are considered herein: As, Cd, Co, Cr(III), Cr(VI), Cu, Mn, Ni, Pb and Zn.

Results and discussion

Principles of heavy metals immobilization

In situ immobilization or in situ solidification/stabilization introduces chemical agents into the original medium (sediments, soils, aquifers, etc.) to trap or immobilize pollutants, such as heavy metals. The aim is to stabilize the heavy metal at the place of the contamination by minimizing the leaching characteristics of the soil matrix as well as to change the heavy metal speciation into a less soluble, less mobile, non-toxic or bioavailable form (Tantawy et al. 2012). There are several approaches how immobilization can be realized: One approach is to change the phase composition by melting and quenching which yields a glassy soil matrix which surrounds and encapsulates the heavy metals and is called vitrification (Dragun 1991). More frequently, additives are added to the soil to provoke precipitation, hydraulic binding reactions or adsorption. In case of precipitation, anion-containing solutions are applied to the soil where they react with the heavy metal ions to form low-soluble mineral phases (Cao et al. 2003). In case of adsorption, materials with high specific surface area such as biochar and fly ash are applied, whereas binders such as cementitious materials and/or blast furnace slag react chemically with water (hydraulic binders) or other reagents to form secondary mineral phases which can trap also heavy metal ions in their structure (Caselles et al. 2020).

Immobilization of heavy metals can be conducted in situ and ex situ (Table 1). Among in situ immobilization, jet-grouting is the state-of-the-art technology (Day et al. 1997; Freitag and Reichenauer 2022). An alternative is the spreading of aqueous solutions containing immobilizing agents, such as phosphorous compounds on the soil's surface (Chen et al. 2003). Ex situ immobilization requires excavation of the soil and mixing it in specific reactors with suitable reagents (Xia et al. 2019).

Metallurgical residues for heavy metal immobilization

The use of industrial wastes or by-products for soil remediation has been studied by various researchers. The utilisation of such materials lowers the environmental impact and is a cost-efficient alternative to segregate heavy metals (Femina Carolin et al. 2017). In this section, the use of red mud and steel slag for in situ immobilisation of heavy metals in soils will be discussed.

Red mud

Red mud is a waste of the alumina extraction from bauxite by the Bayer process. During the Bayer process, bauxite is washed in a hot solution of sodium hydroxide (NaOH), which leaches the aluminium out of the bauxite (Akinci and Artir 2008). The chemical and physical properties of red mud are mainly dependent on the bauxite mineralogy and quality and to a lesser extent on operational conditions of the Bayer process (Paramguru et al. 2005). The amount of already produced red mud is enormous. Power et al. (2011) estimated the annual production of red mud to be 120 million tons and a global inventory of over 2.6 billion tons in 2007. To produce 1 ton of alumina, ~2 to 3 tons of bauxite are needed. By applying a mean ratio of 1.5 to alumina

Table 1 Overview about state-of-the-art Immobilization techniques, mechanisms, advantages and limitations

Technique	Mechanism	Advantages	Limitations/disadvantages	Data source
Jet grouting	Injection of a suspension into the sub-surface, followed by precipitation, adsorption or incorporation into other phases	Stable, long-term immobilization	Negative impact on soil biology, e.g. by pH increase, loss of aeration	Day et al. (1997)
Pouring onto the surface	Dissolving of the immobilizing agent in water and pouring it onto the surface	No drilling required, smaller impact on soil biology	Clogging of pores by phosphate precipitates may hinder penetration of the entire contaminated volume	Chen et al. (2003)
Ex situ immobilization	Excavation of the material and mixing with an immobilization agent in a reactor	Better homogenization of the material and complete reaction with the entire contaminated volume	Additional efforts for excavation and treatment facilities	Xia et al. (2019)



production data (Power et al. 2011), the amount of red mud produced can be determined as up to ~170 million tons in 2015 worldwide. The major alumina producer is China, contributing about 50% to the world's alumina production in 2015.

