QUARTERLY PROGRESS REPORT 3

Title: Effects of Florida Leachates on Geosynthetic Clay Liners (GCLs)

Project Duration: December 1st, 2017 – November 30th , 2018
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PROJECT WEB SITE:
https://www.eng.famu.fsu.edu/~abichou/MSWI%20GCL%20FL%20Project.html

Present Goals:
The main objective of this study is to test the resistance of conventional GCLs from different vendors to synthetic permeant solutions and aggressive leachates from MSW, MSW+ASH, MSW- I landfills and CCPP landfills from Florida and possibly other states in the USA. Further, the intent is to identify conditions where these GCLs might not be adequate (such as negative gradient landfills and fluctuating groundwater table). On the GCLs, conventional tests were utilized (Swell Index, Atterberg limits, 1D Swell Test, hydraulic conductivity). The synthetic permeant solutions and aggressive leachates underwent chemical characterization such as ratio of monovalent and divalent cations (RMD), ionic strength (IC), electrical conductivity (EC), and pH.

Work accomplished during this reporting on 7/31/2018:

Presents Achievements
In the third quarter, we focused on the characterization of different types of leachates collected from landfills and the influence of MSWI bottom ash on the leachate characteristics as well as leachate collection system (LCS) clogging. The basic water chemistry parameters, major cations and anions for all leachate samples were evaluated. The trace metals in the co-disposal and ash monofill leachates were also characterized.

Next, we will showcase some of the work accomplished during this reporting period:
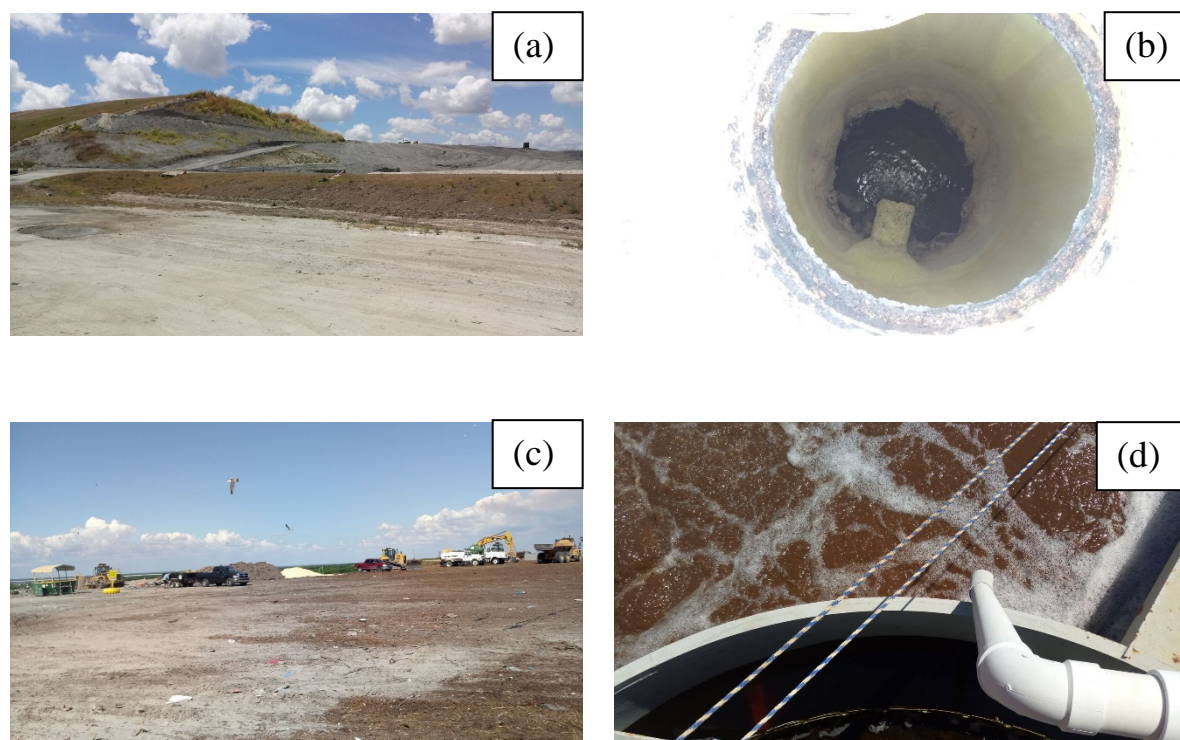
1. Leachate Collection and Characterization

1.1 Leachate Collection

Leachate samples were collected from seven landfills throughout Florida, US. Among these landfills, one received only municipal solid waste (MSW), one received only MSW incineration ash, one is in gradient landfill and four received a mixture of MSW and MSW incineration ash. At least two samples were collected from each facility. Leachate samples from one coal ash landfill (Georgia, US) was also collected. Leachate samples were kept in coolers during transportation and transferred to the refrigerator until characterization. Landfill visit and

leachate sample collection example are depicted in Figure 1. Description of collected samples are listed in Table 1.

Table 1. Leachate source and description.

Abbreviation	Landfill type	Sample description
CA	Coal ash landfill	From Georgia, landfill accepts 100% coal ash
IG	Inward gradient landfill	From Florida,
AM	Ash monofill	From Florida, landfill accepts 100% MSW ash
CD1	Co-disposal landfill	From Florida, landfill accepts 80% MSW ash
CD2	Co-disposal landfill	From Florida, landfill accepts 50% MSW ash
CD3	Co-disposal landfill	From Florida, landfill accepts 20% MSW ash
CD4	Co-disposal landfill	From Florida, ash% varies between 25-75%
MSW	MSW landfill	From Florida, receives only MSW

Figure 1. Landfill visit ((a) & (c)) and leachate sample collection ((b) & (d)).

1.2 Leachate Characterization

1.2.1 Materials and Methods

Collected leachate samples were characterized in terms of major cation concentrations, major anion concentrations, pH, conductivity, total organic carbon (TOC) and chemical oxygen demand (COD). Instruments that are used to characterize the leachate samples are shown in Figure 2. Major cations including calcium (Ca^{2+}), sodium (Na^+), potassium (K^+) and ammonium

Figure 2. Instruments used for the leachate characterization: (a) Multi-parameter meter for pH, conductivity and ammonium measurement; (b) MP-AES for cation measurements; (c) IC for anion measurements; (d) TOC analyzer for TOC measurement; (e) COD reactor and (f) spectrophotometer for COD measurement.

(NH_4^+) were characterized. NH_4^+ was measured using multi-parameter meter (HQ440D, HACH) (Figure 2a), while other cations were measured using MP-AES (4100, Agilent Technologies) (Figure 2b). Trace metals including aluminum (Al), arsenic (As), cadmium (Cd), copper (Cu),

iron (Fe), lead (Pb), magnesium (Mg), nickel (Ni), and zinc (Zn). Anions including chloride (Cl^-), bromide (Br^-) and sulfate (SO_4^{2-}) were measured using ion chromatography (IC) (Dionex Aquion, Thermo Scientific) (Figure 2c). Cations and anions were measured using filtered samples passing through 0.45 µm syringe filter (Acrodisc). TOC was measured using TOC Analyzer (Phoenix 8000, Teledyne Tekmar) (Figure 2d). COD was measured using the spectrophotometer (DR3900, HACH) (Figure 2f). pH and conductivity were measured using multi-parameter meter (HQ440D, HACH) (Figure 2a).

1.2.2 Results and Discussion

Water chemistry parameters of collected samples including pH, conductivity, COD and TOC are listed in Table 2. The pH of collected samples are in the range of 6.5-8.5, which are typical for landfill leachate (Kjeldsen et al., 2002; Townsend, 2015). AM has the highest conductivity which indicates the high ionic strength. The highest TOC in MSW can be contributed by the high organic carbon component (e.g. food waste) in MSW accepted by the corresponding landfill.

