

Properties of biofuel fly ash and capabilities of its use for agricultural needs

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Abstract. The use of various types of biomass for energy production provides great prospects for reducing the consumption of fossil fuels and the negative impact on the environment. However, the use of biomass, in particular agromass for this purpose, results in relatively large amounts of bottom ashes and fly ashes, the composition and properties of which also raise a number of additional environmental problems. The composition and properties of fly ash are investigated in the paper, taking into account the possibilities of utilizing them for soil fertilization and other applications. Fly ash samples were collected from bunkers of flue gas cleaning equipment (electrostatic precipitator and cyclones) installed after water heating boilers, which are firing wood chips and chuffed straw. The composition of fly ash was determined using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) and Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM/EDS) while particle size distribution was obtained using scattered-light aerosol spectrometer. Electrical Low Pressure Impactor (ELPI) was used to separate fly ash into 14 groups by particle diameter, and the analysis of their composition showed differences in the composition of the fly ash collected in cyclones and Electrostatic Precipitators (ESP). An analysis of the composition of samples in regard to the existing heavy metals norms and considering concentrations of elements beneficial to the growth of plants, enables to prepare recommendations for fertilization. The determined alkalinity of fly ash pH 13 confirms the possibility of their use for reducing soil acidity. The analysis of fly ash composition has shown that they contain elements, important for plant growth (Ca, Mg, K, P, N, S), and their concentrations determine the further use for soil quality improvement because the amount of these elements in the acid soils is reduced.

Key words: agromass, biofuel, fly ash, chemical and physical properties.

INTRODUCTION

Biomass is considered as an important renewable energy source the use of which for energy production is constantly increasing. Solid biofuels from wood or waste of agricultures (agromass) make it possible to reduce fossil fuel consumption and greenhouse gas emissions. The quality of solid biofuel, as a raw material, its composition, and the bottom and fly ash that is produced during its combustion

determines the choice of solid biofuel preparation method and the type of incinerator (Olsson et al., 2003). The main cause of concern is the relatively high amount of ash generated by the combustion of solid biofuels produced from agromass, because the chemical compounds contained in the ash causes corrosion of the boiler surfaces, and a large amount of bottom ash increases the equipment surface fouling and the boiler efficiency losses (Zbogar et al., 2009; Frandsen, 2010).

Meanwhile, fly ash particles, which are released into the atmosphere with flue gas, cause environmental pollution and are harmful to human health (Kwauk, 2003; Pui et al., 2014; Xu et al., 2014). Therefore, the constantly tightening emission standards forces to use various methods and equipment, such as cyclones, material filters, electrostatic precipitators and scrubbers, etc., for fly ash deposition. The bottom and fly ash utilization also is a growing concern. The general requirements for the management of non-hazardous waste in Lithuania establish the conditions for the management of the ash of solid biofuel boilers, under which ash can be used in forests and agriculture, for the rehabilitation of damaged areas (Republic of Lithuania... 2011). In general, further use of ash can be carried out in accordance with European Union Directives (European Council Decision 2002; Directive 2006/12/EC) and existing national requirements, which are based on them. However, the use of bottom ash and fly ash collected in flue gas cleaning equipment for forest and agricultural land improvement is only possible if it does not pose a risk to the soil and crop quality and human health (Oberberger & Supancic, 2009). Otherwise, when the maximum permissible concentrations of heavy metals in ashes are exceeded, they must be treated by various methods (e.g. Steenari et al., 1999; Pedersen et al., 2003a; Pedersen et al., 2003b; Mellbo et al., 2008) to reduce their concentrations. When ash is not suitable for fertilization because it does not contain useful elements for plant growth, it can be used in other areas, such as concrete production, road construction, bicycle paths, sidewalks, parking lots, sports grounds, etc. Finally, when the concentration of heavy metals exceeds the limit and it cannot be reduced, collected bottom and fly ash is stored in landfills as hazardous waste.

When ashes are used for fertilization it is important to consider soil acidity, as a key quality indicator, and the elements contained in it. Depending on the elemental composition of ashes which are used for soil fertilization and the acidity of the soil it is possible to increase the yield using the recommended ash rates.

The main purpose of this work is to investigate the elemental composition of the fly ash produced during the burning of wood chips and straw as two major solid biofuel types used for energy production, and to discuss their further use in fertilizing forest and agricultural soils.