The average red mud dry mass contains Fe_2O_3 (41 wt%) and Al_2O_3 (17 wt%), in addition to smaller amounts of SiO_2 (10 wt%), TiO_2 (9 wt%), CaO (9 wt%) and Na_2O (5 wt%) as well as trace elements (e.g. Cr, Cu, Pb, V and Zn) and radionuclides (e.g. U and Th isotopes and their daughter isotopes, and ^{40}K) (Klauber et al. 2011; Somlai et al. 2008; Feigl et al. 2012; Rubinos and Barral 2013; Xue et al. 2016). Though, the composition of the red muds varies due to differences in bauxite composition and treatment applied in the processing plant (Klauber et al. 2011). Red mud is highly alkaline due to incomplete washing of the residue material before disposal, with reported pH values ranging from 9.0 to 13.1 (Kirwan et al. 2013). Due to its highly alkaline nature and high concentrations of potentially toxic elements, red mud can cause environmental problems. On the other hand, it frequently has a large specific surface area, which is suitable for a fast and efficient remediation of metal-contaminated substrates (Friesl et al. 2004). Therefore, red mud has been used for in situ remediation of contaminated soils to neutralize low pH soils and to reduce metal ion mobility through different physicochemical binding mechanisms, such as (ad)sorption and surface complexation with Fe-oxides and clays, formation of inner- and outer-sphere complexes with Fe- and Al-(hydr)oxides and chemical precipitation (Brunori et al. 2005; Santona et al. 2006; Ahn et al. 2015).

The effectiveness of the use of red mud amendment for the remediation of contaminated soils is difficult to assess since a broad variety of published results among different studies exist. In some cases, even an increased metal ion mobility in the soil, especially for As and Cu, was obtained (Hua et al. 2017). The dissolved organic carbon (DOC) was found to increase in the amended soil compared to untreated contaminated soil. Since Cu tends to be strongly adsorbed by soil organic matter, the increased mobility of Cu in red mud-amended soil can be explained (Lombi et al. 2002). The mobility of As can be increased by increasing soil pH, which results in decreased adsorption of As(III) and As(V) to, e.g. goethite (Grafe et al. 2002). This is due to the anionic character of dissolved As species.

Li et al. (2018) suggest red mud-based stabilizers to be effective for the remediation of Cd-contaminated farmland, since its continuous application in paddy soil cultivating two seasons rice could effectively decrease the Cd content in brown rice and had no negative impact on soil microorganisms. To obtain this conclusion, Li et al. (2018) used a mixture of red mud (50 wt%), diatomite (30 wt%) and lime (20 wt%) as stabilizers and added the red mud stabilizer to the

tested ultisol on the test field, where early rice, late rice and brown rice were planted. After the addition of the red mud-based stabilizer, a significant decrease in the Cd contents in brown rice (48%) and late rice (48%) was observed.

The addition of stabilizers to the soil induces a series of chemical and physical reactions (e.g. ion exchange, adsorption/desorption, surface complexation, precipitation, agglomeration and densification), which also changes the soil environment, affecting soil quality and properties (Capuyns 2015; Yin and Shi 2014). Therefore, it is important to consider the effects of the stabilizer on soil texture by cementing finer particles to coarser agglomerates, physicochemical properties, respiration intensity, enzyme activity, biodiversity, structure of microbial communities and reactivation of immobilized metals (Feigl et al. 2017; Hmid et al. 2015; Tang et al. 2016; Xie et al. 2015).

Steel slag

Steel slag is a solid waste or by-product from steel production. It mainly consists of silica (SiO_2), lime (CaO), iron oxide (Fe_2O_3 , FeO), alumina (Al_2O_3), magnesia (MgO), manganese oxide (MnO_2) and phosphorous oxide (P_2O_5); the exact composition varies with the furnace type, steel grades and pre-treatment methods applied (Motz and Geiseler 2001). The density of steel slag lies between 3.3 and 3.6 g/cm^3 . Since steel slag possesses a porous structure and a large surface area and creates a high pH in aqueous solution, it has received increasing attention in the past years in the areas of wastewater treatment and soil remediation. For example, Shi et al. (2011) studied the interaction of Hg-containing seawater with steel slag and observed a high adsorption capacity of steel slag for Hg.

