Heavy Metal Content of Tomatoes Fertilized With A Biosolids.

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ADDITIONAL INDEX WORDS. Milorganite, solanum lycopersicm, natural organic fertilizer

Biosolids re-cycle plant nutrients, but may also have constituents, such as heavy metals, that could contaminate food crops. Tomatoes (Solanum lycopersicm, 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, 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. Fruit was harvested through four months after seeding. 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 USA treat over 150 billion liters day^{-1} of wastewater, generating over 7 million tons (dry weight) yr^{-1} biosolids (USEPA, 2006). The biosolids may be incinerated, placed in landfills, or used as soil amendments and fertilizers.

Using biosolids as fertilizers re-cycles 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 gardener 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. The European Union 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. They have 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 content of most of these elements in tomato fruits grown in natural organic fertilizers, a water-soluble fertilizer, and in the biosolids Milorganite is examined in this paper.

Methods and Materials

Tomatoes (Solanum lycopersicm, var. Better Boy) were grown from seed in a greenhouse in plastic pots 25 cm dia. by 25 cm deep, using a mined sand containing sphagnum peat (90/10 sand/peat, by volume) to avoid root zone-connected heavy metal contamination. Three seeds were planted per pot on Sept. 28, 2013, and thinned to one plant per pot 27 days later. Fertilization was supplied by one of 4 sources, comprising two natural organic fertilizers (Tomato Tone®, Dr. Earth®), one water-soluble fertilizer (Miracle-Gro®), and one biosolids fertilizer (Milorganite®). The fertilizers were thoroughly mixed with most of the root zones prior to planting to provide 4 g N pot^{-1} (1X rate, equivalent to 150 lbs $acre^{-1}$ for a planting rate of 8500 plants $acre^{-1}$). 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^{-1}, and 8.1 g pot^{-1} K_2SO_4 was mixed into 3 of the four biosolids treatment root zones prior to planting to provide 3.5 g K pot^{-1} (4.0 g K_20 pot^{-1}, Table 1). At various intervals, based on the manufacturer's recommendations, fertilizers were applied post plant to total an additional 4 g N pot^{-1} (Table 1). The 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. 200 ml water and drenched into the root zone. Potassium sulfate was surface applied (8.1 g pot^{-1}) 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 (January 28, 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 110C. The vines were cut at soil level, dried at 60C, and weighed. The dried fruits, vines, and fertilizers (other than Miracle Gro) were ground in a Wiley mill, and digested with H_2SO_4/H_2O_2 (Lowther, 1980). 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.

Results and Discussion

<u>Fruit and vine weights</u>. Red fruits were only obtained with the Tomato Tone and 1.5X biosolids + K treatments. With the exception of the unfertilized control treatment, green tomatoes were obtained with all the other treatments. 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 Milorganite 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, 4). Vines fertilized with the biosolids Milorganite in the absence of added K contained less K than the natural organic or water soluble fertilizers (Table 3), and even the Milorganite treatments with added K contained numerically lower vine K than the other treatments, probably because the irrigated sand root zone retained little K against percolation. Milorganite 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 Milorganite treatments because of limited K. 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 Milorganite+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).

<u>Heavy metal content of tomato fruits</u>.

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 Milorganite generally contained greater amounts of many elements than the natural organic products (Table 4, 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 Milorganite in the absence of added K was among the highest (Table 6). In fact, the content of other nutrient and non-nutrient elements also was among the highest for tomatoes fertilized with Milorganite 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 Milorganite +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 Milorganite (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 Milorganite (Table 8). 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.

Literature Cited

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Table 1. Fertilization program

	Pre-plant N		Post-Plant
Fertilizer	Relative	g pot^{-1}	Fertilizations*
None	0X	0	0
Tomato-Tone	1X	4	5
Dr. Earth	1X	4	1
Miracle Gro	1X	4	5
Milorganite+K	0.5X	2	1
	1X	4	1
	1.5X	6	1
Milorganite-K	1X	4	1

* Post-plant N rate totaled 4 g pot^{-1}

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

	N rate	**Tomato yield**	**Vine weight**
Fertilizer	**(g plant^{-1})**	**(g plant^{-1})**	**(g plant^{-1})**
None	0	0f	<1d
Tomato-Tone	8	1018a	81a
Dr. Earth	8	105ef	27c
Miracle Gro	8	128def	32c
Milorganite 0.5X +K	4	227de	34c
Milorganite 1.0X + K	8	387bc	51b
Milorganite 1.5X +K	12	603b	54b
Milorganite 1.0X -K	8	267cd	36c

Values within a column followed by the same letter are not significantly different ($P < 0.05$) by the Duncans Multiple Range Test.

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

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

Values within a column followed by the same letter are not significantly different ($P < 0.05$) by the Duncans 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.

	N	P*	K*	Ca	Mg	Fe	Mn	Zn	Cu	Mo
Fertilizer	**% by weight**									
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**
Milorganite	**5**	**2**	**0**	**1.2**	0.5	**4**	0.1	0.04	0.02	0.0009

* Expressed as oxides

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
Milorganite	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**	**Milorganite 0.5X + K**	**Milorganite 1.0X + K**	**Milorganite 1.5X + K**	**Milorganite 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	7.7bc	5.4d	7.0c	7.5bc	7.3bc	8.5b	10.8a	**
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 the Duncans 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.

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

Values within a row followed by the same letter are not significantly ($P < 0.05$) different by the Duncans 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
Milorganite 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.