**Table 2. Water chemistry parameters of collected leachate samples.**

Sample Abbreviation	pH	Conductivity (ms/cm)	COD (mg/L)	TOC (mg/L)
CA	7.49	15.72	2963	1145
IG	7.34	2.84	172	26
AM	6.71	121.88	4450	109
CD1	7.12	51.32	5215	715
CD2	6.89	25.53	1157	172
CD3	6.46	60.70	1743	124
CD4	8.50	18.5	743	159
MSW	7.53	21.15	8983	1491

The cation and anion concentrations of each sample are depicted in Figure 3. Characterization results suggest Na^+ and K^+ are the major monovalent cations and Ca^{2+} is the major divalent cation in collected leachate samples. Additionally, Cl^- has the highest concentration among the measured anions. Among collected samples, AM has the highest Na^+, Ca^+ and Cl^- concentration which indicates its high ionic strength. This result is consistent with AM's highest conductivity as discussed previously. During combustion process, mineral salts are not volatilized and are concentrated in incineration ash which leads to the high concentration of Na^+, Ca^{2+} and Cl^- in leachate (Townsend, 2015). Comparing with CD and MSW samples, NH_4^+ concentration is low in sample AM since MSW incineration ash has little ammonium containing components left.
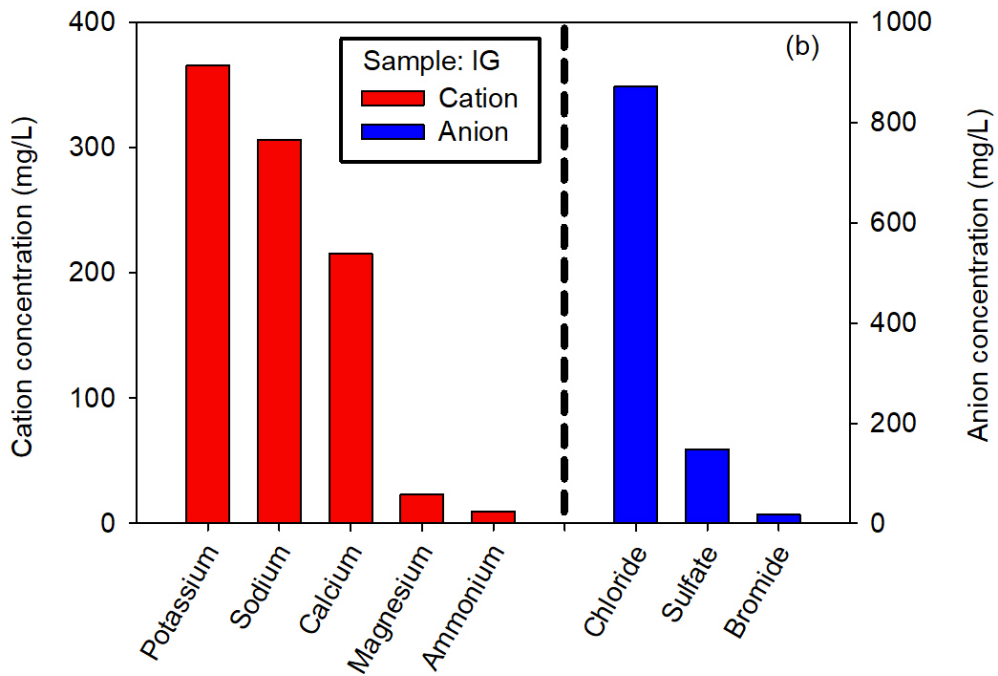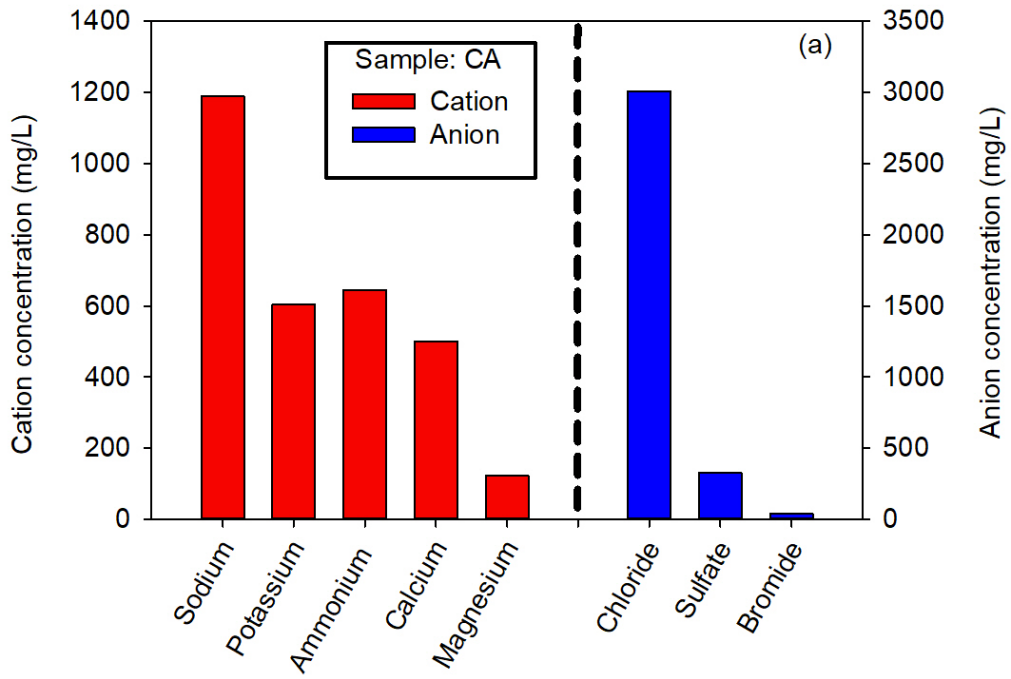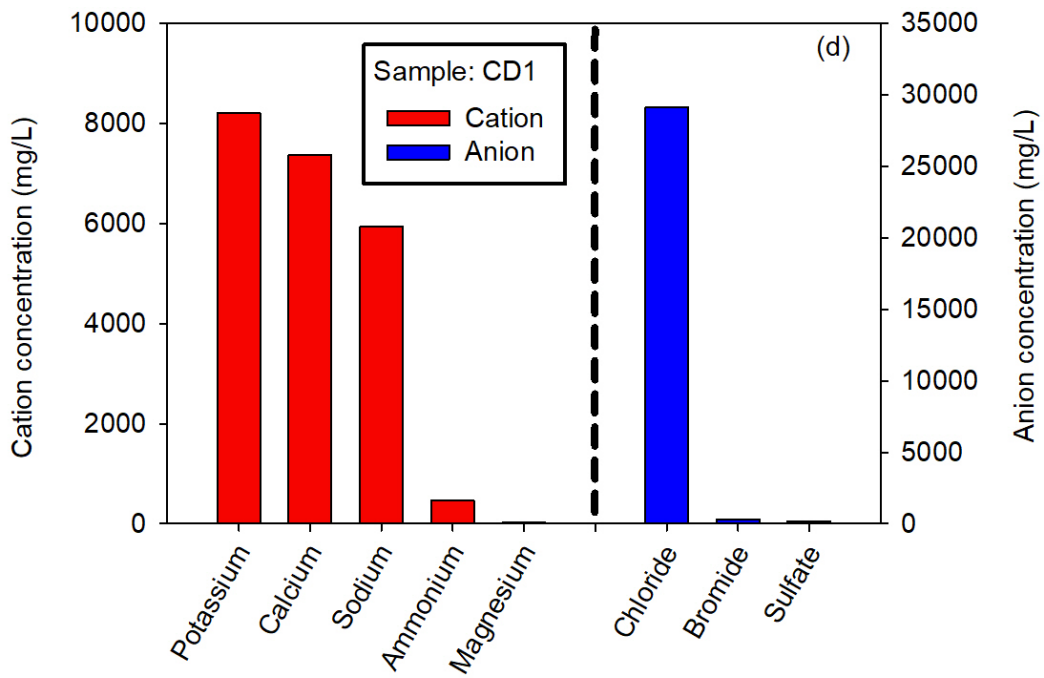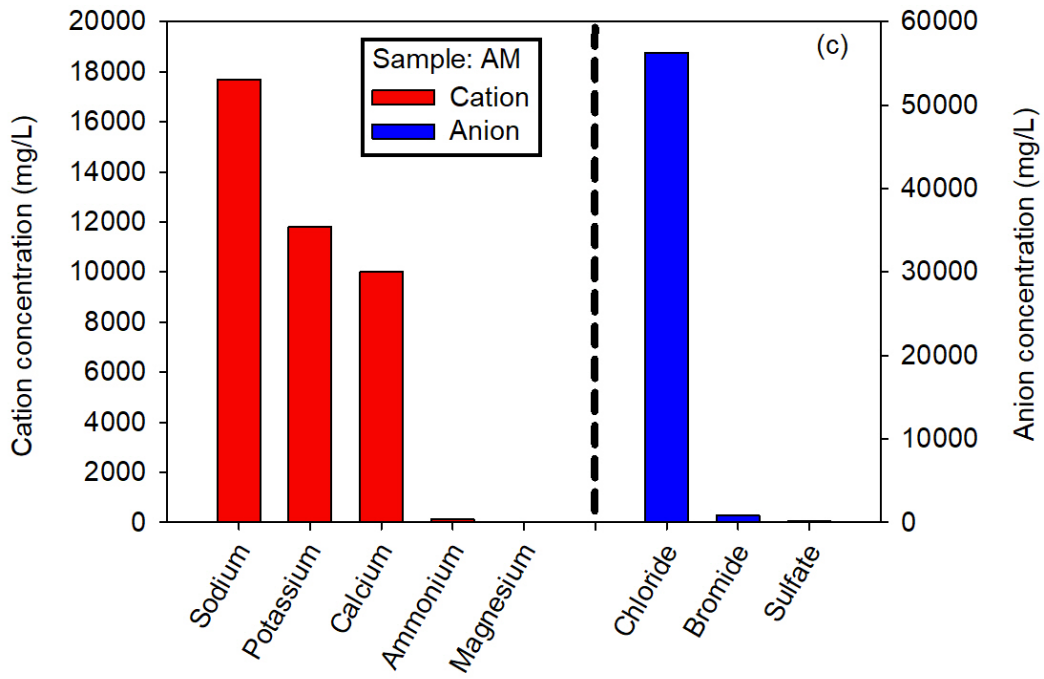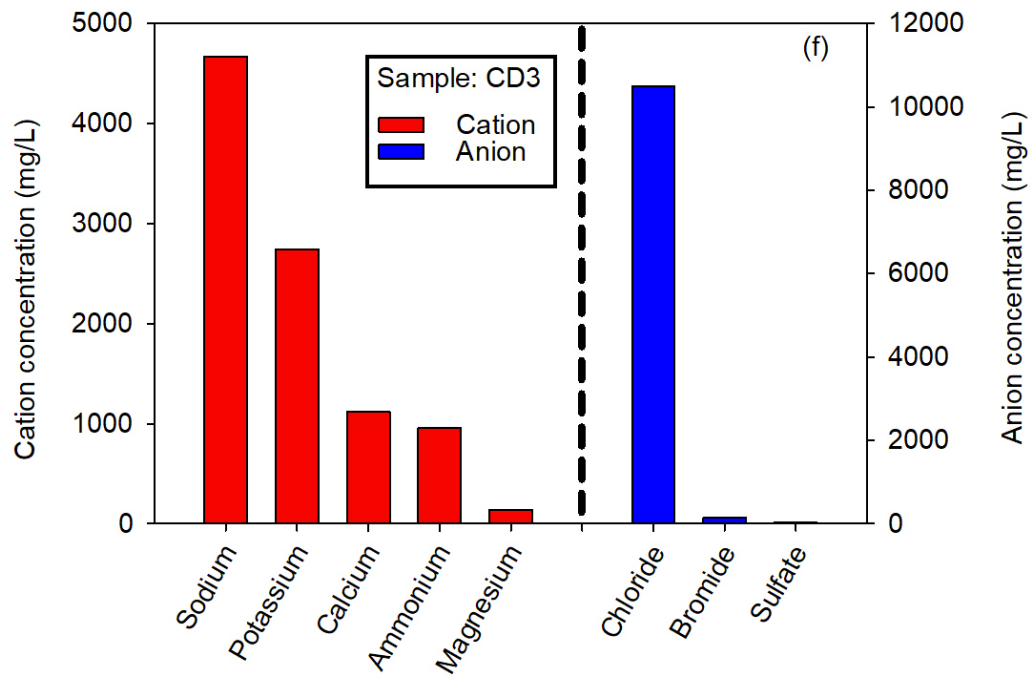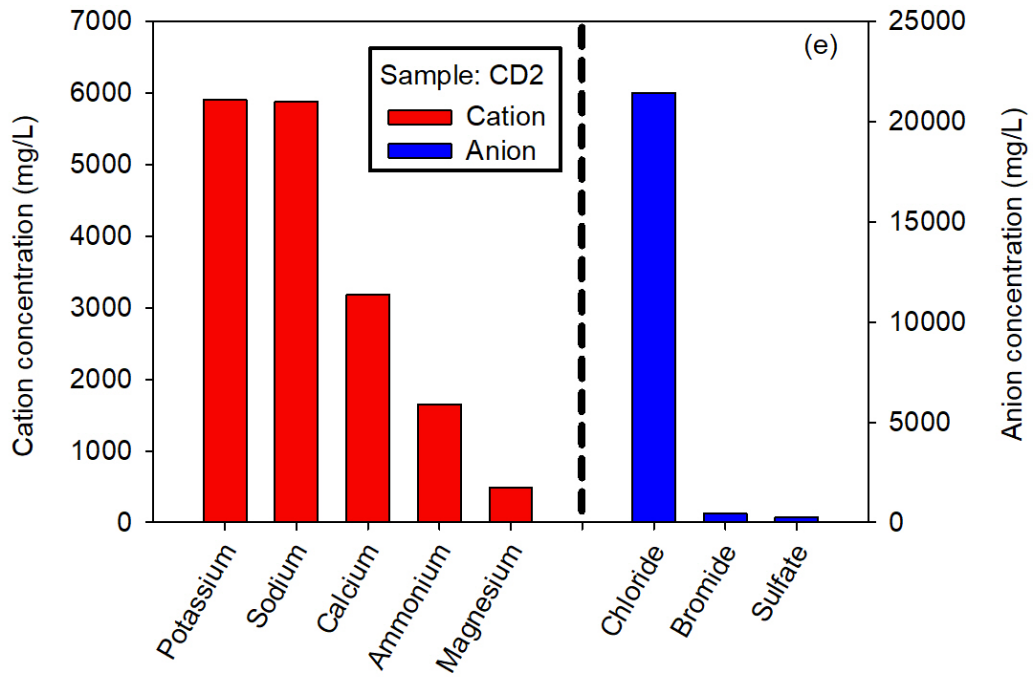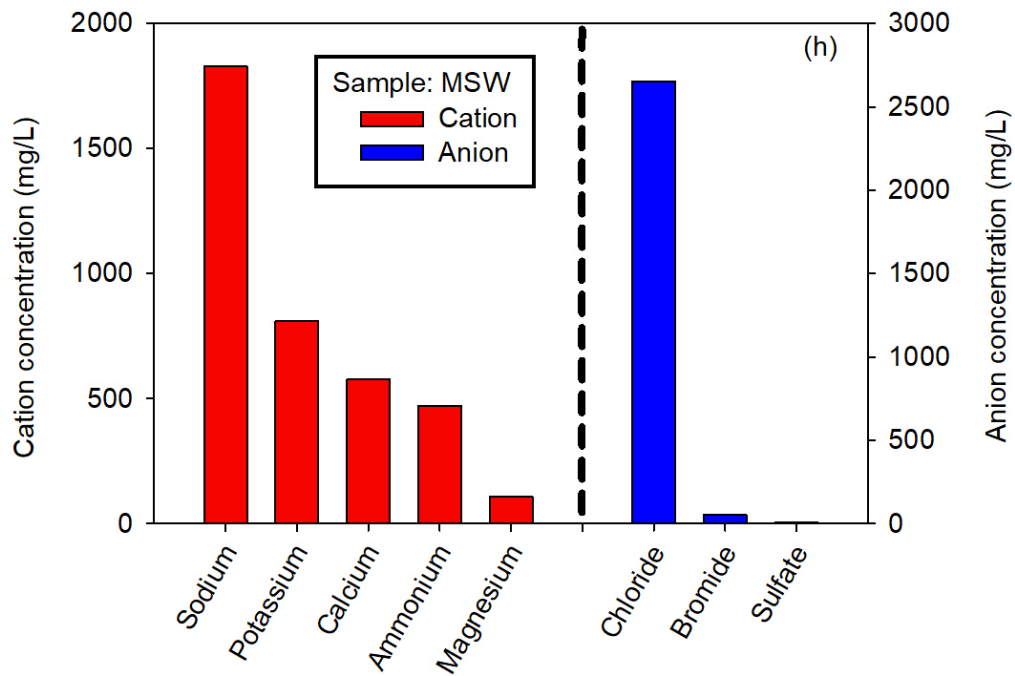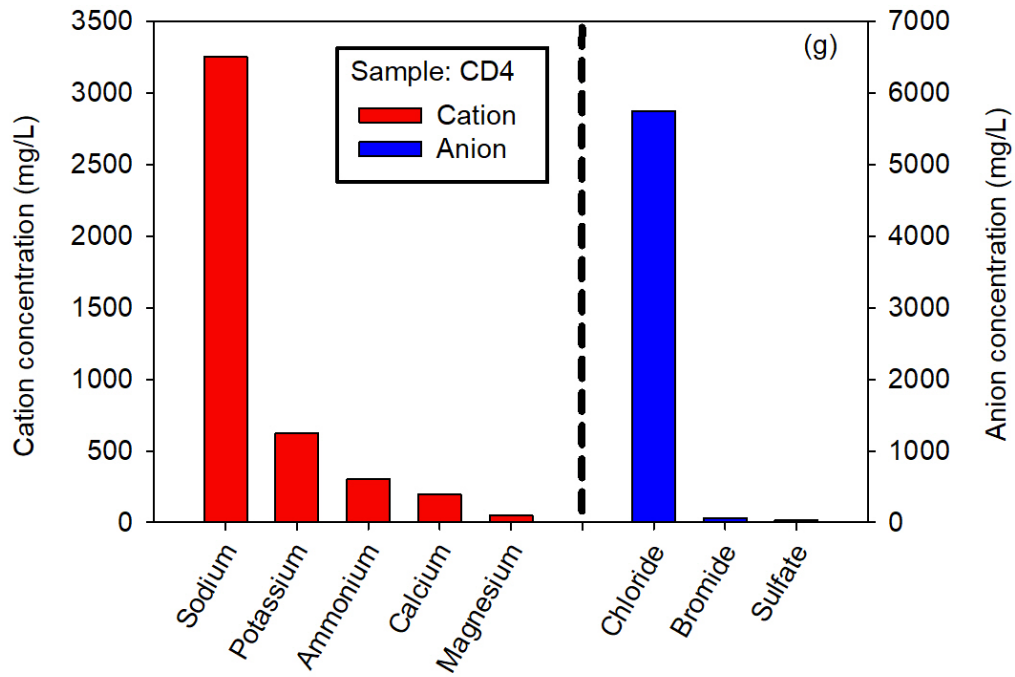
Figure 3. Cation and anion concentrations of sample (a) CA, (b) IG, (c) AM, (d) CD1, (e) CD2, (f) CD3, (g) CD4 and (h) MSW.