Moon et al. (2015) used calcined oyster shells and steel slag to stabilize As-, Pb- and Cu-contaminated soil. As-contaminated soil (obtained from a timber mill site, where chromated copper arsenate was used as a preservative) was mixed with Pb- and Cu-contaminated soil (obtained from a firing range). Oyster shells were used as the main stabilizing agent in their calcined state and steel slag was used as a secondary stabilizing agent, which was produced during the separation of the molten steel from impurities in the furnace. The cooled slag was processed through a conventional aggregate crushing and screening operation. The waste oyster shells were calcined at 900 °C for 2 h to activate quicklime (CaO) from the former calcite shell mineralogy. The reacted steel slag and calcined oyster shells were used to complement each other since Taylor (1997) reported that slag placed in water dissolves to a small extent which can be explained by an increase in the ion activity product $[\text{Ca}^{2+}][\text{H}_4\text{SiO}_4]$ which hampers dissolution of calcium silicates. A silica-rich protective film depleted in Ca quickly forms and inhibits the



further dissolution, but the reaction continues if the pH is kept sufficiently high. The calcined oyster shells provided the high pH and Ca, while the steel slag provided a sufficient iron to form Ca–Fe(III)–arsenates which may have a lower solubility at high pH than simple iron arsenates such as scorodite whose solubility increases with increasing pH (Drahota and Filippi 2009), and to enhance the reactions encapsulating many contaminants (Zhu et al. 2013). As a result, the As, Pb and Cu concentrations in soil solution decreased with increasing calcined oyster shell content and steel slag dosages. Moon et al. (2015) found that As immobilization was mainly achieved by the formation of calcium and iron arsenates. Pb and Cu immobilization could probably be obtained by pozzolanic reaction products, such as calcium silicate hydrates and calcium aluminate hydrates.

Furthermore, Sheridan et al. (2013) investigated sub-surface-flow constructed wetlands (CW) with slag-based matrices for potential use in remediating areas affected by acid mine drainage (AMD). A small-scale CW (200 L) and $a < 19$ mm basic oxygen furnace slag (as bed matrix) were used. The unit was planted with a variety of plants and exposed to an artificial AMD solution. As a result, the system removed almost all of the soluble iron as well as more than 75% of sulphate, while the pH value of the AMD solution was increased from 1.35 to 4.

Comparison of metallurgical residues

Table 2 lists selected studies that focus on the remediation of heavy metals in soils by in situ immobilization. Most studies have been carried out at laboratory scale. All studies show very successful results for heavy metals immobilization.

If the industrial residues red mud and steel slag are compared as in situ immobilizers, some similarities, as well as differences, can be observed. Both materials show a large surface area and produce alkaline conditions upon reaction with an aquatic medium. Moreover, mixing these materials with other components, such as calcined oyster shells (Moon et al. 2015), can help to enhance and boost the reactions responsible for heavy metals immobilization. Furthermore, both residues immobilize Cu, Cd and Pb. However, As was found to be only stabilized by steel slag and not by red mud.

Residues from incineration and pyrolysis for heavy metal immobilization

Fly ash and biochar

The major type of solid waste material developed from thermal plants is fly ash (Bhatnagar and Sillanpää 2010).

According to the American Concrete Institute, fly ash is defined as “the finely divided residue that results from the combustion of ground or powdered coal and that is transported by flue gases from the combustion zone to the particle removal system” (ACI 116R-00 2000). Furthermore, other combustion products, such as incinerator ash, boiler slag and flue gas desulphurization (FGD) wastes, are presently considered to belong in the broader category of fly ash. They show similarities in texture, physicochemical properties and overall behaviour (Dermatas and Meng 2003). Fly ash consists of amorphous and crystalline phases of variable nature, type and content. The latent-hydraulic properties and the pozzolanic reaction of fly ash result in strengthening and hardening characteristics of cementitious materials (ACI 232.2R-96 2002). Nowadays, the application of fly ash in adsorption processes is increasingly considered due to economic reasons and the feasible level of efficiency. The adsorption is mainly enabled by the existence of high levels of reactive silica and alumina in the raw material (Bhatnagar and Sillanpää 2010). Whether fly ash is useable as a substitute for commercial adsorbents is depending on the fly ash source, chemical treatment and reactivity, density, particle size and surface area (Femina Carolin et al. 2017). Another relevant category of residues for immobilization is biochar, which is created through pyrolysis of different types of organic material. Biochar is described as a stable, carbon-rich material. It can either be a by-product of bio-oil production or manufactured specifically out of different organic waste streams, such as sludge or rice husk (Mohan et al. 2014; Zhang et al. 2015; Mahmoud et al. 2011).

