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# Contaminants in Waste Foundry Sand and its Leachate

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Abstract: The environmental characteristics of waste foundry sands (WFS), including chemicals in WFS and its leachate, are essential in understanding the environmental impact, rational disposal and potential development of beneficial applications of this solid industrial waste. Knowing environmental characteristics of waste foundry sand, scientists and engineers are able to make a decision between beneficial reuse, reclamation or waste disposal. This paper presents an assessment of broad-spectrum chemicals (metallic, nonmetallic and organic chemicals) in aspects of their statistics (mean, median and the 95$^{th}$ percentile) in dry-weight WFS and WFS leachates based on laboratory measurements of 594 WFS samples from 123 foundry facilities in US. Results indicate that WFS is basically not hazardous except a risk associated with WFS from copper-based foundry facilities. Leachability of metallic chemicals varies among investigated WFS. A clear delineation between different leaching protocols is implicated. WFS from copper-based facilities contain relatively higher levels of Cu, Pb and Zn in both dry-weight and leaching manners.

Keywords: contaminant, dry-weight, environmental characteristics, leachate, leaching, mean, median, metallic, nonmetallic, organic, toxicity, waste foundry sand.

Biographical notes: Dr. An Deng received his PhD in Civil Engineering from The Pennsylvania State University in 2004 and immediately joined Geotechnical Research Institute at Hohai University. He was entitled as a senior research scholar and undertakes challenging researches in the fields of beneficial reuse of solid waste materials, environmental pollution characterization and minimization. He also taught courses related to geotechnical and geoenvironmental engineering at undergraduate and graduate levels and was awarded in 2005 for his constructive performance in teaching soil mechanics. He is currently the coordinator of international cooperation program in Geotechnical Research Institute in charge of communications and exchange of international affairs. He has published papers in several journals and conference proceedings according to his remarkable research.

## 1 Introduction

Metal casting is a mature industry that is important to the US economy. The estimated metal casting industry shipments reached 12.2 million tons in 2001, with sales valued at $ 18.3 billion (US Census Bureau, 2000a, 2002b). As a solid byproduct of metal casting operations, around 9 million metric tons of waste foundry sand (WFS) were disposed annually (Winkler and Bol'shakov, 2000). As a result, economic and environmental concerns dominate the issue of WFS disposals. The current practice of WFS disposal in landfills is becoming an economic burden for foundries and municipalities as landfills close and regulations grow stricter. One of the solutions is to develop potential beneficial applications (Bhat and Lovell, 1996, 1997; Naik et al., 2001) of WFS.

The environmental characteristics, mainly chemicals in dry-weight and leaching characteristics, of WFS are essential in understanding the environmental impact or toxicity, rational disposal and potential development of beneficial applications of this solid industrial waste. Knowing environmental characteristics of WFS, scientists and engineers are able to make a decision between beneficial reuse, reclamation or waste disposal with respect to WFS destinations. In addition to the benefit concerned with end-application issues, global environmental characteristics of WFS may provide considerable savings. Chemical analysis is a necessity to determine the nature of the material as well as whether the waste will present an environmental threat or benefit. The cost of analyzing the waste can be significant depending on the number of waste streams to be characterized, the number of samples to be tested, the degree of material segregation, the frequency of testing and the number of parameters to be analyzed. Through understanding the environmental characteristics of WFS, the analyzing cost is minimized by identifying densely-detected and over-threshold analytes, eliminating or reducing measurements for non-existent analytes and adjusting analyzing frequency.

Generally, environmental characteristics of a solid waste consist of characterizations of microorganisms, disinfection byproducts, disinfectants, inorganic chemicals, organic chemicals and radio nuclides considering its potential impact to drinking water or underground water. Among them, metallic, nonmetallic and organic chemicals are mostly concerned and densely measured with respect to WFS ascribed to its repetitive exposures to high temperature metal castings and a variety of binding operations. It is investigated that up to 17 metallic chemicals including Ag, Al, As, B, Ba, Be, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Sb, Se and Zn are usually measured for a WFS sample. Eight metallic elements (Ag, As, Ba, Cd, Cr, Hg, Pb and Se) regulated in Resource Conservation and Recovery Act (RCRA) are included. Nonmetallic chemicals mainly include general chemical characteristics: loss on ignition, pH, phenol, total reactive sulfide and total volatile solid etc. Organic chemicals basically span widely, including but not limited to, benzene, chrysene, o-cresol, ethybenzene and naphthalene etc. Quantifications of these elements play a vital role in toxicity assessment, environmental threat and regulatory compliance of a waste stream. Designed workscope includes toxicity assessment of WFS, statistics (mean, median and 95th percentile) of chemicals, leachability of chemicals, effects of leaching protocol and casting operations.

Early-published studies on environmental characteristics of WFS is tracked back to one conducted by Ham and Boyle (1981) who found that no WFS samples in their study produced concentrations above the regulatory levels. Ham et al. (1993) obtained the same findings. Thereafter, various aspects related to environmental characteristics of WFS initiated: a summarization work by Winkler and Bol'shakov (2000) on the binder systems, the metallic contaminants and the leaching of organics, measurements of a wide variety analytes in dry-weight or leached WFS by Regan et al. (1994), Ji et al. (2001), Orkas (2002) or Kendall (2003), a technical database built by Regan and Voigt (1996). All these studies help the understanding of chemical nature of WFS and its impact to living environments per their researched WFS samples. Nevertheless, sound and global studies on broad-spectrum chemicals in dry-weight WFS and its leachates, as of the aim of this study, are not universally documented due to previous limited local samples and characterization data. The expense of conducting a chemical analysis of WFS is high, such as locally US$ 550 for a leaching analysis and varying with locations and numbers of tested chemicals. To execute a nationwide program which intends to retrieve and characterize a great number of WFS samples is basically unaffordable. As an alternative, to collect filed characterization dataset of WFS samples at nationwide levels may not only eliminate the obstacle of insufficient data for statistical implications but also lead to substantial savings both in cost and time.

## 2 Procedure and Methods

US nationwide collection of WFS characterization datasets was implemented. Laboratory datasets of 594 WFS samples, tested between 1997-2001 by independent and quality-controlled commercial laboratories for regulatory compliance and sourced from 123 foundry facilities in US which produce iron, aluminum, steel, copper castings, were pooled into a database characterizing two aspects of WFS: dry-weight and leaching characteristics of metallic, nonmetallic and organic chemicals. Dry-weight characterization is conducted on as-received WFS sample without an extraction or leaching process. It aims at finding out total chemical contents in as-received waste streams, generally in unit of mg/kg, and helps determine whether a leaching program is needed or not. Leaching characterization addresses concentrations of chemicals in a leachate that is extracted from an as-received waste stream by a leaching protocol and is assessed generally in unit of mg/L which is sometimes replaced by part per million (ppm) in datasets and regulations. Leaching assesses the impact of the waste stream posing to environments if exposed to a field extraction condition, such as leaching of rainfall or groundwater which the leaching protocol intends to simulate. In this study, three leaching protocols are involved across collected datasets, namely toxic characteristic leaching procedure (TCLP) developed by US EPA (1985a), synthetic precipitation leaching procedure (SPLP) by US EPA (1985b), and standard test method for shake extraction of solid waste with water (ASTM D3987) by ASTM (2003). Of collected 594 datasets, 123 datasets address dry-weight characteristics and 471 datasets assess leaching characteristics. The latter is furthered partitioned into 234, 151, and 86 datasets corresponding to TCLP, SPLP and ASTM D3987, respectively. Per the casting operations, datasets addressing TCLP characteristics is further partitioned into 193, 24, 49 and 13 corresponding to iron-based, steel-based, aluminum-based and copper-based operations, respectively.

