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CHEMICAL CHARACTERIZATION OF YARD WASTE COMPOST AND MULCH

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ABSTRACT

There are concerns that the composting of yard waste triggered by waste reduction at landfills could damage crops due to possible contaminant transfer from waste to compost. Yard waste compost and mulch samples were collected from 6 districts in Florida and analyzed for metals/metalloids and herbicide content. Zinc (Zn) had the highest concentration (4.01 to 270 mg/kg) while cadmium (Cd) had the lowest (0.28 to 3.44 mg/kg). The concentrations of heavy metals, Zn, copper (Cu), chromium (Cr) and lead (Pb) were higher than other metal/metalloids; arsenic (As), nickel (Ni), selenium (Se), molybdenum (Mo) and Cd. However, the concentrations of all nine metals/metalloids were below USEPA maximum allowable limits. Out of the 14 herbicides tested in compost samples; four herbicides, dalapon, dichlorprop, 2, 4-D and chloramben were not detected in any of the districts. The remaining 10 herbicides had very low concentrations ranging from 0.006 to 12.7 mg/kg. This may be due to degradation of herbicides during composting. The persistent herbicide, picloram was detected in one of the districts at a concentration of 0.007 mg/kg while dicamba was detected in two districts at concentrations ranging from 0.11 to 0.34 mg/kg. Although, the concentrations of the herbicides were far below the soil clean up target levels, some of them might be toxic to plants at low concentrations. Yard waste compost appears to be relatively safe with respect to its metal concentrations while detection of persistent herbicides makes it potentially dangerous to crop growth.

Keywords: yard waste; compost; mulch; metal; herbicides

ISSN: 2455-6939

Volume:02, Issue:04

1. INTRODUCTION

Due to the large amount of waste generated in the state, Florida enacted a law prohibiting landfilling of yard waste to reduce the increasing volume of municipal solid waste (MSW) (Li et al., 2013). The US Environmental Protection Agency (USEPA) proposed that yard waste should be composted since yard waste and food scraps constitute about 20-30% of waste generated in the United States (USEPA, 2016). The USEPA set a goal of reduction of the US municipal waste through source reduction and recycling (including composting) in the late 80's (USEPA, 1989).

Yard wastes including fallen leaves, grass clippings, shrubbery trimmings, and tree limbs are all valuable plant material that can be used to improve soil fertility and conserve soil moisture (McGeehan, 2012). These materials are often separated and processed at collection facilities into compost and mulch. Composting of yard waste is cost effective, leads to resource recovery and is more environmentally favorable than land filling (van Haaren et al., 2010; Sangamithirai et al., 2015).

Composting is a biological process in which microorganisms convert organic matter into a soil like material that has many benefits. Mulch, on the other hand, is coarser-textured compost whose production requires less time compared to compost. Composted yard waste stood at 20 million tons in 2005 compared to 7.0 million tons in 1994 (USEPA, 1995; USEPA, 2005). The possible adverse effects of compost on the environment were not thoroughly investigated as a result of the goal to implement volume reduction of MSW in landfills (Kovacic et al., 1992).

The use of yard waste as compost has increased in the last few years but there is no specific composting legislature for its regulation. It is generally believed that compost from yard waste is safe but if polluted, it could lead to contamination of the food chain when used for crop production or could lead to crop damage (Fricke and Vogtmann, 1994). Ground water contamination is also possible through the leaching of pollutants found in yard waste. Therefore, there is the need to investigate the presence and fate of toxic pollutants in yard waste and its compost products.

The safety issue of the composted materials has been of concern (Buyuksonmez and Rynk, 1999; Buyuksonmez et al., 2000; Strom, 2000; Townsend et al., 2003; Wilson et al., 2003; Aziz et al., 2015). Metals and applied pesticides that may be components of municipal yard waste could pose both health and environmental problems if not thoroughly investigated (Kovacicet al., 1992). Heavy metals are considered grave pollutants due to their persistence, toxicity and non-degradability in the environment. Arsenic is reported to cause skin cancer and kidney failure (Frankenberger, 2002; Meliker et al., 2007; Martinez et al., 2011; Ferreccio et al., 2013) while Pb

ISSN: 2455-6939

Volume:02, Issue:04

has been associated with damage to the central nervous system and learning problems in children (Hou et al., 2013).