MATERIALS AND METHODS

Materials of experiment

Four wood chip and two straw fired plants have been visited to collect the required material for the experiment. Fly ash samples were then taken from flue gas cleaning equipment (ESP and cyclones) bunkers (Table 1). In (A-D) wood chips burning plants, cyclones were installed before the ESP. In (E-F) plants firing straw, only cyclones were installed to clean the flue gas. Dolomite powder was used as a supplement for the straw combustion to increase the melting temperature of the ash in plant F.

Table 1. The sources of investigated wood and straw fly ashes

Biofuel/ fly ash tag	Thermal capacity, MW	Type of separator	Typical temperature of combustion chamber	Approximate composition of burnt solid biofuel	
				Wood chips from forest residue	
A	10	ESP	950 °C	> 90% wood	< 10% bark
B	10	ESP	900 °C	80% softwood and < 20% hardwood	< 10% bark
C	10	ESP	830 °C	80% softwood and < 20% hardwood wood	< 5% bark
D	10	ESP	900 °C	90% softwood and < 10% hardwood wood	< 10% bark
E	1.4	Cyclone	680 °C	Chuffed straw	
F	1.4	Cyclone	1,200 °C		

Determination of ash physico-chemical parameters

The size of the fly ash particles was measured using a scattered light particle spectrometer (Promo 3000). In addition, the size of woody biomass fly ash particles was determined using electrical low pressure impactor (ELPI), which grouped them into 14 fractions in the range of 0.006–10.0 µm during the measurement on-line.

The moisture content of the fly ash particles was determined by weight differences before and after drying (samples were dried for 1 hour at 105 °C) and the density of the test particles was determined according to EN ISO 8130-3 2011, and the bulk density according to EN ISO 60:1977. The morphology of all particles was analyzed using a S-3400N scanning electron microscope (SEM), operated at 5–15 kV accelerating voltage and the angle of repose was determined according to ISO 4324:1977. Since the microscope was equipped with the Bruker Quad 5040 X-ray spectrometer (EDS), additionally information about composition of the analyzed particles was obtained. During the measurements a fully automatic pump and valve control system was used, which provided high vacuum mode, i.e. $\leq 1.5 \times 10^{-3}$ Pa pressure in the sample chamber. The melting temperature of fly ash was determined according to the standard LST CEN/TS 15370-1 :2006 and the pH by hydrogen ion concentration in solution using an ionometer, according to standard LST EN 12176: 2000. The element analyzer (Flash 2000) was used to determine C, H, N, S composition of the solid biofuel and the elemental composition of the fly ash formed during combustion was determined by an inductively coupled plasma optical emission spectrometer (ICP-OES).

RESULTS AND DISCUSSION

The concentrations of basic elements in wood chips and straw (as solid biofuel) and their physical properties (ash content, humidity and lower calorific value) are presented in Table 2. After comparison of this data with analogous literature data (Vesterinen, 2003; Alakangas et al., 2016), a correlation has been established that allows estimating the composition of the selected solid biofuel.

After comparing the (A) and (B) elemental composition of stem without bark, bark, and wood chips from (Vesterinen, 2003; Alakangas et al., 2016) differences in the concentrations of the basic elements were determined which are within the margin of

error and confirm the presence of a stem with bark and branch. The basic elements (C, H, N, S, O and Cl) concentrations of (C) coincided with the composition of the barked stem within the error limits, whereas (D) contained some branches judging from slight differences in O, S, and Cl. Lastly, (E) concentrations of C, H, N, S, O and Cl are close to barley straw, and (F), respectively, close to wheat straw.

The average moisture content of wood chips and straw samples was determined from as received biofuel and correlates with (Alakangas et al., 2016) data. The visible difference between the average moisture content of wood chips and straw in Table 2 is due to the difference in the initial moisture content of the biomass, the difference of which remains when drying biofuel for burning. The average ash content of wood chips and straw samples is related to its composition, inorganic and other mineral substances, which predominantly forms bottom and fly ash. This result correlates with (Alakangas et al., 2016) data and confirms the need for increased researches of ash usages.

