REMEDIATION CONCEPT

Deliverable D.T3.1.2

Version 1

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Prepared by:

Marta Pogrzeba

Jacek Krzyżak

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1. SCOPE OF WORK FOR THIS REPORT

The scope of work includes a remediation concept for Ruda Śląska brownfield, developed basing on the results of the assessment of the environmental status of the brownfield site. The presented concept is considering phytoremediation of the site using aided phytostabilization approach. It includes principles of the aided phytostabilization technique and preliminary tests using pot experiment to select appropriate dose of soil amendments and used grass species to create green cover on the “soil” surface.

2. SUMMARY OF SITE DESCRIPTION (DELIVERABLE D.T.3.1.1)

Conducted site description, based on historical and currently collected data showed serious problem of heavy metal contamination, especially for cadmium and zinc. The environmental impact of dumping site Ruda Śląska is complicated for various reasons. Its influence on surrounding areas results from activities over the last hundred years. Previous studies described that in the 2nd half of XX century the brownfield was revitalized, by covering the wastes with soil layer (of unknown origination). Current studies confirmed that soil and ground of the heap is heavily contaminated with cadmium, lead and zinc, especially in northern and northern-west part. Surface of the heap is covered by grassy vegetation; in the northern-west part some trees are presented. Metallophytes species found at the heap also confirmed heavy metal contamination. Future remediation activities should be focused on metal immobilization, especially cadmium and zinc and replacing current vegetation by grass species which cumulates contaminants in root zone, with limited uptake to above ground parts.

3. PRINCIPLES OF THE REMEDIATION CONCEPT FOR RUDA ŚLĄSKA BROWNFIELD

3.1. Aided phytostablization

Aided phytostabilization is a quite new technology, worked out in the last fifteen years, basing on the use of soil amendments immobilizing heavy metals in the soil with selected plant species. Immobilizing of the contaminants bases on the followed processes: absorption and accumulation in the roots, adsorption on the roots surface or transforming in the non-soluble forms in the soil rhizosphere (Berti et al., 1998; Vangronsveld and Cunningham, 1998; Ruttens et al., 2006). Aided phytostabilization are using biological and chemical processes in the roots zone of the plants. Because of roots secretions and releasing of CO_2, roots zone are the place for pollution precipitation. During the interaction between plants and soil environment, bioavailable forms of heavy metals are converted into less available (Salt et al., 1995; Berti et al., 1998; Sas-Nowosielska et al., 2008). The final goal of the phytostabilization is to create dense plant cover on the soil surface, which protects soil erosion, contaminants migration to deeper layers or runoff with the rain precipitation (Li and Chaney, 1998; Vangronsveld, 1998; Berti and Cunningham, 2000; Knox et al., 2001). Schematic diagram of aided phytostabilization process are presented in Figure 1.