Cadmium is toxic to the kidney and can cause bone demineralization (Bernard, 2008). Although chromium (III) has been reported to be an essential nutrient, chromium (Cr)exposure has been associated with problems in the respiratory system, gastrointestinal system and skin (Wilbur et al., 2012).Persistent organic pollutants (POPs) including dioxins, furans, polychlorinated biphenyls (PCBs), and organochlorine pesticides are carcinogenic and can cause adverse effects on the metabolic, immune, reproductive, nervous, and endocrine systems (Carpenter, 2011; Ruzzin, 2012; Pawelczyk, 2013; Lee et al., 2016).

Few studies have determined metal and pesticide concentrations of yard waste and its compost. Iron (Fe) concentrations of 1990 mg/kg have been reported in yard waste in Florida (Li et al., 2013). Concentrations of copper (Cu), chromium (Cr), and lead (Pb) were higher in rural waste that included yard waste (Guan et al., 2011). Monitoring of 27 pesticides in yard trimmings by Metropolitan Service District of Portland, Oregon revealed low concentrations of pentachlorophenol and chlordane in yard trimming compost samples (Buyuksonmez et al., 2000). Screening of compost made from leaves for 200 pesticides from Croton point facility in Westchester County, New York by Richard and Chadsey (1989) revealed only four of the target pesticides.

Strom (2000) investigated 27 pesticides in samples of finished compost from six New Jersey yard trimming composting facilities. The only pesticide found in detectable level was chlordane, with concentration ranging between 0.29 and 3.23 mg/kg. Townsend et al (2003) investigating impact of chromated copper arsenate (CCA) in wood mulch, detected arsenic concentration of 65.4 μg/l in the leachate of one of the mulch samples purchased from a retail outlet.

Though there are numerous facilities that produce compost or mulch from yard waste in Florida, limited guideline or information is available on the concentrations of pollutants in yard waste compost or mulch. The aim of this study was to investigate the concentration of inorganic and organic pollutants in yard waste composts and mulch.

2. MATERIALS AND METHOD

2.1 Sampling

The yard waste samples were collected from six Florida DEP districts; Northwest, Northeast, Central, Southwest, Southeast, and South districts. The yard waste samplings for heavy metals and herbicides analyses were conducted separately (Tables 1 and 2). A total of 86 samples were collected for heavy metal analysis from 40 facilities located in each district in Florida (Fig. 1a).

ISSN: 2455-6939

Volume:02, Issue:04

For herbicide analysis, 10 samples were collected from 5 districts in Florida (Fig. 1b). Selected compost and mulch piles in the facilities were sectioned into quarters. Five random spots in each quarter were selected and the surface layer (~ 30cm) removed with clean stainless scoop. Enough samples were then collected to fill one gallon clean plastic bags. All the collected samples from a pile were thoroughly mixed to get a composite sample. Sub samples were taken from each composite for analysis.

Table 1. Summary of districts and number of samples collected for heavy metal analysis

Districts	Location by county	Number of facilities	Number of samples	Sample typeand number
S	Fort Myers, Serbing, Cape Coral	8	20	compost - 1 mulch – 19
SE	Medley, Parkland, Delray Beach, West Palm Beach	4	12	compost - 2 mulch – 10
SW	Sarasota, NorthPort, Lecanto, Homosassa, Spring Hill, Brooksville, Odessa, Clearwater, St. Petersburg, San Anthonia	11	20	compost - 1 mulch 19
C	New SmymaBeach, Winter Park, Titusville, Cocoa, Melbourne, Ocala, Tavares	5	18	compost - 2 mulch – 16
NE	Gainesville, Green Cove Spring, AtlanticBeach, Starke, Palatka, Bunnell	14	14	compost - mulch – 14
NW	Pensacola	1	2	mulch – 2
Total			86	

Table 2. Summary of districts and number of samples collected for herbicide analysis

Districts	Location by County	Number of Samples	Sample Type
SE	Miami – Dade	4	compost - 4 mulch - 0
SW	Pork, Citrus	4	compost - 4 mulch 0

C	Marion	2	compost - 2 mulch - 0
NE	Alachua	2	compost - 2 mulch – 0
NW	Escambia	4	compost - 4 mulch – 0
Total		16	

