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Soil Pollution as a Result of Temporary Steel Scrap Storage at the Melt Shop

Tahir Sofilić[1], Blaženka Bertić[2], Vesna Šimunić-Mežnarić[3], Ivan Brnardić[4]*

1 - CMC Sisak Ltd., Braće Kavurić 12, 44010 Sisak, CROATIA
2 - Josip Juraj Strossmayer University of Osijek, Faculty of Agriculture, Kralja Petra Svačića 1d, 31000 Osijek, CROATIA
3 - Bioinstitut Ltd., R. Steinera 7, 40000 Čakovec, CROATIA
4 - University of Zagreb, Faculty of Metallurgy, Aleja narodnih heroja 3, 44103 Sisak, CROATIA
* Corresponding author. brnardic@simet.hr.

Abstract. In this paper pollution of soil (5300 m^2) used as temporarily steel scrap storage at the CMC Sisak Ltd. was investigated. Concentrations of heavy metals, namely Cd, Cr, Cu, Hg, Ni, Pb and Zn in soil were determined after their extraction in *aqua regia*. Concentrations of heavy metals, except Hg, were measured by inductively coupled optical emission spectrometry. Concentration of Hg was determined by atomic absorption spectrometry. For a number of years, steel scrap (raw material for steel production) was temporarily stored in the unprotected steel scrap yard area. To assess pollution level of soil under the scrap yard, comparison was done between levels of heavy metal concentrations in analysed samples and reference sample taken outside the factory ground with the levels representing tolerance for potentially unacceptable risk for industrially used soil according to the Croatian Soil Monitoring Programme. Levels were also compared with the values permitted by some EU member countries. Concentrations of heavy metals in all samples collected from the scrap yard showed higher values of heavy metals compared to the reference sample concentrations. Also, values are higher than those defined as potentially unacceptable risk for industrially used soil according to legislation of some EU member countries. Obtained results qualify analysed soil from the scrap yard as contaminated soil, caused by its use over a long time as a temporarily storage space of steel scrap on unprotected and roofless soil. In future, steel storage areas should be built in a way to prevent soil pollution.

Key words: Soil; Pollution; Steel scrap; Temporarily landfills

Introduction

Industrial manufacturing imposes significant influence on the environment as different types of industries contribute to the air, soil and water pollution with their emissions. In addition, by-products of manufacturing products often negatively affect the entire living and non-living world, especially people and their health. Croatian legislation (Official Gazette of the Republic Croatia, OG 114/08) defines industrial types, which can, with their emissions, cause soil, air, water and sea pollution as well as pollution of other parts of the environment. Those industrial types, to name a few, include energy industry, metal production and processing, mineral industry and chemical industry.

From the environmental point of view, metal production and processing is one industrial branch of particular importance, considering the applied technology

(processes of roasting and sintering of metal orca, processes of raw iron and steel production, production and processing of ferrous and non-ferrous metals). In addition, metal working processes of metal treatment, especially processes with chemical procedures, cannot be ignored.

In real life, one less common is environmental pollution caused by breakdown or major ecological disasters when huge amount of pollutants in the environment is immediately released, but more common is the appearance of long-term emission of lower quantities of pollutants from production processes. Pollutants from the above-mentioned processes, which can harmfully influence air, water and soil, are significant amounts of gas and solid pollutants (EUROPEAN COMMISSION, 2010). In addition, pollutants depend on the technology method used. Most common pollutants occurred as dust, SO_2, NO_x, NH_3, H_2SO_4, HCl, HF, HCN, H_2S, CO, CO_2, CH_4, heavy metals in waste waters and gases (Hg, Pb, Cr, Ni, Zn, Cd, Cu), benzene, phenol, polycyclic aromatic hydrocarbons, polychlorinated biphenyls, polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans, cyanide, oil, grease etc.

In addition to the air and water pollution, metallurgical processes can quite often pollute soil indirectly near the process itself or indirectly at the area used either for the storage of raw material or disposal of produced waste. Contact with wastewater can also cause soil pollution.

