Title: Comprehensive stabilization of all streams of solid residues formed during sewage sludge thermal treatment – case study.

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Abstract

Modern wastewater treatment plants are currently struggling with the problem of processing and ecological management of sewage sludge. Thermal utilization is a frequently used practice of excess sludge utilization especially in large wastewater treatment plants, which seems to be a quite promising concept. However, during thermal treatment, fractions of ashes and dusts are generated. Such wastes, especially the dust fraction, can be potentially harmful to the environment. Therefore, the mentioned wastes should be stabilized. In this paper, a comprehensive method of stabilization is presented. Ash fraction, produced during sewage sludge thermal treatment, is used for dust fraction stabilization, hence no other solid medias, except cheap, commercially available cement, have to be used for stabilization. Laboratory and industrial scale trials were performed. Based on the results of the conducted research it can be stated that dust and ash fractions obtained from one facility can be stabilized together and the final product is safe for the environment since no potentially toxic elements are leached from the materials. There is a potential for improving the proposed method of stabilization to create fully environmentally safe construction materials.

Keywords: sewage sludge ash, sewage sludge dust, stabilization, sewage sludge management, heavy metals

1. Introduction

Nowadays the problem of ecologic sewage sludge management is becoming more and more significant. Since environmental quality standards are becoming more stringent, greater amounts of funds are being allocated to cover the costs of developing environmentally friendly technologies. Just in 1992 the European Union produced 5.5 million tons of dry sludge. After 18 years the production rose to 10 million tons. It is also worth mentioning that almost 17 million tons of sewage sludge was produced by the USA and the EU in 2005 alone. Moreover, it is estimated that by 2020 the production of excess sludge in the EU will have exceeded 13 million tons annually (Garrido-Baserba et al., 2015; Husillos Rodríguez et al., 2012; Pavšič et al., 2014). Those amounts have been rising rapidly over the years due to rapid urbanization and higher processing requirements (Rulkens, n.d.; Wang et al., 2012; Werther and Ogada, 1999).

Sewage sludge management methods based on its thermal processing are gaining popularity, since in most cases sewage sludge from large facilities cannot be used in agriculture due to high concentrations of heavy metals or the occurrence potentially hazardous organic species. The above-mentioned approach may lead to a complete utilization of organic fractions and even a 10-fold reduction of the volume of the produced waste. Those methods are especially attractive for sizeable wastewater treatment plants located in large urban areas where the sludge is produced in greater amounts (Chen et al., 2013; Sabbas et al., 2003). However, the mentioned techniques cannot be considered as fully complete management methods, as in some cases almost 30 % of solid residues need further processing and are often disposed of in landfills (Fytili and Zabaniotou, 2008; Song et al., 2013; Zhu and Zhao, 2011). Such an approach may cause a threat for the environment since many toxic substances can infiltrate into the soil and surface waters. When sewage sludge is combusted in cement plants, toxic substances can be removed and heavy metals are immobilized into a primary matrix material consisting mainly of inert silicates (Lin and Chang, 2006; Lin et al., 2012). Nonetheless, even the aforementioned method does not solve the problem of incomplete waste management. Therefore, landfilling is becoming much more expensive, since suitable site areas are rapidly decreasing in number and in size (Donatello and Cheeseman, 2013; Lin et al., 2012). What is also important, landfilling is at the bottom of the hierarchy of sewage sludge management methods. In some cases, however, it is still the only method possible.

Many researchers are seeking alternative ways to properly manage sewage sludge and waste generated during their processing. Stabilizing solid waste, combined with production of

construction materials, seems to be an environmentally and economically justified concept, especially because the concrete industry is dealing with decreased availability of raw resources (Cieślik et al., 2015; Garrido-Baserba et al., 2015; Pacheco-Torgal et al., 2008; Yang et al., 2013). That is why more and more attention is directed to developing new kinds of artificial lightweight aggregates and other sorts of low strength construction materials (Pacheco-Torgal et al., 2008; Tuan et al., 2013). However, during the implementation of the aforesaid concept, stringent environmental requirements have to be considered, together with local market demand, technologies already implemented in wastewater treatment plants, costs of possible changes or even social acceptance.

Ashes and dust obtained during sewage sludge thermal utilization may contain various toxic species, mainly inorganic, like heavy metals and metaloids, but not exclusively (Donatello and Cheeseman, 2013; Stiernström et al., 2014). While using the aforementioned fraction for the production of construction materials, specified pollutants should be stabilized through form changing, solidification or encapsulation (González-Corrochano et al., 2012; Nishida et al., 2001; Ramesh, A., Koziński, 2001). Cementation of the produced waste together with production of construction materials seems to be very attractive from the economical point of view. Even raw sewage sludge can be stabilized using lime, however ash fraction often contains CaO, which significantly improves the quality of the construction materials that would be produced (Husillos Rodríguez et al., 2012).

The presented study is focused on verifying the alternative method of solid waste stabilization. Ash and dust fractions obtained during industrial combustion of sewage sludge from two Polish wastewater treatment plants were stabilized together with a variety of commercially available cements. A common approach assumes using ashes obtained during the combustion of regular fossil fuels. It appears that sewage sludge ash can be successfully used as a stabilizing agent instead of regular fossil fuel ash. Such an approach allows to save significant funds and grants the possibility of internal processing and management of all streams of solid waste produced during sewage sludge thermal utilization which is used in the majority of large Polish wastewater treatment plants. However, more analyses have to be conducted to prove that it is possible to use such a product as a construction material, since dust fractions have never been considered as a suitable material for such products.

2. Materials and methods

2.1 Origin of the samples and sewage sludge processing technology

Ash and dust fractions were obtained from two sewage sludge treatment plants located in Poland: Group Wastewater Treatment Plant in Łódź (GWTP) and Wastewater Treatment Plant "Wschód" in Gdańsk (WTPW). Each facility processes around 120 000 m^3/d of raw wastewater and produces around 60 000 tons of excess sludge per year. The sludge is subjected to an anaerobic digestion process for 20.5 days and after that is directed to a sewage sludge thermal treatment facility.

