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Leaching of Metals, Organic Carbon and Nutrients from Municipal Waste under Semi-Arid Conditions

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ABSTRACT: A large portion of municipal waste, disposed in landfills, is organic in nature. This organic material starts decomposing soon after the disposal in landfills. The decomposition leads to the generation of in situ leachate, and this leachate can contain substantial amounts of contaminants. Under arid conditions, leachate is solely generated by waste decomposition, as little input of external water occurs due to rain. The objective of this study was to characterize the in situ leachate from typical waste streams in Pakistan. We collected fresh municipal solid wastes typical of household, mixed, and restaurant waste, and analyzed the leachates for volume and water quality parameters. The wastes released up to 30% of their initial moisture as leachate and the leachate contained metals, organic carbon, and nutrients at concentrations many times higher than the environmental water quality standards for waste water discharge. As landfill leachate in Pakistan is usually not contained or collected, the in situ leachate is a source of surface and ground water contamination. Methods have to be developed and implemented to prevent leaching of contaminants into underlying soils and sediments, and subsequently into ground and surface waters.

Key words: Waste, Leachate, Settlement, DOC, Moisture

INTRODUCTION

Landfilling is a convenient and economical way of municipal solid waste disposal, but if done improperly, it adversely affects air, soil, and water quality by liquid and gas emissions (El-Fadel *et al.*, 1997). These emissions are dependent upon the waste composition, initial moisture content, percolation, weather conditions, degree of compaction, and waste decomposition rates. In many countries, particularly the developing countries, municipal solid waste is dumped directly to the open lands without any pre-treatment or processing. This creates aesthetic nuisance for people living around the dumpsite and can deteriorate the groundwater resources through leaching of contaminants (Kanmani and Gandhimathi, 2013). Leachate is the liquid owing from the lower boundaries of waste dumps driven by gravity. This leachate can mobilize contaminants and transport them to the underlying soils and ultimately to the groundwater. An important feature of leachates is the diversity and quantity of contaminants that they contain. Oman and Junestedt (2008) characterized leachates from 12 municipal landfills and identified 90 organic compounds and 50 inorganic elements. These contaminants adversely affect the environment upon exposure to the leachate. Toufexi *et al.* (2013) studied the impact of landfill leachate on mussels and human cells, and concluded that leachate can cause geno- and cyto-toxicity to marine biota as well as humans. Leachate quantity and quality depends on multiple factors in landfills. The major factors affecting the leachate quantity include water intrusions (like rain, snow, and groundwater), landfill operations (compaction, pre-treatment, and daily cover), physical characteristics (initial moisture content, permeability, age, density, and particle size) and internal processes (settlement and organic material decomposition rate). Leachate quality is mainly influenced by waste characteristics (composition and age), site operations (treatment, liquid waste co-disposal, and quality of water entering the landfill), and chemical reactions (biodegradation, adsorption, hydrolysis, dissolution, dilution, contact time, partitioning, and precipitation) (El-Fadel *et al.*, 2002). Kjeldsen *et al.* (2002) reviewed the long-term composition of municipal solid waste leachate and reported that the waste type and internal

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processes affect the leachate quality. Fresh wastes discharge leachates with higher contaminant concentrations (Zhao *et al*., 2013) than the aged waste. As the age of waste is increased, the contaminant concentrations in the leachate tend to decrease (Kulikowska and Klimiuk, 2008). Contaminant concentrations in leachate are strongly affected by the inputs of stormwater or overland flows (Trankler *et al*., 2005; Bendz *et al*., 1997). Leaching from landfills varies seasonally, and high contaminant concentrations are often associated with dry weather (Statom *et al*., 2004; Fan *et al*., 2006; Ziyang *et al*., 2009). When the leachate from waste is studied, the waste is often irrigated or exposed to precipitation (Korfiatis and Demetracopoulos, 1984; Weber *et al*., 2002). Leachate fluxes vary with climate but also with waste type. Leachate fluxes from waste in semi-arid and arid regions are expected to be less than in humid regions as there is limited external water input. Even in absence of external water input, the waste can generate in situ leaching. This in situ leaching can be substantial and can be a source of contamination.