Type of pollution from metallurgical process depends on the nature of technological operation conducted on a given location and their duration. Intensity of pollution depends on the type of pollutant emitted from the process and its interaction with the environment in real life conditions.

For example, steel production in electric arc furnace can negatively influence the environment with emissions in air and water during the production process. Additionally, pollutant influence can be prolonged if by-product and/or waste (raw slag, used heat-resisting material, metal cuttings, different mud, dust from smoke, burnout etc.) are uncontrollably disposed on the unprotected ground. Pollutants can lead to negative physical-chemical properties of soil (pH, availability of biogenic elements, water capacity, air capacity and presence of organic matter in soil) wherewith directly and/or indirectly influence on biogenic and productivity of soil. The largest consequence of polluted soil is transfer of its negative impact on ground water.

Literature commonly (OG 114/08) discusses types of pollution and emission impact from steel production process either during duration of process itself (BARCAN, 2002; SCHULIN *et al.*, 2007; SIKALIDIS *et al.*, 2010; RIZESCU *et al.*, 2011) or of the influence of created waste and of harmful substances they contain on the environment (CHILINGIROVA *et al.*, 2011; LONCNAR *et al.*, 2009; LUXÁN *et al.*, 2000; REMON *et al.*, 2005; RODELLA & CHIOU, 2009; TOSSAVAINEN & FORSSBERG, 2000). However, a limited number of articles discuss the impact raw material for this process has on the environmental pollution (ADAMO *et al.*, 2002; ENE *et al.*, 2011; SALLAKU *et al.*, 2009).

Steel scrap in steel production process with electric arc furnace is used (often and 100%), which is commonly polluted with different inorganic and organic substances. Furthermore, steel scrap, as a basic raw material for steel production, regarding to its physical and chemical characteristics is often categorized according to the European Scrap Grading System (ESGS) in 11 different quality categories (E1-E3, E6, E8, E40, E5H, E5M, EHRB, EHRM and E46) (BAILLET, 2001; GOJIĆ, 2005). According to BAILLET (2001), content of organic and inorganic pollutants in steel scrap should be less than 1.4 % for E1 (light steel scrap prepared for furnace < 6 mm), EHRB (old or new steel scrap prepared for furnace, without Cu, Sn, Pb and their alloys) and 1 % for E3 (heavy steel prepared for furnace, including pipes, hollow profile, without Cu, Sn, Pb and their alloys).

Due to their numerous applications and appearance, unwanted heavy metals can be commonly found in steel scrap. During the melting of steel scrap in a furnace, heavy

metals and other unwanted inorganic and organic ingredients participate in a very complicated reaction of pyrolysis and pyrosinthesis resulting in production of an array of compounds. These potentially very dangerous pollutants are then emitted in the environment in the shape of smoke gases from electric arc furnace. Sometimes those pollutants present in steel scrap enter the environment, especially through soil before they come to the electric arc furnace, i.e. during the time of temporary storage of steel scrap at unprotected soil or during the preparation for cutting to demanded dimensions.

Pollutants and/or unwanted ingredients in steel scrap in the shape of different powders, clusters, sludge etc., either inorganic or organic under the influence of the atmosphere, are washed out from steel scrap or simply scattered, respectively spilled during steel scrap attrition by means of cutting.

In this paper soil under the temporary storage, area for steel scrap in Melt Shop CMC Sisak Ltd. which has been exposed to direct influence of steel scrap (during cutting or temporary storage before melting) for a long period of time was examined. Concentrations of heavy metals in the soil were examined of area so called scrap yard (size of 5300 m^2).

Materials and methods

Sampling. Testing soil was sampled from temporary steel scrap storage at scrap yard, where steel scrap is shipped by railroad and trucks (Fig. 1 and 2). Nearby the steel scrap temporary storage, there are no other facilities that can cause soil pollution. The nearest river is Sava located 1.5 km away in the northeast direction.

