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Operator Exposure to Heavy Metals from Wood Pellet Ash – Risk Assessment

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Abstract

Ash is a by-product of wood biomass combustion that must be removed daily from stoves or fireplaces. Therefore, operators or owners are exposed to the potential impact of ash. The goal of this study was to determine whether heavy metal present in wood pellet ash posed a health concern to stove operators/owners. The risk assessment procedure was carried out in several steps, including exposure evaluation, toxicity evaluation, and risk categorisation. The hazard coefficient (HQ) and non-carcinogenic hazard index (HI) were calculated for Cd, Cr, Cu, Ni, Pb, and Zn. HQ had the highest value for the ingestion pathway ($3.62 \cdot 10^{-6}$), and the value for non-carcinogenic HI was $3.70 \cdot 10^{-6}$. The value HI < 1 suggests that there is no risk to operator health related to heavy metals in analysed wood pellets ash. The carcinogenic risk (CR) was calculated for Ni, Pb, Cr, and Cd, and the values were within the permitted limits. The risk assessment based on HI and CR indicators proved that there was no significant health concern regarding exposure to the analysed ashes.

Keywords

Wood pellets, ash, heavy metals, risk assessment, (non-)carcinogenic risk

1 Introduction

Due to rising natural gas and oil prices, as well as increased environmental awareness, wood is becoming more popular as a source of energy, particularly for thermal energy. In recent years, special emphasis has been placed on the use of wood biomass as an energy source.[1] Advantages of renewable energy sources are widely used, and contribute to environmental protection due to reduced harmful emissions from combustion.[2] Biomass from the industry such as wood waste and residues has been an unused potential in the past. However, in recent times, wood waste and residues are used for the production of especially current forms of wood biomass and pellets.[3]

The waste generated by the utilisation of wood biomass is predominantly ash. Ashes are microparticles of material generated when solid fuels are burned. The type of solid fuel (wood, coal, or other high-energy substances) burned and the boilers used in the process determine the composition of the ash.

Our findings show that the ash obtained by burning wood pellets from the Bosnia and Herzegovina (B&H) market contains heavy metals.[4] Therefore, ash engenders significant environmental and human health problems due to enhanced contents of heavy metals. Because proper maintenance of pellet stoves or fireplaces demands the removal of ashes on a daily basis, we can predict that operators (usually the owners) are most exposed to dust and compounds emitted in the interior air from the ashes.

A health risk assessment is a method for estimating the impact of specific hazardous compounds on human health or the environment. The risk of heavy metals was assessed in different studies for different types of ash like ash from wood pellets,[4,5] solid fuels mixed with municipal waste,[6] and coal.[7]

Therefore, the aim of this study was to assess the potential health effects of heavy metals for operators based on heavy metal levels in wood pellet ash. We calculated the hazard index (HI), total carcinogenic risk (TCR), hazard quotient (HQ), and carcinogenic risk (CR) for heavy metals found in wood pellet ash to determine the corresponding operator's health impacts.

2 Materials and methods

The model used in this work to estimate operator exposure to heavy metals in wood pellet ash was based on models recommended by the US Environmental Protection Agency.[8–10] Health risk assessment for operators was performed for ten samples of ash obtained by combustion of wood pellets (Table 1). Samples of wood pellets for the determination of heavy metals were prepared by wet digestion with HNO_3 (65 %). Atomic absorption spectrometry (Varian AA240FS) was utilised for Pb and Zn, whereas a graphite furnace (Varian AA240Z) was employed for Cd, Co, Cr, Cu, and Ni.[4]

2.1 Non-carcinogenic risk assessment for operators

Non-carcinogenic health risk was calculated for daily intake (DI) by ingestion, inhalation and dermal intake. Eq. (1) was used to calculate the exposure by ingestion (ing). Expo-

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Table 1 – List of analysed wood pellets
Tablica 1 – Analizirani uzorci drvenih peleta

Sample code	Wood type	Origin of the sample
S1	beech and oak	Teslić, B&H
S2	beech and fir	Tomislavgrad, B&H
S3	beech	Misilmeri, Italy
S4	oak, beech, ash tree, and hornbeam	Sarajevo, B&H
S5	oak, beech, ash tree, and hornbeam	Gračanica, B&H
S6	beech and spruce	Foča, B&H
S7	coniferous wood	Gračanica, B&H
S8	beech and spruce	Sokolac, B&H
S9	beech, spruce, and fir	Vitez, B&H
S10	beech, spruce, and fir	Sarajevo, B&H

sure by inhalation (inh) was calculated using Eq. (2), while Eq. (3) was used to calculate dermal-route exposure (der).