TCLP, SPLP and ASTM D3987 are three commonly used leaching protocols developed for simulations of distinctive extraction conditions on liquid, solid, and multiphasic wastes and estimations of inorganic and/or organic contaminants extracted from the wastes. Table 1 lists the major operation criteria of each protocol which include extraction fluid types and pH values, liquid-to-solid ratios, extraction modes and periods, and analytes. TCLP is outlined in US EPA (1985a), with the most recent version of the experimental procedure dated July 1992, revision 0. This leaching protocol is designed to model the leaching behavior of a material disposed in an actively decomposing municipal solid waste landfill in which carboxylic acids are formed from microbial processes (US EPA 1985a). A 100-g size-reduced sample of waste material is mixed with an acetic acid extraction fluid at a liquid-to-solid mass ratio of 20:1. The extraction fluid pH value is set 2.88 or 4.93 depending upon the alkalinity of the waste material. The mixture is rotated for 18±2 hr at 30 rpm. After rotation, the final pH is measured, and the mixture is filtered using a glass fiber filter. The filtrate is collected and analyzed for a number of analytes. If any analyte exceeds its toxic threshold value regulated in CFR Title 40 (2000), the waste is hazardous for the toxic characteristics.

[Insert Table 1]

SPLP is outlined in US EPA (1985b), with the most recent version of the experimental procedure dated September 1994, revision 0. SPLP is developed to simulate leaching under acid rain condition, similar to an industrial waste monofill (US EPA 1985b). SPLP test is performed in the same manner as TCLP, except the extraction fluid. The extraction fluid for SPLP tests is a mixture of two inorganic acids (nitric and sulfuric acid) with pH values 4.22 or 5.0 depending upon geographic locations (Table 1).

ASTM D3987 uses reagent water (distilled deionized water) as extraction fluid and simulates conditions where the solid waste is the dominant factor in determining the pH of the extract (ASTM 2003). Apart from the extracting fluid, ASTM D3987 follows similar operations to those of TCLP and SPLP. It is shown from Table 1 that the difference between three leaching protocols is mainly associated with the selection of extraction fluid and its pH value, which is thought a determinative factor influencing the analyte extraction process.

Fig. 1, a two-dimensional scatter plot, illustrates dry-weight and leaching characteristics of Ba in WFS. Measurements, including below detection limit (BDL) qualifiers and quantified data, are presented in a chart. Horizontal axis represents four characterization attributes: dry-weight characteristics, TCLP, SPLP and ASTM D3987 leaching characteristics. Vertical axes contain dual axes, representing dry-weight level and leaching level, respectively. Points of dry-weight characteristics find their measurements along axis of dry-weight level. Points of TCLP, SPLP or ASTM D3987 find their measurements along axis of leaching level. Distributions of data bodies are able to demonstrate the chemical concentration variations in independent streams. Comparisons run among data bodies implicate leachability of chemicals (dry-weight vs leachates) and protocol effects (TCLP vs SPLP vs ASTM D3987). As analyte Ba is one of RCRA regulated elements, its TCLP toxicity threshold is presented in the legend section to better understand its toxicity. Obviously, as a graphical tool, scatter plots well and objectively illustrate quantified and BDL qualifiers. A straightforward insight into the environmental characteristics of WFS is obtainable.

[Insert Fig. 1]

Leachability of Ba is inferable by comparing and interpreting data bodies demonstrated in Fig. 1. Dry-weight Ba ranges from 0 to 350 mg/kg and mostly between 0 and 50 mg/kg in as-received WFS. TCLP extracted Ba ranges from 0 to 10 mg/L and mostly between 0 to 3 mg/L in leachates. If the solid-to-liquid dilution ratio of 1:20 in a TCLP leaching program is considered, Ba of 0 to 3 mg/L in leachates is equally extracted from 0 to 60 mg/kg Ba on a dry-weight basis, which is approximately comparable to actual dry-weight Ba ranges of 0 to 50 mg/kg. It is thus inferred that Ba in WFS is basically fully extracted by TCLP.

A clear delineation between different leaching protocols is illustrated in Fig. 1. It is indicated that TCLP tends to yield more Ba than SPLP and ASTM D3987. An apparently wider spread of scatter plots of TCLP compared to those of SPLP and ASTM D3987 implicates more Ba extracted by TCLP and detected in TCLP leachates than does by SPLP or ASTM D3987. This is because TCLP uses acetic acid as its extraction fluid and sets a relatively low pH value for the extraction fluid. Nevertheless, SPLP and ASTM D3987 use inorganic acid or reagent water as extraction fluids, respectively. Accordingly, TCLP is more aggressive in extracting most analytes, e.g. Ba, than SPLP and ASTM D3987.

Scatter plots also identify element concentration distributions. In Fig. 1, data do not conform to a uniform or normal distribution, but suggest a lognormal distribution. At low-level tail, most observations concentrate close to zero. At high-level tail, observations become sparse gradually with the increase of concentrations. A lognormal distribution is suitable for modeling element concentration after the distribution is diagnosed and verified according to statistics.

While scatter plot is a definitely useful aid in observing data variation, to quantify data bodies and prove their variation among independent waste streams cannot be addressed by observing a scatter plot. Statistical methodology is required to handle data bodies and to obtain statistical implications. BDL qualifiers become barriers to readily analyze datasets. When an element is not present at the lowest reliable metric of the instrument it is referred to as BDL. Measurements of BDL occur due to instrument quality, testing methods and local regulations and happen frequently in environmental monitoring data as herein. For instance, in a series of measurements of Ba in aluminum-based WFS leachates reported in mg/L, <10, <10, 1.8, 1.62, <1, 0.7, 0.54, 0.39, 0.36, 0.33, 0.32, 0.31, <0.3, <0.2, <0.2, <0.2, 0.185, 0.115, 0.0783 and 0.0251, 13 measurements are quantified as specific numbers including 1.8, 1.62, and 0.7 mg/L etc. The remaining 7 measurements are reported as non-quantified results or as ranges of less than limits, inclusive of <10, <1, and <0.3 mg/L etc., which belong to BDL qualifiers. Essentially actual concentrations of BDL qualifiers exist below their limits. In calculation of estimated mean concentrations of Ba, the upper boundary of mean is the average value of substitutions in which limits are substituted for BDL qualifiers (replace <10 with 10, and so forth), and the lower boundary of mean is the average value of substitutions in which zero is substituted for BDL qualifiers (replace <10 with 0, and so forth). Thus, the upper and lower boundaries of mean concentration of Ba in the sand leachates are calculated as 1.7 mg/L and 0.3 mg/L, respectively, while no point value but a range about mean concentration is determined. The unquantifiable characteristic of a mean concentration impedes proper environmental audit. In this case, if the regulated hazardous threshold for Ba were 1.0 mg/L, it is not reliable to assess average level of Ba either hazardous or non-hazardous.

