Research Article

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Assessment of the mercury contamination of landfilled and recovered foundry waste – a case study

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Abstract: Environmental pollution by mercury is a local problem in Poland and concerns mainly industrial sites. Foundry waste are usually characterized by low mercury content compared to other heavy metals. Spent foundry sands with low content of Hg are the main component of foundry waste. However, Hg may be present in foundry dust, which may also be landfilled. Due to Hg toxicity, even a minimal content may have a negative impact on biota. This study focuses on assessing the mercury content of landfilled foundry waste (LFW), to assess its toxicity. Currently tested waste is recovered and reused as a road aggregate. The results were compared with the mercury content of local soils as the reference level. Waste samples were taken from foundry landfill. The mercury content, fractional composition, organic matter (OM) and total organic carbon content, pH and elementary composition of waste were analysed. It was found that the mercury content in LFW was very low, at the level of natural content in soils and did not pose a threat to the environment. The statistical analysis shows that mercury was not associated with OM of the waste, in contrast to soils, probably due to different types of OM in both materials.

Keywords: foundry waste, landfilling, mercury, fractions, soil

1 Introduction

In the foundry industry, depending on the technological process used, various types of waste are produced. These include mainly spent foundry sands (SFSs), furnace slags, sludges or dusts from dust collectors and small quantities of metallic chips, excess material and others [1]. It is estimated that the mass of waste generated during the production process is similar to the mass of castings produced [2]. Therefore, the most important environmental protection measure taken by the foundry industry is the reduction in the amount of waste generated and its recovery. According to the Polish classification of waste, waste from the iron and steel industry is classified primarily in group 10 “Waste from thermal processes,” subgroup 10 09 “Waste from iron casting.” More than 75 codes have been assigned to foundry waste [2,3]. SFS represents up to 90% of the mass of all foundry waste produced. The sources of SFS are the processes of moulding, knock-out and the purification of castings. Due to high landfill costs, many foundries now use SFS regeneration facilities. The SFS that cannot be regenerated and reused in the foundry may be used in other industrial sectors, e.g. in road building and construction for the production of concrete mortar, Portland cement, road profile filling, road embankments and industrial platforms, as additives to asphalt, brick production, trench filling and ceramics [4]. In many countries such as United States, SFS is used to produce artificial soil substrates for agricultural and horticultural use [5,6]. The use of SFS for the production of artificial soil and technosol must not pose a risk to the environment and human health [7–9]. Therefore, the use of SFS from non-ferrous metal foundries is not recommended due to their contamination with heavy metals [10–13]. In addition to SFS, other foundry waste, such as metal, slag and dust, is also used. Even foundry waste that has been landfilled for many years may be reused [14]. The cost-effectiveness of the use of this waste depends on a number of factors, such as the type of waste, composition, the content of
pollutants, physical properties and availability [15]. The main threat posed by foundry waste is the possibility of leaching and the migration of heavy metals in the environment [10,11,14,16–18]. The degree of contamination of SFS with heavy metals is mainly related to not only the type of cast metal but also the composition of the foundry sands, binders and hardening agents. Among the heavy metals, Hg, Cd and Pb are considered to be the most toxic [19]. However, the content of these metals in foundry waste is usually low [5,20].