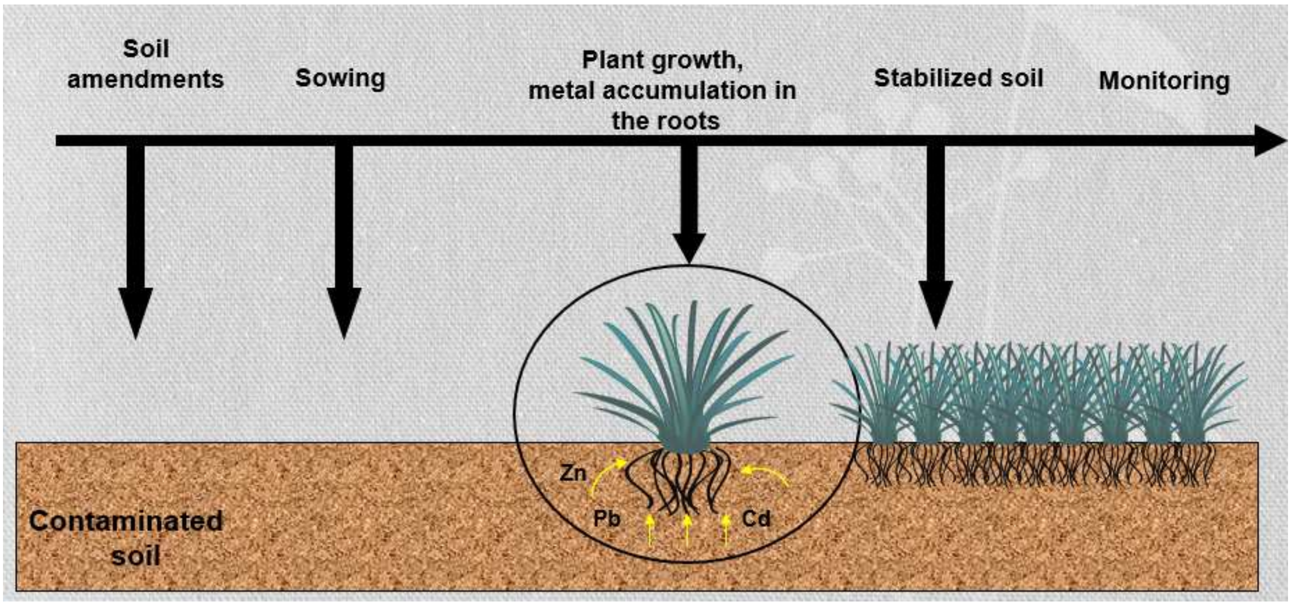

Figure 1. Aided phytostabilization process

3.1.1. Soil amendments for diminishing metals bioavailability

Improvement of soil quality due to application different amendments such as organic matter, lime or phosphorous compounds are well known practice. The most common soil amendments which diminish metals bioavailability in the soil are: phosphorous compounds, natural and synthetic aluminosilicates, ashes, iron oxides, calcium compounds and different types of organic matter (Cunningham et al., 1995; Flathman and Lanza, 1998; Schnoor 2000; Kumpiene, 2010).

Iron oxides diminish arsenic, cadmium, copper, nickel, lead and zinc mobility, due to sorption, precipitation or creating minerals containing mentioned above elements (Berti and Cunnningham, 2000). The surface of iron oxides molecules might be modified, depending on the soil pH, makes them amphotheric and able to sorption both anions and cations (Cornell and Schwertmann, 2003). Nevertheless, iron oxides are compounds diminishing metals bioavailability in the soil and have low negative impact on the soil function, its need to be applied jointly with organic matter (e.g. compost) to improve plant growth on the stabilized soil surface (Ruttens et al., 2006).

Zeolites are natural hydrated aluminosilicate minerals, created during reaction of volcanic dusts with surface or ground water. It could be also created in non-volcanic environment, during interactions between salted soil particles with strongly alkaline solutions (Gworek and Sucharda-Kozera, 1999; Kumpiene 2010). Zeolites are also known as „molecular sieves", because its ability to selective sorption of molecules (Jamil et al., 2010; Blisset and Rowson, 2012). Zeolites could be also easily synthetized from fly ashes (Belviso et al., 2010; Blisset and Rowson, 2012). This soil amendments are highly efficient in the sorption of lead and cadmium (Panuccio et al., 2009; Huang-Ping and Shu-Hao, 2012), as well as copper, zinc and arsenic (Chen et al., 2000; Friesl et al., 2006). Application of the zeolites to the soil rise the pH and metals immobilization (Querol et al., 2006; Mahabadi et al., 2007).

Different types of **fly ashes** are the most efficient for cationic contaminants, such as Cu, Pb, Zn and Ni, while the stabilization effect could be weakened for anionic contaminants, such as arsenate and chromate. Moreover, efficiency of fly ashes are strongly dependent on soil type. The mechanism of contaminant stabilization using fly ashes are basing on the pH rising, precipitation or sorption on fly ash surface and cations exchange (Kumpiene, 2010). The best effectiveness in contaminants immobilization were observed for acidic soils, with low organic matter content (Nachtegaal et al., 2005).