Table 2. Results of solid biofuels analysis and their physical properties

	A	B	C	D	E	F
Composition of basic elements (% of dry basis)						
C	51.8	51.3	51	49.8	49.03	45.6
H	6.1	6.1	6	6.3	5.68	5.8
N	0.3	0.4	0.08	0.13	1.28	0.48
S	0.01	0.02	0	0.02	0.089	0.082
O	41.19	40.85	42.82	43.2	40.25	42.4
Cl	0.0042	0.0076	< 0.0050	0.005	0.069	0.19
Moisture, %	26	24	22	23	1.73	1.75
Ash content, %	1.3	1.9	1.3	2.2	3.67	3.70
NCV ₀ , MJ kg ⁻¹	18.9	19.1	19.2	19	17.9	17.9
Ash melting temperature						
SST, °C	1,175	1,230	1,180	1,135	896	898
DT, °C	1,225	1,240	1,210	1,165	1,042	1,040
HT, °C	1,245	1,245	1,230	1,185	1,058	1,060
FT, °C	1,260	1,290	1,245	1,205	1,081	1,080

Note: NCV₀ – lower calorific value, MJ kg⁻¹; SST – shrinkage starting temperature; DT – deformation temperature; HT – hemisphere temperature; FT – flow temperature.

Determining the concentrations of heavy metal in collected ash samples (A-F) is necessary for the use of ash in soil fertilization or other areas. Concentrations of heavy metals in wood (A-D) and straw (E-F) ash are shown in Fig. 1. The Ba and Sr, as alkaline earth metal, concentrations is an indicator of poor fuel preparation quality, because burned solid biofuel was slightly polluted with soil. Zn concentration in the (A-F) samples was: (A) – 0.34%, (B) – 1.22%, (C) – 0.77%, (D) – 2.45%, (E) – 0.02% and (F) – 0.02%. Similarly, determination of elements useful for plant vegetation was carried out in order to assess their usefulness as fertilizers in forest and agriculture (Fig. 2).

Permissible limits of heavy metal concentrations according to (Republic of Lithuania ... 2011) are exceeded for fly ash samples (A-D) and they cannot be used in forests and agriculture. On the other hand, fly ash (E-F) can be used in forest and agriculture/ rehabilitation of damaged areas, as the amount of heavy metals does not exceed specified limits. The elemental composition of fly ash (A-D) comparison with composition of fly ash of similar solid biofuel in (Vesterinen, 2003; Alakangas et al.,

2016) shows differences of heavy metals in wood parts and its fuels. Therefore heavy metal concentrations in fly ash can be reduced by proper selection of wood as solid biofuel.

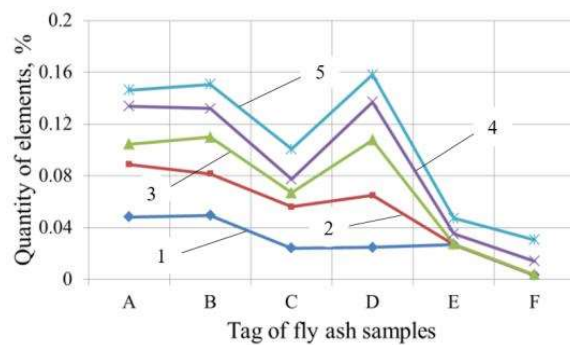


Figure 1. Quantity of heavy metals in fly ash of wood chips – (A-D) and straw – (E-F): 1 – Ba; 2 – $\Sigma(\text{Ba} + \text{Sr})$; 3 – $\Sigma(\text{Ba} + \text{Sr} + \text{Pb})$; 4 – $\Sigma(\text{Ba} + \text{Sr} + \text{Pb} + \text{B})$; 5 – $\Sigma(\text{Ba} + \text{Sr} + \text{Pb} + \text{B} + \text{Cu} + \text{Cr} + \text{Cd} + \text{Co} + \text{Ni} + \text{Mo} + \text{As} + \text{V} + \text{Hg})$.