Mercury is one of the most toxic heavy metals. Hg pollution is a global problem [21]. Therefore, in 2017, more than 120 countries signed the Minamata Convention, which aims to reduce global mercury emissions to the environment. The main anthropogenic sources of environmental pollution by mercury are industry, including the chemical industry, waste and fuel combustion and agriculture [22]. Increased Hg content in soils is found around metallurgical plants and smelters, coal-fired power plants, cement plants and their industrial waste landfills [22]. Industrial waste is also an important source of mercury in the environment [23,24]. Environmental pollution by mercury is a local problem in Poland and concerns industrial sites, mainly those close to mines [25], coal power station [26,27], in the vicinity of transport routes [28,29], in areas with active mining industry [30] and the Pb–Zn metallurgy [24]. In soil, mercury and many other metals are usually bounded with loamy material, sulphates and soil organic matter (SOM) [19,21,29]. Databases and maps of pollutant (heavy metals) distribution in the soils of all continents, including Europe, have been created in recent years [31]. According to these data, the mercury content in European soils varies within limits: agricultural soils <0.003–1.6 mg kg⁻¹ DM and grazing land soil <0.003–3.1 mg kg⁻¹ DM, respectively [31]. In Europe, the mercury pollution of soils occurs locally and is related to human activities, mainly in urban agglomerations, near coal-fired power stations, waste incinerators, chemical plants or metal works. According to GEMAS data, Poland belongs to the group of countries with a low Hg content in soils [31]. Only the border region with the Czech Republic and Germany (Western Sudetes) is polluted by this metal to a greater degree. The cause of this anomaly has not been explained; however, the impact of emissions from the combustion of hard coal was excluded [31]. In Poland, the degree of pollution with mercury and other metals in soils is regulated by Ordinance by conducting the assessment of pollution of the earth’s surface [32]. From the soil, mercury can penetrate into groundwater or accumulate in plants. The use of SFS as a substitute for soil brings about the risk of the contamination of soil substrates and thus the heavy metal contamination of crops [5,6]. However, the content of mercury in foundry waste is generally low, often below the limit of quantification (LOQ); therefore, its determination in SFS is omitted [5,17,20].

Although heavy metal contamination of SFS has been investigated thoroughly, Hg has received relatively little attention. Foundry wastes are usually characterized by low mercury content; for this reason, in the toxicity assessment of this waste the content of this metal is not analysed. However, due to Hg toxicity, even a minimal content may have a negative impact on biota. In previous studies, the content of heavy metals and their leaching from foundry waste were assessed [14,18,33], the purpose of which was to classify and assess the environmental impact of this waste. This study focuses on assessing the mercury content of foundry waste. The results were compared with the mercury content of local soils to assess the degree of waste pollution compared to the reference level. The detailed aims of this study were as follows: (1) the assessment of the degree of mercury contamination of landfilled foundry waste (LFW) from one of the Polish foundries; (2) fractional (particle-size) analysis of waste and the determination of a possible relationship between Hg content, particle size and organic matter (OM); and (3) a comparison of the results with the natural content in soil.

2 Materials and methods

2.1 Foundry characteristics and waste sampling

The foundry is located in south–western Poland, in the Opolskie Voivodeship (N 50°40’20.5853”; E 18°12’44.7181”). The foundry produces steel and iron castings, raw and machined, which are parts for industrial machinery and equipment. The landfill is located next to the foundry (N 50°40’40.0885”; E 18°12’17.1021”). The landfilled waste includes SFS, slag, spent refractories, sludge from dust collectors and sewage treatment plants and metalliferous inclusions. Foundry waste was landfilled from the 1990s to the present day.Previously, foundry waste was landfilled in another landfill, which is located at a considerable distance from the foundry and is no longer owned by the foundry. Currently, the waste material from both landfills is being recovered; this process began in the 2000s, and it is being used for aggregate production. It is estimated that over 3.5 million tonnes of waste was deposited in both landfills (data from 2003).
Due to the recovery and reuse of landfilled waste, in 2015 the landfill area owned by the foundry was reduced from 8.9 to 4.66 ha and modernized. The location of the foundry and landfill is shown in Figure 1. The tested samples were taken from this landfill in 2017. By this time most of the waste had been recovered. Six piles of recovered waste material were created at the landfill, with a height of 3 m and a capacity of approximately 100 m³ each. Test samples were taken from these piles (Figure 2). A total of 10–15 incremental samples were taken from each pile according to Polish standards (PN-EN 932-1:1999; PN-EN 932-2:2001). The samples were mixed and the volume was reduced by quartering to 5 L (laboratory sample). In the laboratory, the tested samples were dried at room temperature. Then, fraction analysis was performed using sieves with a decreasing mesh size (0.2–5.6 mm). Fractional analysis was performed in triplicate for each batch. A sample of 1 kg was used for this purpose. Each fraction was weighed with an accuracy of 0.1 g. The triplicate results were averaged. The other portion of the waste material was ground into a ball mill without pre-sieving.

