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Abstract: Around former glass factories in south eastern Sweden, there are dozens of dumps whose radioactivity and physico-chemical properties were not investigated previously. Thus, radiometric and physico-chemical characteristics of waste at Madesjö glass dump were studied to evaluate pre-recycling storage requirements and potential radiological and environmental risks. The material was sieved, hand-sorted, leached and scanned with X-Ray Fluorescence (XRF). External dose rates and activity concentrations of Naturally Occurring Radioactive Materials from 238U, 232Th series and 40K were also measured coupled with a radiological risk assessment. Results showed that the waste was 95% glass and dominated by fine fractions (< 11.3 mm) at 43.6%. The fine fraction had pH 7.8, 2.6% moisture content, 123 mg kg-1 Total Dissolved Solids, 37.2 mg kg-1 Dissolved Organic Carbon and 10.5 mg kg-1 fluorides. Compared with Swedish EPA guidelines, the elements As, Cd, Pb and Zn were in hazardous concentrations while Pb leached more than the limits for inert and non-hazardous wastes. With 40K activity concentration up to 3000 Bq kg-1, enhanced external dose rates of 40K were established (0.20 □Sv h-1) although no radiological risk was found since both External Hazard Index (Hex) and Gamma Index (Iγ) were < 1. The glass dump needs remediation and storage of the waste materials under a safe hazardous waste class 'Bank Account' storage cell as a secondary resource for potential future recycling.

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30th April, 2019

From : Richard Nasilele Mutafela (MSc), Linnaeus University

To : Jacob de Boer & Shane Snyder

Dear Editors-in-Chief,

Re: Manuscript submission to Chemosphere

On behalf of the authors (Richard N. Mutafela, Juan Mantero, Yahya Jani, Rimon Thomas, Elis Holm and William Hogland), I hereby submit the manuscript entitled ***'Radiometrical and Physico-chemical characterisation of contaminated glass waste from a glass dump in Sweden'*** for your consideration. As authors, we all agree to the submission and declare that the work is original and has not been published in any other journal nor is it under consideration elsewhere.

The investigation focuses on studying the physico-chemical and radioactive potential of wastes including glasses and fine fraction sampled from an old artistic glasswork dumpsite. The characterisation involves waste composition, trace element contents, particle size distribution, trace element leaching and external dose exposure to radionuclides from the waste, among others. It aims to serve as an advisory step in decision-making against such landfilling, and also promotes circular economy application through materials recovery from the excavated wastes as opposed to further landfilling them.

We strongly believe that by publishing this article in Chemosphere, readers would be enlightened about the pollution status of such sites, but also the need to consider potential for future recycling of waste as secondary resources.

Yours sincerely

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Radiometrical and Physico-chemical Characterisation of Contaminated Glass Waste from a Glass Dump in Sweden

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***Highlights (3 to 5 bullet points (maximum 85 characters including spaces per bullet point)**

Manuscript Highlights

- Radiometric and physico-chemical characteristics of glass waste were investigated.
- As, Cd, Pb and Zn were in hazardous concentrations according to Swedish EPA limits.
- Pb leached more than limits for inert and non-hazardous waste unlike As, Cd and Zn.
- External dose rates and gamma measurements showed enhanced levels of ^{40}K.
- External Hazard index and Gamma Index indicated no radiological exposure risks.

Radiometrical and Physico-chemical Characterisation of Contaminated Glass Waste from a Glass Dump in Sweden

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Abstract

Around former glass factories in south eastern Sweden, there are dozens of dumps whose radioactivity and physico-chemical properties were not investigated previously. Thus, radiometric and physico-chemical characteristics of waste at Madesjö glass dump were studied to evaluate pre-recycling storage requirements and potential radiological and environmental risks. The material was sieved, hand-sorted, leached and scanned with X-Ray Fluorescence (XRF). External dose rates and activity concentrations of Naturally Occurring Radioactive Materials from ^{238}U, ^{232}Th series and ^{40}K were also measured coupled with a radiological risk assessment. Results showed that the waste was 95% glass and dominated by fine fractions (< 11.3 mm) at 43.6%. The fine fraction had pH 7.8, 2.6% moisture content, 123 mg kg^{-1} Total Dissolved Solids, 37.2 mg kg^{-1} Dissolved Organic Carbon and 10.5 mg kg^{-1} fluorides. Compared with Swedish EPA guidelines, the elements As, Cd, Pb and Zn were in hazardous concentrations while Pb leached more than the limits for inert and non-hazardous wastes. With ^{40}K activity concentration up to 3000 Bq kg^{-1}, enhanced external dose rates of ^{40}K were established (0.20 μSv h^{-1}) although no radiological risk was found since both External Hazard Index (H_{ex}) and Gamma Index (I_{γ}) were < 1. The glass dump needs remediation and storage of the waste materials under a safe hazardous waste class *'Bank Account'* storage cell as a secondary resource for potential future recycling.

Key words

Glass waste, physico-chemical characterisation, fine fraction, NORM, dose rates, risk assessment

1. Introduction

Due to availability of dumping space and lack of disposal tariffs in the past, landfills and dumpsites were previously considered as one of the cheapest waste management methods globally. Currently, there are over 500,000 landfills in the European Union (EU) alone, most of which are located in semi-urban environments, and 90% are 'non-sanitary' and lack environmental protection measures as they predate the famous 1999 EU Landfill Directive (Nguyen et al., 2018). Sweden, for instance, currently has over 6,000 landfills, both closed and operational, covering over 8000 m^2 of land (Hogland et al., 2010; Mönkäre et al., 2016). Among these are over 40 highly contaminated glass waste dumpsites from decades of crystal glass production in the south eastern part of the country. Most of the sites have been classified as high risk for human health and the environment (Jani & Hogland, 2017). Previously, potential environmental risks of raw materials received little attention, resulting in the use of toxic and radioactive materials in different industrial processes. In crystal glass production, different metal oxides conferred specific properties to the glass depending on a metal's properties (Cesaro et al., 2018). Uraninites and uranium oxides, for instance, were historically used for colouring glass since the 19th century (Strahan, 2001). Use of natural uranium in glass production was later discontinued globally due to its strategic importance during the Second World War, instead depleted uranium was used (Lopes et al., 2008). However, the actual period of discontinuation is uncertain due to glass manufacturers' secrecy regarding their techniques (Brenni, 2007).

