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Manganese Toxicity in Watermelon as Affected by Lime and Compost Amended to a Hawaiian Acid Oxisol

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Abstract. **Manganese (Mn) toxicity in crops is a serious problem in Hawaii, especially Kauai and Oahu, where most soils are highly weathered. To devise a management strategy to control Mn toxicity, a greenhouse experiment was conducted on an acid (pH 4.4) Oxisol (Wahiawa series) having 15g·kg^{-1} total Mn. Factorial combinations of lime (0, 2.0, and 4.0 g·kg^{-1} $CaCO_3$) and two composts (made from chicken manure and from sewage sludge at 0, 5, and 10 g·kg^{-1}) were applied to the soil, which was subsequently planted to watermelon (*Citrullus lanatus* Thunb. 'Crimson Sweet'). Our preliminary results showed that: 1) liming reduced Mn extractability and phytoavailability, but the reduction in Mn per unit increase in pH was much less than predicted by theory; 2) for good watermelon growth, soluble Mn, as extracted by the saturated paste method, should be <2.0 mg·L^{-1} corresponding to a soil pH >5.7; 3) unlike the saturated-paste extractable Mn, the Mehlich3-extractable Mn varied less with pH in a given soil series than between soil series; 4) effects of composts on Mn toxicity varied with compost properties, especially their Ca content and pH altering capacity; and 5) the diagnostic criteria for Mn toxicity in watermelon are tentatively proposed as: leaf Mn >1000 mg·kg^{-1} and leaf Ca/Mn ratio (g·g^{-1}) <25.**

Manganese (Mn) is an essential plant nutrient at low levels, but is toxic when in excess (Marschner, 1995). In fact, Mn toxicity is often more common than Mn deficiency in some regions, such as the humid tropics, where most soils are highly weathered and acidic (Hue et al., 2001). This is because the solubility and availability/toxicity of soil Mn are increased many fold by low pH, and the abundance of electrons (e^-) as shown below (Adams, 1981; Sparrow and Uren, 1987):

$$MnO_2 + 4\,H^+ + 2\,e^- \Longleftrightarrow Mn^{2+} + 2\,H_2O \qquad [1]$$

Acid soils with high Mn content have been found in many parts of the Hawaiian Islands, particularly on Kauai and Oahu, the two geologically oldest islands in the state. A major portion of agricultural land on Oahu consists of soils, mostly Oxisols of basaltic origin, with 10–40 g·kg^{-1} total Mn concentration (Fujimoto and Sherman, 1948). Such Mn concentrations are ≈10× greater than the average soil Mn concentration worldwide (Kabata-Pendias and Adriano, 1995). Total soil Mn, however, only indicates the potential toxicity. Actual Mn toxicity is associated with forms that are either water soluble or easily reducible. Adams (1984) suggested a reducible (presumably NH_2OH-HCl extractable) Mn range of 50–100 mg·kg^{-1} in soil, above which Mn toxicity would occur. To avoid Mn toxicity to soybean (*Glycine max* 'Kahala'), Hue et al. (2001) proposed to keep Mn concentrations in the saturated paste extract below 0.5 mg·L^{-1}, which corresponds to a soil pH of 5.6 or higher. The authors also reported that some low-molecular-weight organic compounds, green manure, and biosolids added to a high-Mn soil increased Mn phytotoxicity.

Different plant species or even varieties within a species have different degrees of tolerance to Mn (El-Jaoual and Cox, 1998; Foy et al., 1988). For example, adverse effects were observed when leaf Mn (in mg·kg^{-1}) exceeded 150 in bean (*Phaseolus vulgaris*), 650 in clover (*Trifolium subterraneum*), and 5000 in lowland rice (*Oryza sativa*) (Hannam and Ohki, 1988). Elamin and Wilcox (1986) reported that watermelon (*Citrullus lanatus* Thunb. 'Sugar Baby') developed Mn toxicity symptoms when leaf Mn exceeded 1300 mg·kg^{-1}; watermelon seemed to be more tolerant of Mn toxicity than was muskmelon (*Cucumis melo*) (Simon et al., 1986). Also, Mn toxicity was alleviated by high levels of other nutrients, particularly Ca (Horst, 1988; Hue et al., 2001), Mg (Elamin and Wilcox, 1986; Goss et al., 1991; Lohnis, 1960), and Si (Horst and Marschner, 1978).