The objective of this study was to evaluate the in situ leachate, both in terms of liquid quantity and contaminant concentrations, from typical municipal wastes in Pakistan under the absence of external water intrusions. We hypothesized that typical municipal waste in semi-arid and arid regions can generate in situ leachate, and that the leachate can contain contaminants in high concentrations, because leachate is not being diluted by external water sources. We collected leachates from freshly collected household, mixed municipal, and restaurant waste. Waste was covered to protect from external water inputs, and in situ leachate was analyzed for water quality parameters, i.e., metals, organic carbon, salts, and nutrients. Concentrations were compared to environmental quality standards to assess the potential risk of environmental contamination.

MATERIALS & METHODS

Three organic streams of fresh municipal solid waste were monitored for leachates: household waste, restaurant waste, and mixed waste. Household and restaurant wastes were collected from primary collection points, whereas mixed waste was taken from the Losar Dumpsite (official dumpsite of Rawalpindi City, Pakistan). Wastes were mixed manually as well as mechanically, and representative samples were taken by the quartering and coning method (ASTM, 2003). To characterize the waste, the waste streams were then sorted by waste component (e.g., yard waste, food waste, plastics), weighed on wet basis for each component, and the moisture content was determined. Moisture content was measured by heating each component at 60oC until the sample reached a constant weight (Wu *et al*., 2012). Then, the overall moisture content of the waste streams was calculated based on a weighted average of the individual components. Three high density polyethylene (HDPE) containers of equal size (0.85 m height, 0.54 m diameter) were used for the leaching experiments. A hole was made at the bottom of each container and a metal strainer was fixed to allow leachate to drain. The wastes were filled into the containers to an equal volume of 0.16 m^3 (i.e., packed to the height of 0.7 m). The containers were then tightly closed with polyethylene sheets to avoid evaporation and intrusion of water from precipitation. The wastes were not compacted; rather the wastes were allowed to settle at their own weights. The wastes were of similar age, i.e., freshly collected. The temperature during the experimental period ranged from 26 - 33°C with an average value of 29°C. Leachate was allowed to pass through the strainers under gravity into polyethylene sampling bottles, which were then collected for chemical analysis. Leachate was collected after every 1.25 L of outflow volume. The parameters analyzed were dissolved organic carbon (DOC), chemical oxygen demand (COD), total phosphorus (tot-P), total nitrogen (tot-N), pH, electrical conductivity (EC), total dissolved solids (TDS), chloride (Cl^-), and metals (Fe, Cu, Cr, Zn, Pb, Mn, and Cd). A closed reflux method (APHA, 2005) was used for measuring the chemical oxygen demand. The samples were filtered (0.45 µm) and digested in HNO3 before analyzing the metals with a Flame Atomic Absorption Spectrophotometer (Phoenix-986, 2010, Biotech Engineering Co. Ltd, UK) with a detection limit of 10 µg/L for all metals. Phosphorus was determined by a spectrophotometer (Spectronic Genesys 5) after digestion. A TOC analyzer (Analytik-jena, multi N/C UV HS with a detection limit of 2 µg/L) was used to measure DOC from 0.45 µm filtered samples. Correlations between waste characteristics and leachate concentrations were analyzed by Pearson correlation coefficients. Statistical analyses were done with R (R Core Team, 2013).

RESULTS & DISCUSSION

The detailed composition of the wastes is shown in Table 2. Household waste consisted of 100% green organic waste, whereas restaurant waste had 95% organics with 5% of plastics and Styrofoam crockery. In contrast, mixed waste consisted of 82% organics and 18% construction and demolition waste. Household waste was all degradable, while restaurant waste had some slowly degradable materials and mixed waste contained an inert fraction along with degradable waste. The initial moisture content of household waste, restaurant waste, and mixed waste was 3.17, 2.02, and 1.74 g of H_2O/g of dried mass of waste, respectively.