Different studies on uranium (U) and potassium (K) containing glasses have indicated weight contents up to 1.4% U and 16.2% K (Kierzek et al., 2000; Lopes et al., 2008; Strahan, 2001). Consideration of such materials from the dosimetric standpoint is essential since radiation may act as a mutual initiator of tumorigenesis even at low doses (UNSCEAR, 2000). Regarding the element K, its radioactive isotope ^{40}K is a beta (1.3 MeV) and gamma (1.46 MeV) emitter. Thus, for an open crystal glass waste dump like Madesjö in this study, a radiometrical investigation is vital. On the other hand

and in terms of leachable elements like As, Cd and Pb, severe environmental contamination has been associated with open dumping practices (Cesaro et al., 2018) due to contaminants leaching to ground water and other aquatic systems as well as through dust emissions (Larsson et al., 1999). Around former Swedish glass factories in particular heavy metal uptake by home-grown vegetables has been reported (Augustsson et al., 2015) while oral intake of locally produced feedstuffs has been described as the main exposure route for residents (Nyqvist et al., 2017) besides exposure through dust emissions (Larsson et al., 1999).

While hazardous glass waste dumps require remediation, it is essential to consider remediation aspects together with potential recycling alternatives of the waste, since the shift towards a more resource efficient circular economy is increasingly becoming inevitable due to resource shortages and other global environmental challenges (Burlakovs et al., 2017). As primary raw material stocks diminish, landfills and dumpsites could be potential sources of secondary raw materials (Machiels et al., 2016; Masi et al., 2014). Both closed-loop and open-loop recycling could be achieved for old glass waste. The closed-loop option would involve use of the glass waste in manufacture of new crystal glass as well as other new glass items such as art and polymeric glasses. Heriyanto et al. (2018), for instance, used broken glass as a primary input in the production of polymeric glass composites with mechanical properties, utility, aesthetic appeal and expected market value similar to natural and engineered stone products widely used as kitchen/bathroom benchtops or floor/wall tiles. The open-loop option, on the other hand, would involve use of the glass in other applications such as in cement mortar, concrete and bricks (Lee et al., 2018; Njindam et al., 2018; Youssef et al., 1998). Nowadays, sustainable construction with focus on reduced raw materials consumption, or their replacement with secondary raw materials, has received much attention. As a secondary resource, glass has been widely studied for reusing as building material aggregates. Lee et al. (2018) proved that concrete with 20% partial replacement of cement with glass could provide longer durability and enhanced mechanical and microstructural properties. Njindam et al. (2018) also showed improved physico-mechanical

properties in produced tiles after using waste glass powder (up to 30%) and clay mixture in the production process.

However, most recycling alternatives require prior decontamination of the waste to acceptable limits of trace elements. Decontamination potential of crystal glass waste has been previously achieved through reduction melting at 99%, 99.9% and 100% efficiency for As, Pb and Cd respectively (Jani & Hogland, 2017), indicating potential for efficient metals recovery for use as secondary raw materials. Therefore, this study focused on assessment of leaching potential and other physico-chemical characteristics of the glass waste to evaluate storage and recycling requirements of the waste. The study also focused on radiometric characterisation of the waste to assess the material's radiological risks to humans and the environment when recycled. Finally, the study discusses management aspects of crystal glass waste in similar unprotected dumps in order to provide support for decision-makers in their management.

2. Materials and methods

2.1. Site Description

The study was conducted at Madesjö open glass dump (56°44′45.9″N 15°52′13.9″E) which is situated in Nybro Municipality, south-eastern Sweden, as shown in Figure 1. Initially in 1960, it was used as a municipal solid waste (MSW) dumpsite for household waste until 1969. Later, glass factory waste began to be temporarily dumped there for later use in a road construction project. The dump is located in a forest area about 200 m away from the nearest residential area and covers an estimated surface area of 38,500 m^2. It has been given a first-class risk status by the county's administrative board (Kalmar County Administrative Board, 2017).

Figure 1. Madesjö dumpsite, Nybro, Sweden. (Map data ©2019 Google)

2.2. Excavation and sampling

2.2.1. Sampling for radiometric measurements

For radiometric measurements, several kilogrammes of glass samples were collected throughout the site and sorted by colour. The colour-based sorting generated six different 1 kg mass fractions of yellow, green, black, blue, white and red glass samples. This set of samples was milled, sieved and homogenised. After homogenization, a fraction of 100 g was packed in a sample container and vacuum sealed to avoid Rn emanation. Gamma measurements were performed after 3 weeks of storage to ensure secular equilibrium between ^{226}Ra and its decay daughters ^{214}Bi and ^{214}Pb.

2.2.2. Excavation and sampling for physico-chemical characteristics

Samples for physico-chemical analyses were collected through excavation of test pits (TPs) using a 5-tonne excavator. The excavation methodology for TPs was adopted from Kaczala et al. (2017) and involved excavation of 1 m deep TPs. Eight TPs were excavated, each with two material stockpiles from 0-0.5 m and 0.5-1 m depths. Samples were collected from the 0.5-1 m depth stockpile according to the Nordtest Method NT ENVIR 004-1996/05. About 70 kg samples were collected per TP and transported to the laboratory for analysis. At the laboratory, samples for leaching tests were temporarily stored at 4 °C and were later analysed in triplicates per TP, resulting in a total of 24 samples.