The objective of this study was to determine how lime and organic materials (i.e., composts) used to amend a high-Mn, acid Oxisol affect soil Mn solubility/availability and watermelon growth, which was severely damaged when planted in the unamended soil. Our preliminary goal was to find a proper management between lime and organic amendments that would increase soil productivity and vegetable production, especially watermelon. (Given the Hawaii's remote location from other U.S. states and nations, fresh local vegetables are always important.)

Materials and Methods

Soil and amendment properties, and treatments. An acid soil with high Mn content (clayey kaolinitic isohyperthermic, Rhodic Eutrustox, Wahiawa series) was selected for the study for the following reasons: 1) the soil represents a large agricultural area in central Oahu, Hawaii, which was recently converted from sugarcane (*Saccharum officinarum*) to vegetables and other crops, such as coffee (*Coffea arabica*) and papaya (*Carica papaya*); and 2) several watermelon crops grown in this soil had consistently shed leaves at or just a few days after flowering, thereby yielding few and low-quality fruit (Hue et al., 1998).

In the unamended state, the soil had a pH of 4.4, 15 g·kg^{-1} total Mn, and 550 mg·kg^{-1} Mn as extracted by the Mehlich-3 solution. However, the soil's Al, as extracted by 1 M KCl, was only 10 mg·kg^{-1}. The X-ray diffraction data of the soil's clay fraction showed kaolinite and

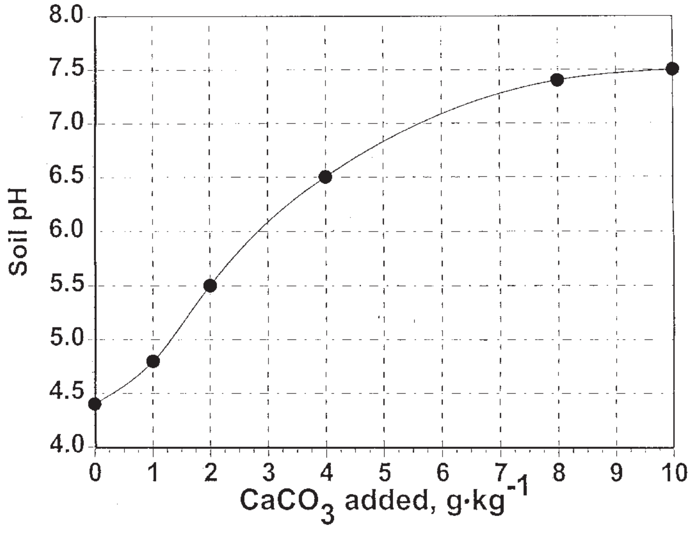

Fig. 1. Lime titration curve of the high Mn, acidic Oxisol (Wahiawa series) used in the watermelon experiment.

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gibbsite as the major mineral phases, and δ-MnO_2 (either vernadite or birnessite) was the predominant Mn oxide mineral. The lime titration curve (Fig. 1) of the soil showed that 2.0 and 4.0 g·kg^{-1} $CaCO_3$ were required to raise the soil pH to 5.5 and 6.5, respectively.

The lime ($CaCO_3$) used was of reagent grade having particle size <0.1 mm in diameter (from Fisher Scientific Co., Calif.). The organic amendments consisted of a chicken manure-based (CM) compost and a sewage sludge-based (SS) compost, which were ground and screened to pass a 2-mm sieve before use. Nutrient contents of the composts are listed in Table 1.

The $CaCO_3$ powder applied at 0, 2.0, and 4.0 g·kg^{-1} was thoroughly mixed with the dry soil, then deionized water was added to bring the soil to the field water holding capacity (≈280 g·kg^{-1} H_2O). The treated soil was air dried on a lab bench for 1 week, then crushed, re-wetted, and re-incubated (until dried) for another week. Two weeks after liming, composts were applied to the soil—limed and unlimed—at 0, 5, and 10 g·kg^{-1} by mixing the screened compost with the soil and subjecting the compost-amended soils to 2 wetting-drying cycles as previously described.

The treatments were a factorial combination of lime (three rates) and two composts at three rates each. There were 15 treatments, each replicated three times, and arranged in a completely randomized design in a greenhouse (Fig. 2). Each pot (replication) contained ≈2.0 kg dry soil and received (throughly mixed with the soil) a basal fertilizer containing (in mg·kg^{-1}) 140 N as urea, 130 K and 103 P as KH_2PO_4, 48 Mg as $MgSO_4$, 5 Cu and 5 Zn as their sulfate salts, 1 B and 0.5 Mo.

