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Heavy Metal Content of Tomatoes Fertilized with Biosolids

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Biosolids recycle plant nutrients, but may contain other constituents, such as heavy metals, that could contaminate food crops through plant uptake. The study was conducted to quantify heavy metal uptake. Tomatoes (*Solanum lycopersicum*, var. Better Boy), grown in a greenhouse in a mined sand containing sphagnum peat [90:10 (sand:peat), by volume], were fertilized with a biosolids fertilizer (Milorganite®), supplemented with K, and with several specialty organic fertilizers (Tomato Tone®, Dr. Earth®), and the soluble fertilizer Miracle-Gro®, all at the same rate of N, except that a 0.5x and 1.5x rate of biosolids was included. A 1x biosolids treatment without added K also was included in the study. The greatest fresh weight of tomatoes was obtained with Tomato Tone® fertilizer. The 1.5x rate of biosolids produced significantly ($P < 0.05$) lower fruit yield, but that treatment, and the 1x biosolids rate, produced greater yield than Dr. Earth® and Miracle-Gro. Although the biosolids contained more As, Cd, Mo, Ni, and Pb than the specialty organic fertilizers, and presumably more than the soluble fertilizer, even at the 1.5x rate of biosolids there were no significant differences ($P < 0.05$) among the four fertilizers in the concentrations of these elements in the tomato fruits. The Dr. Earth® fertilizer contained more Cu, and approximately the same amount of Zn as the biosolids, but the concentration of these two elements was not different in the tomato fruits produced by the four fertilizers.

Biosolids are the nutrient-rich organic materials resulting from the treatment of sewage sludge. They are created by domestic waste water treatment processes designed to reduce discharges into water bodies. More than 16,500 publically owned wastewater treatment facilities in the United States treat over 150 billion L/day of wastewater, generating over 7 million tons/year (dry weight) of biosolids (USEPA, 2006). The biosolids may be incinerated, placed in landfills, or used as soil amendments and fertilizers.

Using biosolids as fertilizers recycles plant nutrients and reduces the need to produce additional fertilizer elements by mining and manufacturing processes. In that regard, it supports sustainability. The Milwaukee (WI) Metropolitan Sewerage District has been marketing biosolids fertilizer under the name Milorganite® since 1926. It is the most commonly available biosolids fertilizer for the home garden market, with approximately 30,000 tons being sold annually for home lawns and gardens.

Many substances enter the sewage stream, including "heavy metals." Actually, there is no universally-accepted definition of "heavy metals." Some prefer the term "toxic" elements, but that term is not well defined either. For example, elements such as Fe, Co, Cu, Mn, Mo, and Zn are essential for humans, but are toxic at elevated levels. Glanze (1996) lists the elements As, Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb, Sn, and Ti as elements of concern. The USEPA regulates As, Cd, Cu, Pb, Hg, Mo, Ni, Se, and Zn in biosolids. This agency has established "ceiling concentrations" and "Exceptional Quality" concentrations for these elements in biosolids used for land application (USEPA, 2012), and Milorganite® meets the Exceptional Quality standards. The study was conducted to quantify the content of most of these elements in tomato fruits grown with natural organic fertilizers, a water-soluble fertilizer, and with the biosolids Milorganite®.

Methods and Materials

Tomatoes (*Solanum lycopersicm*, var. Better Boy) were grown from seed in a greenhouse at the University of Florida, Ft. Lauderdale Research and Education Center, in plastic pots 25-cm dia. x 25-cm deep, using a mined sand containing sphagnum peat [90:10 (sand:peat), by volume] to avoid root zone-connected heavy metal contamination. Four replications of each treatment were arranged in a randomized complete-block design. Three seeds were planted per pot on 28 Sept. 2013, and thinned to one plant per pot 27 d later. Fertilization was supplied by one of four sources, comprising two natural organic fertilizers (Tomato Tone® 3-4-6, Dr. Earth® 5-7-3), one water-soluble fertilizer (Miracle-Gro® 18-18-21), and one biosolids fertilizer (Milorganite® 5-2-0). The fertilizers were thoroughly mixed with most of the root zones prior to planting to provide 4 g/pot of N (1x rate, equivalent to 150 lbs/acre for a planting rate of 8500 plants/acre). However, an unfertilized control was included in the study, and the biosolids (Milorganite®) fertilizer also was applied at a 0.5x and 1.5x rate (Table 1). Dolomite was mixed into all root zones at the rate of 9.5 g/pot, and 8.1 g/pot of K_2SO_4 was mixed into three of the four biosolids treatment root zones prior to planting to provide 3.5 g/pot of K (4.0 g/pot of K_20, Table 1). At various intervals, based on the manufacturer's recommendations, fertilizers were applied postplant to total an additional 4 g/pot of N (Table 1). The