To work with environmental BDL qualifiers generally needs both environmental and statistical knowledge. Statisticians normally call BDL qualifiers censored data. Some simple arbitrary substitutions may convert censored data into specific data, and then conventional statistical methodologies are applied to obtain the statistical summaries. Substitutions include reporting limits, a percentage of the reporting limit, or zero for censored data. Nevertheless, it is difficult to verify this substitution. This method may simply leads to a solution with no statistical basis or quantitative standing and thus is demonstrably false. Further complicated methods mainly include maximum likelihood estimation (MLE) and regression of statistics (ROS), which are basically based on a data distribution model assumption. However, the data distribution of environmental monitoring data is unknown and thus the model assumption is not convincible. Violation of this assumption can lead to invalid applications of a statistical technique. The decisions and conclusions derived from incorrectly used statistics are unreliable.

Survival analysis, a statistical technique initially addressing datasets containing right censored data, was used to quantify statistics of datasets containing BDL qualifiers. Contribution of this technique is the plot of a survival function as illustrated in Fig. 2 which contains three decreasing step functions indicating the survival probabilities $P(X > x)$ of the analyte Ba contents in TCLP, SPLP and ASTM D3987 leachates, respectively. Survival functions demonstrated in Fig. 2 are simply the conversions of corresponding data bodies presented in Fig. 1 and are able to implicate important statistics.

[Insert Fig. 2]

It was proved (Kaplan and Meier, 1958) that mean concentration of a chemical in a waste stream is equal to the geometric area under the chemical's survival function, or say geometric area of the irregular shape bounded by the chemical's survival function and coordinates. By summarizing individual bounded areas, mean estimations of Ba in TCLP, SPLP and ASTM D3987 leachates are 0.639, 0.388 and 0.173 mg/L, respectively. Also by comparing areas of bounded shapes, the descending order of mean estimation is Ba/TCLP, Ba/SPLP and Ba/ASTM D3987. Survival functions not only indicate mean, data ranges and distributions, but are able to quantify median, cumulative probabilities and percentiles by reading coordinates of a point. For instance, it is inferred from the TCLP function in Fig. 2 that 50% observations exceed 0.27 mg/L and that the $95^{th}$ percentile, $P(X \le x)$=0.95, is 2.43 mg/L. Given multiple survival functions plotted in a chart, Kolmogorov-Smirnov (KS) testing, a statistical hypothesis testing technique considering distributions when comparing data bodies, was used to run comparison between survival functions and thus to affirm whether a significant difference exists among two streams or not. KS testing is conducted by treating the maximum vertical distance (D in Fig. 2) between two survival functions (TCLP and SPLP) as a measure of difference between two data bodies. In Fig. 2, the maximum vertical distance D between two functions, i.e. Ba/TCLP and Ba/SPLP, suggests a statistically significant difference. It means that two functions belong to two different distributions and that the leaching effect (TCLP vs SPLP) is significant, or say TCLP and SPLP play different role in extracting Ba.

Using survival analyses, not only statistics including mean, median, $95^{th}$ percentile are estimable, but effects of leaching protocol and casting operations among various streams are testable as well. To depict environmental characteristics of WFS, workscope is divided into two parts as demonstrated in Fig. 3: dry-weight characteristics and leaching characteristics of WFS. The former covers statistics of chemicals per casting operations: iron-based, steel-based, aluminum-based and copper-based castings, and statistics for all-in-one (including all operations) castings. The latter include statistics of chemicals per leaching protocols: TCLP, SPLP and ASTM D3987. For datasets of TCLP, further interest is concentrated on the effect of casting operations mentioned above. By analyzing these statistics of various waste streams, the environmental characteristics of WFS and its toxicity are quantified, which contributes to the decision-making about WFS reuse in actual construction and agricultural works.

[Insert Fig. 3]

## 3 Results and Discussion

### *3.1 Dry-weight characteristics of WFS*

Mean of a chemical is estimable using its survival function. It represents the average value of a data body. Mean estimations regarding pH values of and metallic chemicals contaminated in dry-weight WFS are presented in Table 2. It is indicated that WFS from iron-, steel-, aluminum- and copper-based facilities are moderate alkaline, with pH value ranging on average from 8 to 9. A number of metallic chemicals are found contaminated in WFS at varying levels. Common and not regulated chemicals are most highly concentrated, including Al, Ca, Fe and Mn. Chemicals regulated in drinking water standards and RCRA hazardous provisions are detected at relatively low levels, including Ag, As, Ba, Cd, Cr, Cu, Hg, Ni, Se and Zn. It is also indicated that the mean concentration of each metallic chemical varies among different casting operations. WFS from iron-based facilities is contaminated with Ba, Ca, Fe and Mn at relatively high levels. WFS from steel-based facilities is contaminated with As, B, Cr, Cu, Fe, Mn and Ni at relatively high levels. WFS from aluminum-based facilities is significantly contaminated with Al. WFS from copper-based facilities contain Cu, Pb and Zn at relatively high levels. It is known that iron/steel-based operations usually add iron-related scraps as raw feeds; steel-based operations add chromium as an additive into castings to increase its anti-corrosivity and quality; aluminum-based facilities use aluminum-related scraps as raw feeds; copper-based facilities use copper-related scraps and add Pb and Zn as additives. Accordingly, it is inferred that mean concentrations of metallic chemicals are related to casting operations, specifically its raw feed materials. Reclamation with respect to Fe is suggested as it exists at averagely high levels (1.1% and 6%) in WFS from iron/steel- based operations.

[Insert Table 2]

Median represents the middle value of an ordered set of numerical data. One half of all data is below this value, while one half have a higher value. The median also is known as the 50$^{th}$ percentile which is readable from a survival function. The advantage in using the median is that it is not affected as much by extreme highs or lows in the range of values as is the case with the mean and thus better represent a middle level of environmental monitoring data as herein. Table 3 shows median estimations of pH values and metallic chemicals of WFS at dry-weight basis. Most median values are less than corresponding mean values shown in Table 2, which is ascribed to the lognormal-shape distribution as shown in Fig. 1. Denser observations tend to concentrate at lower levels. As a result, the observation in the middle is more close to zero than the mean. From Table 3, median variation among casting operations is not as significant as that of mean, which suggests the existence of extreme highs. Thus, to avoid a biased implication caused by extreme or isolated observations which may be errors incurred by incorrect analyzing operations or inappropriate sampling, median is superior to mean to indicate the middle levels. In the circumstance that both mean and median of chemicals are high regarding a casting operation, such as Cu, Pb and Zn for copper-based facilities, and Mn for iron/steel-based facilities, it is sound to advise that these chemicals are related with casting operations.