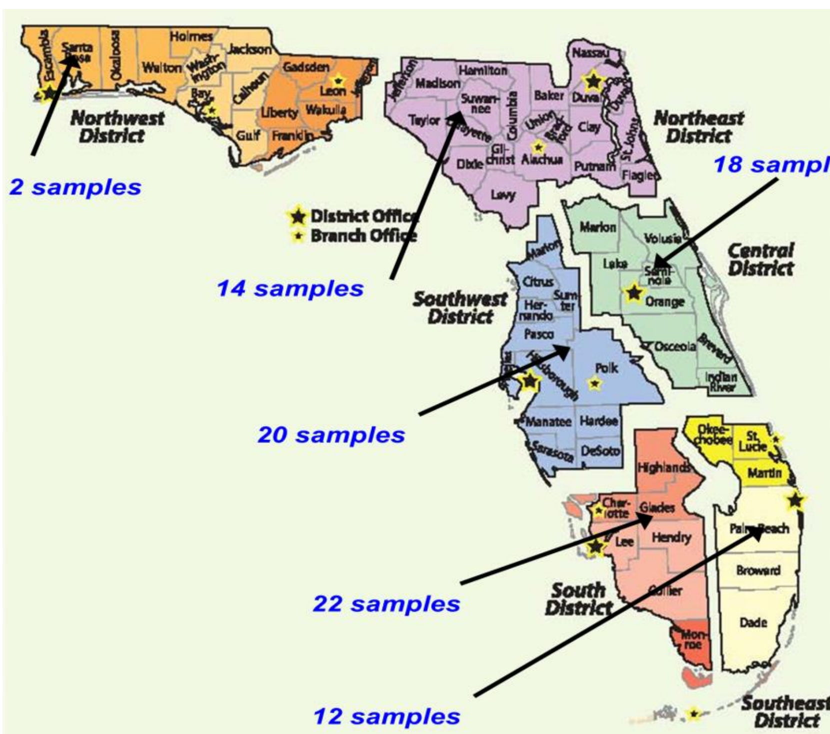

Fig. 1a. Locations and number of samples collected from each district for heavy metal analysis

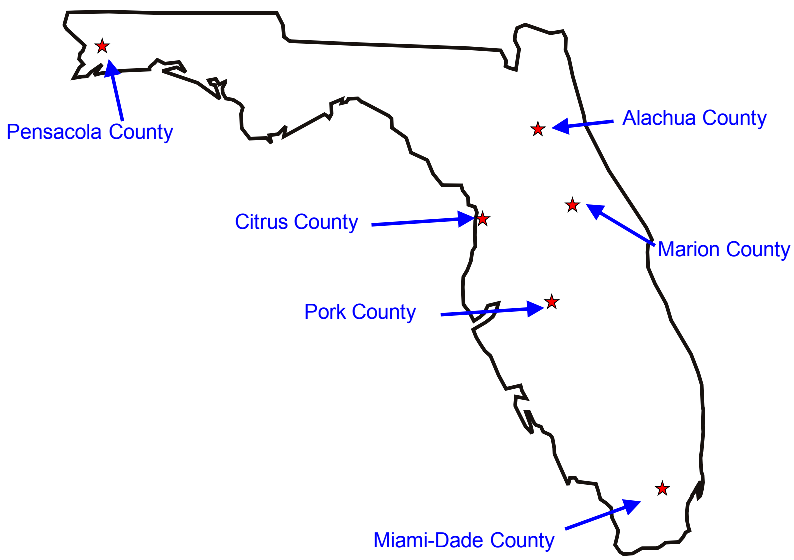

Fig. 1b. Sampling locations for herbicide estimation

2.2 Chemical analysis

Samples for metal analysis were placed in the greenhouse to air dry for 4 weeks. After drying, samples were ground and sieved through a 2-mm sieve. Approximately 0.5g of ground samples were digested using HNO_3/H_2O_2 in a Hot Block Digester using USEPA Method 3050b. The resulting solution was diluted to 50 ml and analyzed for metals with Inductively coupled plasma –optical emission spectrophotometer (ICP-OES) (Perkin Elmer Optima 3200 RL, Massachusetts, MA).