The use of **phosphorous compounds** for metals immobilization base on its ability to precipitation and creating stable forms. Phosphorous compounds are especially recommended for lead immobilization, during the stabilization process, stable, non-soluble compounds (hydroxy pyromorphite) are created (Cao et al., 2008). On the other hand, the use of phosphorous compounds should be limited in case of arsenic, selenium and wolfram contamination. Due to competition of phosphates and arsenates, mobility of arsenic could rise significantly (Geebelen et al., 2002).

Organic matter are essential soil component, which determines physical, chemical and biological parameters. It consists of mixture of humid and fluvic acids. High content of functional groups as -COOH and -OH allows ionic exchange, creating complex with heavy metals. Lack of organic matter are common phenomenon for heavily contaminated soils, because of toxic effect for biological activity and plants growth. Due to that fact, soil amendments consist of organic matter improve soil structure and nutrients holding (Kumpiene, 2010). One of the most popular organic soil amendment is **lignite**. It consists of humid and fluvic acids, among which humid acids are most important from remediation point of view. Humid acids are mainly composed of carbon, hydrogen and oxygen, the second components are nitrogen and sulphur (Vitkova et al., 2011). This compound is characterized by high ion exchange capacity and it was confirmed that the most exchanged ions are sodium, potassium, aluminum as well as copper, lead, zinc and cadmium. Rising the soil pH caused the heavy metals sorption capacity (Havelcova et al., 2009; Doskocil i Pekar, 2012). For example, higher values of soil pH caused higher sorption of Zn cations. Application of lignite to the soil diminishes bioavailability of cadmium and zinc and consequently uptake of this elements by plants (Ociepa et al., 2011, Maciejewska and Kwiatkowska, 2003).

Important factor during the aided phytostabilization process is controlling soil pH for the neutral or slightly alkaline conditions. It could be easily achieved by use of **calcium compounds** such as calcium carbonate, calcium oxides or calcium hydroxides present in lime fertilizers. Lime is the most popular substance in pH controlling, improvement of plants growth and diminishing of metal uptake by plants (Hamon et al., 2002; Bolan et al., 2003; Ruttens et al., 2010).

Summary of soil amendments used in diminishing of metals bioavailability is presented in the Table 1.

Table 1. Soil amendments and its influence on heavy metals mobility (Kumpiene 2010, modified)

Amendment	Influence on metals mobility			Comments
	Positive	Varied	Negative	
Iron oxides	As	Cd, Cu, Ni, Pb, Zn		Immobilisation depends on soil pH, too high manganium concentration are toxic to plants
Manganium and aluminium oxides	As, Zn, Cd, Ni, Cu, Pb		Cr	
Loam	Pb, Cd, Cu, Zn	As		Low efficiency for heavily contaminated soils, might be leached in acidic conditions
Zeolites		Cu, Zn, Mn, As		
Fly ashes	Pb, Cd, Cu, Zn	As		
Phosphorous compounds	Pb, Cu, Zn, Cd		As	
Lignite	Cu, Pb, Zn, Cd			Wide range of applications
Peat	Cu, Pb, Zn, Cd			Low physico-chemical stability
Biodegradable wastes		Cu, Pb, Zn, Ni, Cd, As		Possibility of additional soil contamination from the amendments
Calcium compounds	Cu, Zn, Cd, Pb	As, Cr		Soil pH control is needed

3.1.2. Plant species used for aided phytostabilization of soil contaminated with heavy metals

Selection of appropriate plant species is one of the most important factor which determines effective aided phytostabilization process. Used plants should be characterized by following parameters (Berti et al., 1998; Vangronsveld i Cunningham, 1998; Mench et al., 2010; Zou et al., 2012):

- tolerance to high concentration of contaminants in the soil,
- ability to create dense cover on the soil surface with strong and deep roots system,
- accumulation of contaminants in the roots, with low uptake to the aboveground parts,
- low requirements to the habitat conditions and resistance to the local climate.