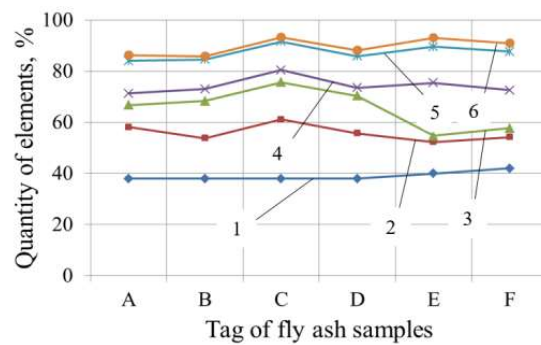


Figure 2. Quantity of elements useful for plant growth in fly ash of wood chips – (A-D) and straw – (E-F): 1 – O; 2 – $\Sigma(\text{O} + \text{Ca})$; 3 – $\Sigma(\text{O} + \text{Ca} + \text{Si})$; 4 – $\Sigma(\text{O} + \text{Ca} + \text{Si} + \text{K})$; 5 – $\Sigma(\text{O} + \text{Ca} + \text{Si} + \text{K} + \text{C} + \text{S} + \text{Cl} + \text{P} + \text{Mg})$; 6 – $\Sigma(\text{O} + \text{Ca} + \text{Si} + \text{K} + \text{C} + \text{S} + \text{Cl} + \text{P} + \text{Mg} + \text{B} + \text{Fe} + \text{Mn} + \text{Na} + \text{N})$.

The basic elements required for plant vegetation (Ca, K, Mg and P) and trace elements required for plant nutrient metabolism (B, Cu, Mn, Mo, Zn) also were determined. Concentrations of the elements B, Cu, Ni and Zn from Fig. 1 are assigned to the group of harmful elements because the concentration of these elements required for plants is very low. Other essential trace elements (N, Na, S etc.) also have been identified. Lastly, Si is considered to be a ballast element in this case, while concentration of C is useful to note, as it generally is an indicator of the quality of combustion and fly ash emissions.

On account of fly ash samples being taken from cyclone and ESP bunkers, it was important to analyse the elemental composition of particles precipitated in cyclone and ESP, since cyclones precipitate largely only coarse particles, whereas only small part of the fine particles pass ESP. This analysis could be performed only for fly ash of burnt wood chips since the cyclone was used as the only flue gas cleaning device for the precipitation of straw fly ash, and comparison by elemental composition of wood fly ash is only approximate due to difference on particle size and elemental composition.

The research of the elemental composition and physical properties of fly ash from a wood burning boiler was carried out by using ELPI and EDS for samples of fly ash collected in the cyclone and ESP bunkers (Fig. 3). Elemental composition studies of 14 ash fractions from ELPI have shown that the main constituents of the particles are O, Ca, K, S, Zn, Mg and P, therefore main compounds are Ca and K oxides and sulphates comprising 83–91% of the samples. In addition, 2.4–4.0% of Zn in fly ash from ESP, and 2.4–2.7% of Mg in fly ash from cyclone and 3.4–6.8% of C, i.e. soot, in all fly ash samples were identified. When using EDS on fly ash particles of $> 1 \mu\text{m}$ diameter, concentration of main elements (Ca and K) and additional elements (Mg, Mn, P, S, and Na) was found (Fig. 3). This result can be explained by the natural small and large particles agglomeration in the turbulent flow. Therefore in this case, the sample of the

fine particles of ESP had to have a higher concentration than that measured by ELPI and much more than in the cyclone sample.

A normalized distribution of the mass concentration of particulate matter from the ESP and the cyclone by particle size is provided in Fig. 4, with an obvious difference between ESP and cyclone precipitated particles. When comparing the distribution of the main elements in the fractions of different size particles, the largest elemental composition changes (Fig. 5) can be seen between 0.25–0.38 μm and 0.6–0.94 μm , because it is transition from fine particle mode of its characteristic composition to coarse particle mode, where particle composition depending on the particle size changes more uniformly.

This composition dependence on the particle size is shown in Fig. 5, where element concentrations normalized to the average of the four particle size fractions. There is a consistent increase in Ca, O and S with increasing particle diameter and significant decreases in C and Zn concentrations throughout the range (Fig. 5). Concentration of K in the 0.25–0.38 μm range was lower than in the 0.6–0.94 μm range but from there on continued to decrease steadily. In contrast, Fe concentration slope was reversed: it was lower between 0.6 and 0.94 μm and then continued to increase steadily. Concentration of Mg in the entire range of 0.6–5.3 μm varied slightly and was higher than the concentration in 0.25–0.38 μm particles. The results from Fig. 4 and Fig. 5 correlates to (Lanzerstorfer, 2015) because concentration of Ca and Mg are increasing while K concentration decreasing in the particle size range from 1 μm to 40 μm correlates with the same element concentration variations which have been determined from 1 μm to 4.5 μm .