2.3. Analyses

2.3.1. Waste composition and particle size distribution (PSD)

The samples were initially weighed and then sieved using a laboratory sieve shaker (Pascall Engineering) and five sieves (Tidbecks Sweden and Giuliani Technologies). From sieve mesh sizes 63 mm, 31.5 mm, 20 mm, 11.3 mm and 4 mm, the resulting particle sizes were classified as course (> 31.5 mm), medium (11.3-31.5 mm) and fine (< 11.3 mm) fractions hereafter in the study referred to as CF, MF and FF respectively. After each complete shaking and sieving round, the materials were weighed

according to particle sizes. This was followed by hand-sorting of the weighed materials into glass, demolition (concrete/cementitious/ceramic material), organic, soil/stones and residual (metals, plastics, etc) fractions per particle size to analyse composition. The hand-sorted materials were also weighed to determine their individual mass contributions to each particle size and ultimately to the whole sample.

2.3.2. Leaching tests

The Swedish Standard method for characterisation of waste (SS-EN 12457-4) was used to leach the FF of the eight samples in triplicates with modified particle size (< 11.3 mm) instead of the standard < 10 mm due to unavailability of 10 mm sieve size. The FF was leached at room temperature with deionized water (18.2 MΩ cm^{-2} Milli-Q™ water) in 2L bottles according to the standard liquid to solid ratio (10 lkg^{-1}). The bottles were steadily agitated at 10 rpm for 24 h (± 30 min) using a Heidolph Reax 20 overhead shaker. After agitation, the bottle contents were allowed to settle, then sieved with 0.45 µm sieves and finally analysed for trace elements content using an Inductively Coupled Plasma Mass Spectrometer (ICP-MS).

2.3.3. Physico-chemical analyses

Trace element contents, moisture content (MC), pH, Dissolved Organic Carbon (DOC), Fluorides (F^-) and Total Dissolved Solids (TDS) were also analysed. Trace elements in solid were analysed in triplicates using an Olympus DS-4000 (Innov-X) which is a portable X-Ray Fluorescence (XRF) analyser. The Swedish Standard SS-EN 14346:2007 was adopted for MC determination, whereas a portable pH meter Radiometer PHM 210 was used for measuring pH. DOC and F^- were analysed by the use and digestion of DR Lange cuvettes in a DR Lange digester HT200S with ultimate analysis of the cuvettes in a DR Lange Spectrophotometer DR 5000. TDS was determined according to the Gravimetric Method 8163 by Hach Lange.

186 2.3.4. Radiometric measurements

187 External dose rates were measured using two detectors; a handheld detector Automess 6150AD
188 connected with a scintillator probe (Range 100 nSv h^{-1} – 10 mSv h^{-1}) for precise measurements and a
189 mobile detector system consisting of a Lanthanum-Bromide crystal. The latter was connected to a GPS
190 receiver and to a laptop with software (courtesy of Swedish Radiation Safety Authority) to create dose
191 maps where the dose rates calculated from the detector were coupled with GPS data.

192

193 Gamma measurements were performed in an extended range Germanium coaxial detector (XtRa) of
194 37.1% relative efficiency and 1.76 keV resolution. The detector system has a 10 cm passive shielding of
195 old Pb and an active shielding made with an organic scintillation detector (Bicron BC-418) that
196 operates in anti-coincident mode with the Germanium detector, resulting in very low background
197 level (Hurtado et al., 2006). In a 200,000 seconds measurement, the system can provide a Minimum
198 Detectable Activity (MDA) of 0.55 Bq for ^{210}Pb (45.6 keV), 0.38 Bq for ^{214}Pb (351.9 keV), 0.30 Bq for ^{214}Bi
199 (609.3 keV) in the ^{238}U series and 1.62 Bq for ^{40}K (1461 keV). Regarding radionuclides in the ^{232}Th series,
200 MDA ranged from 0.5 to 1.5 Bq for ^{212}Pb (238.6 keV), ^{212}Bi (727.3 keV), ^{228}Ac (911.2 keV) and ^{208}Tl (583.2
201 keV). Quality assurance in gamma spectrometry is checked periodically through participation in
202 national and international proficiency tests.

203

204 2.3.5. Radiological indices

205 In case some materials are in close interaction with humans, such as in building materials, different
206 indices are used to classify the materials from the radiological viewpoint: Radium equivalence (Ra_{eq}),
207 External Hazard Index (H_{ex}), Internal Hazard Index (H_{in}), or Gamma Index (I_{γ}) (UNSCEAR, 2000).
208 These indices are based on ^{226}Ra, ^{232}Th and ^{40}K activity concentrations and were used to evaluate the
209 radiological risks of recycling this glass in construction industry applications (concrete, tiles, kitchen
210 and bathroom benchtops, etc). The external radiation hazard due to glasses used as building material
211 can be assessed according to Equation 1:

$$H_{ex} = (C_{Ra}/370) + (C_{Th}/259) + (C_K/4810) \quad (1)$$

Gamma index (I_γ) is also used to evaluate the γ-radiation hazard and follows Equation 2:

$$I_\gamma = C_{Ra}/300 + C_{Th}/200 + C_K/3000 \quad (2)$$

In both Equations 1 and 2, C_{Ra}, C_{Th} and C_K are the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1} respectively. It is recommended that both the H_{ex} and the I_γ be < 1 (UNSCEAR, 2000). Of the two, H_{ex} is more conservative than I_γ. For instance, assuming negligible amounts of Ra and Th, 4810 Bq/kg of ^{40}K are needed to produce $H_{ex} = 1$, while only 3000 Bq of ^{40}K will produce $I_\gamma = 1$. In case $I_\gamma \geq 1$, further studies should be performed depending on how the material is going to be used in order to assess the final dose received by a population.