All reagents used for analysis were trace metal grade (Fisher Scientific, Pittsburgh PA 15275). Stock standard solution was Spex Certiprep (SPEX Certiprep, NJ 08840). USEPA approved Quality Assurance/ Quality Control (QA/QC) plan involving spikes, duplicates, blanks and certified reference materials was followed. Two Certified Reference Material soil samples were analyzed and the measured values of metals showed good precision and accuracy, which was within $\pm$20%.

For herbicide analysis, samples brought from the field were used without drying to preserve volatile compounds. Samples were air-dried in the laboratory for 3 days. Samples were extracted with methylene chloride and acetone using the ultrasonic extractor at pH 2.5. The organic layer was filtered and dried with anhydrous sodium sulfate. The dried extract was concentrated to 5 ml with the aid of a water bath, K-D flask and a concentrator tube. The sample was further concentrated to 1 ml by blowing nitrogen over it. The concentrate was diluted with 1 mL of

ISSN: 2455-6939

Volume:02, Issue:04

isooctane and 0.5 mL of methanol finally made up to a final volume of 4 mL with diethyl ether. The sample was then ready for methylation with diazomethane.

Extracted samples were derivatized before analysis. Through the derivatization process, organic compounds may increase their volatility, enhance separation and improve detectibility on the GC detector. Among several derivatization methods, bubbler method was selected for this research. The bubbler method is described in details somewhere else (EPA SW-846 8151A). Derivatized samples were analyzed using a GC equipped with electron capture detector (ECD). Total 0.5 uL of samples were injected into the GC. Sample peaks were identified using chlorinated herbicide standards (Ultra Scientific, New England).The measured values of herbicides showed acceptable percentage recovery of the method.

The pH of the samples was measured in water (1:20 compost/water ratio) after equilibration for 1 hour (Campbell and Bryant, 1941).The pH was determined using Accumet pH meter model 20 (Fisher scientific, Pittsburgh, PA).

3. RESULTS AND DISCUSSION

3.1 pH of Yard Waste Compost and Mulch

The average pH of all 86 samples was 5.31 which is typical of Florida soils (Fig. 2). It is important to note that 80 out of the 86 samples (93%) were mulches which may have affected the pH. Approximately 80% of the samples had pH less than 6 indicating acidity. The low pH of some of the samples may be due to the production of oxalic acid or hydrogen peroxide (H_2O_2) by wood-decay fungi (Koenigs, 1974; Shimada et al., 1997).Several studies have reported that compost generally have a pH in the neutral range (7.0-8.0) (Kirchhoff et al., 2002; Garnett, 2012; Saad et al., 2013).Saad et al (2013) found that 7 weeks old compost of food and yard waste had pH between 7 and 8 which they attributed to maturation of the compost. According to them, the compost was acidic in the early stages but pH increased to neutral by the sixth week. However, Kirchhoff reported that the model compost specification for pH was in the range 5.0 - 8.5.The addition of food and yard waste compost to soil did not change soil pH in some instances while it either decreased or increased it in some other studies (Kirchoff et al., 2002; Garnett, 2012). The effect of compost on soil pH may depend on soil properties like buffering capacity and compost characteristics.