Table 1. Leachate fluxes produced by municipal solid wastes in different climates

Location	Climate	Waste type (% weight)	External water source	Experimental setup	Leachate flux: Outflow flux[a] (mm/day)	Leachate flux: Leachate Outflow/Inflow Ratio	Leachate flux: Time period (months)	Reference
Thailand	Tropical	Organics (59), plastics (24), glass (7), leather/rubber (5), ferrous metal (1)	Rainfall	Lysimeters	3.24	0.60	35	Trankler *et al.* (2005)
Bangladesh	Subtropical	Food and vegetable waste (93), plastics and polythene (3), leather and rubber (2), others (2)	Rainfall	Lysimeters	0.2	0.16	27	Rafizul *et al.* (2012)
Greece	Mediterranean	Organic (50), paper (18), plastics (7), metals (6)	Rainfall	Landfill	0.29	0.36	12	Tatis and Zouboulis (2002)
Lebanon	Mediterranean	Paper/cardboard (11.09), food waste (63.45), textiles (2.4), plastics (11.3), glass (4.6), metals (2.6), wood (0.15), others (1.5)	Rainfall	Landfill	1.4	0.66	18	El-Fadel *et al.* (2002)
Sweden	Temperate	Mixed household and industrial waste, sewage sludge	Rainfall	Lysimeters	0.16	0.25	78	Bendz *et al.* (1997)
South Africa	Semi-Arid	Paper (34), plastics (7), glass and metals (16), organics (30), ash and dust (2), unclassified (8)	Rainfall	Lysimeters	0.73	0.45	12	Fourie *et al.* (1999)
South Africa	Semi-Arid	Construction and demolition waste (64), paper and cardboard (11), plastics (8), metals (2.5), organics and textiles (2)	Rainfall	Lysimeters	0.08	0.02	61	Blight (2005)
Pakistan	Semi-Arid	Fruits/vegetables (58.4), grass clippings (26.8), shrubs and twigs (14.8)	None	Column	0.96	0.23	1.1	This study
Pakistan	Semi-Arid	Fruits/vegetables (49.9), grass clippings (15.2), shrubs and twigs (11.7), meat and bones (1.1), fabric (4.1), construction and demolition (17.9)	None	Column	1.09	0.28	1.5	This study
Pakistan	Semi-Arid	Fruits/vegetables (31.5), rice and snacks (13.8), meat and bones (17.5), mixed left over food with disposable crockery (32.2), plastics (5.0)	None	Column	0.98	0.30	1.7	This study

[a] Outflow fluxes are average fluxes over the sampling period.
[b] Inflow was zero.

Table 2. Characteristics of the waste types

Waste type	Mass (kg)	Water content (g of H_2O / g of DM)	Organic matter (% weight)	Density (kg/m³)	Composition (% weight)
Household	43.8	3.17	100	274	Fruits/vegetables (58.4), Grass clippings (26.8), and Shrubs and twigs (14.8)
Restaurant	63.7	2.02	95	399	Fruits/vegetables (31.5), Rice and Snacks (13.7), Meat and bones (17.5), Mixed left over food with disposable crockery (32.2), and plastics (5)
Mixed	55.5	1.74	82	348	Fruits/vegetables (49.9), Grass clippings (15.2), Shrubs and twigs (11.7), Meat and Bones (1.1), Fabric (4.1), and Construction and Demolition (17.9)