2.3.6. Statistical analysis

Using GraphPad Prism version 7.0c for Mac (GraphPad Software Inc.), descriptive statistics (minimum, maximum, mean and standard deviation) was calculated $p < 0.05$. For comparisons among different datasets, further statistical analyses involved One-way ANOVA and Tukey's multiple comparison tests. In addition, the data was validated using Grubbs test for outliers ($\alpha = 0.05$) where necessary.

3. Results and discussions

3.1. Physico-chemical characterisation

3.1.1. Waste composition & particle size distribution

Composition of the sampled waste based on the three particle size categories was obtained as shown in Figure 2. Overall, the waste was dominated by glass (95.4%) followed by demolition (3.5%), organic

(0.9%), soil (0.1%) and residual (0.003%) fractions. Demolition fraction decreased with decrease in particle size while soil fraction increased with decrease in particle size. Given the high surface-to-volume ratio in the FF, there is more soil attached to other material fractions which makes them harder to distinguish in the soil-like complex mix. According to a One-way ANOVA ($p < 0.05$) and Tukey's multiple comparisons test, the difference between the glass fraction and other fractions was statistically significant in all three cases. Similarly, the difference between the demolition fraction and other fractions was statistically significant, whereas there were no statistically significant differences among the organic, soil and residual fractions. These results could not be compared with other studies as this is one of the first studies focussing on physico-chemical and radiometric characterisation of glass waste dumpsites. Previous studies on Swedish glass dumpsites have mainly focused on toxicity assessments and remediation aspects (Augustsson et al., 2016; Augustsson et al., 2015; Hagner et al., 2018; Uddh-Söderberg et al., 2015) .

Figure 2. Semi-log graph of abundance of waste fractions grouped according to the three particle size categories ($n = 8$)

As shown in Figure 3, the FF (43.6%) was dominating followed by CF (34.3%) and MF (22.1%). This order of dominance is in line with most studies that have established the biggest amount of materials as belonging to the FF, then CF and lastly MF (Parrodi et al., 2018). The FF is in the same order of magnitude as 38% and 44% observed in two previous studies (Jani et al., 2016; Quaghebeur et al., 2013), although it could be as high as 70.4% (Masi et al., 2014). The MF was lower than the 38% observed by Jani et al. (2016) but similar to the range of 21.8%-31.4% observed by Hogland et al. (2004). On the other hand, the CF was higher than the 24% by Jani et al. (2016) but lower than the 53% by Hogland et al. (2004).

263 The similarities and differences observed in the results might, however, be misleading since different
264 studies set particle sizes for their sieving processes differently (Parrodi et al., 2018). Other factors also
265 play crucial roles in the particle size of excavated waste including waste age, waste type and sampling
266 or excavation procedure (Jani et al., 2016). Further, the inherent heterogeneity of landfills and
267 dumpsites in general is also a vital influencing factor. The high abundance of FF could be attributed to
268 frequent exploratory sampling operations on one hand, since waste-machine interface results in
269 reduced particle sizes. On the other hand, the waste age (half a century in this study) could be
270 responsible since particle sizes decrease overtime due to weathering, biodegradation (in the case of
271 MSW) and mechanical disturbances, thereby increasing the amount of fines (Parrodi et al., 2018). The
272 high standard deviations shown in Figure 3 could be attributed to high heterogeneity in particle sizes
273 across the site, especially in the CF, since single large particles have quite a large influence on the total
274 weight of a particle size category (Kaartinen et al., 2013).
275
276 Waste composition and particle size distribution are important parameters to consider when
277 identifying end-user applications from the circular economy standpoint. For instance, the use of waste
278 glass as a building material aggregate depends on its particle size, where increase in glass particle size
279 increases concrete bleeding and segregation, and decrease in particle size reduces hardened density of
280 concrete blocks (Ling & Poon, 2011; Rashad, 2014). Thus, the particle size should suit each
281 application's specific size requirements. Concerning composition on the other hand, demolition waste
282 has been reported to be one of the most problematic impurities in the thermal extraction of trace
283 elements from waste glass due to its relative resistance to heat (Drumond, 2011), while mixed colour
284 waste glass at certain replacement levels of natural sand in concrete leads to severe concrete bleeding
285 and segregation (Taha & Nounu, 2009). Therefore, these parameters also influence the choice of waste
286 separation techniques for efficient recovery of the desired fractions. Demolition waste is, however,
287 quite low in these findings.
288

289 **Figure 3.** Particle sizes of waste fractions grouped into three particle size categories (n = 8)

290

291 3.1.2. Trace elements content in solid phase (XRF)

292 Averages of elemental concentrations from all eight samples across the glass dump are compared with
293 findings from a previous study (Jani & Hogland, 2017) on crystal glass waste shown in Table 1. The
294 average Pb results in the current study are notably lower than from the previous study. This is
295 because in the previous study crystal glass was sorted and analysed as individual pieces while in the
296 current study glass samples were analysed as they are in the dump (i.e. without sorting the different
297 glass types), thus resulting in a dilution effect on the detected concentration of Pb.

298

299 **Table 1.** A comparison of average trace element concentrations (mean ± standard deviation) of the
300 samples across the dump with that from a previous study based on XRF analyses (Jani & Hogland,
301 2017).

302

303 Total trace element concentrations in solid phase among the eight samples were detected as shown in
304 Table 2. Concentration was in the order Pb > As > Sb > Zn > Ba > Cu > Cd. Some of the elements were
305 in hazardous concentrations according to Swedish EPA limits (Swedish EPA, 2009), the highest in
306 magnitude over the limits being Pb followed by As, Zn and Cd respectively. Contrariwise, Ba, Cu and
307 Sb were below their hazardous waste limits. High standard deviations due to heterogeneity were also
308 observed among the elements Ba, Cu and Zn.