The main mass fractions of oxides in both fly ash and biochar are SiO_2 , Al_2O_3 , Fe_2O_3 and CaO in different ratio (Fig. 1). The main inorganic component in biochar is Fe_2O_3 , whereas fly ash consists mainly of SiO_2 . It should be noted that the chemical composition of the both materials is usually given in oxides form, although the elements most likely occur in different chemical compounds (ACI 232.2R-96 2002).

The composition of these materials depends on source and treatment applied. For example, the composition of coal fly ash from Spain and siliceous fly ash from Poland varies within a single-digit percentage range, while the highest amount (51 wt%) of CaO was found in coal fly ash from China (Querol et al. 2006; Giergiczny and Król 2008; Ma et al. 2018). The influence of the origin of the material is even more evident when looking at various coal fly ash from the USA: bituminous, sub-bituminous, northern lignite and southern lignite coal fly ash show significant differences in composition. For example, the SiO_2 content ranges from 31.1 to 52.9 wt%. Further, the CaO content in bituminous fly

Table 2 Performance comparison of metallurgical residues used for heavy metals immobilization in soil

Material	Country	Test scale	Heavy metals	Dosage	Efficiency	Data source
Red mud (50%), Diatomite (30%), Lime (20%)	China	Field test	Cd	0.3 kg/m ²	20 to 48% Cd reduction in rice grown on an Ultisol	Li et al. (2018)
Red mud	Italy	Laboratory	Zn, Cd, Ni, Co, Cu	20%	Decrease of EDTA-extractable Mn from 100 to 20% of total Mn, Ni from 50 to 30% and Zn from 25 to 10%	Brunori et al. (2005)
Red mud + lime	United Kingdom	Field test (plant uptake)	Zn, Pb, Ni, Cd, Cu	3 to 5% of either material	Decrease of 85–96%, 75–90% and 70–90% water-soluble Zn, Cd and Ni, respectively, short-term decrease of Zn in plants	Gray et al. (2006)
Red mud	Austria	Greenhouse test	Cd, Zn, Ni	5% (w/w)	Reduction of NH ₄ NO ₃ -extractable Cd, Zn, Ni and Pb up to 91%, 94%, 71% and 83%, respectively	Friessl et al. (2004)
Red mud	Italy	Laboratory	Pb, Cd, Zn	1:25 ratio of red mud/metal solution	Adsorption of 1.85 mmol Pb per g red mud from solutions	Santona et al. (2006)
Red mud, red mud + mono-potassium phosphate	South Korea	Laboratory	Cd, Zn, Pb	1:1 ratio amendment to mine tailings	Conversion of plant available to non-plant available Cd by red mud 32%, Pb 88%, Zn 51%	Ahn et al. (2015)
Steel slag + calcined oyster shells	Republic of Korea	Laboratory	As, Pb, Cu	5 to 15%	Decrease in leachability of 71% As, 98% Cu and 99% Pb	Moon et al. (2015)
Nickel smelter slag	Canada	Laboratory	As	Percolation of contaminated water through a soil column at a velocity of 0.3 m/d	99–10% As removal after 65 d; removal capacity 1.04 to 1.05 mg/g	Chowdhury et al. (2014)
Steel slag	South Africa	Constructed Wetland	Fe	Percolation of synthetic acid mine drainage through a slag column at a rate of 30 mL/min	Almost complete removal	Sheridan et al. (2013)
Stainless steel slag + ammonium humate	China	Pot experiment	Cd	Up to 300 mg slag per g soil	16 to 58% removal rate	Zhuo et al. (2012)