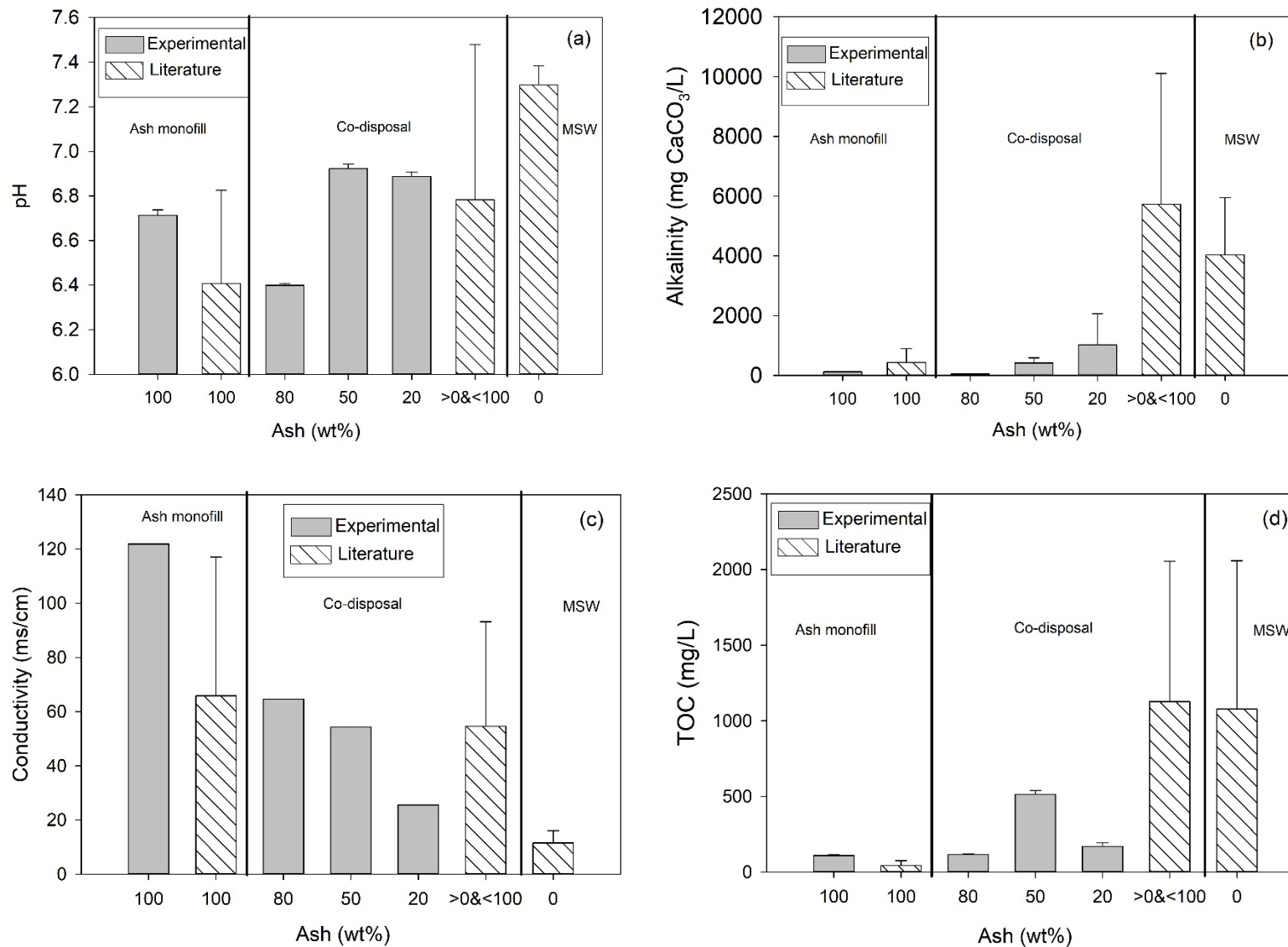1.3 Influence of MSWI Bottom Ash on Leachate Characteristics