[Insert Table 3]

Percentile means a value on a scale that indicates the percent of a distribution that is equal to or below it. For example, a score at the 95$^{th}$ percentile is equal to or less than 95 percent of the scores. A 95$^{th}$ percentile may well indicate the upper boundary of a distribution by properly eliminating potential extreme highs or isolated observations. Comparisons run between 95$^{th}$ percentiles and hazardous thresholds may suggest toxicity of WFS. Table 4 presents the 95$^{th}$ percentiles of pH values of and metallic chemicals in dry-weight WFS and TCLP hazardous thresholds. One the hazardous criteria regarding solid wastes is corrosivity which is assessed according to pH values of waste materials. The hazardous threshold ranges for pH are regulated less than 2 or more than 12.5. The 95$^{th}$ and 5$^{th}$ percentiles of various dry-weight WFS waste streams fall out of the hazardous ranges. It is inferred that WFS is not corrosive with high confidence. US EPA (1985a) states that a leaching program is not mandatory when dry-weight chemicals are less than hazardous thresholds. By comparing results with TCLP toxicity threshold values, it is indicated that most metallic chemicals except Cr and Pb have their 95th percentiles falling within thresholds. It is known that a leaching program uses a solid-to-liquid dilution ration of 1:20. One-twentieth of both Cr and Pb's 95th percentile is less than corresponding thresholds as well. Thus, further toxicity evaluation using TCLP protocol is not necessary for WFS per metallic chemicals.

[Insert Table 4]

Comparisons of point values, including mean, median, maximum and minimum values, and any percentile, for a chemical in two or more waste streams are not statistically sound to differentiate waste streams. A comparison considering distributions at full ranges is advisable. KS testing technique, as demonstrated in Fig. 2, is used to run comparisons between data bodies relating independent casting operations and to implicate effect of casting operations. Selected comparison results are shown in Table 5. It is inferred that casting operation is not a significant factor regarding most metallic chemicals at dry-weight basis. The exceptions are Fe, Mn and Ni which tend to be higher in WFS from iron/steel-based facilities, and Cu, Pb and Zn which tend to be more contaminated in WFS from aluminum/copper-based facilities. Casting operation effects regarding these chemicals are attributed to the raw feed nature aforementioned.

[Insert Table 5]

Besides the metallic chemicals contaminated in dry-weight WFS, nonmetallic chemicals play an important role in depicting environmental characteristics and hazardous threats of WFS. Table 6 presents estimations of mean, median and the 95$^{th}$ percentile of nonmetallic chemicals contaminated in or with WFS at dry-weight basis. It is shown that WFS is averagely being moderate alkaline (pH 8.11). Silica is the dominating mineral, which is consistent with its sand nature. WFS is a sulfide bearing waste with sulfide content well below reactivity threshold 500 mg/kg.

[Insert Table 6]

Organic chemicals in WFS at dry-weight basis are detected and quantified as shown in Table 7, although it is generally thought that organic binders are burned or shaken away in the casting operations and that their remains or decompositions do not exist or present at negligible levels. It is found that most organic chemicals are reported as BDL qualifiers in collected datasets. The remaining quantified organic levels at dry-weight basis suggest the necessity of a leaching program. In RCRA, the regulated organic chemicals include benzene, o-cresol, m-cresol and p-cresol with toxicity thresholds 500 ug/kg for the first one and 200000 ug/kg for the last three. One twentieth of their corresponding levels in Table 7 is far less than thresholds.

[Insert Table 7]

*3.2 Leaching characteristics of WFS*

Leaching characteristics is the only channel assessing the toxicity of a solid waste and thus judging its hazardous threats to human health or the environment. To assess its toxicity, WFS is subjected to a leaching procedure to collect its leachate, which indicates the volume seepaging into surrounding environments. Chemical characteristics of a leachate are analyzed and the results are compared against corresponding TCLP toxicity thresholds. If all regulated chemicals in CFR Title 40 (2000) simultaneously fall within corresponding thresholds, WFS is identified as non-hazardous and has potentials of reuse. A violation of any chemical leads to a judgment that WFS is hazardous and should be disposed in a special landfill.

Three leaching protocols, namely TCLP, SPLP and ASTM D3987, are involved to identify chemicals likely to extract out from as-received WFS. The estimations of mean, median and the 95$^{th}$ percentiles of final pH values of and metallic chemicals in the WFS leachates are presented in Table 8. TCLP and SPLP leachates are on average moderate acid, with final pH values between 5 to 7. Full ranges of TCLP and SPLP leachates span both alkaline and acid properties, with the 95$^{th}$ and 5$^{th}$ percentile being up to 11.7 and down to 2.4. It is inferred that applying acid leaching protocols to WFS on average results in a moderate acid leachate. A low percentage alkaline leachates also exist.

Applying ASTM D3987 to WFS on average leads to an alkaline leachate, with mean and median being 8.27 and 8.9, respectively. Results also indicate that both acid and alkaline leachates are observed. It is known that ASTM D3987 uses reagent water as extraction fluids and is recognized as a neutral leaching protocol. The pH values of extracted waste materials are predominated with respects to leachate pH values.

According to the 95$^{th}$ percentiles, an upper boundary indicator, all leachate streams have regulated metallic chemicals (Ag, As, Ba, Cd, Cr, Hg, Pb and Se) well below TCLP toxicity thresholds. It is inferred that non-toxic characteristics of metallic chemicals put WFS a non-hazardous waste. According to the mean and median in Table 8, middle levels or most metallic chemicals in TCLP leachate are higher than those in SPLP or ASTM D3987 leachates by varying factors up to 50, which is thought associated with TCLP being more aggressive than the other two protocols.

Comparisons run between results in Table 8 and one twentieth of corresponding results in Table 2 and Table 3 indicate the leachability of metallic chemicals. It is inferred that analytes including As, Ba, Cr and Se is fully extracted by TCLP and that the remaining analytes are partly extracted. Extreme high observations may lead to a significant inconsistency between median and mean, such as Cr concentration of steel-based WFS 1664.1 mg/kg (Table 2) and its TCLP concentration (Table 8). It is to note that the leachability not only is related with the absorption of analytes to particles, the reactivity of analytes with extraction fluid, but also affected by the existing forms of analytes, either attached to particle surface or (naturally) embedded into particle mineral composition. If an element is mostly and substantially embedded or crystallized in the molecular structure rather than absorbed to the particle surface, the dry-weight analysis will detects its complete content, but the leaching program only extracts the absorbed one, a minimum part of full element.

[Insert Table 8]

Comparisons between data bodies associated with independent leaching protocols are performed using the KS testing technique. These comparisons are based on the data distributions rather than point values and may indicate the effects of leaching protocols. Results are shown in Table 9. It is inferred that TCLP tends to extract more As, Ba, Fe, Pb, Mn, Ni and Zn than SPLP. TCLP extracts less Hg and more Se than ASTM D3987. TCLP and SPLP extract similar amount of Cr and Cu. TCLP and ASTM D3987 extract similar amount of Cd. SPLP tends to extract less As and more Ba, Fe and Zn than ASTM D3987. SPLP and ASTM D3987 extract similar amount of Al, Cr, Cu, Pb, Mn and Ni. It is suggested that TCLP is more aggressive regarding most metallic chemicals in WFS than SPLP and ASTM D3987.

[Insert Table 9]

Table 10 presents statistics of mean, median and the 95$^{th}$ percentile of organic chemicals in WFS TCLP leachates. Although a variety of organic chemicals are reported in collected dataset as shown in Table 7, less chemicals are detected and quantified in Table 10. The 95$^{th}$ percentiles of chemicals are well below corresponding toxicity thresholds and WFS is suggested as non-hazardous per its organic chemicals.