In both facilities, excess sludge is pre-dried and combusted in a fluidized bed furnace at 850°C. Depending on the daily sludge characteristics the process can be autothermal. In some cases biogas obtained during sewage sludge fermentation is added to support the combustion. Ash fraction is retained at the first set of bag filters and stored in a silo. After the first set of filters, exhaust gases are treated. Different sorbents are injected into a treatment reactor in every wastewater treatment plant. WTPW uses a calcium sorbent mixed with small doses of active carbon. GWTP uses mainly active carbon mixed with sodium bicarbonate as a treatment agent. Solid residues after exhaust gases treatment are retained together with the dust fraction at the second set of bag filters and stored in a silo. Each facility produces around 10 tons/d of ash fraction. GWTP produces approximately 1 ton/d of dust fraction while WTPW produces 6 tons/d of dust fraction. The process scheme is presented in Figure 1. Ash and dust fractions collected for two years from both sewage sludge treatment plants were sampled, transported to a laboratory and stored in HDPE (High Density Polyethylene) containers.
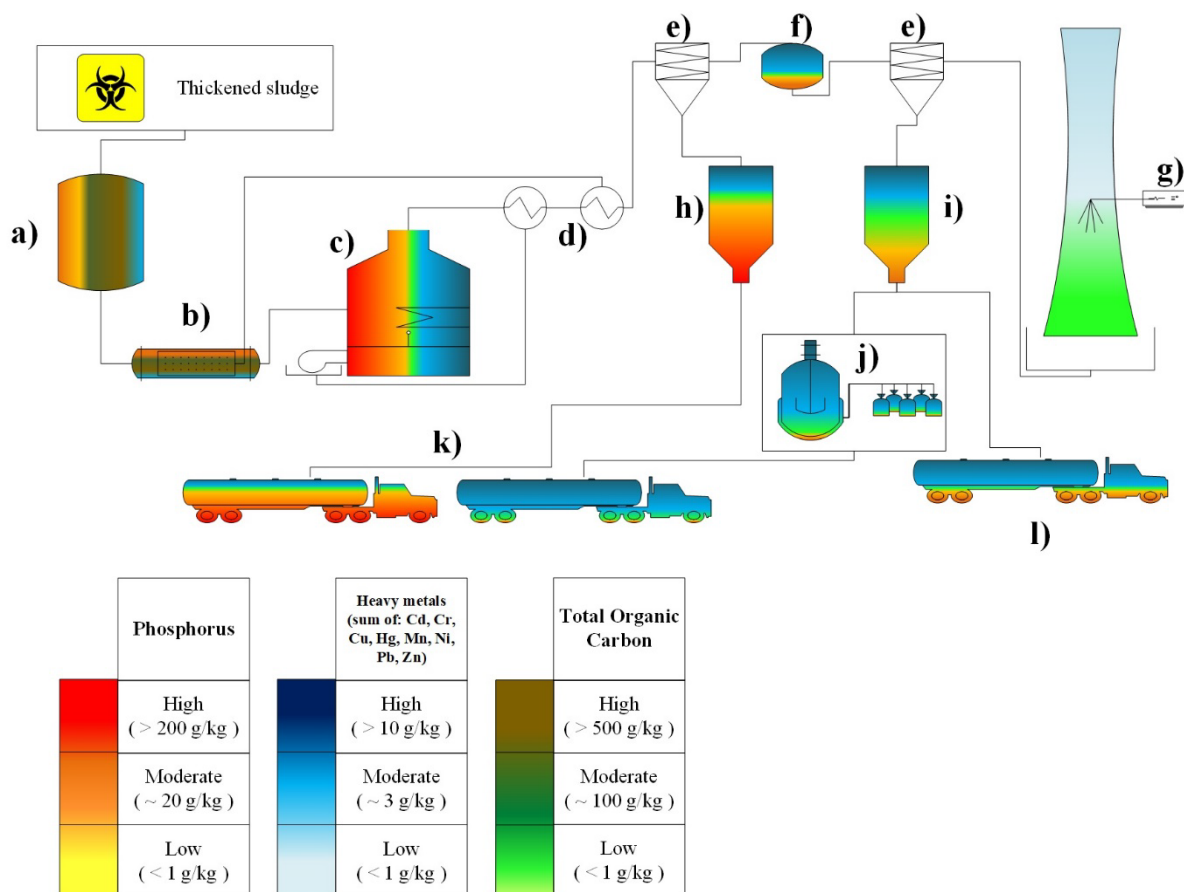
Figure 1. Sewage sludge thermal utilization facilities scheme a) primary tank, b) disc dryer, c) fluidized bed furnace, d) heat exchangers, e) bag filters, f) exhaust gas treatment reactor, g) chimney, h) ash silo, i) dust silo, j) solid residues stabilization facility, k) transport to non-hazardous waste landfill, l) transport to hazardous waste landfill.

2.2 Sample preparation

After ash and dust fraction collection, the obtained material was stabilized according to the proposed procedure. During the first set of laboratory scale tests, the samples were stabilized in 120 mL LDPE (Low Density Polyethylene) containers. After the addition of 29.0 g of dust fraction, 7.0 g of ash fraction and 12.8 g of commercially available Portland cement of different kinds (CEM II BV 32,5; CEM II AV 42,5; CEM II A-S 42,5; CEM III A 32,5; CEM III 42,5; CEM V 32,5), the sample was mixed for 1 min. Next, a mixture of 36.2 g of tap water with 2.7 g of commercially available aldehyde dehydration agent was added to the prepared sample and mixed for 1 min. After that, the series of samples was left for stabilization for 14, 28, 56, 70 and 84 days. Reagents and wastes ratios used for stabilization were adopted from the currently used method of stabilization which was developed for

WTPW and have to be maintained to 2020 when the warranty period passes. After this date the ratios can be optimized and changed.

After the laboratory scale tests, industrial scale trials were conducted. The stabilization was performed in the Stabilization Facility, presented in Supplementary materials 1, located in the WTPW sewage sludge thermal treatment facility. Ratios of reagents used for stabilization were similar to those used during the first set of laboratory scale test. The mixing was performed in the industrial mixer presented in Supplementary materials 2, and the mixing procedure was as follows: addition of 145 kg of dust fraction, 35 kg of ash fraction, 64 kg of Portland cement (CEM III A 32,5), 67 kg of technological water (wastewater after treatment), mixing for about 1 min., addition of 114 kg of technological water with 13.5 kg of aldehyde dehydration agent and further mixing for 1 min. The produced pulp was cast to a "big bag". Three mixing procedures were performed to fill the "big bag" container. In total, six batches of ash and dust fraction were stabilized with the presented method during one day. Consequently, around 9 tons of stabilized material were produced. Only one kind of Portland cement could be used during the industrial scale trial due to technological reasons. Sample "big bags" are presented in Supplementary materials 3. The samples were also left for stabilization for 14, 28, 56, 70 and 84 days.

Afterwards, since there is no possibility to change the ratios of reagents used for stabilization at the industrial scale, a second series of laboratory tests was conducted to check if the currently used method can be optimized and implemented when the warranty period ends. The method of stabilization was similar to the first set of tests, with the exception of amounts and ratios of stabilized media used. The cement used was CEM III A 32,5. The time of stabilization was 28 days. The masses of reagents used for stabilization during the second series of laboratory test are presented in Table 1.

Table 1. Masses of reagents used for stabilization during the second set of laboratory scale tests [g].

SAMPLE NUMBERS	1	2	3	5	6	7	8	9	10	12	13	14	15	16	17	18	20	21	23	24	25
Cement	10	15	10	20	15	10	15	10	15	15	10	15	5	5	15	10	15	15	10	10	15
Ash	25	25	25	25	25	25	25	15	25	25	30	30	25	25	25	25	25	30	30	25	25
Dust	15	15	15	15	15	15	15	25	15	20	20	20	15	25	15	10	15	15	15	15	15
Tap water	26	30	28	30	28	27	26	25	25	30	30	30	27	27	27	27	30	30	30	30	27

(The leftmost label "AMOUNT OF REAGENT" is printed vertically alongside the four reagent rows.)

After the sample collection and stabilization, those reagents were subjected to an analysis described in section "2.3 Analytical procedures".