One week after the basal fertilizer application, ≈100 g of soil was sampled from each pot for chemical analysis. Then one 2-week-old watermelon seedling, 'Crimson Sweet', was transplanted in each pot, and grown for 4 w until the early flowering stage.

The aboveground biomass was harvested, dried in a forced-air oven at 70 °C for 48 h, and dry weight was recorded. The dried leaf tissue was ground to pass a 0.42-mm (60 mesh) sieve for nutrient analysis.

Chemical and statistical analyses

Soil samples. Soil pH was determined by equilibrating 20 g of soil with 20 mL of deionized water and measuring with a pH meter equipped with a pair of glass/calomel electrodes. Saturated paste extracts were obtained by equilibrating 50 g of soil with enough deionized water to form a paste for 30 min, then applying a vacuum suction to collect the liquid. Calcium, Mn, and other nutrients (e.g., P, K, Mg, Zn) in the saturated paste extract were measured with an inductively coupled plasma (ICP) spectrometer. However, only Ca and Mn are reported here. Mehlich-3 extractable Mn was obtained by shaking 2.0 g soil with 25 mL of the Mehlich-3 solution (Mehlich, 1984) for 5 min and filtering through Whatmann No. 6S filter paper. Calcium and Mn were measured with the ICP as previously described.

Table 1. Total nutrient concentrations of the two composts applied to the acidic Wahiawa Oxisol planted to watermelon.

Compost	N	P	K	Ca	Mg	Mn
				g·kg^{-1}		
Chicken manure-based (CM)	19	32	18	175	11	0.9
Sewage sludge-based (SS)	11	5	5	21	16	0.3

Plant tissues. Finely ground leaf tissues (0.20 g of the 4th and 5th leaves from the growing point) were dry ashed at 500 °C for ≈4 h until the ash turned whitish gray. The residue was mixed with 5 mL of 1 M HNO_3 and heated slowly at 120 °C until dryness (this step was taken to ensure a complete dissolution of Mn oxides). The residue was subsequently re-dissolved in 20 mL of 0.1 M HCl and filtered through Whatmann No. 2 filter paper before nutrient analysis (Ca, K, Mg, Mn, P, Zn) with the ICP.

Statistics. Effects of lime and compost on soil properties and watermelon growth were