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post-plant fertilizers were applied on the root zone surface and lightly mixed in, except that the water soluble fertilizer (Miracle Gro®) was dissolved in ca. 500 ml water and drenched into the root zone. Potassium sulfate was surface applied (8.1 g/pot) to the appropriate biosolids treatments at the time of the mid-season biosolids fertilization. Daily irrigation with tap water was used to maintain adequate moisture, but irrigation may have been excessive. Vines were supported by strings and clips. Fruit was harvested through four months after seeding (28 Jan. 2014). Fruits that showed appreciable red color were kept separate from green fruits for chemical analyses. The fruits were weighed fresh, and after drying at 110 °C. The vines were cut at soil level, dried at 60 °C, and weighed. The dried fruits, vines, and fertilizers (other than Miracle Gro®) were ground through a 1-mm screen in a Wiley mill, and digested with H_2SO_4/H_2O_2 (Lowther, 1980) on an Al block (Westco Scientific Instruments, Brookfield CT) at 350 °C. The digestates were analyzed for N and P by automated colorimitry with an AutoAnalzer 3 (Seal Analytical, Mequon, WI). Potassium, Ca, Mg, and heavy metal concentrations were analyzed with an ICP (Perkin–Elmer, Waltham, MA) using EPA method 200.7. Data were subjected to analysis of variance and means were separated by Duncan's multiple range test ($P = 0.05$) using a statistical analysis program package (SAS Institute Inc., Cary NC, ver. 9.3).

Table 1. Fertilization program

	Preplant N		Postplant
Fertilizer	Relative	g/pot	Fertilizations[z]
None	0x	0	0
Tomato Tone®	1x	4	5
Dr. Earth®	1x	4	1
Miracle Gro®	1x	4	5
Biosolids+K	0.5x	2	1
	1x	4	1
	1.5x	6	1
Biosolids-K	1x	4	1

[z]Postplant N rate totaled 4 g/pot.

Table 2. Tomato fresh weight yield and vine dry weight.

	N rate	Tomato yield	Vine wt.
Fertilizer	(g/plant)	(g/plant)	(g/plant)
None	0	0 f	<1 d
Tomato Tone®	8	1018 a	81 a
Dr. Earth®	8	105 ef	27 c
Miracle Gro®	8	128 def	32 c
Biosolids 0.5x + K	4	227 de	34 c
Biosolids 1.0x + K	8	387 bc	51 b
Biosolids 1.5x + K	12	603 b	54 b
Biosolids 1.0x – K	8	267 cd	36 c

Values within a column followed by the same letter are not significantly different ($P < 0.05$) by Duncan's multiple range test.

Results and Discussion

Fruit and vine weights. Red fruits were only obtained with the Tomato Tone® and 1.5x biosolids + K treatments. Green tomatoes were obtained for all treatments except for the unfertilized control. The greatest fresh weight of tomatoes (red + green) was obtained with Tomato Tone® fertilizer, followed by the 1.5x biosolids + K fertilizer (Table 2). The Dr. Earth® and Miracle Gro® fertilizers produced low yields. The 1.5x rate of biosolids produced significantly ($P < 0.05$) lower fruit yield than the Tomato Tone® treatment, but that treatment, and the 1x biosolids rate +K, produced greater yield than Dr. Earth® and Miracle-Gro® (Table 2). The vine weights essentially paralleled the fruit weights (Table 2).

Reasons for the differences in fruit and vine yields are suggested by the nutrient content data of the vines and fertilizers (Table 3, Table 4). Vines fertilized with the biosolids in the absence of added K contained less K than those fertilized with the natural organic or water soluble fertilizers (Table 3), and even the biosolids treatments with added K contained numerically lower

Table 3. Nutrient content of tomato vines on a dry weight basis.

	g·kg-1					mg·kg-1				
Fertilizer	N	P	K	Ca	Mg	Fe	Mn	Zn	Cu	Mo
Tomato Tone®	9.7 bc	3.8	7.3 ab	18.5 a	3.2 b	86	23 d	53 ab	8.2 bc	0.0 d
Dr. Earth®	13.5 ab	5.0	7.3 ab	16.9 a	4.0 a	90	5 e	32 c	6.4 cd	0.4 d
Miracle Gro®	8.2 c	4.0	7.8 a	13.2 b	2.2 c	90	13 de	27 c	5.1 d	0.0 d
Biosolids 0.5x + K	8.0 c	3.6	2.7 bc	18.5 a	3.8 a	96	22 d	35 bc	6.6 cd	1.1 c
Biosolids 1.0x + K	10.1 bc	4.1	2.5 bc	16.5 a	4.0 a	100	50 c	65 a	7.5 bcd	1.8 bc
Biosolids 1.5x + K	12.7 abc	4.4	2.9 abc	16.7 a	4.3 a	97	106 a	65 a	11.1 a	2.7 a
Biosolids 1.0x – K	16.1 a	4.8	1.7 c	16.7 a	4.4 a	108	78 b	61 a	9.8 ab	1.6 bc
Significance	*	NS	*	*	**	NS	**	**	**	**