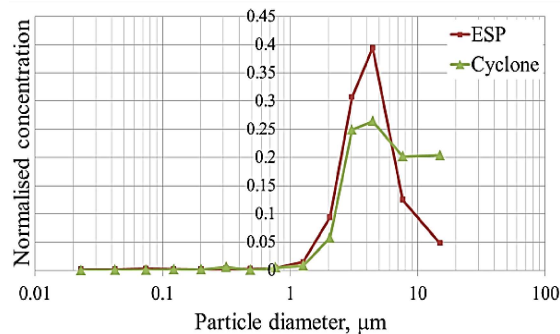


Figure 4. Normalized particle mass size distribution.

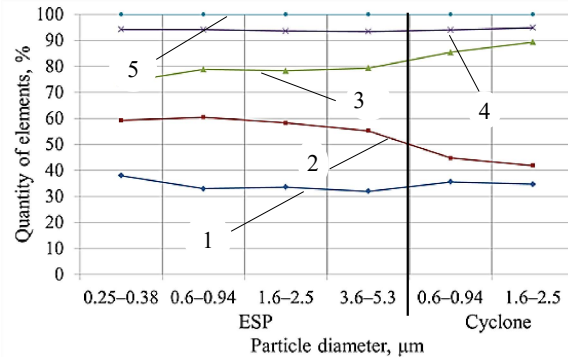


Figure 3. Elemental composition of fly ash samples collected from ESP and cyclone bunker: 1 – O; 2 – $\Sigma(\text{O} + \text{K})$; 3 – $\Sigma(\text{O} + \text{K} + \text{Ca})$; 4 – $\Sigma(\text{O} + \text{K} + \text{Ca} + \text{S} + \text{C} + \text{Zn})$; 5 – $\Sigma(\text{O} + \text{K} + \text{Ca} + \text{S} + \text{C} + \text{Zn} + \text{Mg} + \text{P} + \text{Fe} + \text{Cl} + \text{Mn} + \text{Al} + \text{Na} + \text{Si})$.

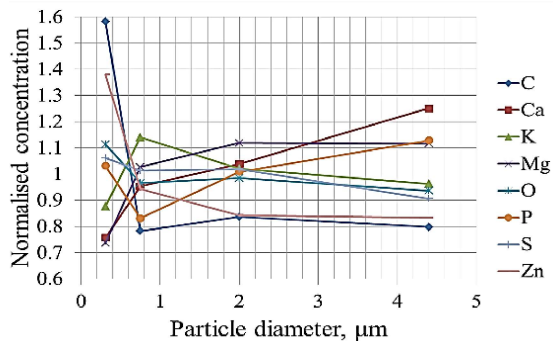


Figure 5. Normalized particle elemental concentration distribution by individual element average.

SEM clearly shows the differences in the particle sizes of the analyzed samples between the different ELPI fraction samples (Fig. 6 (a) and (b)). For many particles that can be clearly distinguished, the measured diameter value corresponds to the particle size range of the individual ELPI fractions (Fig. 6(a) and (c)). From SEM pictures (Fig. 6 (a) and (b)) and Fig. 5 it is obvious that the use of such ashes in the soil will include elements useful for plants (B, Cu, Mn, Mo, Zn) and all heavy metals (As, Cd, Cr, Hg, Ni, Pb) contained in fine particles. Therefore appropriate preparation and spread of either unstabilized or stabilized ash depending on the pH of the soil, the amount of elements in the soil useful for plant vegetation and the ongoing processes is important. This is because unstabilized ash elements get into the soil faster, and stabilized ash elements are washed more slowly, but there is no research on particle size influence on soil and plant element absorption.