ISSN: 2455-6939

Volume:02, Issue:04

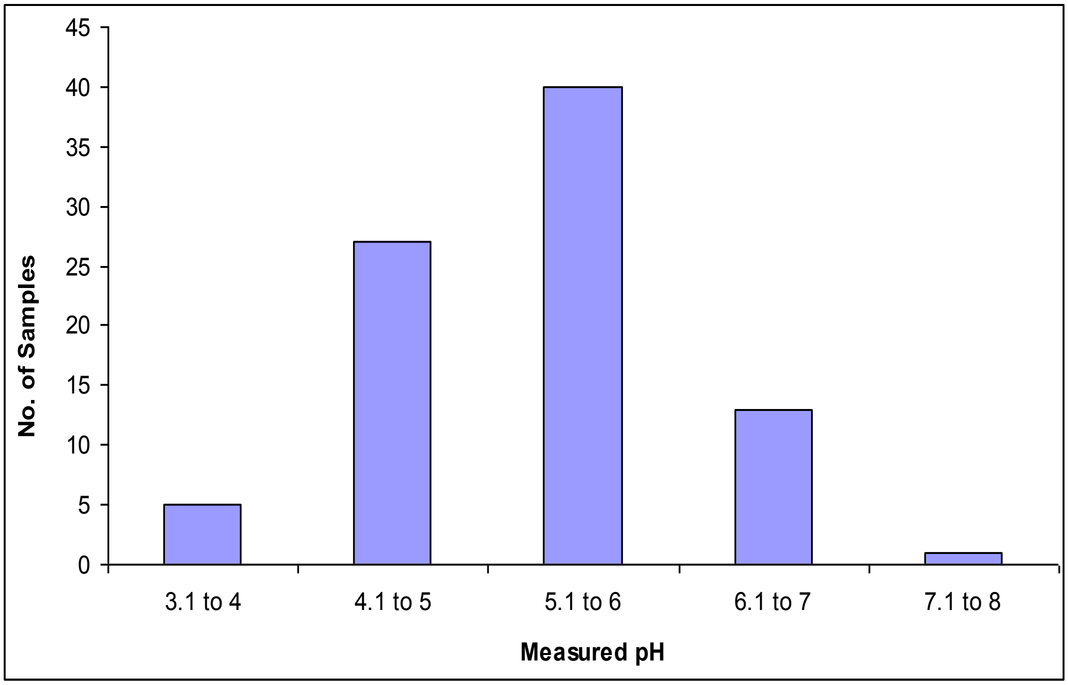

Fig. 2. Frequency distribution of pH in the 86 yard waste samples.

3.2 Metal/Metalloid concentrations in yard waste compost/mulch

Metal/metalloids concentrations were generally low with average concentrations ranging from 0.58 to 21.0 mg/kg (Table 3). The concentrations of metals/metalloids in this study are lower than reported by other studies (Table 4). Sangamithirai et al (2015) reported higher maximum concentrations of Cd, Cr, Cu, Ni and Zn in yard waste composted with different wastes like paper and vegetables. Similarly, Shiralipour (2002) and Ozores-Hampton et al (2005) also reported higher metal/metalloid concentrations in compost.

Table 3: Geometric mean concentrations of metals in yard waste compost

Metal	Number of samples	Number of detects	Average concentrations (mg/kg)	Concentration range (mg/kg)	[a]Maximum allowable (mg/kg)
As	86	58	1.41	Bdl - 9.98	75
Cd	86	88	0.58	0.28 - 3.44	85

Cr	86	87	2.55	Bdl - 18.1	NA
Cu	86	88	8.47	2.57 - 11.6	4300
Pb	86	79	2.36	Bdl - 67.2	840
Mo	86	75	1.20	Bdl - 4.88	75
Ni	86	88	1.34	0.38 - 8.27	420
Se	86	33	1.33	Bdl - 6.27	100
Zn	86	88	21.0	4.01 - 270	7500

a-USEPA (2000) NA-not available Bdl – below detection limit

Table 4: Metal concentrations (mg/kg) in composted wastes

Metal	compost/ mulch	compost	compost	compost	compost
As	1.41	NA	NA	22	NA
Cd	0.58	NA	3	1.5	1.1-3.57
Cr	2.55	NA	NA	24	9.8-50.9
Cu	8.47	8	161	58	18.3-140
Pb	2.36	12	60.2	42	21.9-37.5
Mo	1.20	NA	NA	NA	NA
Ni	1.34	1	8.2	28	4.7-12.7
Se	1.33	NA	NA	NA	NA
Zn	21.0	30	266	240	453-887
Ref	This study	He et al., 2000	Shiralipour 2002	Ozores Hampton et al., 2005	Sangamithirai et al., 2015

NA-not available

On the contrary, compared with this study, Saad et al (2013) reported lower concentrations (<0.2 mg/kg) for cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), mercury (Hg), nickel (Ni) and zinc (Zn) in a mixed food and yard waste compost amended with effective microbes. The metal

ISSN: 2455-6939

Volume:02, Issue:04

content of yard waste compost may vary based on composition, location, environmental conditions and composting method.