For the purpose of soil sampling, temporary storage area, i.e. scrap yard was divided into four almost identical areas (PI, PII, PIII and PIV), size of 1350 m^2 each (Fig. 3). From each area a few soil samples were collected from different depths and composite samples prepared.

Depending of the soil digging possibilities and opening of soil sampling profiles, soil samples were collected as follows; from area PI, 9 soil samples from 3 locations; area PII, 6 soil samples from 2 locations, area PIII 6 soil samples from 3 locations and from area PIV 5 soil samples from 2 locations.

Fig. 1. Shipment of steel scrap by railroad.

Fig. 2. Temporary storage of steel scrap on unprotected scrap yard area.

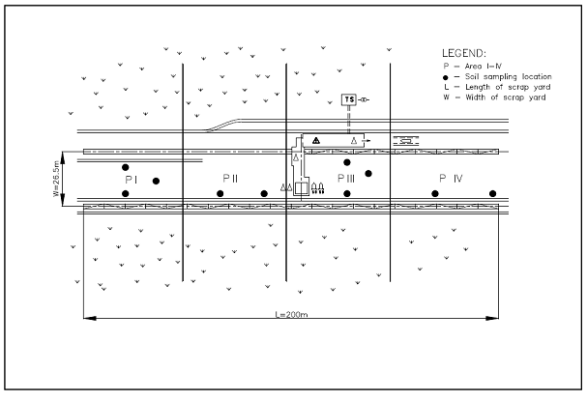

Fig. 3. Soil sampling locations at scrap yard area.

2000g samples were collected using a shovel at 30, 50 and 70 cm depth. For each area, a composite sample was made by mixing identical amounts (1000 g) of individually collected samples, and concentrations of heavy metals were determined.

Presence of water was noticed at a few sampling locations mainly at 0.5–0.7 m depth, and even layering of soil at all locations was noticed as follows:

- surface layer (callow) of sand up to 0.5 m depth (slag and mixture of iron oxides – rust);
- dark colour layer on depths between 0.5 and 1.0 m. The colour probably comes from washing of pollution from used mineral oils on the surface layer, indicated from hydrocarbon essence;
- clay layer on average depth of 1 to 1.2 m.

Water presence at some locations (at 0.5 –0.7 m depths), indicates serious problem in such polluted industrial yards, as pollutants can be washed out by a rainfall to deeper soil levels and can directly pollute underlying groundwater.

One reference sample was collected in a nearby park outside the CMC Sisak Ltd. factory ground approximately 500 m airline in the north-west direction. Activities carried out at the scrap yard have no direct influence on the park were reference sample was collected and as such represent soil image of a wider area (Fig. 4). Reference soil sample was collected in the same manner as samples used for preparation of composite samples.

Sampling locations were marked out with sequence numbers. Additionally, on some locations accordingly to the noticed layering, every soil layer by depth was marked with letters a, b, c (a – surface layer, b – median layer etc.). Preparation of samples for analysis was conducted according to the HRN ISO 11464:2004 (Soil quality –Pre-treatment of samples forphysic-chemical analyses) norm. For preparation microwave oven, MARS Xpress Microwave Digestion System CEM Corporation, was used. After preparing, samples were reserved in desiccators.

Fig. 4. Sampling location of reference soil sample in park outside of the factory.

Cd, Cr, Cu, Hg, Ni, Pb and Zn concentrations

Concentrations of heavy metals (Cd, Zn, Cu, Hg, Ni, Pb and all oxidation states of Cr) after extraction in *aqua regia* according to the HRN ISO 11466:2004 (Soil quality – Extraction of trace elements soluble in *aqua regia*) norm were determined. Heavy metal concentrations, except Hg, were determined by inductively coupled optical emission spectrometry according to the HRN EN ISO 11885:2010 (Water quality - Determination of selected elements by inductively coupled plasma optical emission spectrometry) norm using a PerkinElmer Optima 7000 DV ICP-OESapparatus. Concentration of Hg was determined by atomic absorption spectrometry, HRN EN 1483:2008 (Water quality - Method using atomic absorption spectrometry) norm on a Perkin-Elmer Atomic absorption spectrophotometer with Varian hydride-vapor generation system.