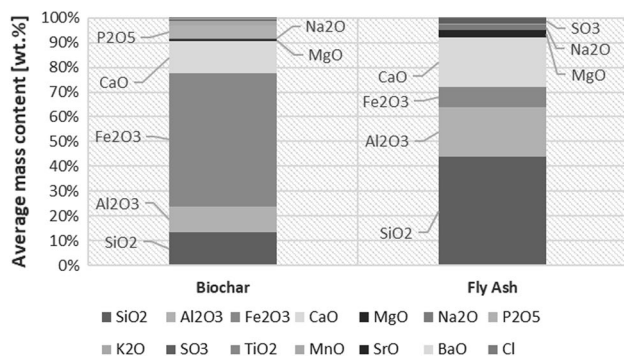


Fig. 1 Average mass contents of oxides in biochar and fly ash of various sources (According to Mohan et al. 2014; Zhang et al. 2015; Tomasevic et al. 2013; Somna et al. 2008; Ma et al. 2018; Querol et al. 2006; Dermatas and Meng 2003; Giergiczny and Król 2008; ACI 232.2R-96 2002)

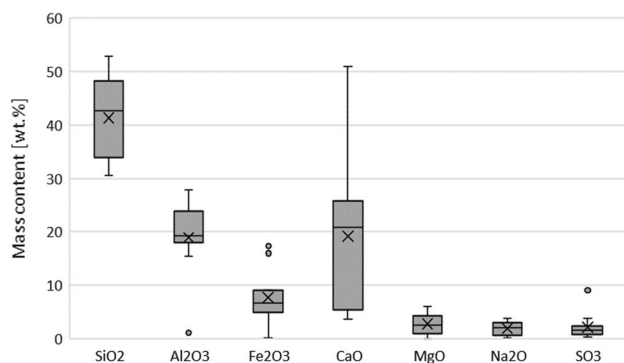


Fig. 2 Range of mass content of major oxides in fly ash using Boxplots with 25%-Quartiles (According to Tomasevic et al. 2013; Somna et al. 2008; Ma et al. 2018; Querol et al. 2006; Dermatas and Meng 2003; Giergiczny and Król 2008; ACI 232.2R-96 2002)

ash is 3.7 wt%, while in northern lignite fly ash, a CaO content of 20.9 wt% were measured (ACI 232.2R-96 2002). The ranges of the composition of fly ashes are given in Fig. 2.

The comparison of the Chinese fly ash with the European fly ash examples shows not only high levels of CaO, but significantly lower levels of SiO₂, Al₂O₃ and K₂O, respectively. This is due to the prior separation of the fly ash in an alumina-refinery, resulting in a calcium-silicate-powder. The study presented the ability of heavy metals removal (Ma et al. 2018). Nevertheless, for a successful immobilization of heavy metals, other properties, such as compressive strength, need to be considered in addition to the theoretical ability of heavy metals removal.

The specific surface areas determined according to Brunauer, Emmett and Teller (BET-SSA) of different residues from incineration and pyrolysis range from 1.9 m²/g in Spanish coal fly ash to 50.7 m²/g in Chinese coal fly ash (Guerrero et al. 2008). Among different types of biochar, the lowest BET-SSA was measured for oak wood biochar (2.1 m²/g), while palm kernel shell biochar had the largest SSA (191 m²/g) (Guerrero et al. 2008). Key factors influencing the SSA and thus the reactivity of biochar include the type of pyrolyzed biomass, particle size, maximum temperature and residence time (Guerrero et al. 2008). However, the sorption capacity is not only dependent on the material's SSA, as other physiochemical characteristics may be also important, such as the pore size distribution, surface site density, ion-exchange capacity, presence or absence of (amorphous) Fe-oxides and soluble Ca-bearing minerals, particle morphology and carbon content. Thus, high temperatures during pyrolysis may cause graphitization of organic carbon due to deoxygenation and dehydrogenation. This results in a significant reduction or loss of the adsorption capacity (Zhang et al. 2013, 2015; Ahmad et al. 2014). This dependence on temperature is not linear: for As sorption with sludge-derived biochar, the SSA first increased with increasing temperature, but at higher temperatures (> 600 °C), a loss of surface active sites and deoxygenation–dehydrogenation was found (Zhang et al. 2015). Other studies (Uchimiya et al. 2010; Yang et al. 2010) found an increasing sorption efficiency only for organic contaminants with increasing temperature due to increasing SSA, micro-porosity and hydrophobicity. In contrast, low temperatures cause oxygen-containing functional groups, electrostatic attraction and precipitation of metal phosphates using the phosphorous provided by manure-derived biochar (Cao and Harris 2010). This enables low temperature biochar for the remediation of inorganic and polar organic contaminants (Ahmad et al. 2014). Furthermore, the controlling mechanism for immobilization depends mainly on the oxidation state of a certain heavy metal. Dermatas and Meng (2003) demonstrated this for chromium in artificially contaminated clayey sand soils. It was found that adsorption is the main mechanism for Cr(VI) retention through pozzolanic-based stabilization/solidification, while the precipitation of chromium hydroxide removed much of the dissolved Cr(III) fraction (Dermatas and Meng 2003). This is because Cr(VI) appears in anionic form, so it is adsorbed at low pH, while Cr(III) appears in cationic form and precipitates at neutral/alkaline pH.



