



Mercury in dumped blast furnace sludge

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HIGHLIGHTS

- Blast furnace sludge (BFS) has been analyzed for Hg for the first time.
- The Hg content of BFS varied between 0.006 and 20.8 mg kg⁻¹ (median 1.64 mg kg⁻¹).
- In comparison to the charge material, Hg was enriched in BFS.
- Hg correlated with non-calcareous carbon content (coke and graphite).

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ABSTRACT

Blast furnace sludge (BFS) is a waste generated in the production of pig iron and was dumped in sedimentation ponds. Sixty-five samples from seven BFS locations in Europe were investigated regarding the toxic element mercury (Hg) for the first time. The charge material of the blast furnace operations revealed Hg contents from 0.015 to 0.097 mg kg⁻¹. In comparison, the Hg content of BFS varied between 0.006 and 20.8 mg kg⁻¹ with a median of 1.63 mg kg⁻¹, which indicates enrichment with Hg. For one site with a larger sample set ($n = 31$), Hg showed a stronger correlation with the total non-calcareous carbon (C) including coke and graphite ($r = 0.695$; $n = 31$; $p < 0.001$). It can be assumed that these C-rich compounds are hosting phases for Hg. The solubility of Hg was rather low and did not exceed 0.43% of total Hg. The correlation between the total Hg concentration and total amount of NH₄NO₃-soluble Hg was relatively poor ($r = 0.496$; $n = 27$; $p = 0.008$) indicating varying hazard potentials of the different BFS. Finally, BFS is a mercury-containing waste and dumped BFS should be regarded as potentially mercury-contaminated sites.

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1. Introduction

Blast furnace sludge (BFS) is a waste generated in the production of pig iron and was dumped in large surface landfills in industrial areas until the commencement of strict environmental laws in the 1960s and 1970s in Europe. During the late 1990s, 6 kg of BFS were generated per ton of pig iron produced (Lopez-Delgado et al., 1998), resulting in 6,300,000 Mg of BFS produced in the European Union alone during the 1990s (World Steel Association, 2013). As these wastes often contain harmful substances, significant hazards to environmental surroundings may arise from former BFS sedimentation ponds.

Pig iron is commonly produced in blast furnaces by smelting several iron (Fe) ores with a high carbon (C) fuel such as coke and flux additives (limestone etc.). Extraction of Fe from its ores and its conversion to alloys is the most important metallurgical

process (Coudurier et al., 1985). During the operation, a gaseous phase (top gas) leaves the top of the blast furnace. Besides the gaseous phases, it contains dragged solid phases such as coke, Fe ores, additives, and their reaction products. For the downstream use, the effluent gas was purified from the dust (30 kg of dust per ton of pig iron produced (Mansfeldt and Dohrmann, 2004)) long before any environmental laws were enacted. As a result of wet purification, a muddy waste referred to as BFS was generated. Besides Fe and C, other elements are also introduced into the blast furnace. Mansfeldt and Dohrmann (2004) investigated 32 samples from an abandoned BFS landfill in the Ruhr area of Germany, specifically examining their elemental composition and identifying four main categories: (i) Fe and C with a median content of >100 g kg⁻¹; (ii) elements such as lead (Pb), magnesium (Mg), zinc (Zn), aluminum (Al), silicon (Si), and calcium (Ca), with contents ranging from 10 to 100 g kg⁻¹ in ascending order; (iii) potassium (K), sulfur (S), manganese (Mn), nitrogen (N), phosphorous (P), and sodium (Na), with contents ranging from 1 to 10 g kg⁻¹; and (iv) minor elements with mean contents <1 g kg⁻¹, such as arsenic (As), cadmium (Cd), and

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many more. All elements were introduced into the process by either Fe ores or coke, and to a lesser extent by flux, in which these elements occur partly as impurities. Due to the presence of significant amounts of As, Cd, Pb, Zn, and cyanides, which are formed during the blast furnace operation, BFS can be regarded as a harmful waste (Trömel and Zischkale, 1971; Van Herck et al., 2000; Mansfeldt and Dohrmann, 2001, 2004; Trung et al., 2011).

Mercury (Hg) is considered as one of the most important environmental pollutants as the element and many of its compounds are highly toxic, persistent, and readily released into the environment due to its high mobility and volatility (WHO, 2005). Considering the enrichment with elements such as Zn, Pb, Na, and K, it is postulated that BFS is enriched with the highly volatile transition metal Hg as well. AMAP/UNEP (2008) estimated a global inventory of Hg to atmosphere to be 1921 Mg. In their Global Mercury Assessment report, the UNEP (2013) stated that the primary production of ferrous metals contributes 45.5 Mg of Hg to the atmosphere being 2% of the global anthropogenic emissions. To the best of our knowledge, no data on Hg in BFS have been published yet, so emissions from BFS are most likely missing in the global inventory.