The most suitable for aided phytostabilization are different grass species. The most popular are: red fescue (*Festuca rubra*), creeper (*Agrostis capillaris*), ryegrass (*Lolium perenne*), tufted-hair grass (*Deschampsia caespitosa*), cordgrass (*Spartina pectinata*) (Kucharski et al., 2005; Gucwa-Przepióra et al., 2007; Cambrolle et al., 2011).

4. PRELIMINARY LAB-SCALE TESTS TO DETERMINE APPROPRIATE DOSE OF SOIL AMENDMENTS AND PLANT SPECIES

To minimize the risk of inappropriate dosage of soil amendments, preliminary lab-scale experiment should be conducted. Too low dose of soil amendments may results in lack of metals immobilization, while to high dose will results high, unnecessary costs of investments. Based on the site characteristics and previous IETU experience with stabilizing soils heavily contaminated with metals, mixture of lignite and lime were selected for aided phytostabilization. *Lolium perenne* STADION was selected as a grass species to create dense cover with strong root system.

4.1. Experiment design

For pot lab-scale experiment soil from the Ruda Śląska brownfield was sampled. Sampling points were selected based on the site characteristics, from the points where highest heavy metals bioavailability were found (for reference see Deliverable D.T3.1.1 – Assessment of the technical and environmental status of the brownfield site). Experimental variants of the pot experiment are as follow:

- Control - no amendments, *Lolium perenne* STADION sieved,
- I – soil amended with 5% of lignite and 0.5% of lime, *Lolium perenne* STADION sieved,
- II – soil amended with 5% of lignite and 0.25% of lime, *Lolium perenne* STADION sieved.

Each variant of the pot experiment is prepared in five replication, to collect enough data for statistical analysis of the results. Soil samples from the pots will be collected, at the beginning of the experiment and two and six weeks after the start of experiment, to control effectiveness of the stabilization process. Soil samples will be analyzed for heavy metals bioavailability, pH and electric conductivity. After six weeks *Lolium perenne* STADION will be sieved on soil surface. Plant biomass will be analyzed for Pb, Cd, Zn and As content in aboveground parts.

Based on the obtained results dose of amendments for field application will be selected.

4.2. Results

Changes in HM bioavailable fraction after amendments addition

Generally addition of lime and lignite significantly decrease bioavailable fraction of all tested metals (Fig. 2, 3, 4), irrespective of the dose. It means that addition of amendments significantly decreased the metals in soil solution in comparison to the control and can decrease also the plant metal uptake.

Directly after addition of lime and lignite about 60% (for dose 0.5% lime + 0.5% lignite) and 80% (for dose 0.25% lime + 0.5% lignite) decrease of bioavailable Pb in comparison to the control was observed (Fig. 2). After two and six weeks of 0.25% lime and 0.5% lignite addition the bioavailable fraction was stable and was lower than control about 88%. Only small amount of Pb in soil solution was bioavailable and can be uptaken by grasses.