309

310 As pointed out by Cesaro et al. (2018), form and time of production are among the many factors
311 affecting glass chemical composition. The composition of crystal glass, for instance, differs from that
312 of other glass types. The concentration results in this study reflect glass production dynamics, where
313 the abundance correlates with the use of the different elements in the crystal glass production (Hynes

& Jonson, 1997). Generally, the heterogeneity of glass waste in terms of composition makes its recycling more challenging, which makes it end up in landfills.

Table 2. Elemental concentrations (mean ± standard deviation) of the samples compared with the Swedish EPA limits values for hazardous waste

3.1.3. Waste and leachate physico-chemical properties

Physico-chemical properties of the waste according to leaching tests and MC analysis are presented in Table 3. Each parameter value has been compared with the Swedish EPA limits for landfilling of inert, non-hazardous and hazardous waste (Swedish EPA, 2009). The classification is a risk assessment-based approach aimed at protecting environments surrounding the wastes' storage and/or containment areas. In case the glass waste is used as construction aggregate, for instance, the EPA limits would evaluate the exposure potential as well as the type of structures that would be built from such an aggregate.

Although its limit for the three waste categories is not specified in the Swedish EPA, pH was around neutral to basic (7.8 ± 0.5), which is in line with the range (7.0 – 8.3) observed in four previous landfill studies (Hogland et al., 2004; Jani et al., 2016; Kaczala et al, 2017; Mönkäre et al., 2016). Landfill waste pH usually correlates with the decomposition phases of the waste with low pH indicating incomplete material biodegradation (acidic phase) and higher pH indicating the methanogenic and humic phases. In the current study, the waste is not affected by these dynamics due to dominance of the glass fraction, hence it ranging around neutral pH. The limit for the three waste categories for MC is not specified in the Swedish EPA either, but the MC was considerably low (2.6 ± 1.3%). The low MC can be explained in terms of low organic fraction (Figure 2) since MC is dependent on the amount of organic fraction, its associated microbial activity and its water retention capacity (Parrodi et al., 2018). In addition, glass waste has a very low water retention capacity. TDS and DOC were also below their

limits for the three categories, as was F^- except it exceeded its inert waste limit. These parameters are all a reflection of the dump's waste composition (95.4% glass and 0.9% organic fraction).

Table 3. Waste and leachate parameters (mean ± standard deviation) against Swedish EPA landfill limits

3.1.4. Leachate trace element contents

The leachate elemental concentrations of the samples are presented in Table 4 along with the Swedish EPA limits for inert, non-hazardous and hazardous waste landfilling for each element. Results for each sample were aggregated from triplicates of that particular sample. The elements As, Ba, Cd and Cu in all eight samples were lower than the three category limits, Cd being lower than its limit of detection in all cases. The low leachability of the elements is in line with the solid phase elemental concentrations in Table 2. However, trace elements in low concentrations in leachate are known for their potential to interact with each other, thereby triggering toxic effects on surrounding organisms (Baścik-Remisiewicz et al., 2009; Cesaro et al., 2018; Tsiridis et al., 2006).

Pb on the other hand was notably higher than the inert and non-hazardous limits in all eight samples. High Pb concentration in the leachate is in line with its high amounts in the waste and implies that the metal poses a contamination hazard to the environment. The results further suggest that any changes in the surrounding pH would lead to risks due to leaching potential of Pb since the dumpsite lacks a protective cover. In this case, low pH would trigger mobility of the leached Pb to the surrounding since cation sorption is quite inefficient at lower (acidic) pH (Uddh-Söderberg et al., 2019). Thus, the material requires safe storage in a hazardous waste-type storage facility, which we refer to as *'Bank Account'*.

365 Safe storage is emphasized due to potential for Pb leaching and also the Swedish government's
366 urgency to move these materials to safe landfills (Högland, 2007), which may jeopardise their
367 recycling potential. The circular economy concept would entail avoiding landfilling. As established in
368 Table 1, elemental compositions (especially Pb) indicate a potential secondary source of metals into
369 the circular economy. According to the U.S. Geological Survey (2018a, 2018b), Pb and Zn are among
370 metals whose production has been on the increase, making the search for their alternative sources
371 significant. Recent metal extraction studies (Jani & Hogland, 2017, 2018) on Pb-crystal glass have
372 indicated extraction efficiencies as high as 99%, although on laboratory scale.
373
374 Decontaminated materials after metal extraction would be potentially useful in glass ceramics, foam
375 glass, new crystal and art glass, and cement and construction industry (Jani & Hogland, 2017),
376 depending on residual trace element contents and each application's specific quality requirements. In
377 cementitious systems, for instance, toxic metals are immobilised by an interplay of factors such as
378 particle size, pH of intergranular solution, existence of calcium silicate hydrates and the cement's
379 capacity to encapsulate contaminants in its hardening structure (Yao et al., 2018). Release of alkalis
380 from the cement hydration process can yield high pH (11 – 13) at which Pb forms stable hydroxide
381 complexes resulting in its immobilisation (Iniaghe & Adie, 2015). Further, the filler effect exerted by
382 fine glass particles enhances higher compactness in cement mortars which renders Pb leachability
383 negligible by hindering water penetration and limiting ion migration (Iniaghe & Adie, 2015; Sua-iam
384 & Makul, 2013). However, the applicability of this particular Pb-crystal glass needs to be
385 independently investigated and optimized according to each potential recycling application with
386 consideration of other environmental and social-economic factors.
387
388 **Table 4.** Leachate trace element concentrations (mean ± standard deviation) of the samples compared
389 with the Swedish EPA landfill limits
390

391 3.2. Radiometric measurements

392 3.2.1. External dose rate

393 Dose rate measurements performed 1 m above the ground inside and around the glass dump ranged
394 from 0.09 to 0.19 μSv/h as shown in Figure 4. The average background dose rate of the area was 0.138
395 μSv/h, which was calculated based on the first 5 points in the top right of Figure 4. There was a clear
396 increase in dose rates when measuring on top of the glass dump compared to the surroundings.
397 According to the gamma ray spectrum from the Lanthanum-Bromide detector, the increase in the
398 intensity of ^{40}K gamma ray in the centre of the dump was around 50% compared to the surroundings.
399 The red spots in Figure 4 indicate higher activity concentration of radionuclides among the glass
400 waste. Naturally occurring radionuclides from the ^{238}U and ^{232}Th series as well as ^{40}K are usually the
401 main contributors to the external dose in the environment. However, to identify and quantify the
402 actual contributing radionuclide(s) in this particular case, glass was sampled for further laboratory
403 analysis while considering the red spots (higher activity concentration spots) also in Figure 4.