Table 3 Overview of different studies concerning heavy metal immobilization using residues from incineration and pyrolysis

Material	Medium	Country	Test scale	Heavy metals	Dosage	Efficiency	Data source
Magnetic oak wood biochar, Magnetic oak bark biochar	Synthetic aqueous solutions, metal concentrations 1–100 mg/L	India	Laboratory	Cd, Pb	10 g/L	100% adsorption of Pb and Cd	Mohan et al. (2014)
Sewage sludge-derived biochar	Synthetic aqueous solutions, 0–30 mg/L As	China	Laboratory	As	2 g/L	Sorption capacity 3.08 to 6.04 mg/g	Zhang et al. (2015)
Wood bark biochar, Copeat biochar, Palm kernel shell biochar	Soil (sandy loam) with Pb contents of 1260 and 1445 mg/kg	Indonesia	Laboratory	As, Pb	5%	Decrease in exchangeable Pb of 77.7 to 91.5% for wood bark biochar	Igalavithana et al. (2017)
Rice husk biochar	Soil (sandy clay loam, aridisol) with 2.86 mg/kg Cd	Egypt	Laboratory, Green-house	Cd	30 t/ha	Removal rate 97%	Mahmoud et al. (2011)
Fly ash	Synthetic aqueous solutions were used to produce a heavy-metal-containing fly-ash-based geopolymer. In a second step leaching from this material was analysed	Thailand	Laboratory	Cd, Ni, Zn	1 to 10% initial heavy metal concentration in geopolymer	Nickel leaching less than 10 mg/L, Cd and Zn leaching about 400 m/L	Somna et al. (2008)
Fly ash (thermal plant)	Stream sediment with 180 mg/kg pseudo-total Ni and 832 mg/kg pseudo-total Zn	Serbia	Laboratory	Ni, Zn	5 to 30%	Reduction in leachability of 90 to 99%	Tomasevic et al. (2013)
Coal fly ash	Synthetic aqueous solutions, initial metal concentrations 200 to 1000 mg/L	China	Laboratory	Co, Cu, Ni, Zn	1 g/L	100% removal of Ni	Ma et al. (2018)
Zeolitic material synthesised from Coal fly ash	Contaminated topsoil, application by manual mixing with the 25 cm top soil	Spain	Full-scale	As, Cd, Co, Cu, Pb, Sb, Ti, Zn	10 to 25 t/ha	Reduction in the leached metal amount of 90 to 99%	Querol et al. (2006)
Coal fly ash Class C	Artificially contaminated clayey sand soils, 4000 mg/kg Cr(NO ₃) ₃ , 4000 mg/kg K ₂ CrO ₄ , 7000 mg/kg PbO	USA	Laboratory	Cr, Pb	25%	Reduction of Cr leaching by 90% and of Pb by 57%	Dermatas and Meng (2003)
Siliceous fly ash and Fluidized bed combustion ash mixtures	Aqueous solutions containing metal ions as soluble salts were added into the binder composition. Subsequently, the leaching of the product was determined	Poland	Laboratory	Cd, Cu, Cr, Mn, Pb, Zn	20% ordinary Portland cement, 30% fluidal fly ash and 50% ground granulated blast furnace slag	Immobilization 86% for Cr(VI) in mortars made on blended binder to 93% in mortar produced on ordinary portland cement, more than 99% for Pb, Cd, Co and Mn	Giergiczny and Król (2008)