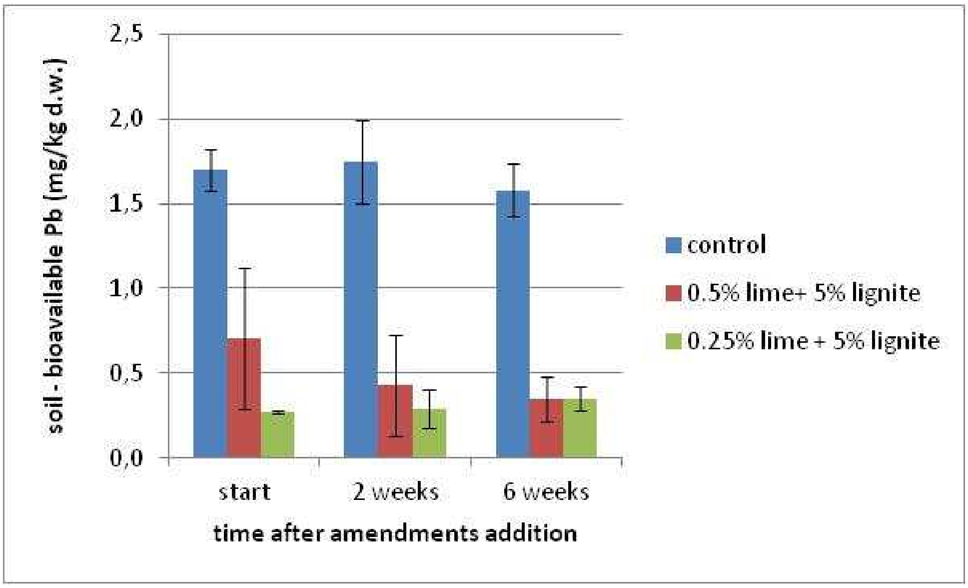

Figure 2. Changes on bioavailable Pb fraction after addition of amendments to the soil. Values are means of five replicates ± SD

The same trend was observed after amendments addition for Cd bioavailable fraction (Fig. 3). It was assessed that both tested variants of amendments (0.5% lime + 0.5% lignite; 0.25% lime + 0.5% lignite) are able to decrease bioavailable Cd in comparison to control, about 85% and 91%, respectively. After six weeks of amendments addition no difference between tested variants of amendments was found, it could be concluded, that lower dose of lime (0.25% w/w) can be used in the field demonstration and we can expect the same effectiveness in comparison to the higher dose of lime (0.5% w/w).

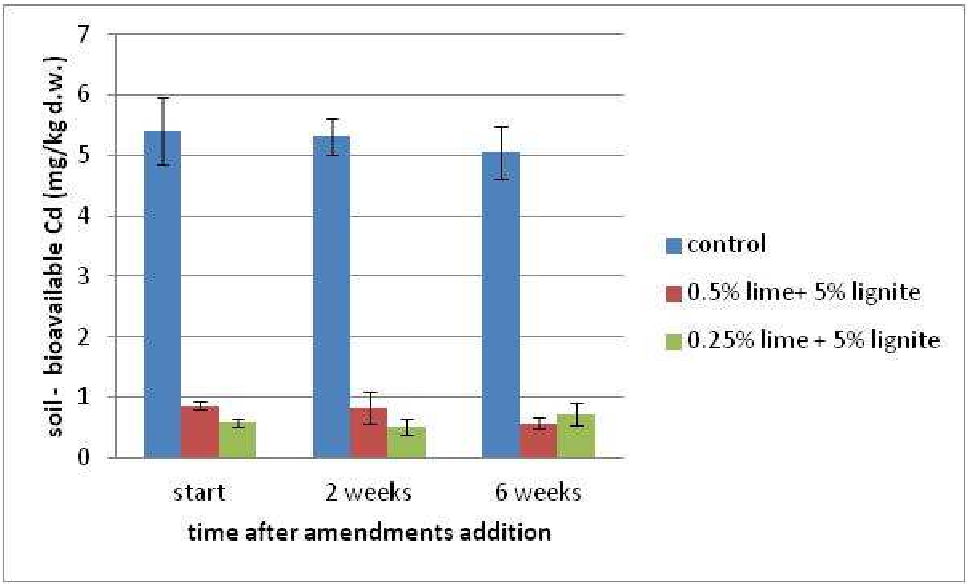

Figure 3. Changes on bioavailable Cd fraction after addition of amendments to the soil. Values are means of five replicates ± SD

The highest decreasing of bioavailable fraction after amendments addition was observed in case of Zn (Fig. 4). More than 90% of bioavailable Zn was converted into no soluble forms.