Values within a column followed by the same letter are not significantly different ($P < 0.05$) by Duncan's multiple range test.
*, **, and NS refer to $P < 0.05$, 0.01, and $P > 0.05$, respectively.

Table 4. Nutrient content of fertilizers based on guaranteed analysis (bold) or analysis by authors.

	% by weight									
Fertilizer	N	P[z]	K[z]	Ca	Mg	Fe	Mn	Zn	Cu	Mo
Tomato Tone®	**3**	**4**	**6**	**8**	0.7	0.06	0.03	0.04	0.03	0.0004
Dr. Earth®	**5**	**7**	**3**	**8**	0.3	0.02	0.01	0.01	0.002	0.0002
Miracle Gro®	**18**	**18**	**21**	**0**	**0.5**	**0.1**	**0.05**	**0.05**	**0.05**	**0**
Biosolids	**5**	**2**	**0**	**1.2**	0.5	**4**	0.1	0.04	0.02	0.0009

[z]Expressed as oxides.

vine K than the other treatments, probably because the irrigated sand root zone retained little K against leaching. The biosolids contains too little K to be included in the guaranteed analysis (Table 4). It is likely that tomato and vine yields were reduced in the biosolids treatments because of limited K. Potassium deficiency symptoms were apparent in lower leaves (Fig. 1, Fig. 2). Vines fertilized with Miracle Gro® contained less Ca and Mg than the other treatments (Table 3). Miracle Gro® contains no Ca (Table 4). Although it contains Mg, because of its high N content it was applied at a much lower rate of product than the natural organic and biosolids fertilizers, resulting in a lower rate of Mg fertilization. The Miracle Gro® fertilized vines also contained less Zn and Cu than the higher yielding Tomato Tone® and 1.5x biosolids+K fertilized vines (Table 3). Vines fertilized with Dr. Earth® contained less Mn than any of the other treatments (Table 3), and Dr. Earth® fertilizer contains less Mn than the other fertilizers (Table 4), which may have lowered yields (Table 2).

Heavy metal content of tomato fruits. Since green tomatoes were produced by all treatments with the exception of the control, green tomato fruits were analyzed for heavy metals, other elements of concern, nutrient elements, and selected other elements. Data are presented on a dry-weight basis. Dry weight of green tomatoes averaged 4.6% of fresh weight (CV = 17.8), and for red tomatoes the corresponding figures were 5.3% (CV = 14.3). Although the biosolids generally contained greater amounts of many elements than the natural organic products (Table 4, Table 5), with the exception of Cu, there were no significant differences in content of heavy metals and elements of concern of tomato fruits among plants fertilized with natural organic fertilizers, the chemical fertilizer, or the biosolids fertilizer (Table 6). In agreement with the data for the vines, the Cu content of tomatoes fertilized with Dr. Earth® was among the lowest observed, and the Cu content of the tomatoes fertilized with biosolids in the absence of added K was among the highest (Table 6). In fact, the content of other

Fig. 1. Biosolids-fertilized plant on left received supplemental K, whereas the one on the right did not, and the lower leaves of the latter appear to display K deficiency 6 weeks before the final harvest.

Fig. 2. At the time of final harvest, lower leaves of both biosolids-fertilized plants receiving supplemental K (right) and those not receiving K (left) display K deficiency symptoms.

Table 5. Selected elemental content of the non-water soluble fertilizers (mg·kg-1).

Fertilizer	As	Cd	Co	Cr	Hg	Li	Ni	Pb	Se	Sr	Ti	Tl
Tomato Tone®	1.0	0.1	1.0	15	0	7.4	6	0	0.3	152	74	0.1
Dr. Earth®	0.3	1.1	0.7	27	0	7.4	6	0	0.7	115	66	0
Biosolids	1.9	0.8	4.0	113	0	4.4	24	5	0	217	250	0

Table 6. Dry-weight content of heavy metals and metals of concern in green tomatoes (mg·kg-1).