2.2 Soil sampling

In order to assess the reference level, soil samples were taken from 20 measuring points located at different distances from the landfill. The sampling locations were the same as they were in previous studies [33] (Figure 3). The soils originated from arable land (No. 2, 3, 4, 7, 8, 16, 17, 19 and 20), meadows (No. 1), wasteland (No. 6, 9 and 14), allotment gardens (No. 5, 11, 12, 13 and 15) and forest soil (No. 10 and 18). Soil samples were taken and prepared according to Polish standards (PN-ISO 10381-2:2007; PN-ISO 10381-5:2009). Similar to waste samples, the soil was collected in November 2017. The samples were dried at room temperature, in air-dry conditions, triturated in a mortar and sieved (<1, <0.2 mm). No fractional analysis was performed for soil samples as it was not the aim of the study. The granulometric composition and classification of soils were performed in previous studies [33]. The mercury content in local soils as a natural background was compared with the landfilled waste.

3 Methods

The pH was measured in an aqueous extract and in 1 M KCl (1/5, v/v) using a glass electrode with pH-conductometer CPC 501 (Elmetron) according to Polish standard (PN-ISO 10390). The content of total organic carbon (TOC) (<0.2 mm) according to the modified Tyurin
titrimetric (classical) method [34] was analysed. The CHNS (carbon, hydrogen, nitrogen, sulphur) in the milled <0.1 mm averaged waste lot were determined using Elemental Vario Macro Cube elemental analyzer. In every fraction of waste and soil samples (<1 mm), the content of OM and SOM at 550°C and Hg was analysed. Mercury content was determined using the cold vapour atomic absorption spectrometry method for solid samples using an analyzer AMA 254 (Altec Ltd). All samples were carried out and analysed in triplicate.

### 3.1 Correlation analysis and quality control

The arithmetic mean and standard deviation from triplicate data were calculated using Software Statistica version 13.3, considering a significance level of $p < 0.05$. The simple correlation coefficients ($r$) ($p < 0.05$) between the LFW fractions, OM, TOC, total carbon (TC) and Hg of foundry waste and between SOM and Hg content in soil were calculated using Pearson’s test. For quality control certified reference materials (CRMs) were also analysed: for Hg content “Metals in soil” (SQC001, Merck) and “Rock” (DC73303, NCS) and for CHNS content “Alfaalfa” (Cert. No. 41505). The accuracy of the methods was assessed by testing the degree of CRM recovery. Acceptable recovery range for CRM was 90–110%. The LOQs for each method were as follows: pH < 0.1; OM, TC, and TOC < 0.01 wt%; H, N, and S < 0.01 wt%; Hg < 0.0005 mg kg$^{-1}$.

**Ethical approval:** The conducted research is not related to either human or animal use.

### 4 Results and discussion

#### 4.1 Foundry waste

The basic characteristics of the analysed waste are presented in Table 1. The waste was characterized by a neutral and moderately alkaline pH with a mean $\text{pH}_{\text{H2O}}$ of 7.9 (range 7.3–8.5). Similarly, Dayton et al. found a neutral pH of leachate from foundry waste [6]. It is known that the pH value affects the leaching of contaminants from foundry waste; a low pH increases heavy metal mobility [5,20]. In the current study, mean OM and TOC contents in waste were 4 and 2.2 wt%, respectively. The main component of OM in foundry waste is organic binders which are present in spent casting moulds after the metal casting process. OM in SFS may contain toxic substances, for example, phenol, formaldehyde directly from phenol-formaldehyde binders or polycyclic aromatic hydrocarbons generated from organic binders after contact with liquid metal at a high temperature [6,11]. Among the analysed elements from elemental analysis (C, H, N, S), the highest percentage of carbon was found. The percentage of other elements (HNS) was low (0.04–0.09 wt%) (Table 1).

In the case of sulphur, its forms that dissolve in water, i.e. sulphates, may be a problem. An excessively high concentration of sulphates in the eluates may disqualify the waste from being recovered [28,29]. However, the percentage of sulphur in the examined waste was small (mean 0.09%); therefore, it may be assumed that the waste will not pose a threat to the environment.