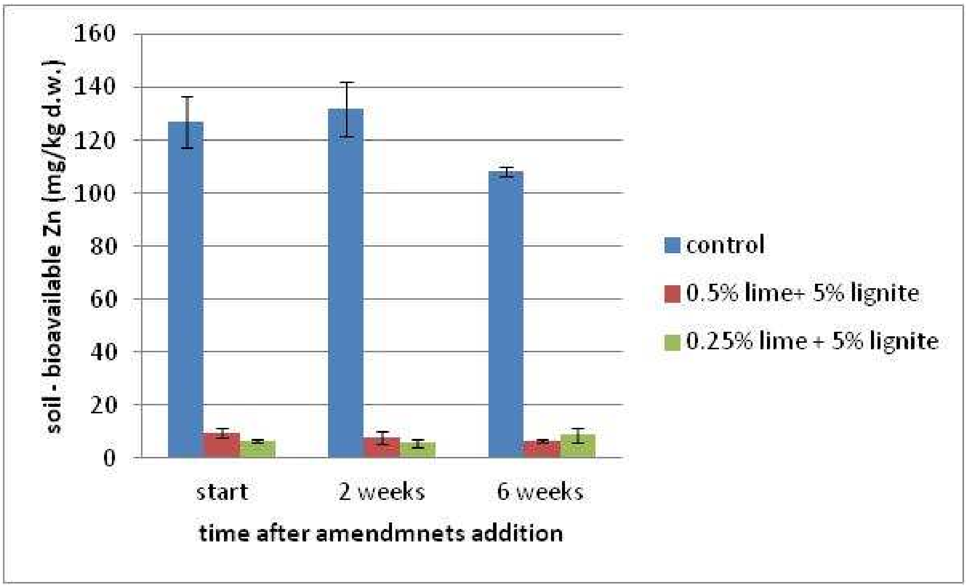

Figure 4 Changes on bioavailable Zn fraction after addition of amendments to the soil. Values are means of five replicates ± SD

Changes in physical soil parameters after amendments addition

Generally addition of lime and lignite irrespective of the dose significantly increased soil pH and did not change soil electrical conductivity (Table 2). Observed changes were from 0.2 to 0.5 unit and finally after 6 weeks of amendments addition was near neutral. Range of measured pH between 6.61 (for dose 0.5% lime + 0.5% lignite) and 6.68 for dose 0.25% lime + 0.5% lignite) can enhance also plant growth development.

Table 2. Changes in soil pH and electrical conductivity after amendments addition

Experimental variants	pH (H_2O)			pH (KCl)			EC (μS/cm^3)		
	Time after amendments addition								
	start	2 weeks	6 weeks	start	2 weeks	6 weeks	start	2 weeks	6 weeks
Control	6.56±0.03	6.42±0.03	6.53±0.01	5.94±0.07	5.97±0.01	6.02±0.01	157.2±12.46	162.4±9.75	145.6±4.80
0.5%lime +0.5% lignite	6.85±0.01	7.04±0.01	7.20±0.01	6.41±0.03	6.46±0.01	6.61±0.03	122.0±18.41	143.6±15.50	147.6±20.95
0.25%lime +0.5% lignite	6.85±0.02	7.05±0.02	7.14±0.01	6.37±0.06	6.54±0.03	6.68±0.01	157.2±11.20	121.2±11.56	159.0±3.17

Values are means of five replicates ± SE

Influence of amendments addition on HM uptake by plants and plant biomass production

Low concentration of Pb (from 25 to 28 mg/kg d.w.) in biomass of *Lolium perenne* was measured. No changes in Pb concentration in grass biomass in all experimental variants were observed after the first harvest (Fig . 5).