Metal concentrations varied with location in the six different districts sampled (Table 5). The highest Se was recorded in the South; highest Cu and Mo in the Southeast; highest Cd and Pb in the Southwest; highest As and Cr in the Central; and highest Zn in the Northwest. The lowest As, Cd, and Cr was recorded in the Northwest while the lowest Cu, Pb, Ni and Zn was recorded in the South district. One major source of Cu, As and Cr in yard waste is chromated copper arsenate (CCA) treated wood. This may be responsible for the high As and Cr in the Central district. The low heavy metal concentration in the South district may be due to the absence or presence of few industries or factories in the region.

Table 5: Mean metal concentration (mg/kg) in the six districts in Florida

District	As	Cd	Cr	Cu	Pb	Mo	Ni	Se	Zn
South	1.55	0.62	3.56	9.36	2.65	1.04	1.22	**1.41**	19.8
Southeast	1.37	0.53	5.71	**15.5**	3.09	**2.32**	1.86	1.29	30.2
Southwest	1.15	**0.68**	3.35	13.2	**6.88**	1.35	1.80	1.20	35.5
Central	**2.57***	0.62	**5.94**	9.99	4.86	1.74	1.55	0.54	30.9
Northeast	2.51	0.63	5.23	11.5	3.79	0.82	1.39	0.84	23.2
Northwest	0.39	0.52	2.36	10.1	4.49	1.27	**2.75**	1.26	**57.3**

*The highest metal concentrations among the 6 districts are highlighted in bold.

Generally, the heavy metals, Zn, Cu, Cr and Pb had higher concentrations than other metals/metalloids arsenic (As), nickel (Ni), selenium (Se), molybdenum (Mo) and Cd. This is consistent with previous research which reported high concentrations of Cu, Zn and Pb in composts (He et al., 1992). In this study, Zn had the highest concentration with values ranging from 4.01 to 270 mg/kg while Cd had the lowest concentrations with values ranging from 0.28 to 3.44 mg/kg. Comparing the results to the soil background metal concentrations in Florida (Ma et al., 1997), the geometric mean concentrations of Cr, Pb and Ni were all lower than their respective background concentrations. The concentration values of As, Cd and Cu were 1.3, 2.8 and 2.3 times respectively higher than the background soil concentrations.

Cadmium concentrations in yard waste samples were lower than other metals, with about 99% of the samples less than 1 mg/kg. Geometric mean concentrations of Moin yard waste samples were relatively higher than Cd with 37 samples (49.3%) being < 1 mg kg^{-1}. Compared to Mo,

ISSN: 2455-6939

Volume:02, Issue:04

geometric mean concentrations of Niin yard waste samples were higher with 68 samples(79%) being <1 mg kg^{-1}. Similar to nickel, selenium concentrations in yard waste samples were relatively low with 60 samples (70%) being <1 mg kg^{-1}. Arsenic concentrations in the yard sample were low, with 56% being <1 mg kg^{-1}, similar to Asconcentrations in Florida soils (Chen et al., 1999). Even though the geometric mean concentration for As was lower than the soil cleanup target level (SCTL), 30% of the samples with As were above the residential SCTL for As (2.1 mg kg^{-1}).

Unlike cadmium, chromium concentrations in yard waste samples were relatively higher, with 15 samples (17%) exceeding 9 mg kg^{-1}. Copper concentrations in yard waste samples were relatively higher than chromium with 38 samples (44%) exceeding 9 mg kg^{-1}. Geometric Pb concentrations in yard waste samples were relatively lower than Cr, Cu and Zn with 9 samples (11.4%) exceeding 9 mg kg^{-1}. Unlike Pb, zinc concentrations in yard waste samples were relatively higher with 71 samples (83%) exceeding 9 mg kg^{-1}. However, the metal concentrations were far below the maximum allowable for compost by the United States Environmental Protection Agency (USEPA, 2000) showing they are relatively safe for use as compost and mulch.

3.3 Herbicides in yard waste compost

Crop damage by herbicide contaminated compost has been reported in Washington State, USA (Rynk, 2000). Tomatoes and other horticultural crops were damaged by herbicides, in particular, picloram and clopyralid. Similarly, herbicide residues in compost damaged vegetable crops in Vermont, USA (Greene et al., 2013). It is necessary to screen composts for their herbicide content before use as compost to prevent crop damage and harvest losses. Yard waste compost samples were collected from five districts in Florida. However, the concentrations of most of the herbicides in the yard waste samples were very low or not detected (Table 6).