#### 4.2 Fractional analysis, particle size of waste

The percentage of every fraction in the analysed waste is shown in Figure 4. A wide range of results was obtained for most of the fractions. This proves the heterogeneity of the landfilled material. Mass-wise, the highest percentages were found for the largest fraction above 5.6 mm and for fractions with a particle size between 0.2 and 0.6 mm. The largest fraction (>5.6 mm) consisted of metallic elements, sinter, slag and others. The percentage of other fractions did not exceed 5%. The percentage of the clay fraction (<0.2 mm) was low in the tested waste. This fraction is responsible for the binding properties of waste, which is important for applications in construction, but less so as an aggregate of foundry waste. The fractional analysis showed that the waste consisted mainly of sand fractions (<2 mm). Dayton et al. [6] obtained similar results. On the other hand,
Siddique et al. [16] stated that the main fraction of foundry waste (85–95%) are particles of size between 0.15 and 0.6 mm, while the fine fraction (<0.075 mm) can reach up to 12%. The diversity of the composition and its specific structure makes it possible to use foundry waste in agriculture. The use of SFS in agriculture and horticulture is popular in countries such as Brazil, the United States and South Africa [5,6,16]. In 2007, the US EPA confirmed the benefits of using sands from iron and aluminium foundries in agricultural, horticultural and geotechnical applications [35]. Several US states have allowed the use of SFS for the production of artificial horticultural soils and Technosols [36,37].

4.3 Mercury and OM content in the waste fraction

The mercury content in most fractions was similar and fell within the range of 0.023–0.031 mg kg\(^{-1}\) DM (Figure 5). The highest Hg content was found in the finest fraction (<0.2 mm), on average 0.065 mg kg\(^{-1}\) DM. The landfilled waste contained dust from dust collectors, which may have been the main component of the finest fraction. Foundry dusts, especially electric arc furnace dust (EAFD) is characterized by a high content of heavy metals including Hg compared to SFS [38–41]. Therefore, it may be assumed that the source of mercury in the finest fraction of the tested waste was foundry dusts with a high Hg content. Another explanation for this phenomenon may be the association of Hg with the loamy matter of this waste, just like in soils. Similarly, Klojzy-

Klojzy-Karczmarczyk and Mazurek [26,28,30] and Kicińska [42] found the highest content of mercury and other heavy metals in the finer fraction of the mining waste, which was the effect of association with OM and the loamy material of this waste.

Figure 6 presents the percentage of OM in all fractions of the tested waste. The highest percentage of OM was found in the smallest fraction <0.2 mm (median 4.8%). In subsequent fractions, the percentage of OM decreased to 2.3% (fraction >0.6 mm), then increased to 4.5% for fractions >2.8 mm. The highest differences in the results were obtained for the largest fraction (>5.6 mm); OM varied between 3 and 15.5 wt%. This fraction was non-heterogeneous and consisted of various materials,
hence the OM value differences. The composition of OM in foundry waste depends on the type of organic binders used in the production of foundry moulds. Binders may remain on sand particles after metal casting or may be ground off when separating the mould from the casting during shock grating or during mechanical regeneration of SFS [43].

### 4.4 Correlations analysis

A positive correlation means that both variables vary in the same way, when one increases the other increases. For a negative value of the correlation coefficient, the relationship between the variables is inverse. In the current study, no statistically significant correlation coefficients for the relationship between the Hg content and other parameters such as particle size – fractions, OM, TOC and TC were found. Therefore, it can be concluded that the mercury content did not depend on these parameters, unlike the soil. While a relationship between particle size and OM and carbon compounds (TC and TOC) in these waste was found. A low positive correlation between these parameters was obtained. The expected result was a high positive correlation for the relationship between OM, TOC and TC in the tested waste.

### 4.5 Local soil characteristic

This section presents the test results of soil around the foundry landfill. The aim was to compare the results of mercury content in waste with local soils being used as a natural reference. The physicochemical characteristics of the soil samples are presented in Table 2. An analysis of particle size shows that the soils of the studied area were light soils. The pH value in the analysed soils varied between pH KCl 5.8 and 7.5. The soils in the Opole region, as in Poland, are moderately acidic or acidic soils. The SOM content in the soils ranged from 1.77 to 5.25% and the TOC content from 0.59 to 2.14%. For comparison, the content of SOM is on average 1.94% (range 0.62–6.62%) and TOC 1.12% (range 0.36–3.84%) in Polish soils (https://www.gios.gov.pl/). The analysed soils were characterized by a moderate content of these components. In order to assess the degree of soil contamination of heavy metals, soil type, pH and TOC or SOM content is usually taken into account. Polish Regulation [32] considers Hg in soils depending on the soil group according to the purpose of their use. The permitted mercury content in Polish soils and the classification of the tested soils are also shown in Table 2.