1.3.1 Influence of MSWI Bottom Ash on Water Chemistry Parameters

Water chemistry parameters including pH, alkalinity, conductivity and TOC were compared for leachate samples at different ash percentages. The influence of bottom ash on these parameters are shown in Figure 4. Results from literature (Townsend, 2015) were also added for a comparison. Error bars in the figure represent the standard deviation of the corresponding parameter. Results suggest that ash percentage has little influence on the pH of leachate. Literature results show the large variation in pH of leachates where MSWI bottom ash is added at the associated landfill. When comparing the pH of co-disposal and ash monofill leachates to the MSW leachate, the MSW leachate has relatively higher pH. However, this is inconsistent with the results from laboratory leaching tests on MSWI bottom ash, which suggest the leachate pH increases as the MSWI bottom ash percentage increases (He et al., 2017). There are two possible reasons for the lower pH observed in the co-disposal and ash monofill leachates from real leachate: (1) the low pH of co-disposal and ash monofill leachates can be caused by the carbonation process, which involves the adsorption of carbon dioxide (CO_2) and precipitation of carbonate minerals. The source of CO_2 may be from the atmosphere as well as the biodegradation of the organics in the landfill (Belevi et al., 1992); (2) rainwater, which can low the pH of leachate, has more influence on co-disposal and ash monofill than MSW landfill. Considering the large amount of precipitation in Florida, leachates from the co-disposal and ash monofill are diluted and the pH can be reduced. However, MSW leachate may have stronger buffer capacity than co-disposal and ash monofill leachates due to the higher CO_2 amount caused by MSW biodegradation. This can reduce the effect of rainwater on the pH of MSW leachate.

Comparing to MSW leachate, co-disposal and ash monofill leachates have lower alkalinity (Figure 4 (b)). The low alkalinity in co-disposal and ash monofill leachate reflects the leachate's weak buffer capacity caused by the lack of biodegradation process occurred at associated landfill (Naveen et al., 2014). The lack of significant amount of organic compositions (e.g., food wastes) in the co-disposal and ash monofill inhibits the biodegradation process that can produce biocarbonate/carbonate and therefore result in the low alkalinity. Literature results have shown the co-disposal landfill leachate has the highest average alkalinity, however, its variation is large. As discussed above, the low alkalinity in co-disposal and ash monofill leachates contributes to the low pH by magnifying the negative effect of rainwater.

Influence of ash percentage on conductivity is also depicted in Figure 4(c). The conductivity of the leachate increases with the increase of ash percentage in associated landfill, suggesting the total concentration of dissolved salts and metals in the leachate increases as the ash percentage increases. Studies have also shown the conductivity is correlated with Cl, Na and K, which are the major cations and anion in the leachate (Vadillo et al., 1999). Literature have also shown the similar increasing trend of the conductivity from MSW leachate to leachate obtained from landfill where MSWI bottom ash is disposed of, although there is large variation in the conductivity of co-disposal landfill and ash monofill leachate.

TOC was also investigated in terms of ash percentage in this study. The TOC of ash monofill leachate is lower than 110 mg/L and the TOC of co-disposal landfill leachates vary but are relatively higher than that of ash monofill leachate. Literature have shown that there is a large variation in the TOC of MSW leachate, with an average of 1200 mg/L. Many factors such as MSW composition and age of the waste can contribute to the large variation in TOC (Ragle, et al., 1995; Statom et al., 2004).

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Figure 4. Change of water chemistry parameters including pH (a), alkalinity (b), conductivity (c), and TOC (d) in the leachate at different associated ash percentages. Literature data is from Townsend (2015).

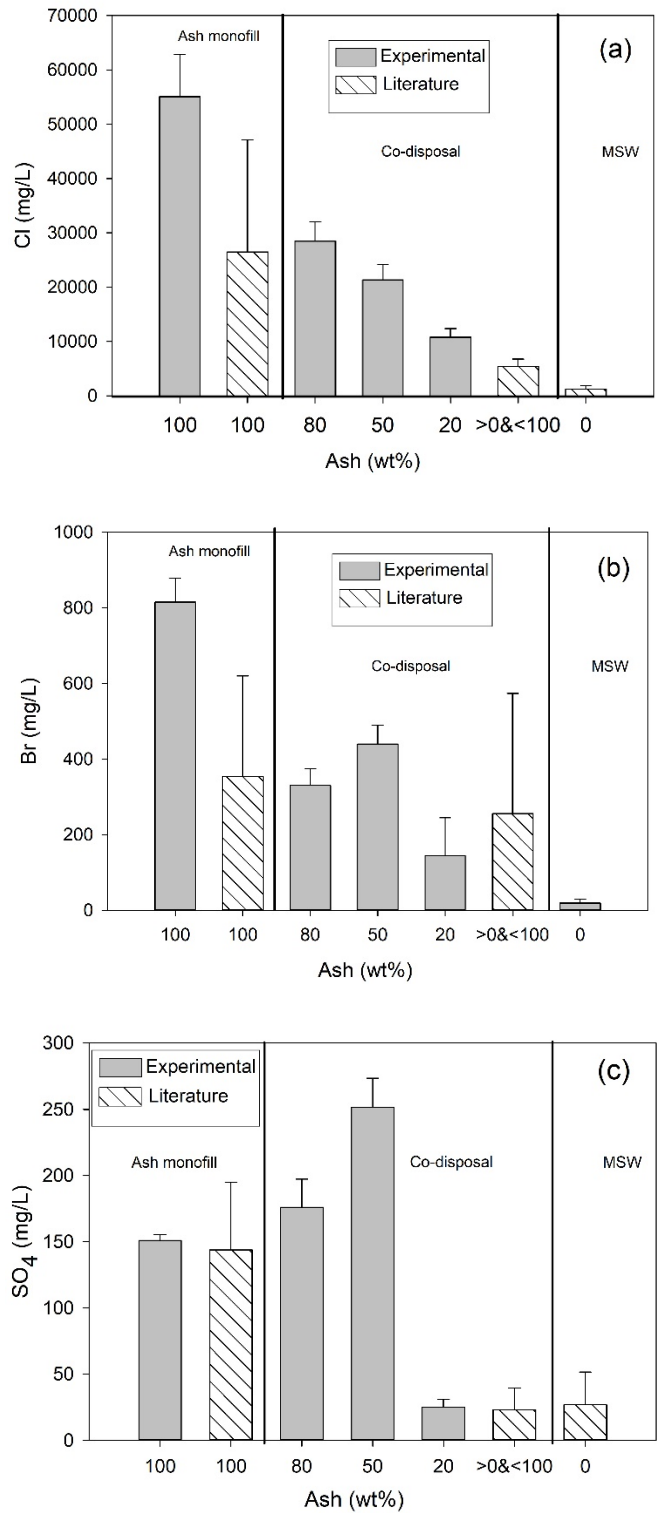*1.3.2 Influence of MSWI Bottom Ash on Major Cations*