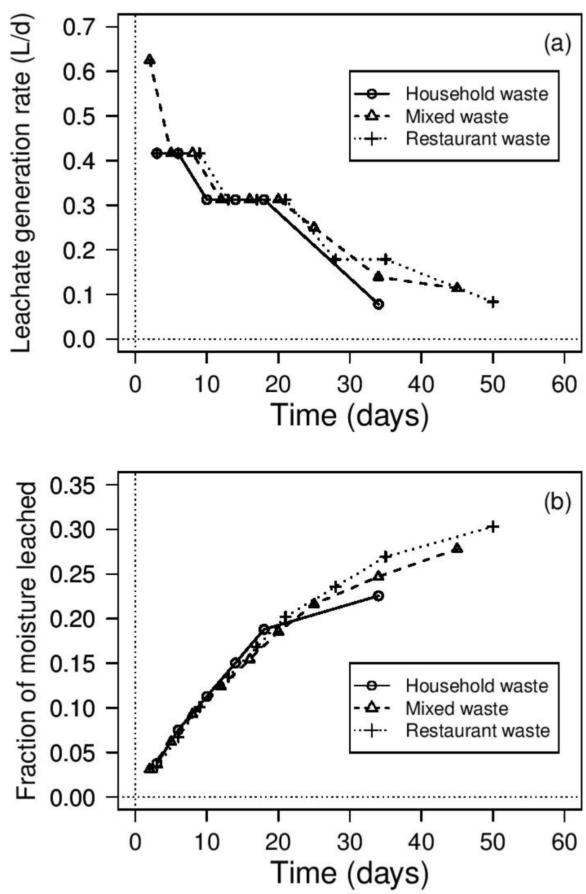

Fig. 1. Leachate outflow from the three different waste streams during the experiment period: (a) leachate fluxes, and (b) cumulative leachate volume normalized by the initial moisture content

Fig. 1a shows the leachate fluxes (mm/day) from different waste streams plotted against time elapsed after filling the waste containers. In the beginning, leachate fluxes were high for all wastes, but then the fluxes decreased until the leachate generation eventually ceased. The total volume of leachate generated by household waste, mixed waste, and restaurant waste was 7.5 L, 11.25 L, and 11.25 L, respectively. Drainage ceased after 34, 45, and 50 days for household waste, mixed waste, and restaurant waste, respectively. The cumulative leachate was normalized with the initial water content to measure the amount of volume leached (Fig. 1b). The total leachate volume ranged from 20 to 30% of the initial

Table 3. Settlement parameters of the three waste types

Waste type	Time (days)	Volume reduced (%)	Settlement height S_t (m)	Coefficient of settlement C_α
Household	34	74.4	0.52	0.49
Restaurant	50	40	0.28	0.24
Mixed	45	33.1	0.23	0.20

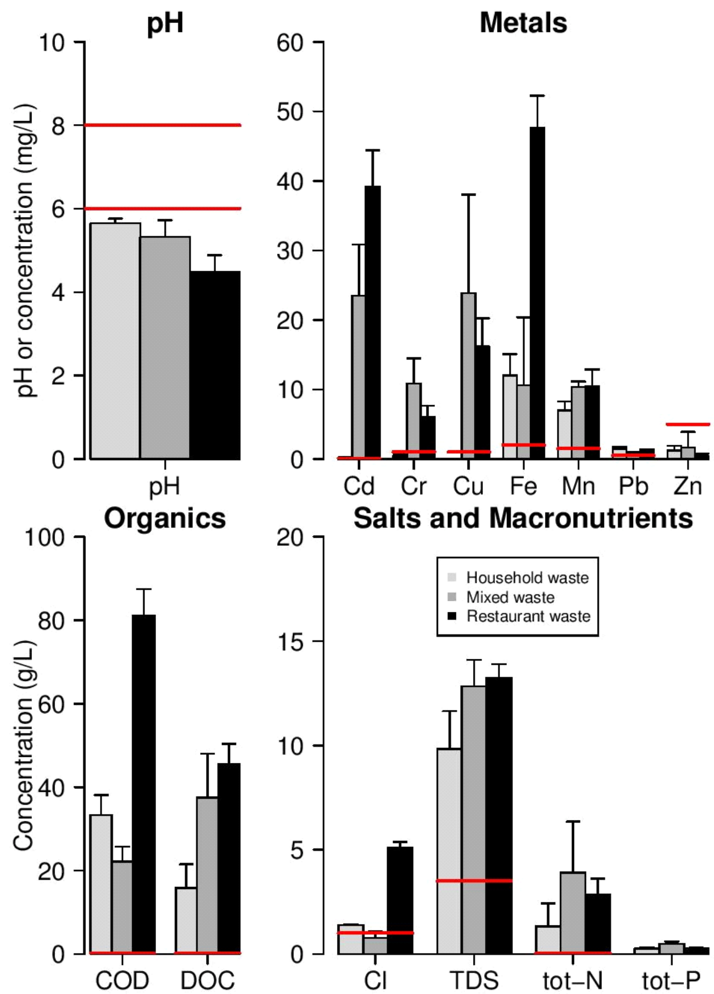

Fig. 2. Water quality parameters of the different waste leachates. Error bars indicate one standard deviation; solid red lines indicate range (pH) and limits of environmental quality standards