This study aimed at the determination of (i) the total Hg content and (ii) the easily mobilized fraction of Hg in BFS samples from different locations in Europe. Furthermore, (iii) the Hg content of some charge materials was investigated in order to estimate a possible enrichment with Hg. Overall, the aim of the study is to provide first insights about Hg in BFS and in addition to that to provide primary information about potential Hg emissions for a future global inventory of Hg including these emissions. Overall, this study provides first insights about Hg in BFS and contributes primary information to the global inventory of Hg regarding this waste.

2. Materials and methods

2.1. Sampling sites, sampling, and sample preparation

Samples were obtained from six former BFS sedimentation ponds and one operating BFS deposit in Europe: Herne and Dinslaken (operating) in the Ruhr area (Germany), Eisenhüttenstadt (Germany), Lübeck (Germany), Nowa Huta (Krakow, Poland), Esch-Belval (Luxembourg), and Nancy (France).



Fig. 2. View of former blast furnace sludge sedimentation ponds in Esch-Belval (Luxembourg).

Esch-Belval (Luxembourg), and Nancy (France) (Figs. 1 and 2). Sampling was performed in three ways: first, near-surface samples (0–10 cm) were taken (Lübeck, Eisenhüttenstadt, Nowa Huta, Esch-Belval), and second, samples were taken at different depths from fresh pits up to 1.0 m depth (Herne, Nancy). Additionally, core samples from 3.9 to 12 m depth were obtained (Herne). In total, 65 samples (42 from Herne, 1 from Dinslaken, 4 from Eisenhüttenstadt, 4 from Lübeck, 3 from Nowa Huta, 6 from Esch-Belval, 6 from Nancy) were collected. The field-moist samples were dried at room temperature, sieved to a size fraction <2 mm, and manually homogenized. No material >2 mm was present. For the analysis of element contents, subsamples were ground to analytical grain size in an agate ball mill (PM 400, Retsch). Furthermore, an exemplarily loading of a blast furnace was investigated for its Hg content: 3 coke samples from Poland ($n = 1$) and Germany ($n = 2$), 1 Fe ore sample from South Africa, 1 Fe ore pellet sample from Canada, 1 sinter product sample, and 4 additive samples (olivine, bauxite, gravel, ilmenite). The samples were dried at room temperature and pre-ground by a jaw crusher (BB1, Retsch) before

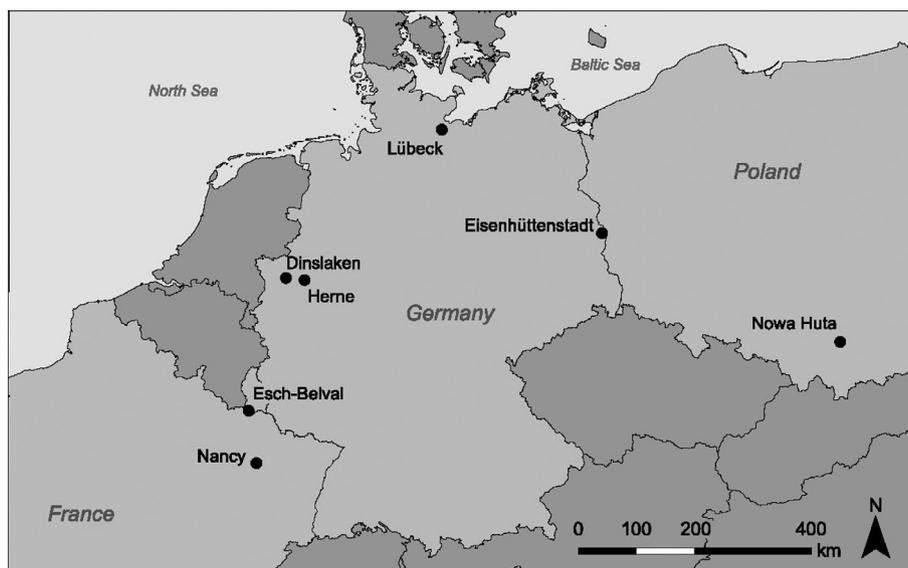


Fig. 1. Sampling sites in Europe.

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