ISSN: 2455-6939

Volume:02, Issue:04

Table 6. Average concentrations of selected herbicide concentrations of each district.

	Herbicide Concentration Detected in Samples (mg/kg)					SCTL (mg/kg)
	NW	SW	NE	Central	SE	(Residential)
Dalapon	<0.002	<0.002	<0.002	<0.002	<0.002	
3,5-dichlorobenzoate	<0.004	0.134	<0.004	0.464	<0.004	3.5
4-nitroanisole	<0.004	0.006	<0.004	<0.004	<0.004	
Dicamba	<0.001	0.107	<0.001	0.344	<0.001	2300
MCPP	<0.070	<0.070	<0.070	<0.070	0.6255	64
MCPA	<0.045	12.721	<0.045	<0.045	<0.045	55
Dichlorprop	<0.016	<0.016	<0.016	<0.016	<0.016	370
2,4-D	<0.001	<0.001	<0.001	<0.001	<0.001	770
Pentachloroanisole	<0.002	0.101	0.029	<0.002	<0.002	
Chloramben	<0.042	<0.042	<0.042	<0.042	<0.042	960
Dinoseb	<0.010	0.051	<0.010	0.154	<0.010	65
Bentazon	<0.086	0.773	<0.086	<0.086	<0.086	2100
Picloram	<0.003	0.007	<0.003	<0.003	<0.003	
Acifluorfen	<0.041	0.281	<0.041	<0.041	<0.041	28

This may be due to the degradation or decomposition of applied herbicides either during application or during the composting process (Pichtel, 2014). MCPA had the highest concentration of 12.7mg/kg which was still much lower than the soil clean up target level (SCTL). MCPA, a selective phenoxy herbicide, was only detected in the Southwest district indicating more usage of the herbicide in the region.

Four herbicides; Dalapon, Dichlorprop, 2,4-dichlorophenoxyacetic Acid (2,4-D) and chloramben were not detected in the samples. Out of the remaining ten herbicides detected, 3,5-dichlorobenzoate was the most frequently detected while bentazon was the least detected.

Herbicides detected within the detection limits are widely used as ingredients of weed controller. These include MCPP, MCPA, dinocep and picloram. Picloram, one of four persistent herbicides of concern (Coker, 2015) was detected in only the Southwest district at a very low concentration (0.007mg/kg). However, picloram is highly toxic in the soil and residues as low as 0.25 µg/kg soil can be toxic to sensitive broadleaf crops (Jotcham et al., 1989). Greene et al (2013) reported that levels of picloram as low as 5ppb in compost is known to cause harm to crops.

Dicamba was detected in two districts; Southwest and Central, at concentrations below 0.5 mg/kg which is substantially lower than the soil clean up target level (SCTL). Dicamba was among the top five herbicides used in the home and garden sector (Kurenbach et al., 2015).Dicambais commonly used as weed control in lawns which may lead to contamination of yard waste compost (Hamid et al., 2011).

No herbicide was detected in the Northwest while only one herbicide was detected in Northeast and Southeast. Nine herbicides were detected in the Southwest while three were detected in the Central district. This may reflect more usage of herbicides in the Southwest and Central districts. Higher detection of herbicides in Southwest and Central districts may also be due to lower rates of degradation of the herbicides in these two areas.

Although the number of samples collected from each district may be insufficient to make a clear conclusion, the results implied that higher herbicide residues had been left in the yard wastes of Central and Southern Florida probably due to their use in greater amounts. The levels of all of the 14 herbicides were however below the soil cleanup target levels.

4. CONCLUSION

This study was conducted to address potential concerns of metals and herbicide contamination of yard waste compost and mulch. Generally, the heavy metals, Zn, Cu, Cr and Pb had higher concentrations than other metals/metalloids As, Ni, Se, Mo and Cd. However, the metal concentrations were far below the maximum allowable by the United States Environmental Protection Agency (USEPA, 2000) showing they are relatively safe for use as compost and mulch. Among 14 types of herbicides investigated in this study, 4 were not detected in any district. However, the concentrations of the detected herbicides were very low though it has been reported that low concentrations can be harmful to crops, especially picloram. Though the metal concentrations showed that compost/mulch are not highly contaminated, care should be taken when using yard waste compost especially in the Southwest district where several herbicides were detected.

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