2.3 Analytical procedures

In total, 43 samples of ashes and 39 samples of dust were analyzed. Considering all laboratory scale tests and industrial scale trials, 70 samples were stabilized and analyzed. Moreover, by using a leaching procedure described in subsection 2.3.2, 158 extracts were prepared and analyzed. Every single measurement was performed 3 to 4 times for every obtained extract or mineralized sample. The average is calculated based on 9 to 12 measurements, which is schematically presented in Figure 2. Precision as repeatability and intermediate precision, linearity and range of measurements together with the limits of detection and quantification was checked for every method.
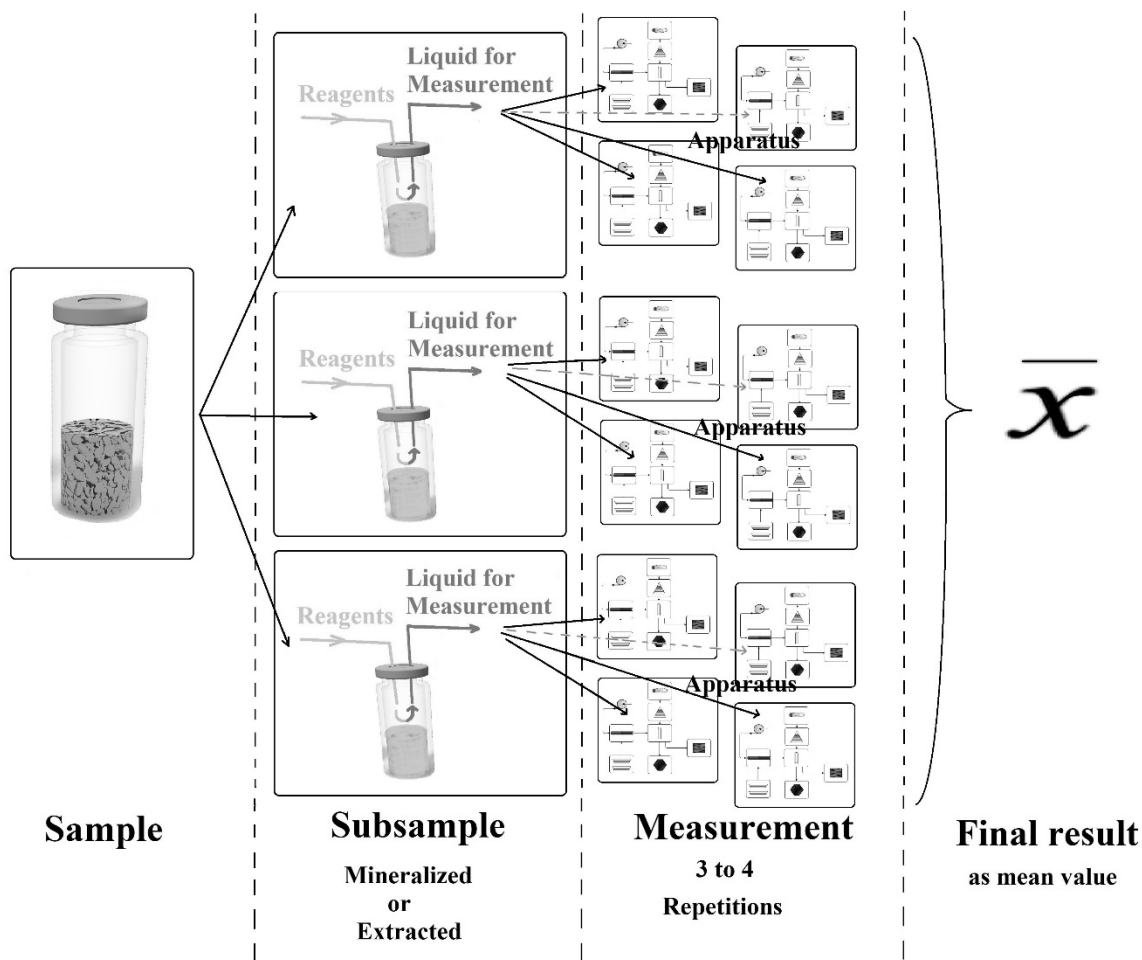
Figure 2. Scheme of sample preparation and measurements procedure.

2.3.1 Microwave digestion

For mineralization of the samples, Multiwave GO digestion system supplied by the company Anton Paar was used. Ashes, dusts and stabilized materials were all subjected to mineralization in order to carry out determination of trace elements. For each sample 3 independent subsamples were mineralized which is also presented in Figure 2. The mineralization program was as follows: 15 min. ramp heating to 110°C and hold for 20 min, next 30 min. ramp heating to 200°C and hold for 30 min. For mineralization, a mixture of oxidizing agents was used: 10 mL of 65 % nitric acid, 2 mL of 36 % hydrochloric acid, 2 mL of perhydrol. All reagents were of Suprapur grade and supplied by Merck.

2.3.2. Leaching

A leaching procedure which was used for extracts preparation was adopted from EN 12457-2006 "Characterization of waste. Leaching. Compliance test for leaching of granular waste materials and sludge" with the following changes: Liquid to solid ratio was equal to ¼ or

1/10; Time of extraction was 24 h ± 0.5 h or 72 ± 1 h; mixing with 160 rpm. Only stabilized/cemented materials were subjected to leaching to estimate the retention of elements in produced media.

2.3.3 Atomic absorption spectroscopy

In order to determine heavy metals at moderate concentration, Flame Atomic Absorption Spectrometer SensAA, , with dual beam optical system and air acetyl flame, supplied by GBC Scientific equipment Pvt. ltd (Australia), was used. Deuterium lamp for background correction and hollow-cathode lamps as a radiation source were installed. For low concentration heavy metals determination, Graphite Furnace Atomic Absorption Spectrometer Savant AAZ, supplied by GBC Scientific equipment Pvt. ltd (Australia), with Zeeman background correction, was used. Technical grade argon was supplied as a carrier gas, and hollow-cathode lamps were installed as a radiation source. Mercury/MA-3000, supplied by Nippon Instruments Corporation (NIC, Japan), was used to analyze mercury by cold vapor technique and pure oxygen was used as the carrier gas.

2.3.4 X-ray fluorescence

To carry out simultaneous determination of elemental species in solid samples (ashes, dusts, and stabilized materials), Thermo Scientific Niton Xl3t GOLDD+ XRF Analyzer was used.

3. Theory

A lot of researchers conduct analyses of ash fraction after sewage sludge thermal utilization. However, since dust fraction is often produced in much lower amounts, results of their analyses are often neglected. Very little information about the mentioned dust fraction can be found. Yet, there is a lot of data concerning the quality of sewage sludge ashes. It was stated that ash fraction consists predominantly of Si, Al, Ca and Fe. Because of high temperature processing, these elements occur mostly in the form of SiO_2, Al_2O_3, CaO, and Fe_2O_3 (Anderson, 2002; Donatello et al., 2010; Donatello and Cheeseman, 2013; Smol et al., 2015). Such substances are commonly available in ordinary Portland cements. Unfortunately, in some of the studied cases sewage sludge ash contained relatively low concentrations of the aforementioned SiO_2 and Al_2O_3, which are responsible for pozzolanic activity in cement based materials (Baeza-Brotons et al., 2014). It is important to mention that Al and Fe concentrations in sewage sludge ash highly depend on the sewage sludge composition and the method of phosphorus precipitation, since commercially available precipitation agents are

based on Fe or Al compounds (Wang et al., 2012). The above-mentioned ashes may contain a wide variety of other, potentially toxic, trace elements. Cd, Pb, Mo, Sb, or Se are considered as pollutants of the highest concern since their concentration may even prevent the material from being safely landfilled (Donatello and Cheeseman, 2013). Sometimes even concentrations of essential macro elements like Ca and K can be present at such a high level, that one can observe the eco-toxicity of leachates from ash fraction used for production of construction materials. However, long term toxicity observed in crustaceans is possibly triggered by difficulties with acclimatization of the tested organisms to rapidly changing conditions, rather than the toxic effect of high K concentration in the environment (González-Corrochano et al., 2012; Stiernström et al., 2011).

3.1. Possibility of production of construction materials from solid waste obtained during thermal utilization of sewage sludge

Using waste as an additive for construction materials is a common practice nowadays. There are reports stating that it is even possible to apply waste as the main ingredient in the production of construction materials. One of the oldest investigated ways of ecological management of ashes after thermal utilization of sewage sludge was the manufacturing of bricks and tiles (Anderson, 2002; Donatello and Cheeseman, 2013). Unfortunately, in most cases if blocks or other construction materials are produced from 100 % of recycled aggregates, products exhibit lower durability than those manufactured with natural ones (Smol et al., 2015). However, in another research, even 85 % of the studied samples show compression strengths equal to or greater than the minimum accepted standards (Baeza-Brotons et al., 2014). Other researchers stated that an addition of over 30 % of sewage sludge ash leads to a significant decrease in compressive strength (from 46 to 20 MPa), so the mentioned approaches require implementation of some process adjustment and may affect the quality of the obtained products (Donatello and Cheeseman, 2013; González-Corrochano et al., 2012). There are also reports showing that 10 % substitution of cement with sewage sludge ash leads to 25 % decrease in durability of the obtained material, despite the fact the hydraulic properties of sewage sludge ash are very similar to the properties of Portland cement (Chen et al., 2013). Other researchers claim that a 10 % addition does not influence the pore structure of the pastes (Chen and Poon, 2017).

Dried or wet sludge has been used as an additive to cement mortar or concrete (Valls et al., 2004; Yang et al., 2013). Sewage sludge can also be used for stabilized/solidified admixture of cement–jarosite/alunite or cement–bentonite production (Katsioti et al., 2008) or stabilized

and solidified with cement and aluminate $12CaO \times 7Al_2O_3$ (Zhen et al., 2012). It is also possible to obtain controlled low-strength material by stabilizing sewage sludge with biomass ash. The compressive strength of the obtained materials was between 0.5 to 2.5 MPa (Pavšič et al., 2014). Nevertheless, using wet sewage sludge may cause lumps in the produced mixtures, mainly because of high organic load and water content (Valls and Vàzquez, 2000; Yang et al., 2013).