Leaching studies have also shown Ca, Na and K are the major cations present in ash leachates (Karoline, 2012; Steenari et al., 1999) and they were measured as major cations in this study. Experimental results suggest that with the increase of ash percentage, concentrations of Ca, Na and K increase in the leachate (Figure 5). This is due to the increasing concentration of these cations as the MSWI bottom ash proportion increases in the landfill. Literature have also shown the increasing trend of Ca, Na, K concentrations with ash from MSW leachate to co-disposal and ash monofill leachates although large variation is present. In the ash monofill leachate investigated in this study, average concentration of Ca, Na and K can reach 10000, 17700 and 11790 mg/L, respectively. However, ash characterization results suggest that the mass sequence of the major cations are Ca>Na>K (Data not shown). XRD results suggest that a certain amount of Ca is in the form of calcite (Data not shown) which are not mobilized and trapped in MSWI bottom ash. Other soluble Ca species including calcium hydroxide, calcium sulfate, calcium oxide and Cl can migrate to leachate depending on the physico-chemical parameters such as pH and ionic strength. (Rendek et al., 2006; Lo and Liao, 2007). Na and K are in the form of soluble mineral and can easily migrate to leachate. Leaching tests on MSWI bottom ash have shown the rapid dissolution of Na and K until exhaustion (Kirby and Rimstidt, 1994; Dijkstra et al., 2006).

1.3.3 Influence of MSWI Bottom Ash on Major Anion

Anion concentration including Cl, Br and SO_4 in leachates at different ash percentages were measured. Cl concentration increases with the ash percentage as shown in Figure 6. Cl is in the form of soluble salt (Dijkstra et al., 2006) and can migrate to leachate easily. Br concentration has the similar trend as Cl, since Br is also a soluble anion. Although leachate associated with 80% MSWI bottom ash is an exception, the Br concentration in it is close to that in leachate associated with 50% MSWI bottom ash. Literature results have also shown ash monofill leachate has the highest concentration of Br, while the MSW leachate has the lowest, although large variation is present. SO_4 concentration in leachate is higher when associated ash percentage is equal to and above 50%. Leaching of SO_4 is controlled by the precipitation/dissolution of various sulfate-containing minerals (e.g., gypsum and ettringite). Specifically, sulfate-calcite relationship cannot be described only by precipitation/dissolution but a more complex relationship (Piantone et al., 2004). It is possible a certain amount of the SO_4 is trapped in waste with metals at high ash percentage (> 50% MSWI bottom ash), causing the decreasing trend of SO_4 concentration in the leachate with ash percentage. The low content of SO_4 in the waste at low ash percentage (< 50% MSWI bottom ash) is likely to result in the low concentration of SO_4 in the leachate.
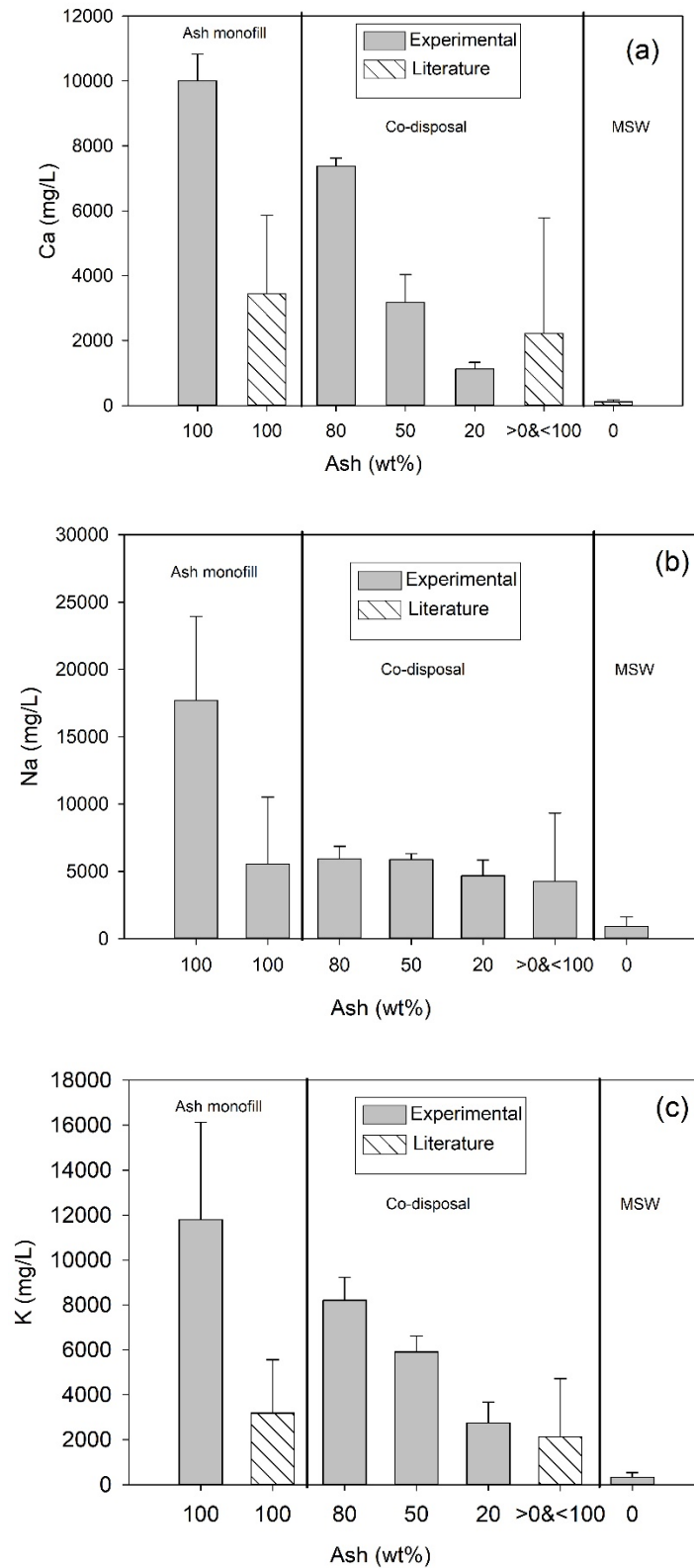
Figure 5. Change of major metals including Ca (a), Na (b), and K (c) in the leachate at different associated ash percentages. Literature data are from Townsend (2015).
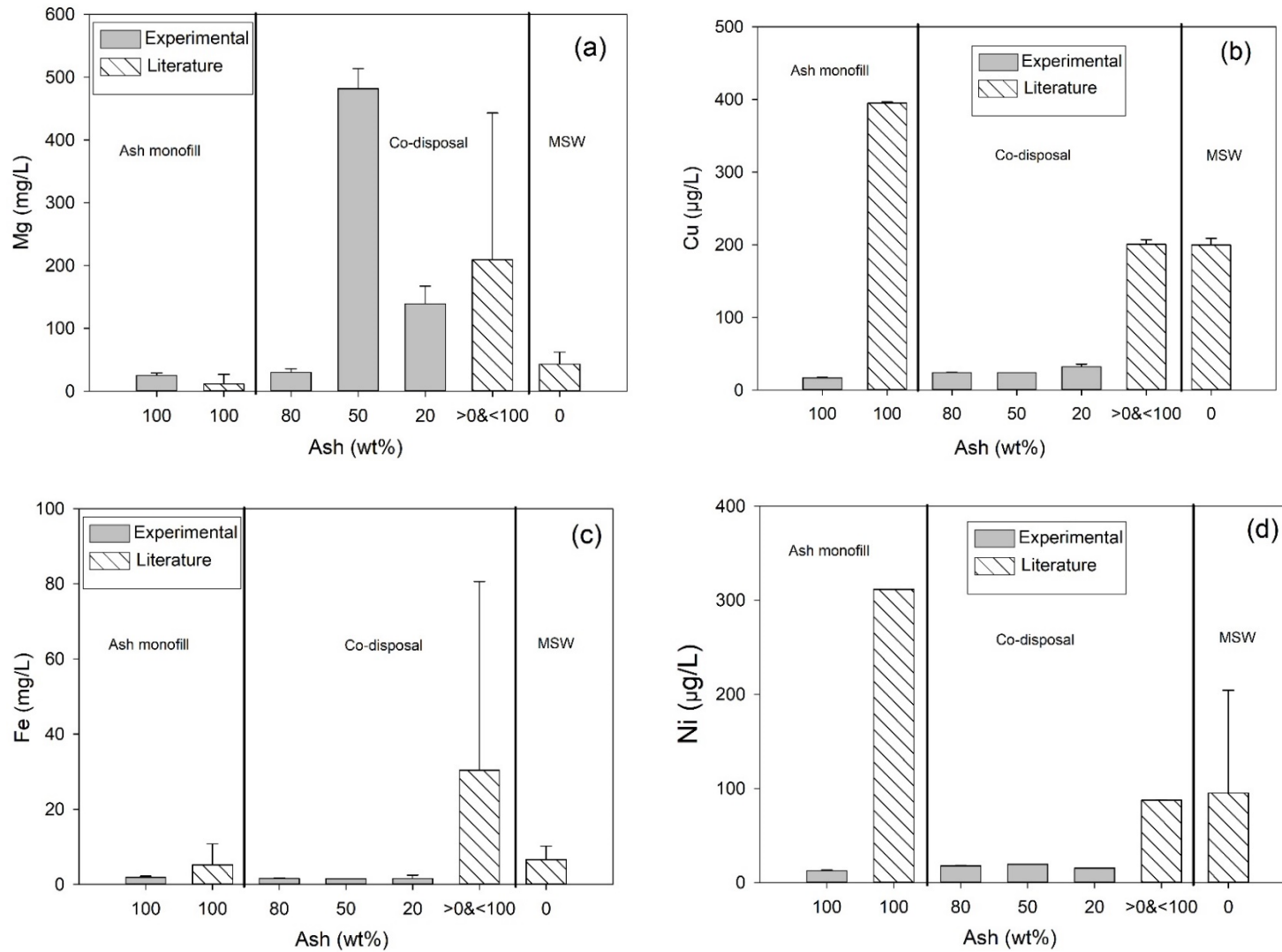
Figure 6. Change of major anions including Cl (a), Br (b), SO₄（c）in the leachate at different associated ash percentages. Literature data are from Townsend (2015).