Comparison of residues from incineration and pyrolysis

In Table 3, an overview of the assembled studies concerning remediation of heavy metals is presented. The test scale was mainly laboratory; a full-scale implementation of coal fly ash was carried out only in one case to realise the remediation of soil after a pyrite slurry spit. In that case, zeolitic material was synthesized from coal fly ash and manually applied to the top 25 cm of the soil. The leaching of Cd, Co, Cu, Ni and Zn was decreased. This is due to the permanent negative charge of the cage structure of zeolites which makes them an excellent cation exchanger. As for As, Pb, antimony (Sb) and thallium (Tl), no improvements were found (Querol et al. 2006). Most studies present a good potential for immobilization of the investigated heavy metals. One study investigated the usability of siliceous fly ash and fluidized bed combustion ash mixed with binders at different ratios for the immobilization of heavy metals. Immobilization degrees up to 99.99% were achieved depending on the concentration of Cd, Cu, Mn, Pb and Zn. Only for Cr(VI), the removal was limited to 88.74% (Giergiczny and Król 2008). The reason for this is the anionic form of Cr(VI) which favours adsorption at low pH and the generally weak adsorption of chromate onto oxides (Gonzalez-Rodriguez and Fernandez-Marcos 2021).

Although many properties of different biochar made from residues were found to be suitable, the implementation of this material for remediation of landscapes contaminated by heavy metals seems to be less common than fly ash. Good results for Cd and Pb removal from the aqueous solutions were found for magnetic oak wood, which is produced by magnetization of oak wood biochar (Mohan et al. 2014), and oak bark biochar as well as sewage sludge-derived biochar. Zhang et al. (2015) interpreted these results as valuable for the relevance in the implementation for contaminated soils. Studies with wood bark biochar, cocopeat biochar and palm kernel shell biochar in contaminated lowland paddy and upland agricultural soils confirmed the usability of biochar for Pb decontamination, whereas an increased mobilization of As was found in the same soils. A possible explanation for this behaviour might be the high alkalinity and phosphorous content generated by the biochar which allows precipitation of lead phosphates (Mohan et al. 2014; Zhang et al. 2015; Igalavithana et al. 2017), but also the different behaviour of cationic Pb and anionic As may be a reason. The implementation of biochar for Cd decontamination was further examined in another study using lacustrine soil samples from Egypt. In this study, both isotherm experiments and greenhouse experiments were conducted to investigate

the influence of rice husk biochar on metal ion removal from aqueous solution. The results showed an increased Cd immobilization, which may be caused by carboxylic-carbon and aromatic-OH functional groups and a higher SSA and pore volume, besides the formation of organic and inorganic metal aquo-complexes (Mahmoud et al. 2011).

Conclusion

The utilization of industrial waste materials may decrease the environmental impact of remediation and is a cost-efficient alternative to segregate heavy metals, if the benefits (resource efficiency, heavy metals removal capacity) exceed the risks resulting from the introduction of additional contaminants released from the residues into the soil and (ground)water. However, most recent studies only focus on the desired immobilization of the contaminants already present in the soil, but do not investigate the fate of those contaminants (e.g. Cr, vanadium (V), molybdenum (Mo), tungsten (W) and fluorine (F) in steel slags) in the soil environments. Red mud and steel slag are able to immobilize As, Cd, Cu and Pb among others, which makes them useable for a wide variety of applications. Both wastes are able to neutralize low pH solutions and exhibit versatile physicochemical mechanisms, such as immobilization of metals through adsorption and surface complexation. The feasibility of using partly reacted fly ash and biochar for heavy metals immobilization was mostly assessed at laboratory scale, where fly ash showed a high potential for Cd, Co, Cr(III), Cu, Mn, Ni and Zn removal and biochar was successful in the immobilization of Cd and Pb. As the addition of a reactive treatment agent to a contaminated site severely affects the soil environment quality, it is important to consider the effects of the stabilizer on soil texture (i.e. by cementing finer particles to coarser aggregates), physicochemical properties, respiration intensity, enzyme activity, biodiversity and structure of microbial communities and reactivation of immobilized metals. The possible adverse environmental effects arising from the use of industrial residues, as well as apparent limits due to the legal framework, need to be evaluated for each application site, where industrial residues are to be used for heavy metals immobilization.

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Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

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