moisture content. Some of the initial moisture of municipal solid waste is likely used for biological degradation and oxidation/reduction reactions (Pommier and Lefebvre, 2009). The quantity of leachate generated by municipal solid waste is dependent upon the amount of rainfall or any other source of water entering into the landfill and the evaporation rates in that area (Blight, 2005). Table 1 shows leachate fluxes produced by various waste types in different climates. The leachate generation rates tend to decrease in drier areas. However, as our results here show, the waste itself produces a measurable amount of leachate without any other source of water entering into the waste. The initial leachate fluxes from the waste itself can be substantial (Fig. 1), although over longer periods of time the average leachate fluxes diminish, as the leaching ceases. Initially, the bulk density of the waste was low, but as the waste settled, the density increased and the proportion of smaller pores increased. This leads to an increasing moisture holding capacity of the waste (Wu *et al.*, 2012). The unsaturated hydraulic properties of waste with respect to waste depth and age was studied by Wu *et al.* (2012), and they found that over time, the residual water content and the field capacity of organic waste increased. Kjeldsen *et al.* (2002), in their review article on composition of landfill leachate, point out that leachate discharges are high in the beginning when settlement of waste occurs. In our experiments, a significant reduction in waste depth was observed during the leaching period. Table 3 shows the volume reduction and settlement heights. The settlement of municipal solid waste leads to higher leachate volumes especially in fresh wastes (Kjeldsen *et al.*, 2002). Dixon and Jones (2005) reviewed the mechanical behavior of municipal solid waste. They concluded that heterogeneity of waste and rate of decomposition strongly affect the settlement behavior. As there was no external pressure applied for settlement in our experiments and if we assume that the total height of the waste is uniform, the settlement can be quantified by a coefficient of compression (C_α) from a one-dimensional consolidation model (El-Fadel *et al.*, 1999):

$$C_\alpha = S_t / [H_o \log(t/t_r)] \quad (1)$$

Where C_α is the coefficient of compression, S_t is the settlement (m), t is the settlement time (days), t_r is a reference time (days), and H_o is the initial waste thickness (m).