404

405 **Figure 4.** Dose map over the dump. The black line delimits the border of the visible glass dump. (Map
406 data ©2019 Google)

407

408 3.2.2. Activity concentration of natural radionuclides

409 Table 5 shows the radionuclide concentrations found in the six samples sorted by colour. ^{137}Cs was
410 included here as it is a common anthropogenic radionuclide in the environment. However, the
411 combination of rinsing the glass samples prior to the analysis and the inertness of glass showed that
412 the glass is not contaminated with ^{137}Cs. Thus, in all samples ^{137}Cs was below MDA. Similarly, gamma
413 emitting radionuclides in the ^{232}Th series were all below MDA. ^{40}K had the highest concentrations
414 among all the radionuclides analysed and was quite stable among the different coloured samples. The
415 activity concentration was about twice the levels found in Swedish soils, which ranges between 560-
416 1150 Bq kg^{-1} (UNSCEAR, 2000). Assuming the ratio between ^{40}K and stable K to be 0.0117, calculation

of stable K yields 8.5%. Thus, measurement of ^{40}K is an efficient way to determine both the stable and radioisotope form of K in large sample sizes, for in-situ measurements and with minimal sample preparation and treatment.

In the ^{238}U series, ^{214}Pb and ^{214}Bi (which are in secular equilibrium with the parents ^{222}Rn and ^{226}Ra) were in low concentrations in all samples. For comparison, the average levels of each radionuclide in the ^{238}U series in Swedish soil is about 70 Bq kg^{-1} (Evans & Eriksson, 1983). Uranium levels in the glass samples were unexpectedly low, especially for the yellow coloured glass fraction since yellow colour was sometimes achieved through addition of uranium compounds (Strahan, 2001). However, the second highest activity concentration came from the ^{210}Pb which belongs to the ^{238}U series, and is higher than ^{214}Pb and ^{214}Bi in the same series. This indicates the use of Pb-containing compounds in manufacture of the glass as established by XRF results also, since all Pb compounds contain some part of the radioactive isotope ^{210}Pb. It should be mentioned that ^{210}Pb decays to ^{210}Po which is one of the most significant radionuclides among the NORMs in relation to radiation dose to humans when ingested or inhaled. Since the half-life of ^{210}Pb is about 22 years, and assuming the glass in the dump was produced in 1980, the activity concentration at the time of manufacturing would be approximately 45 Bq kg^{-1}. This value is still lower than the average concentration found in Swedish soil.

Table 5. Gamma results in glass samples (activity concentration in Bq kg^{-1}). High uncertainties in ^{238}U series radionuclides due to levels being close to the MDA. The uncertainty is given as 1-σ.

3.2.3. Radiological indices

To assess the External Hazard Index (H_{ex}) and Gamma Index (I_γ) according to Equations 1 and 2, the activity concentrations of ^{214}Bi and ^{214}Pb can be used when in secular equilibrium with ^{226}Ra. Contribution from the ^{232}Th series in the studied glass samples was negligible as the activity

concentration was lower than the MDA (0.5 to 1.5 Bq kg^{-1}). However, Th has been used in glass production due to its high refractive index, in applications such as camera lenses which could contain between 5 and 10% Th (Schirmer et al., 2016). The H_{ex} and I_γ for each glass colour are shown in Table 6. Several glass samples (white, red and blue colours) had $I_\gamma > 0.90$ (mainly due to ^{40}K contribution) but H_{ex} was lower than 0.62. From these results, it could be concluded that these glass materials would pose no radiological risk if used in construction applications, since both H_{ex} and I_γ are < 1. However, white glass samples are close to the threshold, $I_\gamma = 0.99$, compared to the yellow glass fraction, $I_\gamma = 0.77$, indicating the differences in radionuclide contents even in the same glass dump. Thus, it is important to perform both a radiometric and an elementary characterisation of the glass material in order to fully assess the possible health risks of recycling the material.

Based on Equation 1, it can be further concluded that although the material contains negligible amounts of both ^{226}Ra and ^{232}Th, a radiological risk can be posed if the ^{40}K activity concentration is 4810 Bq kg^{-1}. This in conjunction with the ratio of ^{40}K to stable K being 0.0177, an elementary analysis of stable K concentrations higher than 9.6% would give rise to a $I_\gamma = 1$. Therefore, stable K could be used as a 'parameter' to exclude (or at least to require further investigations concerning its use) any material from being used for building purposes from the radiological point of view. It has to be noted that if the glass waste is to be used in combination with some other materials, it is the final mixture that should fulfil this threshold of H_{ex} or I_γ indices.