Analyses of other tests results indicate that construction materials obtained from only sewage sludge ash can lead to higher stabilization factors and strength than materials produced from dried or wet sludge. However, sewage sludge thermal utilization is always connected with high operational and investment costs, and higher consumption of energy during production (Wang et al., 2009). There are many more approaches to producing eco-construction materials from waste from sewage sludge treatment plants (Smol et al., 2015; Valderrama et al., 2013), however they are outside of the scope of this study.

It is important to mention that solidification and stabilization technology may be successfully used to decrease the leachability of organic and inorganic pollutants. By the mentioned application, potentially hazardous waste can be transformed to nonhazardous and safely landfilled or even used for the production of eco-materials (Song et al., 2013; Ucaroglu and Talinli, 2012). For some years, stabilization and solidification techniques were considered as the "best demonstrated available technology" (BDAT), for example by institutions like the United States Environmental Agency (Singh and Pant, 2006; Song et al., 2013).

The previously mentioned chemical composition of the used waste has great influence on the quality of the produced construction material. As an example, enhancement of early hydration and incensement of early strength of the eco-cement pastes can be obtained by a properly increased amount of aluminum oxide, which is one of the main ingredients of ashes after sewage sludge thermal utilization. On the other hand, decreased concentrations of Al and Fe may cause lower early strength of such materials. What is more important, higher concentrations of Cr, Cu and Zn can influence the initial setting, hydration of the cement and strength of the produced eco-cement mortar (Lin et al., 2004, 2012).

3.2 Leachates from stabilized materials

Before the implementation of the proposed concept, that is production of construction materials with the use of solid waste produced during thermal utilization of sewage sludge, it is important to prove the ecological safety of the presented approach. Even before the

landfilling of the produced waste it is necessary to estimate the leachability of certain species. Sequential leaching of produced materials is a common method for evaluating the availability of potentially harmful elements and thus for proving the ecological safety of the tested stabilization method (Huang et al., 2007). Materials produced with the addition of sewage sludge ash contain significantly higher amounts of heavy metals and thus the leachability of the aforementioned pollutants might be greater (Lin et al., 2012). However, there are researchers proving that sewage sludge ash can be used for Pb stabilization in soil (Li and Poon, 2017). Moreover, most researchers prove that the leaching ability can be significantly decreased after stabilization, solidification, cementation or other encapsulation technologies. In most cases, heavy metals availability is negligible (Hunce et al., 2012; Lee, 2007; Lin et al., 2012; Luna Galiano et al., 2011; Song et al., 2013; Ucaroglu and Talinli, 2012). Cement based solidification and stabilization is widely reported and allows binding of harmful pollutants, such as heavy metals, in solidified material. Potentially toxic species can be stabilized in produced materials in one or more of the following ways: adsorption, chemisorption, inclusion, incorporation, ion-exchange, passivation, precipitation, or surface complexation (Song et al., 2013). Major elements, the previously mentioned Ca, Si, Al and Fe, also remain immobilized in the structure of the produced materials (Lin et al., 2012).

If the produced material is solidified by applying an acidic reagent, the leaching ability of heavy metals can be decreased as compared to solidification without acidic reagent treatment. Stabilization seems to be especially effective for Cu, Ni, Pb and Zn species. Moreover, acidic pretreatment of the raw material can result in compressive strength increase in the cemented waste material (Cieślik and Konieczka, 2017; Moon and Dermatas, 2005). However, low pH of the extraction agent and extraction temperature growth can increase the release of toxic species (Song et al., 2013). In some cases, sintering of the materials can improve immobilization of metals like Be, Co, Cr, Cu, Mn, Ni, Pb and Zn and most of the rare earth elements (González-Corrochano et al., 2012). However, such approaches are considered highly expensive. In many sewage sludge treatment plants, where stabilization technologies are already implemented, the process can be easily improved to obtain even better stabilization of potentially toxic elements. Moreover, it is possible to obtain simultaneous decrease in processing costs due to the use of the produced sewage sludge ash fraction instead of buying regular fossil fuel ashes.

4. Results and discussion

After determining the selected elements in the samples of ashes and dusts collected from the two wastewater treatment plants, the stabilization of waste was performed. Stabilized materials were subjected to a leaching procedure and the analysis of the eluted fraction was done. The results and the conclusions which were drawn from the analysis are described in subsequent sections. For the analysis of the obtained results mainly boxplots and histogram analyses were used.

4.1 Ash fractions

Mean concentrations of Ba in ash fraction differ slightly between the two wastewater treatment plants. They are visibly higher in GWTP. Moreover, the concentrations of Cr, Mo Sb, Zn in GWTP are significantly greater than in WTPW. Cr concentrations were higher even by an order of magnitude, while Mo, Sb and Zn concentrations differences reached ½, 4 and 4 fold respectively. Mean concentrations of Cu do not differ statistically significantly, but in several cases those values were even 2-3 times higher in GWTP compared to those from WTPW. A similar phenomenon was observed for Ni concentrations, with the exception that the concentrations of Ni in GWTP were even 6-fold higher in several cases. In contrast to previously observed trends, higher concentrations of Pb and Sn were generally observed in WTPW. The differences here reached the factor of 5. The concentrations of Fe do not differ between the two facilities, but the amounts of this element in ash fraction can differ greatly in time. The observed phenomenon depends on the amount of phosphorus precipitation agent added at the step of wastewater treatment. Moreover, the concentrations of As, Cd, Hg, Mn, Se, Co and Cl species also do not differ between the two facilities in a statistically significant way. Example boxplots showing the differences in concentrations of the selected elements are presented in Figure 3. All boxplots for ash fractions are presented in Supplementary materials 4.
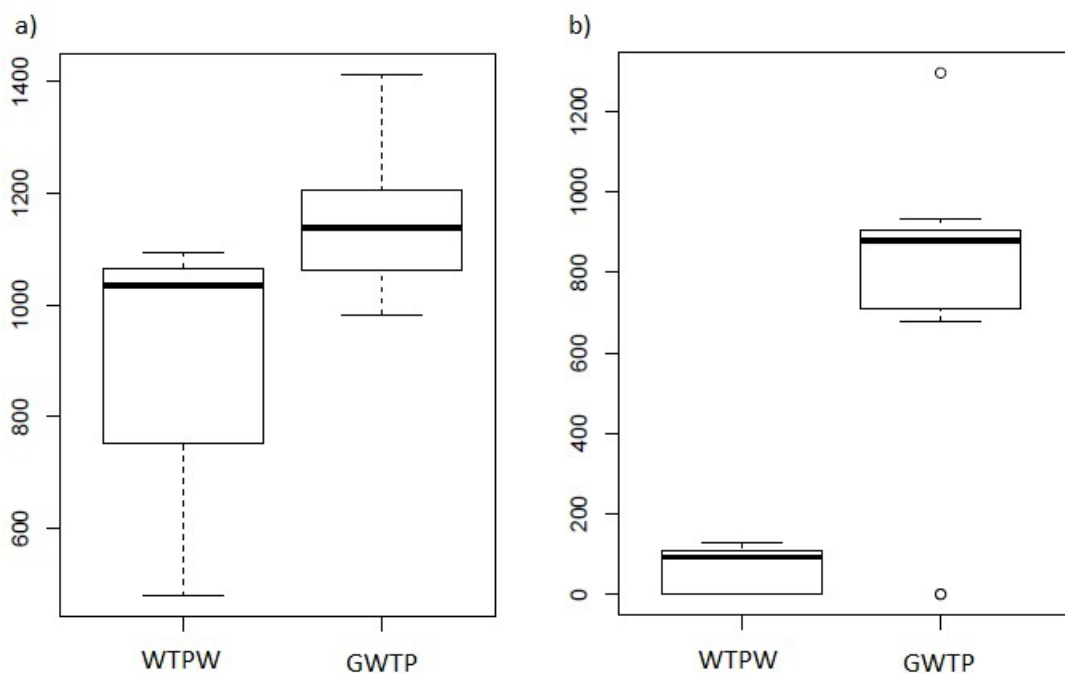
Figure 3. Boxplots presenting mean concentrations, quartiles and outliers for a) Ba, b) Cr in ash fraction from WTPW and GWTP. Values in [mg/kg].