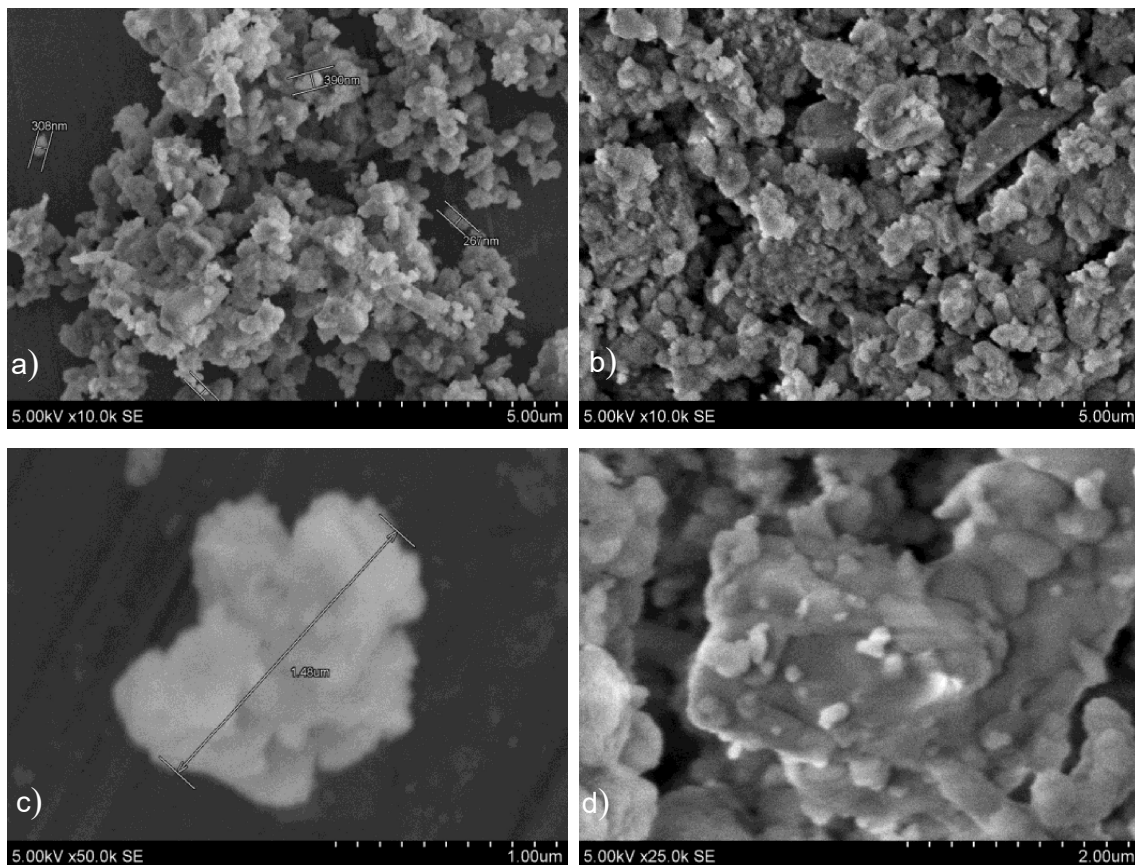


Figure 6. SEM images of particle size distributions from source A: a) 0.25–0.38 μm , 10,000x; b) 3.6–5.3 μm , 10,000x; c), 1.6–2.5, μm , 50,000x; d), 1.6–2.5, μm , 25,000x.

The physical properties of dry fly ash samples (A-F) (Table 3), as well as the elemental composition previously established, are important for determining additional requirements for combustion equipment, storage tanks, soil fertilization and other areas. Therefore, in this case, it is important to know the sample pH, particle size distribution, density of particles and bulk density of the samples.

Table 3. Physical properties of fly ash

Parameter	Fly ash					
	A	B	C	D	E	F
pH	12.9	12.5	13.1	12.8	13.3	13.3
Mass median diameter d_{50} (μm)	3.1	3.2	5.2	4.8	4.0	1.6
Spread of the size distribution	3.5	4.0	3.5	3.8	4.0	3.6
Density (kg m^{-3})	1,934	1,988	1,937	1,938	1,959	1,960
Bulk density (kg m^{-3})	160	100	140	130	320	290
Voidage	0.74	0.96	0.95	0.95	0.86	0.88
Angle of repose (degree)	50	49	49	50	50	43

The average pH of the fly ash was the same for the (A-D) and (E-F) samples. Meanwhile, the average mass distribution of (A-D) fly ash particles was $4.1 \pm 0.8 \mu\text{m}$ and in the case of (E-F) it was $2.8 \pm 0.8 \mu\text{m}$. The mean particle size distribution of (A-D) fly ash was 3.7 ± 25 and 3.8 ± 25 for (E-F). (D-E) fly ash particles has a similar mass distribution, but the distribution of particle size (F) is twice as large as (D). The wide particle size distribution correlates with the cyclone and ESP installation of the flue gas cleaning system and its operation, so there are obvious differences between (A-D) and (E-F) samples particles that correlate with the corresponding density data (van Loo & Koppejan, 2008).