Table 6. Radiological indices in the glass samples

4. Conclusions

The radiometrical and physico-chemical characteristics of glass waste at Madesjö glass dump were studied to evaluate pre-recycling storage requirements and radiological risks of recycling it. The glass waste had hazardous concentrations of As, Cd, Pb and Zn, although the leachate concentrations of As,

Cd and Zn were lower than the Swedish EPA limits for inert, non-hazardous and hazardous wastes. Potential use of this waste glass as a secondary resource requires methods that tackle both the resource deficiency challenge and the contamination problem. Due to Pb leaching higher than inert and non-hazardous waste limits, the glass dump needs urgent remediation and storage of the waste under a hazardous waste class *'Bank Account'* storage cell as a potential secondary resource for future recycling. The glass deposit also had some enhanced levels of ^{40}K, but based on the presented radiological indices, it is not expected to induce a radiological risk. However, as much as is reasonably achievable it is ethical to aim at a low radiation exposure of the population. This study has highlighted the need to consider physico-chemical and radiometric characterisations in combination to explore the fate of some radioactive materials used in the production of old glass. The results could be a valuable source of information in planning of handling and management alternatives of such contaminated waste streams.

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Conflict of Interest

The authors declare that they have no conflict of interest regarding the research, authorship and/or publication of this article.

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List of Tables (all created with the tables function in Microsoft Office Word)

Table 1. A comparison of average trace element concentrations (mean ± standard deviation) of the samples across the dump with that from a previous study based on XRF analyses (Jani & Hogland, 2017).

Table 2. Elemental concentrations (mean ± standard deviation) of the samples compared with the Swedish EPA limits for hazardous waste

Table 3. Waste and leachate parameters (mean ± standard deviation) against Swedish EPA landfill limits

Table 4. Leachate trace element concentrations (mean ± standard deviation) of the samples compared with the Swedish EPA landfill limits

Table 5. Gamma results in glass samples (Activity concentration in Bq kg^{-1}). Uncertainties in ^{238}U series radionuclides are high because the levels found are close to the MDA. The uncertainty is 1-sigma criteria.

Table 6. Radiological indices in the glass samples

Table 1

Elements	Mean value (± SD), present study (mg kg^{-1})	Mean value (± SD) from Jani & Hogland (2017) (mg kg^{-1})
As	13,138 (451)	19,761 (705)
Ba	1,221 (46)	878 (30)
Cd	394 (25)	847 (38)
Cu	551 (44)	856 (69)
Pb	200,102 (4634)	323,977 (8,524)
Sb	4233 (141)	4,373 (108)
Zn	3680 (97)	1,616 (106)

Table 2

Samples	**Elements (mg kg^{-1})**						
	As	**Ba**	**Cd**	**Cu**	**Pb**	**Sb**	**Zn**
S1	11,960 ± 430	964 ± 48	331 ± 25	372 ± 46	203,111 ± 4931	3,408 ± 130	2,905 ± 94
S2	15,454 ± 546	643 ± 34	460 ± 27	< LOD	269,347 ± 6525	3,496 ± 132	2,584 ± 91
S3	10,225 ± 354	1,469 ± 47	245 ± 19	750 ± 48	182,066 ± 3998	4,495 ± 130	5,120 ± 108
S4	16,989 ± 614	426 ± 38	588 ± 33	493 ± 52	303,669 ± 7653	3,700 ± 152	1,213 ± 64
S5	12,083 ± 398	1,476 ± 45	390 ± 23	124 ± 36	200,997 ± 4480	5,313 ± 158	4,528 ± 103
S6	14,391 ± 461	1,061 ± 37	431 ± 25	221 ± 32	225,194 ± 553	3,733 ± 124	2,756 ± 73
S7	10,032 ± 328	2,474 ± 69	279 ± 19	955 ± 42	171,439 ± 3507	5,199 ± 152	6,443 ± 141
S8	13,968 ± 474	1,255 ± 46	429 ± 26	945 ± 54	227,791 ± 5428	4,521 ± 152	3,889 ± 98
SEPA Limits	**1000**	**10000**	**100**	**2500**	**2500**	**10000**	**2500**

LOD is 'Limit of Detection' (0.5 mg kg^{-1}); n.s. is 'not specified'

Table 3

Parameter	**Value**	**Swedish EPA Landfill Limits**		
		Inert	Non-hazardous	Hazardous
pH	7.8 ± 0.5	n.s.	n.s.	n.s.
Moisture content (%)	2.6 ± 1.3	n.s.	n.s.	n.s.
Fluorides (mg kg^{-1} DS)	10.5 ± 7.9*	10	150	500
Total Dissolved Solids (mg kg^{-1} DS)	123 ± 45	4000	60000	100000
Dissolved Organic Carbon (mg kg^{-1} DS)	37.2 ± 5.5	500	800	1000

** Values higher than the statutory limit; n.s. is 'not specified'; DS is 'dry substance'*

Table 4

Samples & Swedish EPA Landfill Limits	Elements (mg l^{-1})						
	As	**Ba**	**Cd**	**Cu**	**Pb**	**Sb**	**Zn**
S1	0.04 ± 0.01	0.07 ± 0.01	< LOD	0.01 ± 0	6.26 ± 0.53*	0.05 ± 0.01	0.05 ± 0.01
S2	0.05 ± 0.01	0.10 ± 0.01	< LOD	0.01 ± 0	7.92 ± 0.94*	0.07 ± 0	0.09 ± 0.07*
S3	0.03 ± 0	0.03 ± 0	< LOD	< LOD	4.84 ± 0.56*	0.05 ± 0	0.02 ± 0
S4	0.02 ± 0	0.02 ± 0	< LOD	< LOD	4.85 ± 0.56*	0.04 ± 0	0.01 ± 0
S5	0.04 ± 0	0.25 ± 0.01	< LOD	0.01 ± 0	8.36 ± 1.92*	0.10 ± 0.02*	0.13 ± 0.01*
S6	0.03 ± 0	0.04 ± 0.01	< LOD	0.01 ± 0	5.77 ± 0.92*	0.06 ± 0.01	0.03 ± 0.01
S7	0.02 ± 0	0.12 ± 0.01	< LOD	0.01 ± 0	4.85 ± 0.65*	0.06 ± 0.01	0.04 ± 0.01
S8	0.03 ± 0	0.03 ± 0.01	< LOD	< LOD	5.30 ± 1.20*	0.05 ± 0.01	0.03 ± 0
Inert	**0.06**	**4**	**0.02**	**0.60**	**0.15**	**0.10**	**1.20**
Non-hazardous	**0.3**	**20**	**0.3**	**30**	**3**	**0.15**	**15**
Hazardous	**3**	**60**	**1.7**	**60**	**15**	**1**	**60**