As mentioned before, concentrations of some elements show temporal variability. The amounts of Cr, Cu, Sb and Zn increased slightly in both facilities during the sampling time. The increases were around 10-fold, 3-fold, 2-fold and 2 fold, respectively. Concentration growth of some elements was significantly slower and reached only around 10 - 25 %. Those were Ba, Fe, Hg, Mn. The amounts of other elements were classified as time independent. Those trends could be caused by the treated wastewater quality. Both facilities observed slow rises of heavy metals load in the received stream of the collected wastewater.

4.2 Dust fractions

The trends observed for the concentrations of the selected elements in the dust from WTPW and GWTP are similar compared to the trends for the ash fraction. In most cases the concentrations of the determined heavy metals are significantly lower for WTPW compared to GWTP. Such a phenomenon was observed for As, Ba, Cr, Cu, Fe, Hg, Mn, Mo, Pb, Se, Zn, Cl and Sn. However, Cd concentrations seem to be at a similar level and do not differ in a statistically significant way. Also Ni concentrations do not show high differences over time, but this statement concerns only the mean value. Generally, concentrations of Ni in dusts from WTPW are considered to be slightly lower, as shown in Figure 4. The concentrations of Sb show a different trend since the dusts from WTPW show higher concentrations than those from GWTP. The concentrations of Co are similar for dusts from both facilities. Lower

concentrations of the determined elements in dust from WTPW result from different methods of purification of exhaust gas in both facilities. In case of GWTP, relatively low amounts of sodium bicarbonate and active carbon are used for exhaust treatment. In the WTPW thermal utilization facility, much greater amounts of calcium based sorbent are used. That is the reason why ash to dust fraction ratio in the latter facility can reach even 5:3. Therefore, noticeably lower concentrations of heavy metals in dust fractions from WTWP result from a high dilution factor of the obtained solid fraction with the reagents, rather than much different characteristics of the utilized sewage sludge from the two facilities. Figure 4 shows examples of boxplots for the selected elements. All boxplots for dust fractions are presented in Supplementary materials 5.
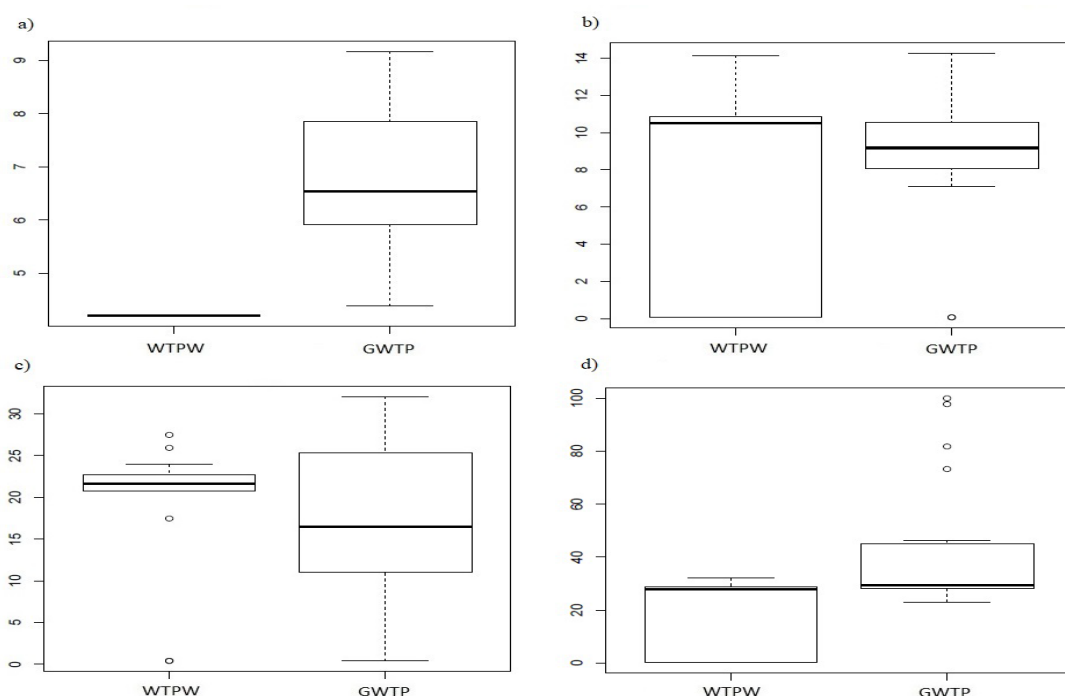

Figure 4. Boxplots presenting mean concentrations, quartiles and outlier for a) As, b) Cd, c) Sb and d) Ni in dust fraction from WTPW and GWTP. Values in [mg/kg].

Concentrations of some elements in dust fractions from both sewage sludge treatment plants also slowly increase. It is particularly noticeable for Cr, Hg, Mo, Pb and Cl, where increases were around 10-fold, 4-fold, 4-fold, 10-fold and 5-fold respectively, as in the case of the ash fraction. Other elements like Cu also show a slow growth in time, close to around 10 %. However, in most cases concentrations of the determined elements are considered to be stable over time.

4.3 Relations between elements content in ashes and dusts

After comparing the results of elemental analyses of dusts and ashes, the following conclusions were formed. The concentrations of determined elements were at substantially higher levels in collected ash fractions than in dust fractions in both facilities. Such a phenomenon was observed for As, Ba, Cr, Cu, Fe, Mn, Ni, Pb, Zn, Co and Sn. Moreover, the differences between concentrations of Cu, Fe and Zn in dusts from WTPW are considerably greater than those between the same fractions from GWTP. However, in case of Sn concentrations the tendency is reversed. Those significant discrepancies are presented in Figure 5. Furthermore, the concentrations of Cd, Hg, Mo, Sb and Se can be considered as being at the same level both in ashes and in dusts. Nonetheless, such a phenomenon occurs in all of the mentioned cases only in solid waste collected from WTPW. Dust fractions collected from GWTP are characterized by higher concentrations of Se and Hg than in ash fractions. That may be caused by the fact that the mentioned elements are considered as more volatile than the other analyzed species. Moreover, the exhaust gas treatment system from GWTP, which applies mainly active carbon and small additions of sodium bicarbonate, is probably more effective for the retention of volatile species. A similar tendency can be observed for Cl concentrations. Dust fractions collected from WTPW contain around 2500 mg/kg of the mentioned element, while dust fractions collected from GWTP may contain even around 8500 mg/kg. Ash fractions from both facilities contain less than 400 mg/kg.
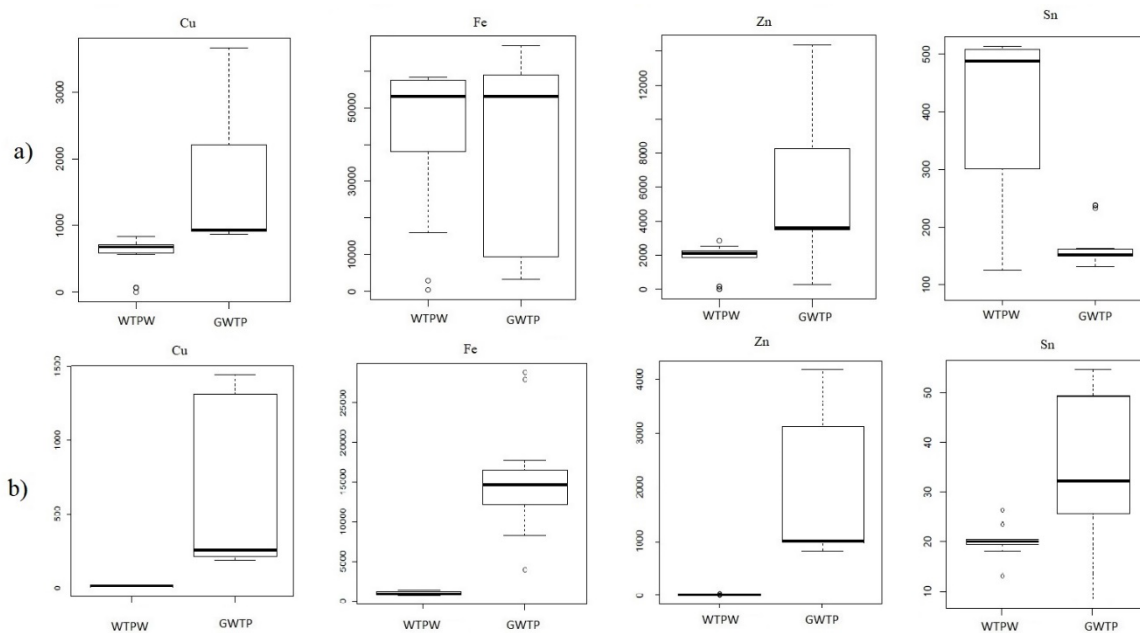
Figure 5. Boxplots presenting the mean concentrations, quartiles and outliers for Cu, Fe and Zn for a) ash fraction and b) dust fraction in WTPW and GWTP. Values in [mg/kg].