$$\mathrm{DI}_{\mathrm{ing}} = \frac{C_{\mathrm{UCL}} \cdot \left(R_{\mathrm{ing}} \cdot F_{\mathrm{exp}} \cdot T_{\mathrm{exp}} \cdot 10^{-6}\right)}{\mathrm{ABW} \cdot T_{\mathrm{avrg}}} \quad (1)$$

$$\mathrm{DI}_{\mathrm{inh}} = \frac{C_{\mathrm{UCL}} \cdot \left(R_{\mathrm{inh}} \cdot F_{\mathrm{exp}} \cdot T_{\mathrm{exp}}\right)}{\mathrm{PEF} \cdot \mathrm{ABW} \cdot T_{\mathrm{avrg}}} \quad (2)$$

$$\mathrm{DI}_{\mathrm{der}} = \frac{C_{\mathrm{UCL}} \cdot \left(\mathrm{SAF} \cdot A_{\mathrm{skin}} \cdot \mathrm{DAF} \cdot F_{\mathrm{exp}} \cdot T_{\mathrm{exp}} \cdot 10^{-6}\right)}{\mathrm{ABW} \cdot T_{\mathrm{avrg}}} \quad (3)$$

Reference values of health risk assessment parameters are shown in Table 2.

C_{UCL} (exposure point upper confidence limit – UCL) is an evaluation of the reasonable maximum exposure which is the upper limit of the 95 % confidence interval for the mean.[11–13] Eq. (4) was used to determine the C_{UCL}:[9,13]

$$C_{\mathrm{UCL}} = X + \left[Z_{\alpha} + \beta\left(1 + 2 \cdot Z_{\alpha}^{2}\right) \cdot \left(6\sqrt{n}\right)^{-1}\right] \cdot \left[\frac{\mathrm{STD}}{\left(\sqrt{n}\right)}\right] \quad (4)$$

where X (mg kg^{-1}) is the mean concentration of each heavy metal, Z_{α} is $(1-\alpha)^{\mathrm{th}}$ quantile of the standard normal distribution for the 95 % confidence level (1.645), β is the skewness, n is the number of samples, and STD is the standard deviation.

The hazard coefficient (HQ) was used to calculate the possible non-carcinogenic risk for each heavy metal. Equation (5) was used to calculate hazard coefficient:

$$\mathrm{HQ} = \frac{\mathrm{CDI} \cdot \mathrm{BAF}}{\mathrm{RfD_O}} \quad (5)$$

Table 2 – Reference values of variables used for health risk assessment of heavy metals from ash samples
Tablica 2 – Referentne vrijednosti varijabli upotrijebljenih za procjenu zdravstvenog rizika od teških metala iz uzoraka pepela

Variable (abbreviation)	Value used	Units	Ref.
C_{UCL} – exposure-point upper confidence limit content concentrations	–	mg kg^{-1}	5
R_{ing} – ingestion rate	30	mg day^{-1}	11
R_{inh} – inhalation rate	20	m^3 day^{-1}	10, 11
F_{exp}* – exposure frequency	2.5	day year^{-1}	5
A_{skin} – skin area	5700	cm^2	8
SAF – skin adherence factor	0.1044	mg cm^{-2}	9, 10
DAF – dermal absorption factor	0.001	–	10
PEF – particle emission factor	$1.36 \cdot 10^9$	m^3 kg^{-1}	10
ABW – average body weight of operators	70	kg	9–11
T_{avrg} – average time; for non-carcinogens	8760	day	10

* Calculated as: 20 min *per* day for 180 days a year as the average exposure frequency

where BAF is the ratio of metal content that is bioavailable to the total content in the analysed samples,[5] and $\mathrm{RfD_O}$ is the reference dose of a specific heavy metal (mg kg^{-1} day^{-1}).