The calculated C_α values are shown in Table 3. The reference time (t_r) used to calculate C_α was 1 day, as suggested by El-Fadel *et al.* (1999). In our study, C_α represents overall compression during the leachate drainage period. Household waste, having the highest organic proportion, had the highest C_α value (C_α = 0.48), which is consistent with the findings of El-Fadel *et al.* (1999), who attributed the higher values of C_α as a consequence of enhanced biodegradation of organic waste. The C_α values of the other two waste types were within the typical range (C_α = 0.02 to 0.35) reported by Babu *et al.* (2010). The volume reduction in restaurant and mixed waste was not as significant as in the household waste due to the presence of bulky materials like construction and demolition waste and plastics. Fig. 2 shows the water quality parameters of the leachate in comparison with the Pakistan National Environmental Quality Standards (NEQS) for wastewater discharge. The pH of the leachates was acidic and below the environmental quality standard (Fig. 2). Leachate from restaurant waste was the most acidic (average pH 4.5), and it generally also contained the highest amounts of metals. The low pH dissolves metals in waste (Kjeldsen *et al.*, 2002), thereby causing the highest concentrations of metals in the restaurant leachate. Most metals exceeded the environmental quality standards, in many cases by several orders of magnitude. The only metal not exceeding the standards for all three waste streams was Zn. Leachate from household waste generally had the lowest metal concentrations. The metal concentrations in Fig. 2 represent dissolved metals, i.e., metals filtered through a 0.45 µm filter. This excludes larger colloids, but includes metals complexed with fulvic or humic substances. Gounarls *et al.* (1993) observed that colloids play an important role in heavy metal mobilization in landfill leachates. The metal-laden colloid concentrations are most pronounced in early stages of waste decomposition, and decrease with time (Gounarls *et al.*, 1993). In anaerobic decomposition, metals are reduced and are mobilized by forming complexes with organic compounds. Xiaoli *et al.* (2013) reported higher re-activity between heavy metals and humic substances in leachates produced by anaerobic as compared to semi-aerobic degradation. As most bivalent metals tend to form complexes with dissolved organic matter (McBride, 1994), it is likely that a large fraction of the metals leached in our experiment were associated with dissolved organic matter. Indeed, the highest metal concentrations in leachate were observed in restaurant and mixed wastes, which also had high amounts of dissolved organic matter. For household waste leachate, Cd, Cr, and Cu did not exceed the environmental quality standards, however for the other two waste streams, these metals exceeded the standards by up to 300 times. Construction and demolition waste, meat/bones, and mixed leftover waste present in mixed and restaurant waste can contain higher values of Cd and Cr. Holm *et al.* (1995) found a strong correlation of Cd with dissolved organic carbon. They characterized the dissolved Cd speciation in organic-rich leachates from various sources and concluded that Cd forms organic complexes which can mobilize Cd.

The leachate contained large amounts of organics, which was evident by DOC and COD having values exceeding the environmental quality standards by up to 800 times (Fig. 2). Higher values of both COD and DOC are indicative of initial phases of waste degradation with an elevated rate of decomposition (El-Fadel *et al.*, 2002). Our results were similar to those of Zhao *et al.* (2013) who analyzed leachates from fresh wastes. COD and DOC were highest in the restaurant waste. Restaurant waste generally has high density and high proportion of organic material, resulting in elevated COD and DOC values in its leachate. As COD is a composite parameter for the oxygen requirement of both organics and inorganics in a solution, the representation of organics can be

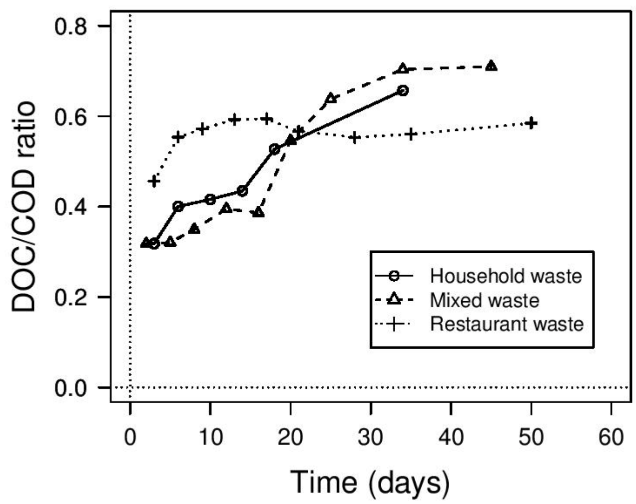

Fig. 3. DOC/COD ratios in leachates during t

Table 4. Pearson correlation coefficients of various waste characteristics with total contaminant loads in leachate (chemical oxygen demand (COD), dissolved organic carbon (DOC), total dissolved solids (TDS), total nitrogen (tot-N), and Cu)

Waste characteristics	Units	COD	DOC	TDS	Tot-N	Cu
		Pearson correlation coefficients				
Initial moisture content	g of H_2O/kg of DM	0.76	0.67	0.74	0.96	0.96
Total leachate volume	m^3/m^3 of available moisture	1.00	1.00	1.00	0.88	0.88
Initial density	kg/m^3	0.98	1.00	0.99	0.81	0.81
Organic proportion	kg/kg of DM	1.00	1.00	0.98	0.93	0.93
Volume reduction	m^3/m^3 of DM	-0.91	-0.93	-0.87	-0.99	-0.99