In most cases, ash and dust samples from GWTP contain higher amounts of potentially toxic elements. However, those differences do not influence the leaching ability of stabilized waste from the subject facilities. However, based on the ash and dust elemental analyses it seems that the presented technology could be particularly interesting for WTPW, where the obtained materials should probably contain higher amounts of Ca and lower amount of Cl species and other elements which may affect the quality of the produced material.

Unfortunately, slow rise of some of the analyzed pollutants in time was observed in ashes and dust fractions from both studied facilities. The recognized trends result in an increased interest in modern methods of sewage sludge management for both sewage sludge treatment plants since "no waste" approaches are rather rarely presented and are considered as the most promising ones (Cieślik and Konieczka, 2017; Pavšič et al., 2014; Smol et al., 2015). That was the main reason for organizing industrial scale trials of the proposed stabilization method, allowing to form cement aggregates using all streams of solid waste produced during thermal utilization of sewage sludge, that is ash and dust fractions. The proposed approach can be especially interesting for WTPW, even in the case of the landfilling of the produced materials, since it can decrease the costs of the currently used process.

4.4 Stabilized/cemented materials

After the elemental analyses of the stabilized ashes and dust fractions, with the use of commercially available cement, it became clear that the concentrations of most heavy metals

were significantly lower because of purposeful mass increase. The obtained materials were characterized by 2 – 5 fold lower concentrations of As, Ba, Cu, Fe, Mn, Pb, Zn and Sn compared to ash fraction. The statement concerns both facilities as well as materials produced in laboratory scale tests and during industrial scale trials. All boxplots are presented in Supplementary materials 6. The concentrations of Cd, Ni, Sb, Se and Co are considered negligible. The elemental composition of the materials produced during both laboratory scale test and industrial scale trials is considered to be almost identical. The possibility of contamining the environment with these elements in the case of introduction of the produced materials into it, is recognized as highly improbable for the material from all three trials. On the other hand, the mentioned products exhibit even a 2-fold increase of Ba, Cr, Hg, Mo and Sn compared to non-stabilized dust fractions. However, all the mentioned elements are considered to be stabilized while the leaching factor was negligible in a vast majority of the studied cases. Although, contrary to what was expected, Cl concentrations in the produced materials were at a relatively high level of even around 1000 mg/kg. Such high concentrations of Cl species can influence the durability of the obtained products.

Widely understood durability tests of the produced materials were conducted by a private laboratory and are not the subject of this research since there will be no permission to publish them. What is important, all of the produced samples were characterized by shear strength and compressive strength over the minimum value indicated in former local legislation concerning the landfilling non-hazardous waste ("Polish Ordinance of the Ministry of Economy of 8 January 2013, on the criteria and procedures for the disposal on certain type landfills). The current legislation ("Polish Ordinance of the Ministry of Economy of 16 July 2015, on the acceptance of waste for landfill") which was implemented from EU directives ("Council Decision of 19 December 2002, establishing criteria and procedures for the acceptance of waste at landfills pursuant to Article 16 of and Annex II to Directive 1999/31/EC") requires only the leachability tests. The only information that can be mentioned is that the durability is far above the former legal limit (≥ 25 kN/m^2). However, the value is insufficient to prove the suitable durability of the obtained samples. Although, there is a possibility of increasing the durability of the stabilized material through previous implementation of phosphorus recovery from ash fraction based on acidic extraction (Cieślik and Konieczka, 2017; Moon and Dermatas, 2005). There is a vast variety of phosphorus extraction methods with the use of acidic reagents (Gorazda et al., 2017; Wzorek et al., 2006). Nevertheless, it is worth mentioning that there are methods of phosphorus recovery which are not based on acidic

extraction from ash fraction (Li et al., 2017). To the best of the authors' knowledge, no work has been done to study the effects of phosphorus precipitation agents on chemical and mineralogical composition of sewage sludge ash.

4.4.1 Extracts from cemented materials

During the first set of laboratory scale tests it was found that the kind of commercially available cement used for the stabilization does not influence the retention factor in a statistically significant way. Moreover, it was impossible to perform a similar test during industrial scale trials. Furthermore, it was found that the time of stabilization also does not influence the retention of the determined species in a statistically significant way. It was observed for both the first set of laboratory tests and the industrial scale trials. Therefore, during the second set of laboratory scale tests it was decided to conduct the stabilization only for 28 days with the use of one kind of cement – CEM III A 32,5.

In a vast majority of cases the concentrations of leached species were below the limit of detection. However, such information was sufficient to compare the leaching abilities with the above-mentioned regulatory standards, and to prove the environmental safety of the proposed method. In case of both Cl^- and SO_4^{2-} leaching, it turned out that the determined species concentrations are below the lowest acceptable values for materials other than hazardous mentioned in the legislation. The determined anions are probably extracted together with non-hazardous elements like Mg, Na, and Ca, while heavy metals are practically retained in stabilized materials. Organic fractions are flushed from cemented materials in negligible amounts, which is shown in Figure 6. That is due to the fact that a high percentage of organic fraction is decomposed during the thermal utilization process. Moreover, none of the reagents used for cementing contain high organic loads, except the dehydration agent which is added in small amounts, below 4.0 % w/w. The possibility of contaminating the environment with the organic fractions is considered highly improbable, despite the fact that some potentially toxic organic species, such as some of polychlorinated biphenyls, can occur. The concentrations of Hg in the extracts were determined at a relatively high level, exceeding the more restrictive values of the legislation in the case of the samples from the first set of laboratory scale tests and the industrial scale trials. However, during the second set of laboratory tests the Hg concentration was negligible, even 3 order of magnitude lower than for the previously mentioned samples. It was probably the increased amount of ash fraction in the stabilized material that improved the retention of Hg. It is important to mention that the Hg content in materials stabilized during the second set of laboratory scale tests does not differ significantly