The potential non-carcinogens risk to the operators/owners through intake of heavy metals was determined using hazard index (HI) given by Eq. (6).

$$\mathrm{HI} = \sum_{k=1}^{n} \mathrm{HQ}_k \quad (6)$$

2.2 Carcinogenic risk assessment

The carcinogenic risk (CR) can be used to calculate the probable cancer risks associated with exposure to a specific amount of heavy metal in ashes of wood pellets. The CR is defined as the cumulative risk of acquiring any type of cancer throughout a lifetime as a result of the operator's exposure to a carcinogenic hazard over ashes of wood pellets. For calculating the lifetime cancer risk, the following equation was used:

$$\mathrm{CR} = \mathrm{CDI} \cdot \mathrm{BAF} \cdot \mathrm{SLF} \quad (7)$$

where SLF is slope factor, values are from Regional Screening Levels.[5,14]

The total cancer risk (TCR) associated with exposure to certain carcinogenic heavy metals is the sum of the individual cancer risks.

3 Results and discussion

Operator exposure to heavy metals (Cd, Cr, Cu, Ni, Pb, and Zn) from ash, as well as the potential impact on their health was estimated using calculations based on the concentrations of heavy metals in wood pellet ash.

The concentrations of heavy metals in these samples are shown in Fig. 1.[4] As may be expected, there are significant differences in the concentrations of heavy metals depending on the type of wood pellet the ash of which was analysed. The content of heavy metals in wood pellets depends on the type of wood biomass from which the wood pellet is made, but also on the pollution caused by the production process and the use of chemically treated wood biomass.[15] Heavy metals are present in very small concentrations in woody biomass, although most of them are increased due to pollution. Such pollution could be due to the growth of wood in the urban, industrial zone of the city. Therefore, some European countries (Sweden, Denmark, Germany) have defined the permissible concentrations of heavy metals in the ash by legislation.[16]

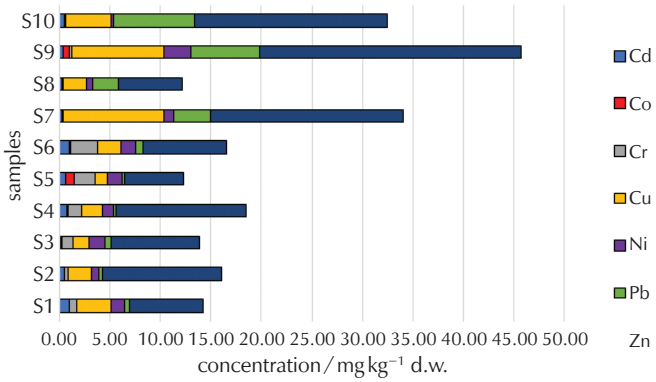

Fig. 1 – Concentrations of heavy metals in ash of wood pellets.[4] Abbreviation d.w. refers to dry weight
Slika 1 – Koncentracija teških metala u pepelu drvenih peleta.[4] Kratica d.w. označava suhu masu

The exposure to heavy metals from ash is evaluated for non-carcinogenic (Hazard Index) and carcinogenic effects (Carcinogenic Risk Index).

Because different metals are commonly found in wood ash, humans, especially operators, are exposed to heavy metals *via* different pathways. If operators or owners of wood pellet stoves or fireplaces are not adequately protected, they are frequently exposed to the possible impacts of heavy metals from ashes during maintenance and cleaning.

Therefore, the calculation was realised for non-carcinogenic and carcinogenic health risks for three different routes: ingestion, inhalation, and dermal.

The obtained results for non-carcinogenic risk (HQ) for three different routes of exposure to metals are presented in Fig. 2. The calculated HQ values are less than 1. Thus, heavy metals from analysed ash posed no direct risk for operators.