[Insert Table 10]

Table 11 presents statistics of nonmetallic chemicals in the WFS leachates extracted by ASTM D3987. Comparison run between dry-weight (Table 7) and leaching chemicals (Table 11) may indicate the leachability of organic chemicals. Comparable chemicals include phenol, oil/grease and total petroleum hydrocarbon, contents of which indicate partial extractions of these chemicals.

[Insert Table 11]

The estimations of mean, median and the 95th percentiles of final pH values of and metallic elements in WFS TCLP leachates per casting operations are presented in Table 12. Leachate final pH values are on averae acid. No significant difference is found among the final pH values of WFS from four casting operations. WFS leachates of iron-based facilities present relatively higher level of Fe. WFS leachates of steel-based facilities present relatively higher level of Ni. WFS leachates of copper-based facilities present relatively higher level of Pb. No significant difference is found associated with the remaining chemicals. This finding is consistent with the finding aforementioned with respect to casting operation effects on dry-weight chemicals. It is inferred that casting operation is not a significant factor regarding most metallic chemicals in TCLP leachates.

[Insert Table 12]

## 4 Conclusions

Through statistics (mean, median and the 95th percentile) and comparisons among various waste streams (leaching related and casting operations related) with respect to dry-weight and leaching characteristics (metallic, nonmetallic and organic chemicals) of WFS based on 594 chemical characterization datasets, environmental characteristics (contamination) of WFS is depicted at global and quantifiable levels. Following points are summarized.

WFS is non-hazardous supported by both dry-weight and leaching characteristics, while wide-scale chemicals are contaminated in WFS and its leachate. To subject WFS to a leaching protocol and to measure full-spectrum of chemicals are not mandatory except those from copper-based facilities where a risk associated with Pb still exists. Data is likely lognormal distributed although not yet verified and median is better to indicate a middle level of a chemical. TCLP is a more aggressive leaching protocol and tends to yield more metallic chemicals than SPLP and ASTM D3987. A suitable selection of a protocol is advisable in toxicity assessment depending upon the simulated extraction conditions. TCLP and SPLP leachates are on average moderiate acid. ASTM D3987 are on average moderate alkaline. Leachability varies among chemicals. Analytes As, Ba, Cr and Se are identified being fully extracted from WFS into TCLP leachates. The remaining analytes are partially extracted. WFS from copper-based facilities contain relatively higher levels of Cu, Pb and Zn than from other facilities at both dry-weight and leaching basis. WFS from iron/steel-based facilities contain relatively higher levels of Fe and Mn than from other facilities at both dry-weight and leaching bases.

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**List of Tables**

Table 1: Operation criteria of TCLP, SPLP and ASTM D3987

| Operation criteria | TCLP | SPLP | ASTM D3987 |
|---|---|---|---|
| Extraction fluid | Acetic acid solution | Sulfuric/nitric acid solution | Reagent water |
| pH value of extraction fluid | Non-alkaline materials: $4.93 \pm 0.05$, alkaline materials: $2.88 \pm 0.05$ | East of Mississippi River: 4.20, west of Mississippi River: 5.00 | N/A |
| Extraction fluid and mode for volatile analytes | pH 4.93, zero-headspace extraction vessel | Deionized water, zero-headspace extraction vessel | N/A |
| Particle size reduction | <9.5 mm | <9.5 mm | Not required |
| Liquid-to-solid ratio | 20:1 (by mass) | 20:1 (by mass) | 20:1 (v/m) |
| Mode of extraction<br>Extraction period | Rotary agitation<br>$18 \pm 2$ hrs at 30 rpm | Rotary agitation<br>$18 \pm 2$ hrs at 30 rpm | Rotary agitation<br>$18 \pm 0.25$ hrs at 30 rpm |
| Leachate-solid separation | Glass fiber filters of pore size 0.6 to 0.8 µm | Glass fiber filters of pore size 0.6 to 0.8 µm | Glass fiber filters of pore size 0.45 µm |
| Collection time | One | One | One |
| Operation temperature | Room temperature | Room temperature | Room temperature |
| Analytes | Inorganic and organic analytes | Inorganic and organic analytes | Inorganic analytes |

Table 2: Estimations of mean of pH and metallic chemicals in dry-weight WFS

| Chemicals | Mean (mg/kg) | | | | |
|---|---|---|---|---|---|
| | Iron-based | Steel-based | Aluminum-based | Copper-based | All-in-one |
| pH | 7.96 | 8.68 | 7.95 | 8.68 | 8.11 |
| Ag | - | - | - | - | .47 |
| Al | 1327 | 1576 | 12931 | 1190 | 3326 |
| As | .76 | 6 | .34 | .39 | .86 |
| B | 9.86 | 276.2 | - | - | 38.53 |
| Ba | 24.31 | 7.77 | 6.53 | 3.97 | 14.95 |
| Be | - | - | - | - | .08 |
| Ca | 4842 | 1366 | 927 | 2481 | 3309 |
| Cd | - | - | - | - | .22 |
| Cr | 16.23 | 1664.1 | 4.63 | 3.21 | 114.03 |
| Cu | 30.32 | 404.73 | 17.68 | 653.2 | 103.60 |
| Fe | 11163 | 60819 | 1898 | 1986 | 10911 |
| Hg | - | - | - | - | .04 |
| Mg | - | - | - | - | 1881 |
| Mn | 483 | 523 | 26.66 | 14.33 | 257 |
| Mo | - | - | - | - | 38.84 |
| Ni | 6.85 | 1664.6 | 44.17 | 9.86 | 107.94 |
| Pb | 19.81 | 8.42 | 3.62 | 88.6 | 15.72 |
| Sb | - | - | - | - | 4.34 |
| Se | - | - | - | - | .64 |
| Tl | - | - | - | - | .43 |
| Zn | 131 | 64.1 | 31.6 | 691.2 | 102.48 |

"-" result not available.

Table 3: Estimations of median of pH and metallic chemicals in dry-weight WFS

| Chemicals | Median (mg/kg) | | | | |
|---|---|---|---|---|---|
| | Iron-based | Steel-based | Aluminum-based | Copper-based | All-in-one |
| pH | 8.3 | 8.73 | 8.27 | 8.55 | 8.3 |
| Ag | - | - | - | - | .1 |
| Al | 920 | 1200 | 1100 | 480 | 1400 |
| As | .49 | .56 | .38 | .48 | .49 |
| B | 6.93 | 29 | - | - | 6.93 |
| Ba | 14 | 5 | 7.1 | 4.51 | 8 |
| Be | - | - | - | - | .079 |
| Ca | 880 | 719 | 574 | 279 | 738 |
| Cd | - | - | - | - | .039 |
| Cr | 2.5 | 5.6 | 2.6 | 3.02 | 3.7 |
| Cu | 5.89 | 7.2 | 3.3 | 410 | 12 |
| Fe | 3490 | 2900 | 1300 | 1220 | 3700 |
| Mg | - | - | - | - | 310 |
| Mn | 43.7 | 71 | 12 | 10 | 49 |
| Mo | - | - | - | - | .1 |
| Ni | 2.21 | 4.5 | 1.6 | 8.9 | 4.5 |
| Pb | 2.81 | 3.03 | 1.7 | 61 | 3.1 |
| Sb | - | - | - | - | - |
| Se | - | - | - | - | - |
| Tl | - | - | - | - | - |
| Zn | 8.63 | 5.3 | 23.1 | 250 | 13 |

"-" result not available.