compared to the materials obtained during the first set of laboratory tests and industrial scale trials (differences between 0 and 25 %). What is also worth mentioning, such high retention capabilities were observed even for the more aggressive leaching procedure presented in subsection 2.3.2. Concentrations of the mentioned species determined in the prepared extracts are presented in Figure 6 with the comparison to the values from the previously mentioned legislation. Differences between the concentrations of Hg in the samples prepared during the first set of laboratory scale tests, the industrial scale trials and the second set of laboratory tests are presented in Figure 7.
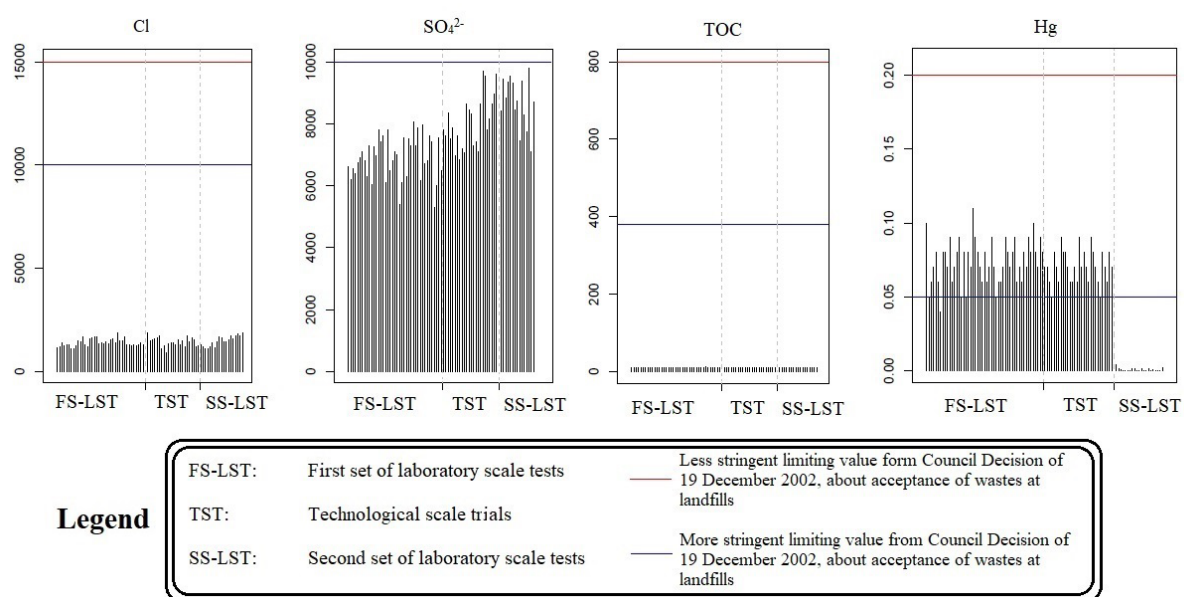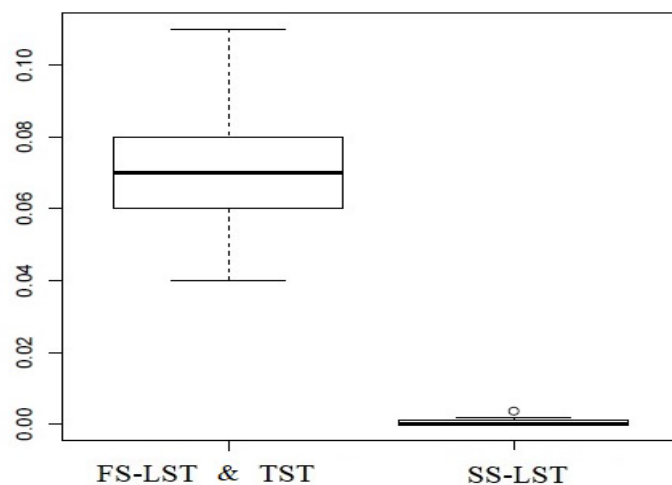

Figure 6. Concentrations of selected parameters in extracts after leaching of cemented materials and values from current legislation concerning landfilling. a) Cl b) SO_4^{2-} c) TOC concentrations d) Hg concentrations. Values calculated as in the mentioned directive in mg of pollutant / kg of leached solid material [mg/kg].

Figure 7. Boxplots showing differences in concentrations of Hg in extracts from samples prepared during industrial scale trials and laboratory tests. These values were calculated as in the mentioned directive in mg of pollutant/kg of leached solid material [mg/kg].

As mentioned before, most elements like Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Zn, Co, and Sn are leached from stabilized materials at a level below 1.0 % for samples stabilized during the second set of laboratory scale tests and the industrial scale trials. Specific values are shown in Table 2. It is important to mention that those values in most cases were compared with the limits of detection for every element determined in the extract fraction. Therefore, the real stabilization degree can be even higher than the one presented here. The specific values were presented only if it was possible to detect a given element in the stabilized material. Values which were presented as n.d. could not be calculated, since the concentrations of the selected elements were below the limit of detection in materials stabilized during the second set of laboratory scale tests.

Table 2. Mean percentage degree of leaching of specific elements from stabilized materials produced during industrial scale trials and the second set of laboratory scale tests [%]

Element	Cd	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Sb	Zn	Cl	Co	Sn
Industrial scale trials [%]	0.8	0.051	0.045	0.00091	0.84	0.015	0.22	0.2	0.23	0.0057	75	0.32	0.0011
Second set of laboratory scale tests [%]	n.d.	n.d.	0.076	0.00097	0.000054	0.019	0.49	0.38	n.d.	0.01	n.d.	n.d.	0.0021

It is worth paying attention to Fe leaching degree, which is equal to 0.00091 % for the industrial scale trials and 0.00097 % for the second set of laboratory scale tests. The stabilization factor is significant despite the fact that the concentrations of Fe in ash fractions are extremely high and the mean values for all analyzed samples are above 50 g/kg. That directly influences the concentrations of Fe in the stabilized material, which were determined at around 20-30 g/kg. A similar phenomenon is observed for Zn, whose concentrations in a stabilized material are around 800 mg/kg and the leaching degree is 0.0057 % for the industrial scale trials but only 0.010 % for the second set of laboratory scale tests. Degrees of stabilization for materials obtained during the second set of laboratory scale tests are generally a bit higher than for materials produced on industrial scale, with the exception of Hg. However, such a phenomenon can be caused by the fact that in most cases the concentrations of elements in extracts were at the level of the limit of detection. It is highly possible that the realistic leaching ratios would be even lower. What is also important to mention, such a high retention level was obtained for every ratio of reagents tested. No significant differences in concentrations of the determined heavy metals in extracts were found regardless of the ratios of waste used for stabilization. Therefore, it is possible to implement the proposed method for stabilization of all streams of produced ashes and dusts from both facilities: WTPW and GWTP.

5. Conclusions

The proposed method of comprehensive stabilization of all streams of solid residues produced during thermal utilization of sewage sludge from the two surveyed sewage sludge treatment plants can be considered environmentally safe. The leaching degree of potentially toxic elements and heavy metals from stabilized media is negligible. In most cases the mentioned degree is lower than 0.01 %. The proposed method can be implemented in the already existing facility in WTPW or adapted in GWTP. Implementation of the presented method in

other facilities is possible, however it has to be tested first, since the obtained sewage sludge ash and dust differs, at least slightly, in every sewage sludge treatment plant. The stabilization allows production of safe materials which may be improved and used as a basis for the production of ecological low strength construction material. The fact that the strength of the material can be enhanced justifies continuing the research. Moreover, the owners of GWTP and WTPW can even consider selling the materials for minimum price, since the savings which can be made due to total elimination of landfilling costs would be enough of a reimbursement. At the moment the material can be easily improved to fulfill regulation standards and be safely disposed of at the non-hazardous materials landfills. Another series of industrial scale trials should be carried out to prove the effectiveness of the improvements.