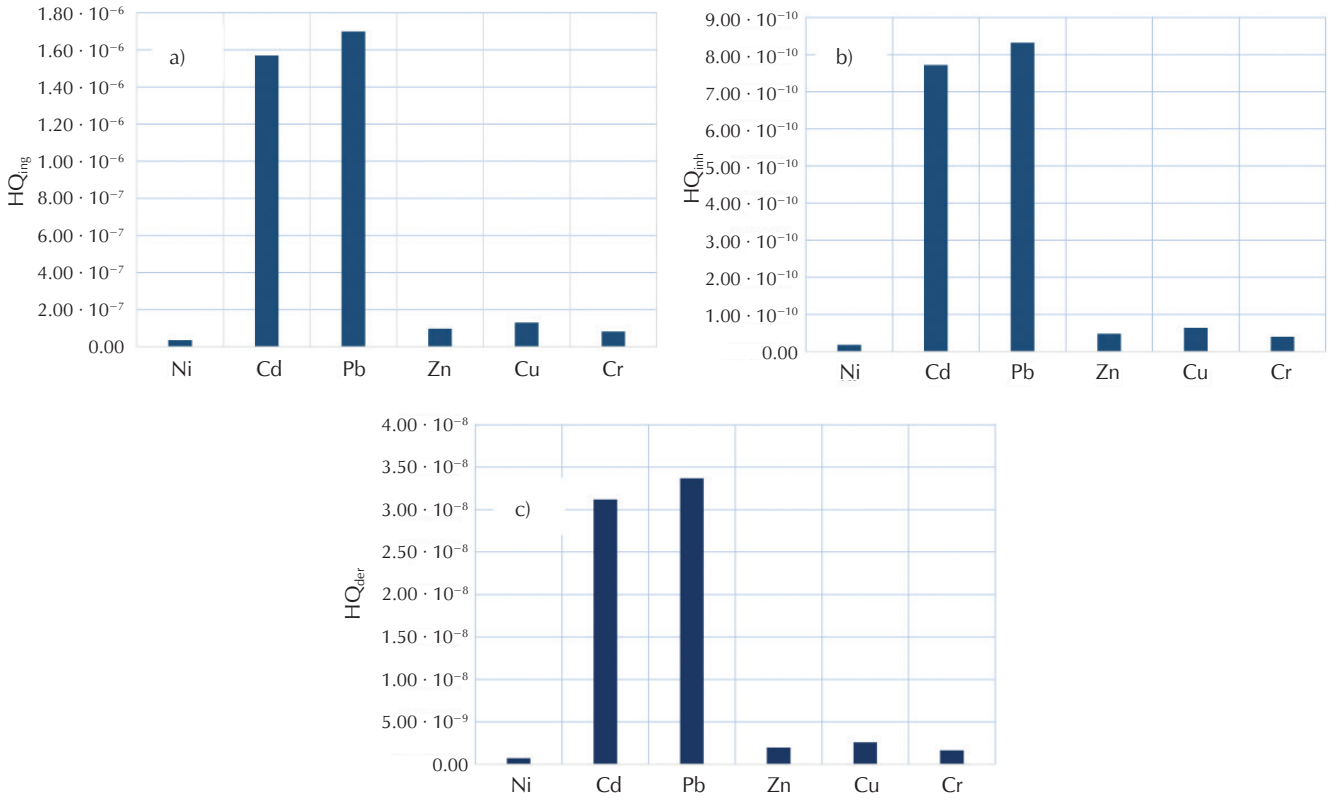

Fig. 2 – Hazard coefficients (HQ) for three different routes of exposure: (a) ingestion, (b) inhalation, and (c) dermal exposure
Slika 2 – Koeficijent opasnosti (HQ) za tri različita puta izlaganja: (a) gutanjem, (b) udisanjem i (c) preko kože

Table 3 – Daily intake (DI), non-carcinogenic hazard index (HI), carcinogenic risk (CR) and total carcinogenic risk (TCR) values for operator exposure by ingestion (ing), inhalation (inh), and dermal route (der)

Tablica 3 – Dnevni unos (DI), indeks opasnosti (HI), karcinogeni rizik (CR) i ukupni karcinogeni rizik (TCR) za operatore izložene gutanjem (ing), udisanjem (inh) i preko kože (der)