Table 4: Estimations of the 95th percentile of pH and metallic chemicals in dry-weight WFS

| Chemicals | 95th percentile (mg/kg) | | | | | TCLP hazardous thresholds (mg/L) |
|---|---|---|---|---|---|---|
| | Iron-based | Steel-based | Aluminum-based | Copper-based | All-in-one | |
| pH | 9.91 / 3.82* | 10.4 / 6.99 | 9.6 / 5.26 | 10 / 7.7 | 10.08 / 4.41* | <2 or >12.5 |
| Ag | - | - | - | - | .21 | 5 |
| Al | 3800 | 1200 | 15000 | 480 | 4300 | - |
| As | 1.8 | .56 | .67 | .49 | 1.6 | 5 |
| B | 39 | 29 | - | - | 88.9 | - |
| Ba | 52 | 18 | 12 | 4.51 | 28 | 100 |
| Be | - | - | - | - | .229 | - |
| Ca | 12200 | 3600 | 2100 | 278 | 11000 | - |
| Cd | - | - | - | - | .25 | 1 |
| Cr | 67 | 5.6 | 19.3 | 4.82 | 65 | 5 |
| Cu | 154 | 7.4 | 64.3 | 1860 | 350 | - |
| Fe | 25000 | 340000 | 4600 | 5750 | 25000 | - |
| Hg | - | - | - | - | .06 | .2 |
| Mg | - | - | - | - | 5240 | - |
| Mn | 548 | 2800 | 109 | 33 | 480 | - |
| Mo | - | - | - | - | 67.1 | - |
| Ni | 29 | 12.3 | 3 | 17.8 | 58 | - |
| Pb | 29.8 | 30 | 19 | 64 | 53 | 5 |
| Sb | - | - | - | - | 31 | - |
| Se | - | - | - | - | .49 | 1 |
| Tl | - | - | - | - | - | - |
| Zn | 42 | 7.4 | 99 | 620 | 216 | - |

"-" result not available. "*" 5th percentile of pH.

Table 5: Comparisons between casting operations

| Chemicals | Statistical comparison results | Testing significance *p* |
|---|---|---|
| Al | Iron=Steel, Steel=Aluminum, Aluminum=Copper | >.2, >.2, >.15 |
| As | Iron=Steel, Steel=Aluminum, Aluminum=Copper | >.2, >.2, >.2 |
| Ba | Iron=Steel, Steel=Aluminum, Aluminum=Copper | .17, >.2, .14 |
| B | Iron=Steel | >.2 |
| Ca | Iron=Steel, Steel=Aluminum, Aluminum=Copper | >.2, >.2, >.2 |
| Cr | Iron=Steel, Steel=Aluminum, Aluminum=Copper | >.2, >.2, >.2 |
| Cu | Iron=Steel, Steel=Aluminum, Aluminum<Copper, Iron<Copper, Steel<Copper | >.2, .2, .008, .005, .01 |
| Fe | Iron=Steel, Steel>Aluminum, Aluminum=Copper | >.2, .2, >.2 |
| Pb | Iron=Steel, Steel=Aluminum, Aluminum<Copper, Iron<Copper, Steel=Copper | >.2, .2, .008, .008, >.2 |
| Mn | Iron=Steel, Steel>Aluminum, Aluminum=Copper | >.2, .02, .2 |
| Ni | Iron=Steel, Steel>Aluminum, Aluminum<Copper | .2, .04, .04 |
| Zn | Iron>Steel, Steel<Aluminum, Aluminum<Copper, Iron<Copper, Steel<Copper | <.05, .04, .008, <.005, .01 |

Note: A $p$ value close to zero, or $p < 0.05$, means a significant factor effect.

Table 6: Estimations of mean, median and the 95th percentile of nonmetallic chemicals in WFS

| Chemicals | Mean | Median | 95th percentile |
|---|---|---|---|
| pH | 8.11 | 8.3 | 10.08 |
| Moisture (%) | 1.27 | .62 | 4.4 |
| Silica (%) | 77.2 | 76 | 99 |
| Loss on ignition (%) | 2.54 | 2.33 | 5.54 |
| Oil and grease (mg/kg) | 1744 | 560 | 3240 |
| Total petroleum hydrocarbon (mg/kg) | 223.52 | 71 | 1100 |
| Formalhyde (mg/kg) | 9.21 | 4.21 | 27.6 |
| Phenol (mg/kg) | 21.45 | 4.2 | 124 |
| Total reactive sulfide (mg/kg) | 37.02 | - | 36 |
| Total volatile residual (mg/kg) | 20995 | 23000 | 35000 |
| Hexavalent chromium (mg/kg) | .08 | .022 | .59 |
| Total volatile solid (mg/kg) | 1.90 | 1.79 | 3.52 |

"-" result not available.

Table 7: Estimations of mean, median and the 95th percentile of organic chemicals in dry-weight WFS

| Chemicals | Mean (ug/kg) | Median (ug/kg) | 95th percentile (ug/kg) |
|---|---|---|---|
| Acenaphthylene | 100 | 45 | 54 |
| Anthracene | 370 | 19 | 2240 |
| Benzene | 120 | - | 750 |
| Benzo-(a)anthracene | 10 | 5.7 | 19 |
| Benzo-(b)fluornathene | 10 | 9.6 | - |
| Benzo-(k)fluornathene | 2 | 2.3 | - |
| Chrysene | 27 | 20 | 46 |
| o-Cresol | 2722 | 170 | 13300 |
| m_p-Cresol | 1119 | 5910 | 0 |
| 2,4-Dimethylphenol | 1194 | 170 | 5790 |
| Ethylbenzene | 46 | - | 88 |
| Fluoranthene | 306 | 24 | 465 |
| Fluorene | 37 | - | - |
| 1-Methyl naphthalene | 328 | 300 | 511 |
| 2-Methyl naphthalene | 406 | 480 | 669 |
| Naphthalene | 435 | 300 | 1030 |
| Phenanthene | 1604 | 120 | 8440 |
| Pyrene | 47 | 2.4 | 57 |
| Styrene | 7796 | 51 | 23000 |
| Toluene | 45 | 10 | 210 |
| Xylene | 61 | - | 300 |

"-" result not available.