The average density of (A-F) fly ash particles was $1,950 \text{ kg m}^{-3}$. The average density of (A-D) fly ash was 133 kg m^{-3} , and 305 kg m^{-3} from (E-F), which may be due to different sampling points for fly ash particles. The average voidage (A-D) and (E-F) is the same, the distribution is very similar and only in the case of (A) the porosity is lower. Finally, in the case of (A-D), the angle of the repos is very narrow ($49\text{--}50^\circ$), and in the case of (E-F) it is broader ($43\text{--}50^\circ$) and determined by the coincidence of (A-D) and (E) angle of repose. When the elemental composition and physical properties of ash is determined, it becomes possible to evaluate the use of ash in forest and agriculture/rehabilitation of damaged areas, the reduction of soil acidity and as a fertilizer additive, e.g. (Muse & Mitchell, 1995). In some cases, ash can be used to improve the composition of sewage sludge suitable for fertilization, composting of organic waste or even paper.

In general, ash can be either untreated or treated reducing heavy metal concentrations to meet the national standards using various leaching methods. In the case at hand, collected volatile ash samples (A-D) should be treated with a variety of leaching methods, as concentrations of heavy metals (B, Cd, Cr, Cu, Pb and Zn) are exceeded, and if it's not possible then they would have to be landfilled. Furthermore, even if concentrations of some heavy metals can be reduced by the use of various leaching methods, the elements that are useful for fertilization can be washed off as well making the ash unsuitable as fertilizers.

Soil improvement with unstabilized (dusty) ash activates soil processes, intensifies the mineralization of organic nitrogen and makes an increase in migration of other ions into deeper soil layers and groundwater possible. However, if the ash is used in forestry and spread up to 5 tonnes of ash per hectare, then some heavy metals do not exceed the maximum permissible levels according to regulatory documents. Meanwhile, the impact of stabilized (granulated) ash, according to forest ecosystem studies, has an opposite effect on the soil which can last from 30 to 60 years (Silverberg & Hotanen, 1989;

Korpilahti et al., 1998). Therefore, depending on the leaching of materials from ash granules even if ash doses of 11–44 t ha⁻¹ are used for fertilization, the groundwater is not contaminated with either heavy metals or with Ca²⁺, K⁺, SO₄²⁻ and other ions (Williams et al., 1996). In the event that even treated ash is not suitable for fertilization, it can be utilized in alternative ways which, unfortunately, are not currently regulated in Lithuania. As a result, landfill disposal is the only treatment even though it is economically disadvantageous.

Alternative use of ash is applied in a number of European Union (EU) countries (e.g. Sweden, Germany, Denmark, France) but it is the most widely developed in Sweden, as ash is used for road construction, landfill reclamation, filling of old shafts or in the manufacture of concrete products including the installation of parking bases. Each alternative use of ash depends on proper dosing, physical properties and elemental composition, as the performance of the product depends on these characteristics. Considering the possible areas of use of bottom and volatile ash, investigated methods of altering their elemental composition it can be expected that developed ash processing and utilization will reduce their amount in landfills and local environmental pollution.

CONCLUSIONS

Samples (A-D) of the wood chips supplied to the boiler plants in operation show that chips are prepared from a fairly diverse range of wood as solid biofuel including tree waste, without paying due attention to their quality. Therefore, the incineration of such chips results in increased amounts of heavy metals in fly ash, which are higher than the permissible limits for soil fertilization set by the Lithuanian minister of environment order No. D1-572, June 25 2014. Such fly ash cannot be used for soil fertilization. In contrast, when the straw is burned, heavy metal concentrations in fly ash are under permissible limits and ash can be used for soil fertilization.

The average pH and porosity of volatile ash are similar between samples and equal to 13 and 0.9 respectively, while (E-F) particles are on average 2.3 times larger than (A-D) volatile ash particles. Average particle density of (A-F) was 1,950 kg m⁻³, whereas the mean bulk density (E-F) for particles was 2.3 times the average bulk density of (A-D) particles.

Major elements in the fly ash are Ca, K, Zn, S, C and Mg the quantities of which depend on the particle size. The large particles that are retained by the cyclone have higher Ca, Mg and C concentrations. On the other hand, fine particles, retained by the ESP, contain higher concentration of K, Zn, S and other elements. However, in the flow after the boiler particles are affected by turbulence and agglomerate, therefore it is difficult to precisely link the composition of the particles to their size.

The ESP also does not precipitate some of the finest particles, which remain a source of environmental pollution and a threat to human health. Therefore, the research on fine particle agglomeration methods is promising.

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