depicted by the ratio of DOC/COD. Fig. 3 shows the dynamics of the DOC/COD ratio during our study period. For house- hold and mixed wastes, the DOC/ COD ratio was initially around 0.3 and increased with time to 0.6 to 0.7. This indicates that during the experimental time, degradation processes converted organic material more and more into DOC. On the contrary, the restaurant waste showed rather constant DOC/COD ratios, probably because of higher levels of non-carbon oxygen-demanding substances initially present in waste or due to slowly degradable organics that retard the degradation process. Ziyang *et al*. (2009) assessed the long-term variation in components of COD in the different phases of the degradation process and concluded that organic carbon is the major portion of COD in the early stage of degradation. Chloride exceeded the environmental quality standards in household and restaurant waste leachates; however, remained within the limits in the mixed waste leachate (Fig. 2).

The amount of total dissolved solids in the leachates from three waste types was almost three times higher than the environmental quality standard, which itself is even more saline than seawater. The contribution of chloride in TDS is less than ten percent, and other species like carbonates, sulfates, nitrates, and phosphates have major influence on the salinity of leachates. Total nitrogen also exceeded the environmental quality standards in all waste types. High nitrogen concentrations in leachate are common. Cheng and Chu (2011) even proposed to use leachates high in nitrogen as fertilizer. Total phosphorus showed high values of 0.25, 0.48 and 0.26 g/L for household waste, mixed waste, and restaurant waste, respectively. No environmental quality standards for phosphorus are available in Pakistan, but our measured concentrations exceed standards of point source phosphorus discharges in the US by a factor of 200 to 500 (e.g., the standard for phosphorus discharge from waste water treatment plants in the state of Wisconsin is 1 mg/L (Wisconsin Department of Natural Resources, 2012)).

The normalization equation used by Rafizul *et al*. (2012) describes the total loading of a particular contaminant in the leachate per unit weight of the waste:

$$L = CV / M_w \quad (2)$$

Where, L is contaminant load (mg/kg of the waste), C is concentration of contaminant in leachate (mg/L), V is leachate volume (L), and M_w is total mass of the waste (kg). Equation (2) was used to calculate the total loads of individual contaminants with respect to leachate volume discharged and the initial weight of the wastes. These values were then correlated with waste characteristics.

Table 4 shows the correlation coefficients of normalized waste characteristics with total contaminant loads of the leachate (COD, DOC, TDS, tot-N, and Cu). The initial moisture content (g of H_2O/g of DM) significantly correlated with tot-N and Cu loads, but COD, DOC, and TDS were not significant. All the contaminants strongly correlated with the fraction of leachate volume per unit available moisture in the wastes. Similarly, the initial waste density (kg/m^3), and organic proportion (kg/kg of DM) had a strong effect on all the contaminant loads in the leachate.

Waste settlement was negatively correlated with the contaminant loads: the higher the settlement, the lower was the contaminant load. Pommier and Lefebvre (2009) studied the impact of moisture content on biodegradation of heterogeneous solid waste and reported that the retention of moisture is dependent upon the field capacity of the waste. Heterogeneity of the waste affects waste settlement and water retention behavior and thereby affects the leachate volume. The settlement increases the leachate volume hence diluting the original contaminant concentrations.

CONCLUSIONS

Our results show that typical waste deposited in landfills in Pakistan leaches considerable amounts of contaminants, even in the absence of external water inputs as is the case in semi-arid and arid regions. The concentrations of metals, organic materials, and nutrients exceeded the environmental water quality standards manifold, indicating that waste leachate poses a threat to surface and ground water. This is of particular concern because most landfills in Pakistan and other developing countries, are not lined, so that leachate will seep unhindered into the underlying soil. The semi-arid climate does not prevent leaching of contaminants, but rather accentuate the problem as contaminant concentrations are even higher because there is no dilution with rainwater. Current landfill management practices in Pakistan therefore need to consider the toxic nature of leachate from municipal waste, and methods have to be developed to either contain contaminants in the waste or collect and treat the leachate before it is discharged to surface and ground waters.

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