Table 8: Estimations of mean, median and the 95th percentile of final pH and metallic chemicals in WFS leachates

| Chemicals | Mean (mg/L) | | | Median (mg/L) | | | 95th percentile (mg/L) | | | TCLP toxicity threshold (mg/L) |
|---|---|---|---|---|---|---|---|---|---|---|
| | TCLP | SPLP | ASTM D3987 | TCLP | SPLP | ASTM D3987 | TCLP | SPLP | ASTM D3987 | |
| pH | 6.48 | 6.24 | 8.27 | 5.1 | 6.1 | 8.9 | 11.7 / 4.07* | 10 / 2.4* | 10.1 / 4.66* | - |
| Ag | .004 | - | - | .0006 | - | - | .008 | - | - | 5 |
| Al | 1.785 | 2.698 | 2.094 | .62 | .205 | .33 | 4.51 | 12 | 6.6 | - |
| As | .031 | .001 | .003 | .0572 | - | - | .07 | .002 | .005 | 5 |
| Ba | .639 | .388 | .173 | .31 | .17 | .044 | 2.4 | 1.9 | .6 | 100 |
| Cd | .004 | - | .0003 | .018 | - | .0002 | .018 | - | .0004 | 1 |
| Cr | .042 | .002 | .007 | .11 | - | .001 | .11 | .003 | .018 | 5 |
| Cu | .521 | .061 | .087 | .01 | .005 | .004 | .4 | .18 | .033 | - |
| Fe | 61.78 | 6.095 | 1.245 | 1.1 | .689 | .292 | 5750 | 30 | 5.02 | - |
| Hg | .0002 | - | .0002 | - | - | .0002 | .00004 | - | .0003 | .2 |
| Mn | 1.009 | .215 | .061 | .095 | .009 | .0042 | 5.72 | .928 | .19 | - |
| Na | - | - | 15.043 | - | - | 1.3 | - | - | 56.3 | - |
| Ni | .183 | .029 | .006 | .025 | .06 | .0011 | .875 | .06 | .026 | - |
| Pb | .222 | .009 | .008 | .006 | - | .0037 | .66 | .02 | .018 | 5 |
| Se | .041 | - | .002 | .006 | - | .001 | .1 | - | .0026 | 1 |
| Zn | 1.006 | .264 | .177 | .28 | .06 | .03 | 5.29 | .84 | .41 | - |

"-" result not available. "*" 5th percentile of final pH.

Table 9: Comparisons between leaching protocols

| Chemicals | Statistical comparison results | Testing significance $p$ |
|---|---|---|
| Al | TCLP ≠ SPLP, SPLP=ASTM | <.01, <.1 |
| As | TCLP>SPLP, SPLP<ASTM | .025, <.005 |
| Ba | TCLP>SPLP, SPLP>ASTM | <.005, <.005 |
| Cd | TCLP=ASTM | >.2 |
| Cr | TCLP=SPLP, SPLP=ASTM | >.2, >.2 |
| Cu | TCLP=SPLP, SPLP=ASTM | >.2, >.2 |
| Fe | TCLP>SPLP, SPLP>ASTM | <.025, <.005 |
| Hg | TCLP<ASTM | .005 |
| Mn | TCLP>SPLP, SPLP=ASTM | <.005, >.2 |
| Ni | TCLP>SPLP, SPLP=ASTM | <.005, >.2 |
| Pb | TCLP>SPLP, SPLP=ASTM | .01, >.2 |
| Se | TCLP>ASTM | .02 |
| Zn | TCLP>SPLP, SPLP>ASTM | <.005, <.025 |

Note: A $p$ value close to zero, or $p < 0.05$, means a significant factor effect.

Table 10: Estimations of mean, median and the 95th percentile of organic chemicals in WFS TCLP leachates

| Chemicals | Mean (ug/L) | Median (ug/L) | 95th percentile (ug/L) | TCLP toxicity thresholds (ug/L) |
|---|---|---|---|---|
| Acetone | 265.5 | 86 | 115 | - |
| Benzene | 1.46 | - | 1.8 | 500 |
| Cresol_m | 22.33 | - | 160 | 200000 |
| Cresol_o | 46.4 | - | 351 | 200000 |
| 2_4-Dimethylphenol | 30.59 | - | 218 | - |
| Ethyl benzene | 5.78 | 1 | 33 | - |
| Methyl isobutkl ketone | 50.5 | - | - | 200000 |
| Naphthalene | 71.69 | - | 616 | - |
| Phenanthrene1 | 6.99 | - | 14.8 | - |
| Styrene | 57.3 | - | 430 | - |
| Toluene | 51.92 | 4.2 | 20 | - |
| Trichloro-ethylene | 7.23 | 1 | 38 | 500 |
| Xylene_total | 3.73 | - | 5.9 | - |

"-" result not available.

Table 11: Estimations of mean, median and 95th percentile of nonmetallic chemicals in WFS ASTM D3987 leachates

| Chemicals | Mean (mg/L) | Median (mg/L) | 95th percentile (mg/L) |
|---|---|---|---|
| Acidity | 11.8 | - | 86 |
| Alkalinity | 50 | - | - |
| Ammonia | 4.511 | .541 | 12.6 |
| Chemical oxygen demand | 439 | 79 | 2400 |
| Chloride | 8.003 | 3 | 23.34 |
| Cyanide | .003 | - | .007 |
| Fluoride | .734 | .338 | 2.5 |
| Nitrate | 1.266 | .12 | 4.12 |
| Nitrite | .021 | - | .066 |
| Oil and grease | 14.700 | 1.5 | 122 |
| Phenol | .992 | .061 | 5.64 |
| Sulfate | 56.596 | 12.11 | 241 |
| Total dissolved solid | 347 | 126 | 1200 |
| Total oxygen halogen | 6.221 | .015 | 35.4 |
| Total petroleum hydrocarbon | 2.047 | .24 | 11 |

"-" result not available.

Table 12: Estimations of mean, median and the 95th percentiles of final pH and metallic chemicals in WFS TCLP leachates

| Chemicals | Mean / Median / 95th percentile (mg/L) | | | |
|---|---|---|---|---|
| | Iron-based | Steel-based | Aluminum-based | Copper-based |
| pH | 6.02 / 5.03 / 9.79 / 3.84* | 5.43 / 4.99 / 7 / 3.68* | 6.96 / 7.6 / 9.09 / 4.46* | 5.62 / 5.05 / 8.18 /4.89* |
| Al | 1.793 / .58 / 11 | 4.239 / 1.7 / 21 | .816 / .4 / 3.5 | 1.393 / 1 / 2.7 |
| Ba | .672 / .294 / 2.4 | .582 / .208 / 1.48 | .413 / .31 / 1.62 | .589 / .028 / 1.68 |
| Cd | .003 / - / .012 | .009 / - / .06 | --- | --- |
| Cr | .044 / - / .11 | .063 / .006 / .39 | --- | --- |
| Fe | 106.7 / 3.31 / 285 | 23.8 / .24 / 72.2 | 3.44 / .68 / 4.23 | --- |
| Hg | .0004 / - / .002 | .0006 / - / .004 | .0005 / - / .001 | --- |
| Mn | 1.409 / .12 / 7.03 | 1.307 / .34 / 4.31 | .447 / .12 / 2.04 | --- |
| Ni | .198 / .019 / .875 | .455 / .327 / 1.28 | .071 / .025 / .439 | --- |
| Pb | .123 / .0043 / .24 | .023 / .0056 / .068 | .174 / .04 / .49 | .73 / .35 / 2 |
| Se | .042 / - / .191 | .021 / .0077 / .07 | --- | --- |
| Zn | .785 / .27 / 2.54 | .318 / .21 / .52 | .715 / .26 / .65 | --- |

"-" result not available. "*" 5th percentile of final pH.