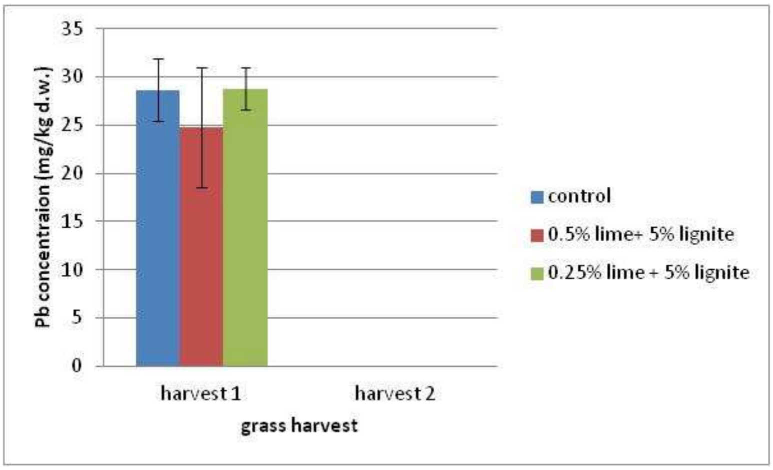

Figure 5. Concentration of Pb in grasses grown under different soil amendments. Values are means of five replicates ± SD

Addition of amendments significantly decreased the bioavailable Cd (Fig. 2) in soil solution and also significantly diminished plant Cd uptake in comparison to control in the first harvest (Fig. 6). Near to 60% diminishing of Cd concentration in grass biomass collected in the first harvest were assessed in comparison to control. No differences in Cd concentration in grass biomass grown under different amendments were observed.

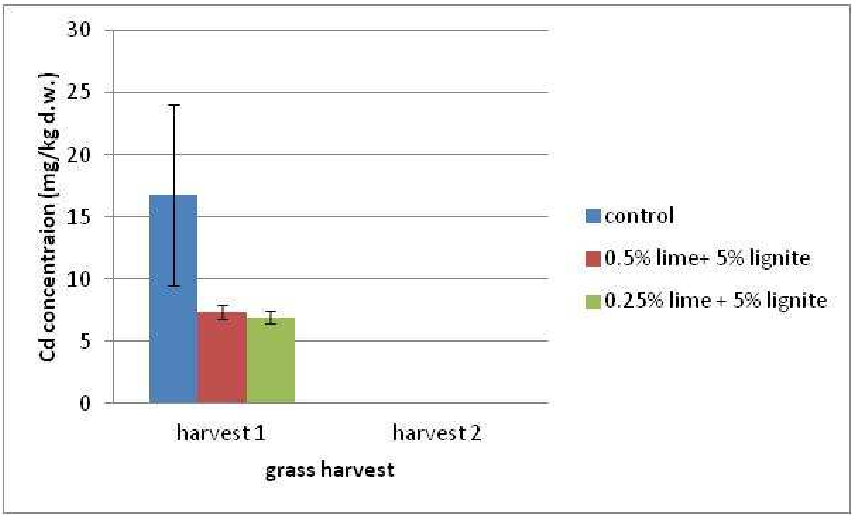

Figure 6. Concentration of Cd in grasses grown under different soil amendments. Values are means of five replicates ± SD

Addition of amendments significantly decreased the bioavailable Zn (Fig. 4) in soil solution and also significantly diminished plant Zn uptake in comparison to control in the first harvest (Fig. 7). From 20 to 40% diminishing of Zn concentration in grass biomass collected in the first harvest were assessed in comparison to control. No differences in Zn concentration in grass biomass grown under different amendments were assessed.