6. Acknowledgements

7. References

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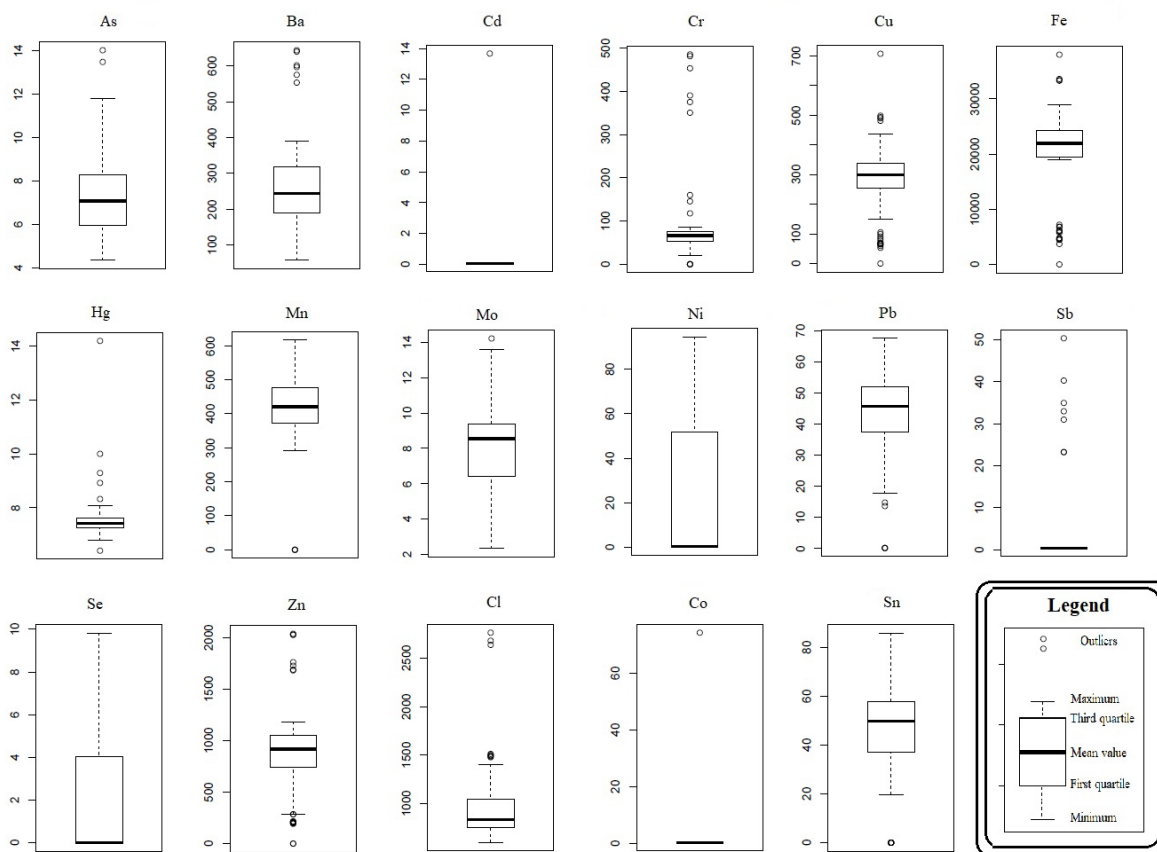
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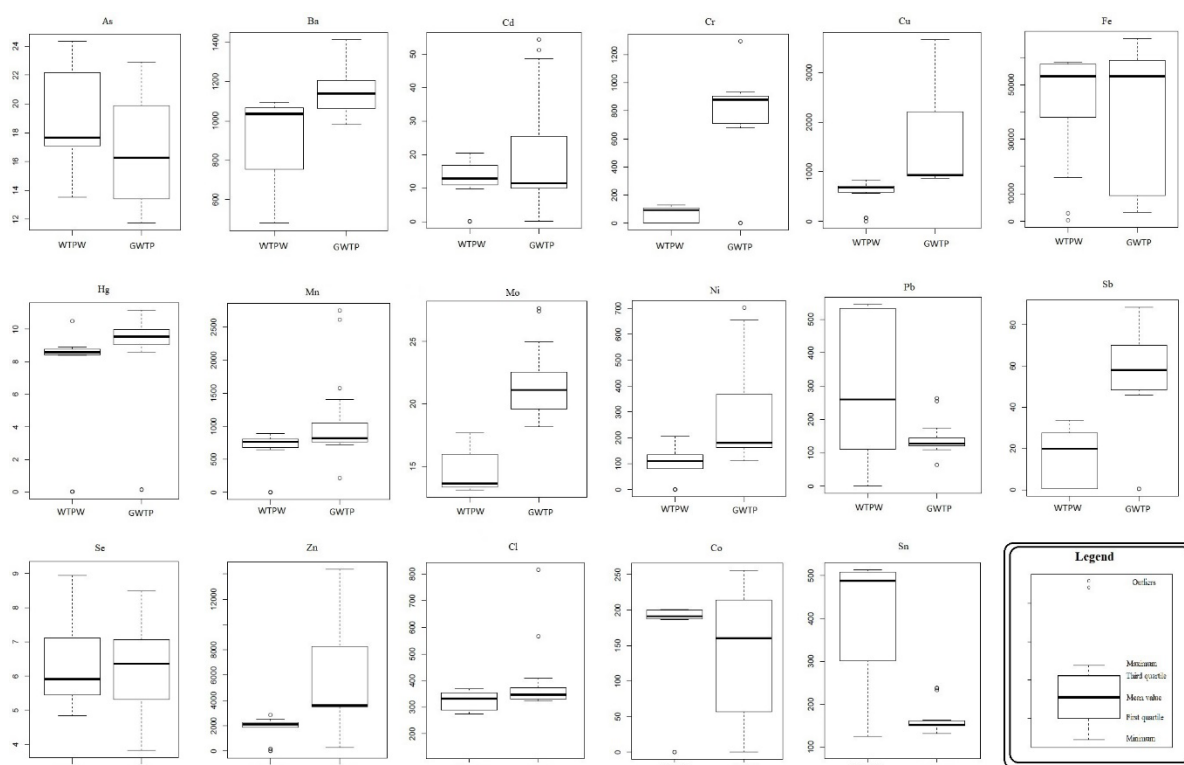
Supplementary materials 4. Boxplots presenting mean concentrations, quartiles and outliers for all analyzed elements in ash fractions from WTPW and GWTP. Values in [mg/kg].
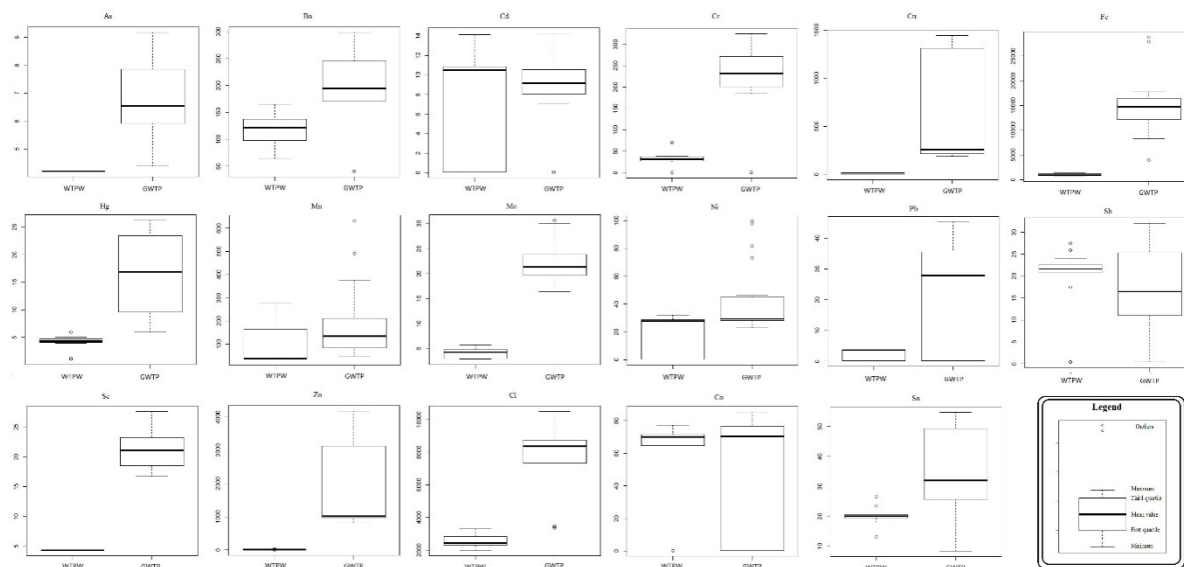
Supplementary materials 5. Boxplots presenting mean concentrations, quartiles and outlier for all analyzed elements in dust fractions from WTPW and GWTP. Values in [mg/kg].

Supplementary materials 6. Boxplots presenting mean concentrations, quartiles and outliers for all analyzed elements in stabilized materials. Values in [mg/kg].

Element	Cd	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Sb	Zn	Cl	Co	Sn
IDL [mg/kg]	2.6	4.0	10	8.4	0.0004	3.3	8.4	7.6	16	4.4	100	8.8	0.04

Supplementary materials 7. Instrumental detection limits (IDL) for presented elements [mg/kg]