**List of Figures**

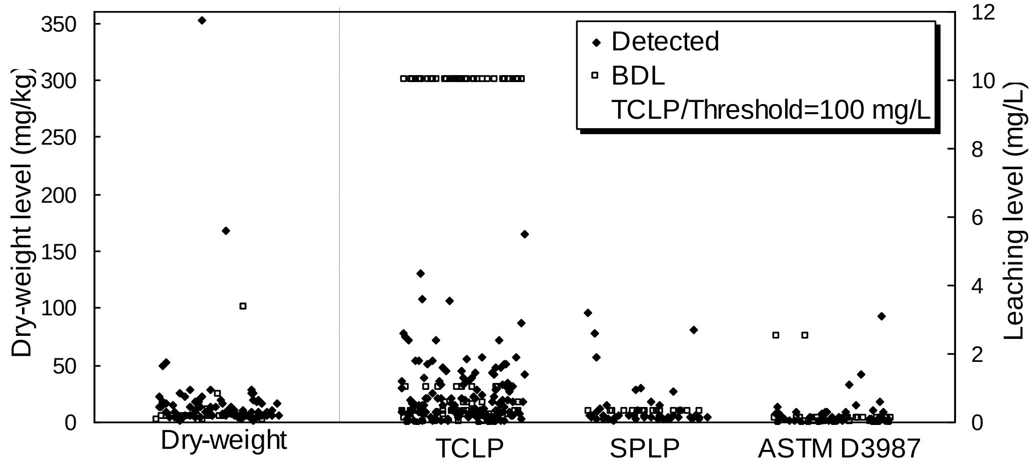


Fig. 1: Dry-weight and leaching characteristics of Ba in WFS

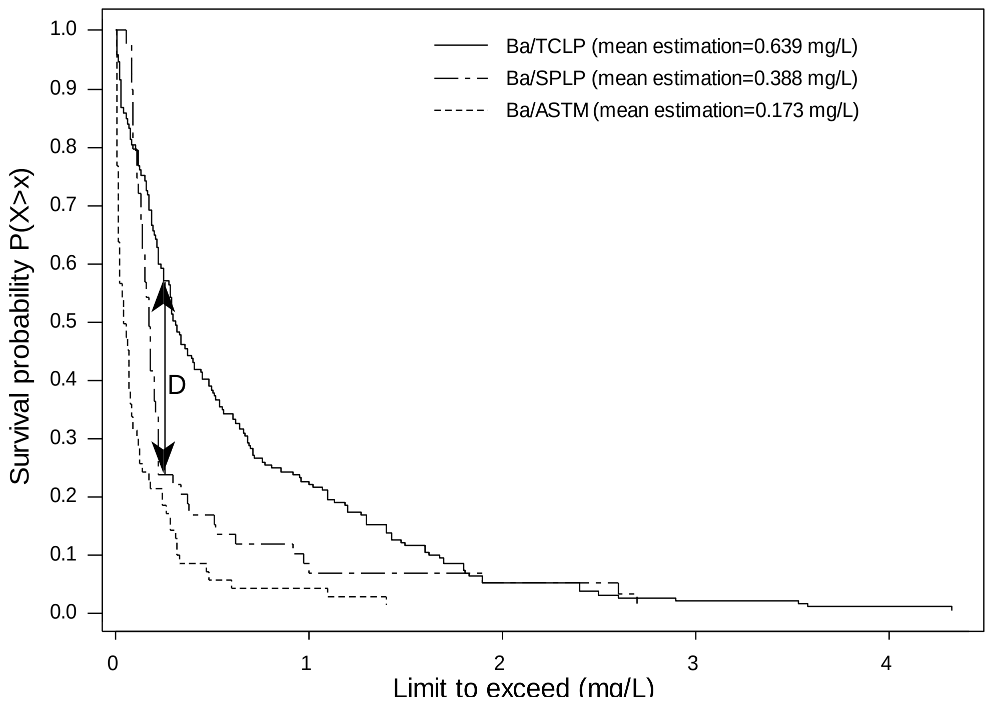


Fig. 2: Survival functions of Ba in TCLP, SPLP and ASTM D3987 leachates

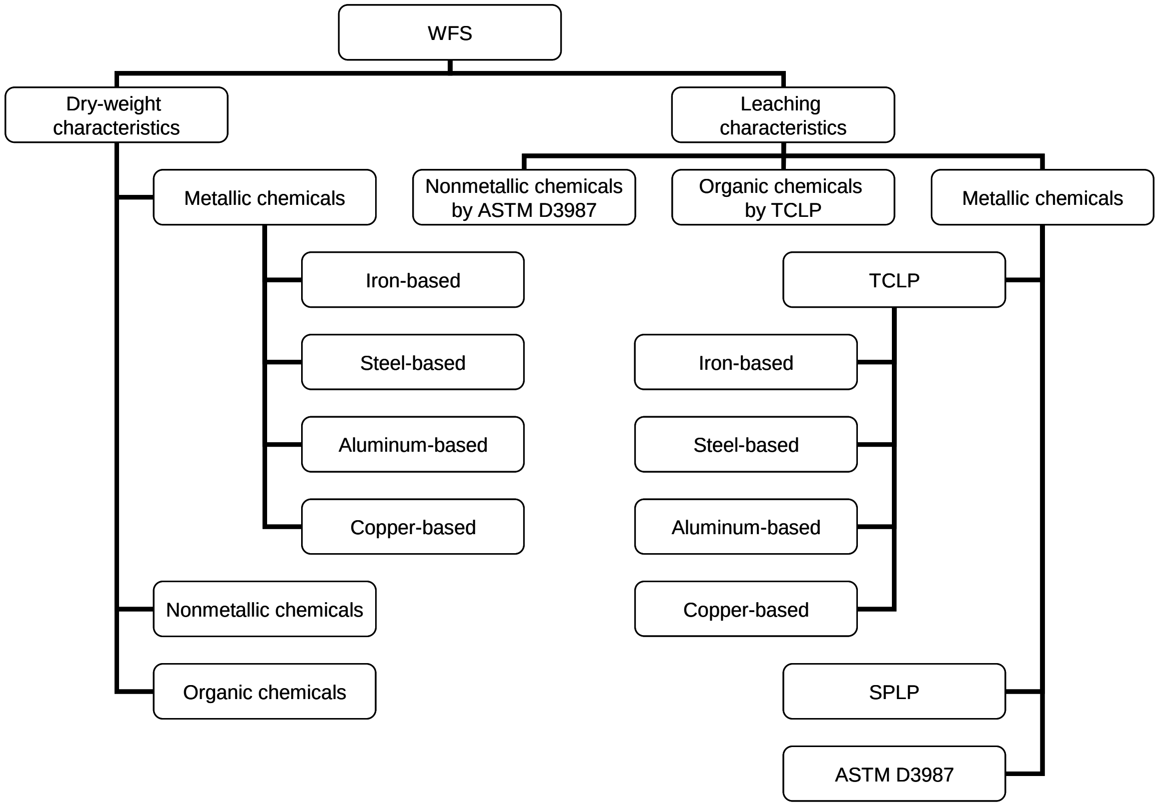


Fig. 3: Workscope of depicting contaminates in WFS and its leachate