Results and Discussion

Soil as part of an ecosystem is, because of its complexity, a more dynamic system in comparison to the air and water; therefore, defining pollution is more complex. Soil buffering capacity is determined by its

physical, chemical and biological properties, which influence the "vulnerability"to pollution. Because of big differences between soils, there can also be significant differences in chemical composition. To determine the pollution level of any ecosystem component including soil, it is necessary to determine pollution influence on human health. This is done in order to determine the limiting value of pollution level of a particular pollutant for which, based on scientific knowledge, there is not even the slightest possible risk of harmful effect on the human health and/or the environment.

Croatian legislation defines Regulation on protection of agricultural soil from pollution (OG 32/10) as the only regulation that regulates quality of agricultural soil. Limiting values of pollutants, including the limits for concentrations of heavy metals, for soil used for industrial purposes are not prescribed. In Croatia, there is no legal regulation which is directly related to monitoring of soil condition and collection of data regarding potentially polluted or polluted soil. Problems with potentially polluted or polluted soil are only indirectly mentioned in legislation. Some EU member countries regulate limited values of pollutants in the soil (CARLON, 2007) according to the land usage (agricultural, residential, recreational, etc.), principle that was adopted in preparation of Croatian soil monitoring programme (CROATIAN ENVIRONMENT AGENCY, 2008).

The objective of the measurements conducted during the research was to determine if the temporary storage of steel scrap at the unprotected area present potential local source of anthropogenic soil pollution (despite its usage for industrial purposes). Croatian soil monitoring programme (CROATIAN ENVIRONMENT AGENCY, 2008) was used for the identification and interpretation of results. Based on potential sources of soil pollution and type of possible pollutant emissions listed in Programme (CROATIAN ENVIRONMENT AGENCY, 2008), in this paper, concentrations of Cd, Cr, Cu, Hg, Ni, Pb and Zn as pollutants originating from metal industry were determined.

The results of soil analysis from scrap yard area were compared to the recommended limited values (LV) of pollutants in soil allocated for industrial and commercial usage according to Croatian soil monitoring programme (CROATIAN ENVIRONMENT AGENCY, 2008) (Table 1).

Table 1. Limited values (LV) of pollutants in soil (metals extracted in *aqua regia)* according to land usage.

Sort of pollutions in soil	**Soil for agricultural production**	**Playground**	**Resident areas**	**Park and recreation areas**	**Areas for industrial and commercial usage**
	(mg/kg dry matter)				
Cd	2	5	10	30	50
Cu	60	60	100	300	500
Ni	50	50	70	200	500
Pb	100	100	100	500	1000
Zn	200	200	300	700	1200
Cr	100	100	200	500	750
Hg	2	5	10	30	50

Results for determination of heavy metals in composite samples of soil from scrap yard area (Table 2) show that concentrations were mostly below limited values (LV) of pollutants for industrially and commercially used soils according to Croatian soil monitoring programme (CROATIAN ENVIRONMENT AGENCY, 2008).

Exceptions were concentrations of Pb in soil samples from areas PII, PIII and PIV, Cu from PI, Cr from PIII and Zn from PII, PIII and PIV.

Table 2. Results from composite sample analysis from scrap yard area compared to the LV of pollutants in soil allocated for industrial and commercial usage according to Programme.