	Fertilizer source							
Element	Tomato Tone®	Dr. Earth®	Miracle Gro®	Biosolids 0.5X + K	Biosolids 1.0X + K	Biosolids 1.5X + K	Biosolids 1.0X – K	Statistical significance
As	0.13	0	0.15	0.3	0.03	0	0.03	NS
Cd	0	0	0	0	0	0	0	NS
Co	0	0	0.2	0	0	0.1	0.2	NS
Cr	0.30	0.23	0.40	0.28	0.33	0.43	0.30	NS
Cu	7.7 bc	5.4 d	7.0 c	7.5 bc	7.3 bc	8.5 b	10.8 a	**
Hg	4.8	1.0	0.4	0.0	3.0	1.6	0	NS
Mo	1.0	0.9	4.7	1.3	1.7	1.8	2.1	NS
Ni	0.5	4.2	22.3	1.2	0.4	0.5	1.3	NS
Pb	0.1	0.3	0.5	0.1	0.1	0.7	0	NS
Se	0	0	0	0	0	0.08	0.05	NS
Ti	0.18	0.33	0.18	0.15	0.20	0.18	0.20	NS
Zn	20.3	28.4	22.6	17.4	31.8	32.1	29.4	NS

Values within a row followed by the same letter are not significantly ($P < 0.05$) different by Duncan's multiple range test.
** and NS refer to $P < 0.01$ and $P > 0.05$, respectively.

Table 7. Dry-weight content (mg·kg^{-1}) of various elements in green tomatoes for which significant ($P < 0.05$) differences were observed among fertilizer treatments.

Element	Fertilizer source							
	Tomato Tone®	Dr. Earth®	Miracle Gro®	Biosolids 0.5X + K	Biosolids 1.0X + K	Biosolids 1.5X + K	Biosolids 1.0X – K	Statistical significance
Ca	1215 a	849 b	975 ab	986 ab	999 ab	1003 ab	1221 a	*
Li	0 c	0 c	0 c	0 c	0.05 c	0.20 b	0.28 a	**
Mg	1539 ab	1338 c	1324 c	1342 c	1377 bc	1473 bc	1665 a	**
Mn	8.5 bc	5.1 c	9.7 bc	9.6 bc	10.0 b	15.2 a	16.2 a	**
Sr	5.4 ab	3.4 c	5.7 a	4.4 bc	4.1 c	4.4 bc	5.9 a	**

Values within a row followed by the same letter are not significantly ($P < 0.05$) different by Duncan's multiple range test.

Table 8. Dry-weight content (mg·kg^{-1}) of various elements for which significant ($P < 0.05$) differences were observed between green and red tomatoes.

Fertilizer	Tomato type	Ca	Co	Hg	Mg	Sr	Ti	Tl
Tomato Tone®	Green	1215	0	4.8	1539	5.4	0.18	0.13
	Red	1110	0.15	0	1484	5.1	0.50	0.20
Biosolids 1.5X+K	Green	1003	0.13	1.6	1473	4.4	0.18	0.13
	Red	690	0.15	0	1292	3.1	0.30	0.50
Significance	Fertilizer	**	*	*	*	**	NS	NS
	Tomato type	**	**	**	*	**	*	*
	Interaction	NS	NS	*	NS	*	NS	NS

*, **, and NS refer to $P < 0.05$, 0.01, and $P > 0.05$, respectively.

nutrient and non-nutrient elements also was among the highest for tomatoes fertilized with biosolids in the absence of added K (Table 7). No significant ($P < 0.05$) differences in the elemental composition of green tomatoes among fertilizer treatments were observed for Al, Be, Fe, Sb, Tl, and V (data not presented).

Red tomatoes were only obtained for the natural organic fertilizer Tomato Tone® and for biosolids+K at the 1.5X N rate. For these two treatments, the content of 7 elements were observed to differ between red and green tomatoes (Table 8). Tomatoes fertilized with Tomato Tone® appear to have more Ca, Mg, and Sr than those fertilized with biosolids (Table 8). Green tomatoes had more Hg than red tomatoes, with the greatest amount being in the plants fertilized with Tomato Tone® (Table 8). Red tomatoes had more Co, Ti, and Tl than green tomatoes, with the greater amount being found in tomatoes fertilized with biosolids (Table 8).

Tomato fruits produced with biosolids fertilization generally did not contain greater quantities of heavy metals than those produced with natural organic or water-soluble fertilizers. The heavy metal results are in general agreement with a previous investigation involving heavy metal uptake of tomatoes grown with Milorganite® (Kussow and Iyler, 1996), but quantitative data from that study has not been published. However, that study was conducted in Wisconsin on a high-K Plano silt loam, and no fruit yield response was reported for supplemental K fertilization.

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