	DI_{ing}	DI_{inh}	DI_{der}	HI	CR_{ing}	CR_{inh}	TCR
Fe	$9.08 \cdot 10^{-7}$	$4.45 \cdot 10^{-10}$	$1.80 \cdot 10^{-8}$	–	–	–	–
Mn	$1.88 \cdot 10^{-7}$	$9.21 \cdot 10^{-11}$	$3.73 \cdot 10^{-9}$	–	–	–	–
Ni	$4.67 \cdot 10^{-9}$	$2.29 \cdot 10^{-12}$	$9.26 \cdot 10^{-11}$	$3.73 \cdot 10^{-8}$	$6.67 \cdot 10^{-10}$	$3.02 \cdot 10^{-13}$	$6.67 \cdot 10^{-10}$
Co	$1.20 \cdot 10^{-9}$	$5.90 \cdot 10^{-13}$	$2.39 \cdot 10^{-11}$	–	–	–	–
Cd	$2.11 \cdot 10^{-9}$	$1.04 \cdot 10^{-12}$	$4.19 \cdot 10^{-11}$	$1.60 \cdot 10^{-6}$	–	$4.86 \cdot 10^{-12}$	$4.86 \cdot 10^{-12}$
Pb	$1.27 \cdot 10^{-8}$	$6.20 \cdot 10^{-12}$	$2.51 \cdot 10^{-10}$	$1.73 \cdot 10^{-6}$	$5.05 \cdot 10^{-11}$	$1.22 \cdot 10^{-13}$	$5.06 \cdot 10^{-11}$
Zn	$4.91 \cdot 10^{-8}$	$2.40 \cdot 10^{-11}$	$9.73 \cdot 10^{-10}$	$1.00 \cdot 10^{-7}$	–	–	–
Cu	$1.76 \cdot 10^{-8}$	$8.62 \cdot 10^{-12}$	$3.49 \cdot 10^{-10}$	$1.34 \cdot 10^{-7}$	–	–	–
Cr	$4.26 \cdot 10^{-9}$	$2.09 \cdot 10^{-12}$	$8.44 \cdot 10^{-11}$	$8.40 \cdot 10^{-8}$	$1.23 \cdot 10^{-10}$	$4.96 \cdot 10^{-12}$	$1.28 \cdot 10^{-10}$
Σ	$1.19 \cdot 10^{-6}$	$5.82 \cdot 10^{-10}$	$2.36 \cdot 10^{-8}$	$3.70 \cdot 10^{-6}$	$8.40 \cdot 10^{-10}$	$1.02 \cdot 10^{-11}$	$8.50 \cdot 10^{-10}$

The highest HQ values were observed for ingestion ($HQ_{ing} = 3.62 \cdot 10^{-6}$); they were lower for dermal contact ($HQ_{der} = 7.18 \cdot 10^{-8}$), and lowest for inhalation ($HQ_{inh} = 1.77 \cdot 10^{-9}$). Our results for HQ values are consistent with the results of *Kicinska*[6] and *Bourliva*[17]. Therefore, the most relevant exposure pathway in terms of health concerns appears to be ingestion. This can be related to the fact that the actual daily intake was determined by the value of the hazard quotient in each pathway, and parameters used to calculate the exposure dose for each exposure pathway were different.[18]

The total hazard index for non-carcinogenic substances was $3.70 \cdot 10^{-6}$ (Table 3). HI < 1 suggests a very low risk for adverse health impact on operators exposed to wood pellet ash during cleaning of pellet stoves in confined environments. The fact that the greatest values for HQ (ingestion, inhalation, and dermal) are calculated for Pb ($1.73 \cdot 10^{-6}$) and Cd ($1.60 \cdot 10^{-6}$) can be concerning, even while the result for HI was within the acceptable levels. Therefore, operators may be exposed to the harmful effects of Pb and Cd from ash when maintaining and cleaning wood pellet stoves if they do not wear appropriate protective equipment. Pb is one of the most common heavy metals found in different types of ashes.[19] It is moderately toxic, it affects the intestine and central nervous system when swallowed, and causes anemia.[20]

Cd is an element that has no function in the body. It is a cumulative poison that enters the body, is deposited in the kidneys, and remains almost a lifetime. Lung cancer, prostatic proliferative lesions, bone fractures, kidney dysfunction, and hypertension are all possible side effects of chronic Cd exposure.[21] Therefore, the permissible concentration of Cd in ash is defined in some countries, and is 30 and 20 $\mu g\, g^{-1}$ for Sweden and Denmark, respectively.[22]

Considering the cancer risk, the lowest CR value was calculated for Pb and inhalation ($1.22 \cdot 10^{-13}$), and the highest for Ni and ingestion ($6.67 \cdot 10^{-10}$). Chronic exposure to small doses of Ni, especially through inhalation or ingestion of its dust, can cause cancer.