Element	Content of metals in soil, mg/kg dry matter				
	P I	**P II**	**P III**	**P IV**	**LV**
Cd	1.37	25.30	<0.001	<0.001	50
Hg	0.58	2.43	3.23	1.42	50
Pb	558	**2538**	**2620**	**2724**	1000
Ni	25.27	321	477	263	500
Cu	155	**2239**	722	643	500
Cr	158	438	**1333**	437	750
Zn	508	**14851**	**2457**	**2907**	1200

Noted differences in concentrations of metals between individual areas of scrap yard, for example concentrations of Zn and Pb in PI are significantly lower than those in other areas, can be explained by the shortest contact of PI area with steel scrap. Namely, area PI was rarely exposed to direct influence of pollutants contained in the steel scrap as steel scrap was rarely stored on that area. The newly brought steel scrap has mainly been unloaded on areas PIV and PIII, which are closest to the factory hall and the electric arc furnace.

Before comparison of concentrations of heavy metals in composites soil samples from scrap yard area with the concentrations of heavy metals outside of the factory, first it is necessary to compare results obtained for reference samples with the LV in soil allocated for industrial and commercial usage (Table 3).

Values of heavy metals in reference sample shown in Table 3 are significantly lower than the LV of tested metals for industrially and commercially used soil according to Croatian soil monitoring programme (CROATIAN ENVIRONMENT AGENCY, 2008).

Results obtained from the analysis of reference sample were compared with the maximum allowed values for heavy metals in agricultural soil (Table 4), according to the Regulation on protection of agricultural soil from pollution (OG 32/10). Concentrations of all metals, except Zn, were under the maximum allowed values or within the permitted maximum values.

Table 3. Contents of heavy metals in reference sample compared to the LV for in soil allocated for industrial and commercial usage.

Element	Contents of metal in soil, mg/kg dry matter	
	Reference sample	**LV**
Cd	<0.001	50
Hg	0.162	50
Pb	49.82	1000
Ni	34.14	500
Cu	21.09	500
Cr	57.84	750
Zn	284	1200

Results from determination of heavy metal content in composite samples (Table 5) point to a significant difference in permitted levels when compared to the content determined in reference samples and some of monitored metals in relation to the LV. This can be assigned to the contamination of soil with the heavy metals in the scrap yard area as a consequence of long-term temporary storage of steel scrap at unprotected and unroofed soil under different weather conditions. For pollution assessment, comparison

of results for heavy metals of composite soil samples from all four areas of scrap yard with values for potentially unacceptable risk for soil allocated for industry regulated by some EU member countries is given in Table 5.

Table 4. Content of metals in reference samples compared to regulated boundaries of maximum allowed values in agricultural soil.

Soil sample	Maximum allowed values, mg/kg dry matter						
	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Sandy soil	0.0-0.5	0-40	0-60	0.0-0.5	0-30	0-50	0-60
Dusty – clay soil	0.5-1.0	40-80	60-90	0.5-1.0	30-50	50-100	60-150
Clay soil	1.0-2.0	80-120	90-120	1.0-1.5	50-75	100-150	150-200

Table 5. Comparison of results of heavy metals content in composite soil samples with values for potentially unacceptable risk for soil allocated for industry regulated by some EU member countries.

Element	Scrap yardarea	LV of metals in soil, mg/kg dry matter							
		Sample	Reference sample	Belgium /Bruxelles/	Belgium /Wallonia/	Finland	Italy	Poland	United Kingdom
Cd	P I	1.37	<0.001	30	50	20	15	13	1400
	P II	25.30							
	P III	<0.001							
	P IV	<0.001							
Hg	P I	0.58	0.162	30	84	5	5	27	480
	P II	2.43							
	P III	3.23							
	P IV	1.42							
Pb	P I	558	49.82	**2500**	**1360**	**750**	**1000**	**600**	**750**
	P II	**2538**							
	P III	**2620**							
	P IV	**2724**							
Ni	P I	25.27	34.14	700	500	**150**	500	**285**	-
	P II	**321**							
	P III	**477**							
	P IV	**263**							
Cu	P I	155	21.09	**800**	**500**	**200**	**600**	**600**	-
	P II	**2239**							
	P III	**722**							
	P IV	**643**							
Cr	P I	158	57.84	**800**	**700**	**300**	**800**	**475**	5000
	P II	**438**							
	P III	**1333**							
	P IV	**437**							
Zn	P I	**508**	284	**3000**	**1300**	**400**	**1500**	**1650**	-
	P II	**14851**							
	P III	**2457**							
	P IV	**2907**							

Comparison of heavy metals content in composite soil samples from scrap yard area with the values for potentially unacceptable risk for industrial soil regulated by some EU member countries points out that concentrations of Cd and Hg were under the value for potentially unacceptable risk regulated in Belgium, Finland, Italy, Poland

and the United Kingdom i.e. all considered countries which have regulated LV for those heavy metals.