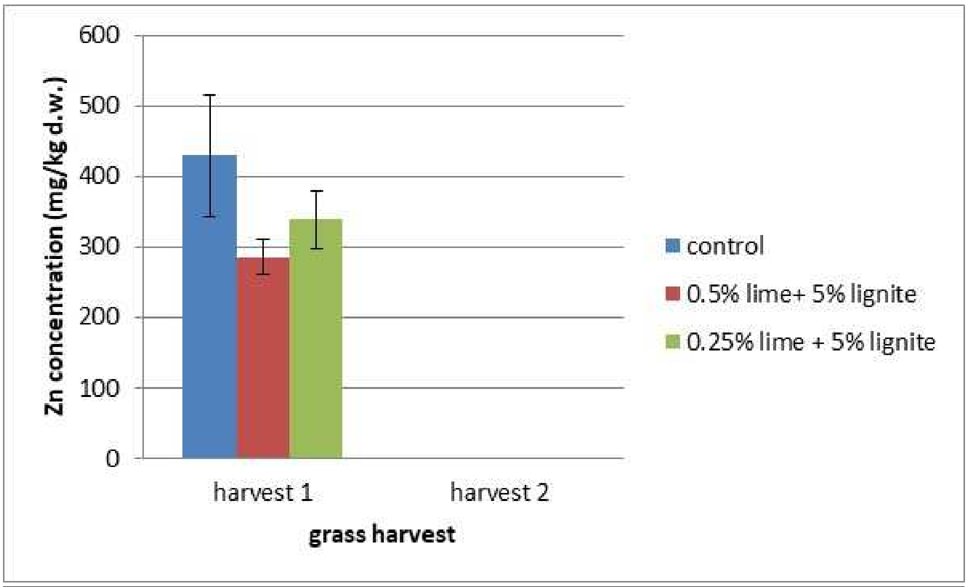

Figure 7. Concentration of Zn in grasses grown under different soil amendments. Values are means of five replicates ± SD

Significantly higher biomass production after amendments addition in comparison to control were found (Fig. 8). Tested grass species after addition to the soil 0.25% lime and 0.5% lignite were able to produce 20% higher biomass in comparison to control in the first harvest, while in the second harvest the biomass production was 120% higher.

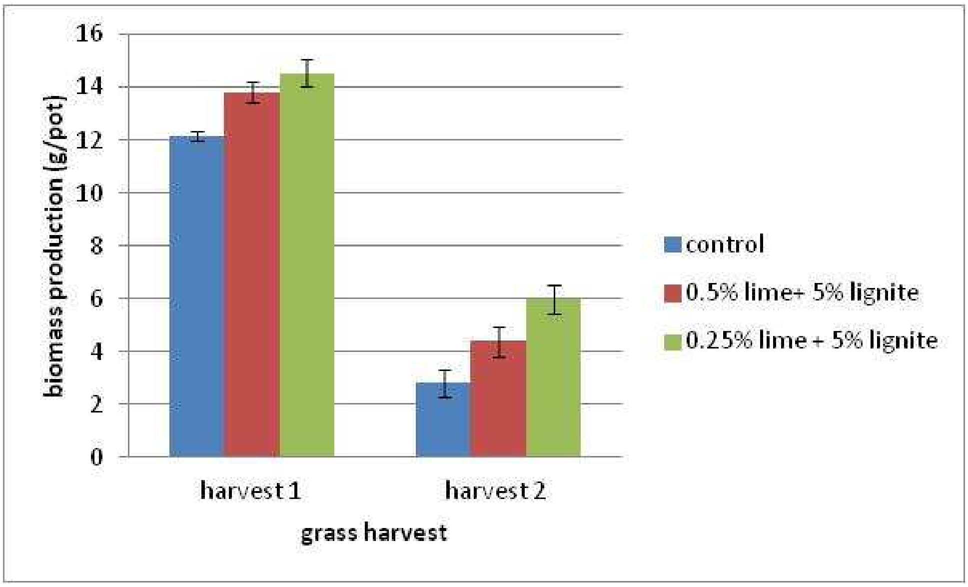

Figure 8. Biomass production under different soil amendments. Values are means of five replicates ± SD

5. CONCLUSIONS

It could be concluded that amendments addition:

1. significantly diminish soil metal bioavailability (mainly Cd and Zn),
2. significantly enhanced plat biomass production, in comparison to control.

For full scale application to diminish the cost of buying and application the dose of 0.25 % lime and 0.5% lignite are proposed. Also tested grass species – *Lolium perenne* cv. STADION can be used successfully for phytostabilisation application on Ruda Śląska brownfield.

6. LITERATURE

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