4 Conclusion

Heavy metals in ashes, like other hazardous substances, cause serious health consequences. Operators who clean and maintain stoves on a daily basis may be exposed to heavy metals from the ash produced by burning wood pellets. Therefore, a health risk assessment was performed for the three routes of exposure: ingestion, inhalation, and dermal exposure. In general, HQ and HI were lower than the permitted level of 1. The highest values for HQ were obtained for Pb and Cd, which are toxic elements, so further monitoring is required. In addition, operators and owners should use adequate protective equipment. The obtained result shows that the cancer risk levels of Ni, Cd, Pb, and Cr are lower than the acceptable range, indicating that the carcinogenic risks of mentioned metals for indoor wood pellet ash are negligible. Consequently, based on the results of the non-carcinogenic and carcinogenic risks for the metals analysed in the current study, the health risks from wood pellet ash for operators in indoor environments are not significant.

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List of abbreviations
Popis kratica

ABW – average body weight of operators
– prosječna tjelesna težina operatera

A_{skin} – skin area
– područje kože

BAF – ratio of metal content
– omjer sadržaja metala

CR – carcinogenic risk
– karcinogeni rizik

C_{UCL} – exposure-point upper confidence limit
– gornja granica intervala pouzdanosti točke izloženosti

DAF – dermal absorption factor
– apsorpcijski faktor kože

DI – daily intake
– dnevni unos

DI_{der} – dermal daily intake
– dnevni unos preko kože

DI_{ing} – ingestion daily intake
– dnevni unos gutanjem

DI_{inh} – inhalation daily intake
– dnevni unos udisanjem

F_{exp} – exposure frequency
– učestalost izlaganja

HI – non-carcinogenic hazard index
– indeks nekarcinogene opasnosti

HQ – hazard coefficient
– koeficijent opasnosti

HQ_{der} – hazard coefficient for dermal
– koeficijent opasnosti za izloženost preko kože

HQ_{ing} – hazard coefficient for ingestion
– koeficijent opasnosti za gutanje

HQ_{inh} – hazard coefficient for inhalation
– koeficijent opasnosti za udisanje

PEF – particle emission factor
– faktor emisije čestica

RfD_o – reference dose
– referentna doza

R_{ing} – ingestion rate
– brzina gutanja

R_{inh} – inhalation rate
– brzina udisanja

SAF – skin adherence factor
– faktor prianjanja na kožu

SLF – slope factor
– faktor nagiba

STD – standard deviation
– standardna devijacija

T_{avrg} – average time; for non-carcinogens
– prosječno vrijeme; za nekarcinogene tvari

TCR – total carcinogenic risk
– ukupan karcinogeni rizik

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SAŽETAK

Procjena rizika izloženosti operatora teškim metalima iz pepela drvenih peleta

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Pepeo je nusproizvod izgaranja drvne biomase koji se svakodnevno mora uklanjati iz peći ili kamina. Stoga su operateri ili vlasnici izloženi potencijalnom utjecaju pepela. Ovo istraživanje ima cilj procijeniti zdravstveni rizik izloženosti operatera/vlasnika pepelu drvenih peleta zbog sadržaja teških metala. Postupak procjene rizika proveden je u nekoliko koraka uključujući procjenu izloženosti, procjenu toksičnosti i kategorizaciju rizika. Izračunati su koeficijent opasnosti (HQ) i indeks nekarcinogene opasnosti (HI) za Cd, Cr, Cu, Ni, Pb i Zn. HQ je imao najveću vrijednost za izloženost putem gutanja ($3.62 \cdot 10^{-6}$), a vrijednost za nekarcinogeni HI bila je $3.70 \cdot 10^{-6}$. Vrijednost HI < 1 ukazuje na to da sadržaj teških metala u analiziranom pepelu ne predstavlja rizik za zdravlje operatera. Karcinogeni rizik (CR) izračunat je za Ni, Pb, Cr i Cd, a vrijednosti su bile unutar dopuštenih granica. Rizik procijenjen primjenom HI i CR pokazatelja potvrdio je da ne postoji značajna opasnost za zdravlje osoba koje dolaze u kontakt s analiziranim pepelom.

Ključne riječi
Drveni peleti, pepeo, teški metali, procjena rizika, (ne)karcinogeni rizik

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