** Values higher than at least one threshold; LOD is 'limit of detection' (0.0001 mg l^{-1} for Cd and 0.001 mg l^{-1} for Cu)*

Table 5

Radionuclides	Glass samples activity concentration (Bq)					
	Yellow	Black	White	Green	Red	Blue
^{214}Pb (^{226}Ra)	2.2 ± 1.0	2.1 ± 1.6	3.9 ± 0.9	MDA	MDA	MDA
^{214}Bi (^{226}Ra)	2.9 ± 1.8	3.1 ± 1.4	3.7 ± 0.7	MDA	MDA	MDA
^{210}Pb	MDA	14.3 ± 2.6	17.4 ± 2.2	10.6 ± 2.2	13.4 ± 2.8	12.0 ± 7.0
^{40}K	2290 ± 132	2643 ± 136	2919 ± 148	2696 ± 138	2735 ± 141	2751 ± 141
^{137}Cs	MDA	MDA	MDA	MDA	MDA	MDA

MDA is 'Minimum Detectable Activity' (^{210}Pb =0.55; ^{214}Pb =0.38; ^{214}Bi =0.30 in the ^{238}U series; ^{40}K = 1.62; ^{137}Cs = 0.11)

Table 6

Sample	Yellow	Black	White	Green	Red	Blue	**Average**
H_{ex}	0.49	0.56	0.62	0.56	0.57	0.57	**0.56**
I_{γ}	0.77	0.89	0.99	0.90	0.91	0.92	**0.90**

Credit Author Statement

1. Richard N. Mutafela – Physico-chemical
    - Funding acquisition, Investigation, Methodology, Data Analysis, Writing (original draft + review and editing)
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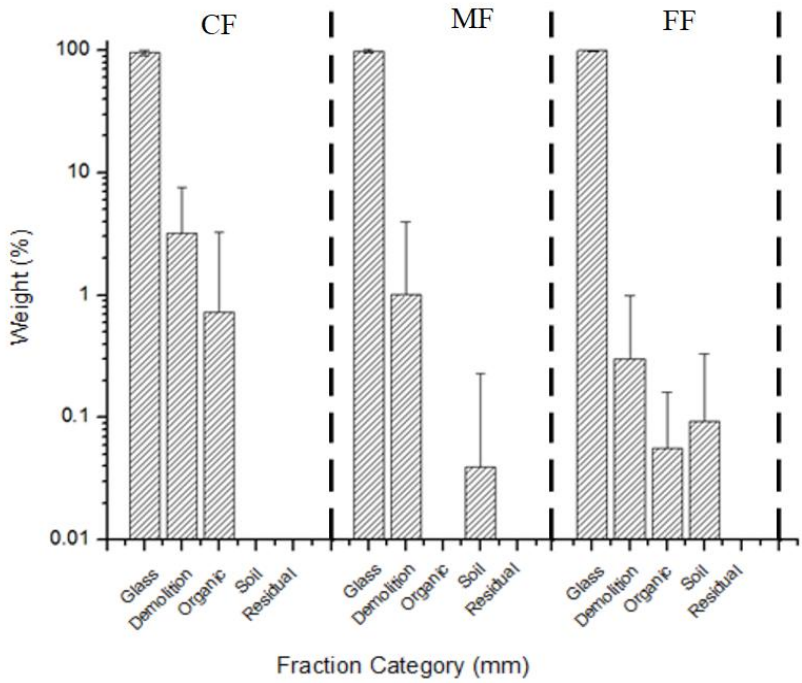

Figure 3

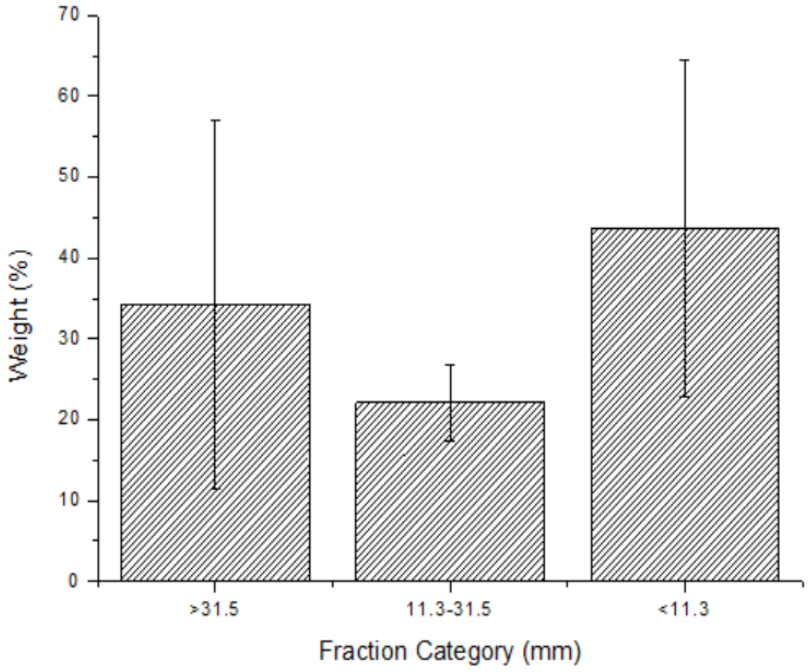

Figure 4