Current research shows that the mercury content in the soils of the studied area was very low and similar to its content in the tested waste. The mercury content in the soils studied was significantly lower than the limit values [32]. The Hg content in most soil samples was below 0.05 mg kg\(^{-1}\) DM, which is the average level for Polish soils [19,24]. Only a few samples were characterized at a slightly higher Hg content. In some regions of Poland, the Hg content in soils is increased due to industrial activities [22,24], along transport routes [28], in areas adjacent to a coal-fired power plant [26]. Mercury in the soil is known to have a strong affinity with SOM, mainly thiol and clay minerals, as it binds to colloids because of its large specific surface area and the presence of surface functional groups [19,21,29]. For this reason, the correlation between SOM, TOC and Hg content in the tested soils was determined (Table 3). A positive correlation was found between these components in the soil.

### Table 2: Characteristics of soil samples

| Samples                         | Arable land and meadows | Allotment gardens | Forest soil | Wasteland |
|---------------------------------|-------------------------|-------------------|-------------|-----------|
| Types of land use\(^a\)         | I – II                  | II – II           | III         | IV        |
| Hg (mg kg\(^{-1}\) DM)          | 0.018–0.092             | 0.038–0.056       | 0.017–0.019 | 0.028–0.064|
| Limit Hg content\(^b\) (mg kg\(^{-1}\) DM) | 2                       | 4                 | 10          | 30        |
| Granulometric composition       | Sand/loamy sand         |                   |             | Sand      |
| pH KCl                          | 6.0–6.7                 | 6.6–6.9           | 6.8–7.5     | 5.8–6.7   |
| TOC (wt%)                       | 0.72–2.14               | 1.25–1.71         | 0.59–0.71   | 0.71–1.82 |
| SOM (wt%)                       | 2.08–5.25               | 2.96–3.62         | 1.77–2.03   | 1.83–5.11 |

\(^a\)According to Polish classification (Journal of Law 2016 item. 1395), Group: I – arable land, orchards, meadows, pastures, allotment gardens, protected areas; II – forests, wooded areas; IV – industrial areas and traffic areas; II-1 and II-2 – subgroups according to pH and SOM values.
5 Conclusions

- The mercury content in the LFW was very low, similar to the natural content in soils. Obtained results show that the tested foundry waste was not contaminated with mercury, which should not negatively affect the environment during its landfilling or its reuse. An environmental problem with the use of LFW may be the high content of other heavy metals or toxic organic substances. The studies showed that the assessment of the mercury content in LFW, containing mainly SFS, may be omitted due to the very low content of this metal. However, when using LFW containing a high percentage of foundry dust, e.g. EAFD, it may pose an environmental problem, as shown in previous studies.

- Fractional analysis showed that the tested waste was characterized by the highest percentage of sand fraction (<2 mm). The percentage of the finest fraction (clay fraction), which is responsible for the binding properties, an important parameter for building materials, was low. A high percentage of the largest fraction (>5.6 mm) consisted of metallic inclusions, sinter, slag and other materials was found. This fraction was also characterized by the largest differences in the content of OM due to the diversity of its composition. The highest percentage of OM for the finest (<0.2 mm) and the largest (>5.6 mm) fraction was found.

- Statistical analysis showed no statistically significant correlation between the Hg content and other parameters of the tested waste. Low positive correlations between OM, TOC, TC and the particle size of the LFW were determined. In contrast to waste, a positive correlation between mercury content and SOM and TOC for the soil samples was found. This is probably due to the difference in the type of organic substances in waste and soil.

Abbreviations

- CHNS carbon, hydrogen, nitrogen, sulphur
- CRM certified reference material
- EAFD electric arc furnace dust
- LFW landfilled foundry waste
- LOQ limit of quantification
- OM organic matter
- SFS spent foundry sand
- SOM soil organic matter
- TC total carbon
- TOC total organic carbon

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Data availability statement: The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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