3.3 Influence of MSWI Bottom Ash on Trace Metals

Besides the major cations studied above, trace metals including Al, As, Cd, Cu, Fe, Mg, Ni, Pb and Zn were also investigated. Results indicate that Cu, Fe, Mg and Ni are existed in all leachates (Figure 7). Leaching behavior of trace metals from leachate has been studied and several leaching mechanisms have been reported, including (1) mineral dissolution/precipitation; (2) complexation to inorganic ligands such as calcite, aluminum hydroxides, iron hydroxide and other minerals; and (3) complexation to organic ligands such as fulvic acid (Dijkstra et al., 2006; Meima and Comans, 1997; Zhang et al., 2008; van Zomeren and Comans, 2004). Other factors such as waste particle size (Lo, 2005), pH and weathering time (Dijkstra et al., 2006) can also contribute to the leaching of trace metals.

The co-disposal landfill leachate (associated with 50% and 20% MSWI bottom ash) has high concentration of Mg, while the MSW leachate and ash monofill leachate have low concentration of Mg, which is consistent with the results from literature (Figure 7). It has been reported that Mg is existed in the form of brucite ($Mg(OH)_2$), the secondary mineral such as sepiolite ($Mg_2Si_3O_6(OH)_4 \cdot 1.5H_2O$) (Meima and Comans, 1997) and Mg-Al-layered double hydroxide (Dijkstra et al., 2006) in weathered MSWI bottom ash. This can be a combined effect of Mg content in the landfill and leachability of Mg which can be controlled by the dissolution and precipitation of $Mg(OH)_2$ and secondary Mg-containing minerals (Meima and Comans, 1997; Dijkstra et al., 2006). At higher ash percentage, it is possible that a certain amount of Mg is in the form of insoluble Mg-containing minerals and cannot transport to leachate.

Ash percentage has little influence on Cu, Fe and Ni concentrations in studied leachates, and these concentrations are lower than that reported in the literature. This can be caused by the variation in the landfill waste composition, pH and wreathing time. MSWI bottom ash characterization results suggest the Cu, Fe and Ni contents are 1.8 mg/g, 12.2 mg/g and 85.3 µg/g dry ash, respectively (Data not shown). While, the Cu, Fe and Ni concentration in associated leachate are 16.76 µg/L, 1.85 mg/L and 12.63 µg/L, respectively. The leaching of Cu from MSWI bottom ash can be described by the surface complexation to Fe and Al containing minerals (Dijkstra et al., 2006) and organic ligands (Meima and Comans, 1999; Meima et al., 1999). Studies have shown Cu is largely complexed to dissolved organic carbon (Meima et al., 1999; Meima and Comans, 1998 and 1999; van Zomeren and Comans, 2004) and the low concentration of total organic carbon in the MSWI bottom ash can probably contribute to the less leaching of Cu. It has been reported that leaching of Fe from weathered MSWI bottom ash is controlled by the complexation to organics such as humic and fulvic acid (Dijkstra et al., 2006). The Ni leaching has been reported to be controlled by precipitation/dissolution of nickel hydroxide and surface complexation to hydrous ferric oxide (Zhang et al., 2008).

Figure 7. Change of trace metals including Mg (a), Cu (b), Fe (c), and Ni (d) in the leachate at different associated ash percentages. Literature data is from Townsend (2015).

Al, As, Pb and Zn were also measured and they were detected in at least one of the leachate samples investigated in this study (Table 3). Leaching of Al, As, Pb and Zn can be a combined effect of availability of these heavy metals in associated landfill and their leachability. Al concentration determined in the co-disposal leachate (with 50% ash) and ash monofill leachate are less than 1 mg/L, which is consistent with literature results (Table 3). MSWI bottom ash characterization indicates that Al in the ash has a concentration of 80.45 mg/g which is the second highest (Data not shown). However, Al concentration in associated ash monofill leachate is not high as other metals such as Fe. The leaching of Al is pH dependent and controlled by solubility behavior of Al-containing minerals such as gibbsite (Dijkstra et al., 2006). Precipitation of gibbsite can occur in the landfill when pH is in the range of 4-8 (Dijkstra et al., 2006), causing less leaching of Al from studied ash monofill. As, Pb and Zn are present in the leachate associated with 20% of MSWI bottom ash, but not in other co-disposal and ash monofill leachates. Leaching of Pb can be controlled by the complexation to organic ligands such as humic acid and fulvic acid (Dijkstra et al., 2006) and leaching of Zn is controlled by the precipitation/dissolution of zinc carbonate ($ZnCO_3$) in the studied pH range (6-8) (Zhang et al., 2008). At low ash percentage, the relatively high concentration of organics can contribute to the leaching of Pb and Zn, by forming the soluble complexation to Pb and forming $ZnCO_3$, respectively. The leaching of As from landfill is less studied and the leaching mechanism is unclear. It is possible the organics in the leachate associated with low ash percentage facilitates the leaching of As.