Lead concentration was lower only in part of PI area, compared to the area (PII – PIV) where it was higher than the values marked as potentially unacceptable risk regulated by some countries. Regarding to its Pb content area scrap yard could be considered polluted.

Concentration of Ni in PI was also lower than the values considered as potentially unacceptable risk regulated by the mentioned EU member countries, while concentrations of Ni from PII – PIV were lower than the values considered as potentially unacceptable risk regulated by Belgium and Italy, and higher than those regulated by Finland and Poland.

Pollution of scrap yard area with the Cu, Cr and Zn was more pronounced, indicating determined concentrations which were, except in PI, higher than values for potentially unacceptable risk regulated by Belgium, Finland, Italy and Poland for soil allocated for industrial and commercial use.

Conclusions

Soil pollution in the Republic of Croatia is defined only for agricultural soil by Regulation on protection of agricultural soil from pollution (OG 32/10), while the quality of soil allocated to industrial and other purpose is not regulated.

Soil pollution assessment was carried out at the scrap yard area that was used for temporary storage of steel scrap during a number of years in CMC Sisak Ltd. Obtained results for heavy metals were compared with the values marked as potentially unacceptable risk for industrial soil according to the Croatian Soil Monitoring Programme and regulated values by some EU member countries. Results were also compared with content of heavy metals in reference soil sample in the vicinity of storage, as well as to all obtained data, thus that following conclusions could be drawn:

- concentrations of heavy metals in all composite soil samples exceed concentrations from the reference sample, which can be assigned to soil contamination at the scrap yard area as a consequence of long term temporary steel scrap storage at unprotected and unroofed soil;
- Cd and Hg concentrations were lower than values for potentially unacceptable risk regulated in all considered countries;
- Pb concentrations were lower only in a part of PI area, opposite to areas (PII – PIV) where they were higher than values for potentially unacceptable risk regulated by considered countries. Regarding to Pb levels determined in composite sample area, the scrap yard could be considered polluted;
- Concentration of Ni from PI were also lower than values considered as potentially unacceptable risk regulated by mentioned EU countries, while concentrations from PII – PIV were lower than values for potentially unacceptable risk regulated by Belgium and Italy, and higher than values regulated by Finland and Poland.
- Pollution of scrap yard area with Cu, Cr and Zn was more pronounced than with other metals, indicating determined concentrations which were, except in PI, higher than values for potentially unacceptable risk regulated by Belgium, Finland, Italy and Poland for soil allocated for industrial and commercial purposes.
- On the base of determined differences in metal content in-between individual areas of the scrap yard, PI area has been least polluted with heavy metals, because steel scrap was rarely stored in that area.

As a result, concentrations of heavy metals in all composite soil samples taken from scrap yard area mainly exceed values of same metals in the reference sample, and values for potentially unacceptable risk for industrial soil as regulated by some EU countries, implying this soil can be defined as contaminated. Therefore it is necessary to take measures as specified in Article 8 Regulations on waste management (OG 23/07, OG 111/07) for the purpose of soil

protection and prevention of further soil pollution with following methods:

- area for temporary steel scrap storage should be built from solid material, fully sealed or enclosed roofed space protected from rainfall;
- Floor of the storage/temporary landfill should be leak-tight and resistant to effects of stored steel scrap and polluted ingredients present in it;
- Storage/temporary landfill should be equipped to avoid emission of dust, noise, smell and other emissions in the environment.

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