Table 3. Trace metals detected in at least one of the leachate samples.

Ash (wt%)	Al (mg/L)	As (µg/L)	Pb (µg/L)	Zn (µg/L)
Experimental				
100	0.8	<20	<10	<10
80	<0.01	<20	<10	<10
50	0.68 (0.54)[a]	<20	<10	<10
20	<0.01	133 (11)	56.5 (7.9)	36.0 (30.3)
Literature[b]				
100	0.70 (0.47-0.93)[c]	79.7 (16.4-285)	77.9 (1.20-77.9)	50.8 (50.8-386)
>0&<100	0.56 (0.54-0.58)	201 (7.81-418)	10.5 (4.08-47.2)	93.7 (43.7-163)
0	1.92 (0.60-6.98)	55.9 (1.22-368)	76.6 (1.35-77.5)	39.8 (12.4 -1.20×10³)

a. In experimental data, values in the parentheses represent standard deviation.
b. From Townsend (2015).
c. In literature data, values in the parentheses represent the range of associated parameter.

Although detected in the MSWI bottom ash (Data not shown), concentration of As, Pb and Zn are lower than quantification limits in associated ash monofill leachate. Besides the low concentration of organics in ash monofill, other parameters not considered in this study such as particle size and weathering time may also inhibit the leaching of As, Pb and Zn by adsorbing the heavy metals to the waste (Meima and Comans, 1999; Shim et al., 2003). Cd is below the quantification limit in all studied leachates. However, chemical composition characterization of MSWI bottom ash indicates the presence of Cd (58 µg/g dry ash) in the ash (Data not shown). Comparing the MSWI bottom ash characterization results to the chemical compositions of associated leachate, toxic metals that bring environmental and health concern such as As, Cd, Pb and Zn in the leachate are lower than quantification limit, while other trace metals such as Cu (16.8 µg/L) and Ni (12.6 µg/L) can meet the sewer discharge limitations regulated by counties in Florida (Code of Ordinance of Miami-Dade County, 2014; Code of General Ordinary of the City of Tallahassee, 2017). Because of the low concentration, these trace metals in ash monofill leachate can serve as nutrients to microorganisms when discharged at wastewater treatment plant, instead of exerting inhibitory effect. However, some factors such as waste particle size and weathering time that may influence the leaching of heavy metals are not considered in this study. To fully understand the leaching of metals from landfill and the influence of ash percentage on associated leachate, these factors need to be considered in further study.

2. Influence of MSWI Bottom Ash on LCS Clogging

Precipitations associated with Ca-containing minerals such as calcite has been identified as the main reason for LCS clogging. The Visual MINTEQ software 3.1 (Gustafsson, 2016) was applied to calculate the saturation index (SI) of minerals which can be the possible precipitations associated with calcium in the leachate at equilibrium (He et al., 2017; Herck et al., 2000). Leachate characteristics including pH, dissolved inorganic carbon including carbonate and bicarbonate which are calculated based on alkalinity, concentrations of major cations and anions measured in above section were added as the model input and the SI, as the model output, was calculated. The positive SI indicates the possibility of precipitation formation.

Saturation indexes of possible precipitations associated with Ca were calculated using Visual MINTEQ and listed in Table 4. Positive SI indicates there is a risk of precipitation of the corresponding minerals. The higher the SI, the more risk of the precipitation (He et al., 2017). Results suggest aragonite, calcite, dolomite, vaterite and magnesites can be the reason for LCS clogging. These possible precipitations are also observed by other geochemical modeling studies (Bennett et al., 2000; Cardoso and Levine, 2009). With the increase of ash percentage in the landfill, there is a higher risk of LCS clogging. Specifically, ash monofill has the highest risk of LCS clogging in terms of precipitations including aragonite, calcite and vaterite. Landfill associated with 80% of the MSWI bottom ash has the highest risk of dolomite precipitation. MSWI bottom ash addition is likely to result in the increasing concentration of Ca and Mg in the leachate, while MSW degradation can lead to the formation of dissolved inorganic carbon (e.g., bicarbonate and carbonate). The increasing risk of the clogging is a result of combined effect of increasing concentration of Ca and Mg and decreasing concentration of dissolved inorganic carbon in the leachate associated with high ash percentage.

Table 4. Saturation index of the minerals in leachate at different ash percentages.

Mineral	Chemical formula	Ash percentage (%)			
		100	80	50	20
Aragonite	$CaCO_3$	2.295	1.756	0.675	0.459
Calcite	$CaCO_3$	2.439	1.899	0.819	0.603
Dolomite (disordered)	$CaMg(CO_3)_2$	1.032	1.125	0.542	0.065
Dolomite (ordered)	$CaMg(CO_3)_2$	1.582	1.675	1.092	0.615
Gypsum	$CaSO_4 \cdot 2H_2O$	-0.283	-0.659	-0.628	-1.972
Vaterite	$CaCO_3$	1.872	1.333	0.252	0.037

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Information Dissemination Activities: We are working on Draft Papers to showcase the testing completed on the Geosynthetic Clay Liners and leachate characterization.

Metrics:

1. List of graduate student or postdoctoral researchers **funded** by **THIS** Hinkley Center project

Last name, first name	Rank	Department	Professor	Institution
Bently Higgs		Civil & Environmental Engineering	Dr. Tarek Abichou	FAMU-FSU College of Engineering
Christian Wireko		Civil & Environmental Engineering	Dr. Tarek Abichou	FAMU-FSU College of Engineering
Dr. Liang Li		Civil & Environmental Engineering	Dr. Tarek Abichou & Dr. Youneng Tang	FAMU-FSU College of Engineering

2. List undergraduate researchers working on **THIS** Hinkley Center project

Past Undergraduate Researchers
- Name: Alyssa Schubert
  Department: Environmental
  Science
  Professor: Dr. Tarek Abichou, Ph.D, P.E.
  Institution: FAMU-FSU College of
  Engineering

- Name: Nora Sullivan
  Department: Environmental Science
  Professor: Dr. Tarek Abichou, Ph.D,
  P.E.
  Institution: FAMU-FSU College of Engineering

Present Undergraduate Researchers

- Name: David Carbajal
  Department: Civil and Environmental
  Engineering Professor: Dr. Tarek Abichou,

Ph.D, P.E. Institution: FAMU-FSU College of
Engineering

- Name: Tristan Wahl
  Department: Mechanical Engineering
  Professor: Dr. Tarek Abichou, Ph.D, P.E.
  Institution: FAMU-FSU College of
  Engineering

- Name: Avery VanRussel
  Department: Civil and Environmental
  Engineering Professor: Dr. Tarek Abichou,
  Ph.D, P.E. Institution: FAMU-FSU College of
  Engineering

3. List research publications resulting from **THIS** Hinkley Center project (use format for publications as outlined in Section 1.13 of this Report Guide).

4. List research presentations (as outlined in 1.13.6 of this Report Guide) resulting from **THIS** Hinkley Center project.

5. List who has referenced or cited your publications from this project?
    .

6. How have the research results from **THIS** Hinkley Center project been leveraged to secure additional research funding?

7. How have the results from **THIS** Hinkley Center funded project been used (**not** will be used) by FDEP or other stakeholders? (1 paragraph maximum).

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