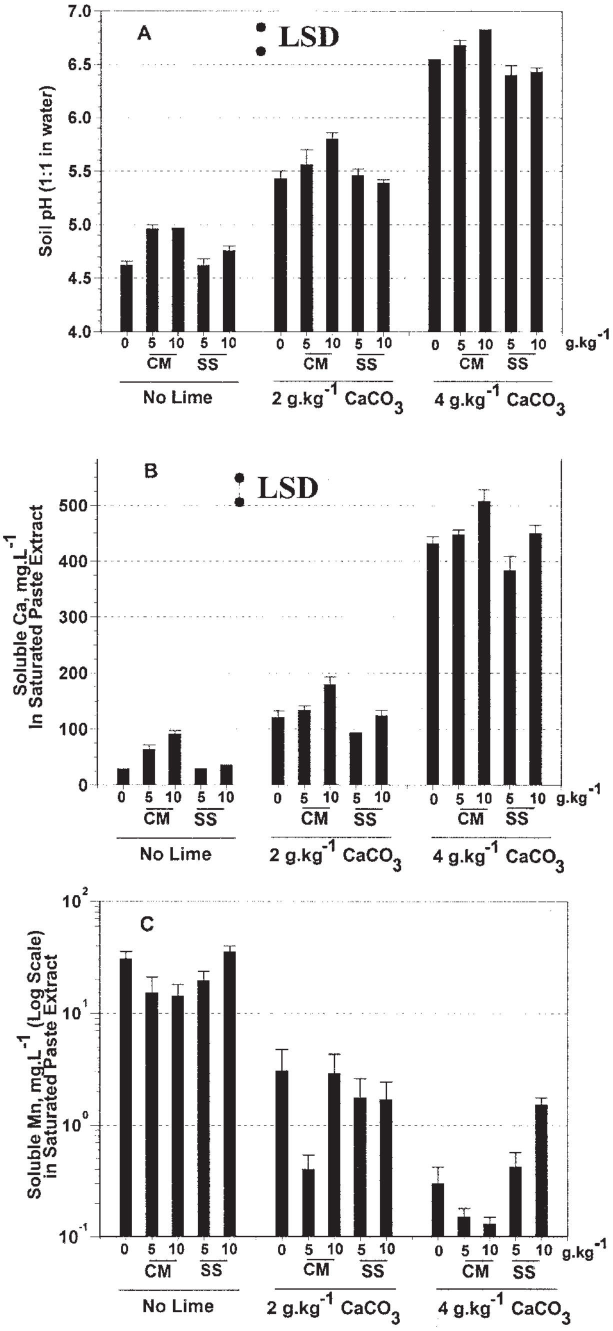


Fig. 2. Effects of lime and composts on (**A**) soil pH, (**B**) soluble Ca, and (**C**) soluble Mn of the Wahiawa Oxisol. CM = chicken manure-based compost; SS = sewage sludge-based compost. Vertical T's are SE of the means. LSD = least significant difference.

analyzed with the analysis of variance (ANOVA) and least significant difference (LSD) mean comparisons, using the SAS® software (SAS Inst., Cary, N.C.). Regression analysis (linear and nonlinear) was performed with the PLOTIT® software (Scientific Programming Enterprises, Haslett, Mich.).

Results and Discussion

Soil Mn and related properties as affected by lime and compost amendments. Soil pH increased by ≈1 unit for each 2 $g \cdot kg^{-1}$ $CaCO_3$ addition (Fig. 2A). The increase agreed well with the lime titration curve (Fig. 1). More interestingly, at each lime level, the CM compost raised soil pH ≈0.2–0.4 units whereas the SS compost slightly lowered soil pH (Fig. 2A). The differential effect on soil pH of the composts was likely a result of the higher liming potential of the CM compost than the SS compost as reflected by the former's higher total Ca content (Table 1) and high Ca concentration in the saturated paste extract (Fig. 2B).

Soluble Mn clearly decreased with increasing pH (Fig. 2C). The decrease, however, was not as fast as would be predicted by theory. In theory (Havlin et al., 1999; Hue et al., 2001), if the soil system is poised (i.e., pH + pe = constant), then solution Mn should decrease by 100-fold for each unit pH increase as illustrated below:

$$Mn^{2+} + 4\,OH^- <==> MnO_2\,(solid) + 2e^- + 2H_2O \qquad [2]$$

Equilibrium constant (K_{eq}) of reaction [2] can be expressed as

$$K_{eq} = (e^-)^2/[(Mn^{2+})\,(OH^-)^4]$$
$$= [(e^-)^2\,(H+)^4]/[(Mn^{2+})\,K_w^{\,4}]$$

where K_w is the dissolution constant of water, which is 10^{-14} at 25 °C.

Thus,

$$\log K_{eq} = 2\log(e^-) + 4\log(H^+) - \log(Mn^{2+}) - 4\log K_w$$

Rearranging yields

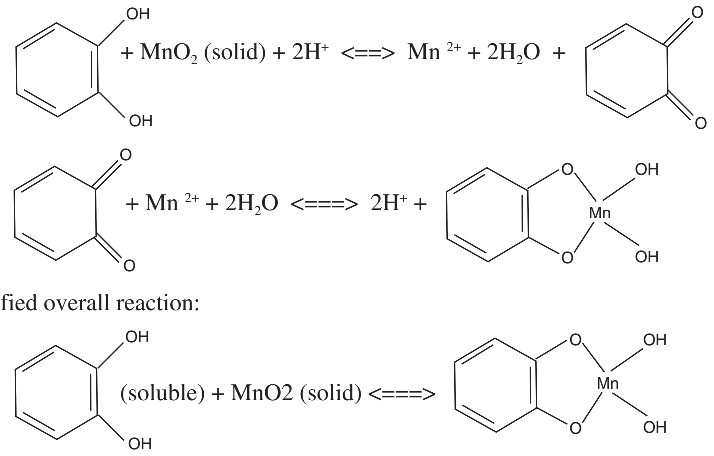
$$\log(Mn^{2+}) = -4\log K_w - \log K_{eq} + 2\,[\log(e^-) + \log(H^+)] + 2\log(H^+)$$

or

$$\log(Mn^{2+}) = K' - 2\,pH$$

$$\text{where } K' = -4\log K_w - \log K_{eq} + 2\,[\log(e^-) + \log(H^+)]$$
$$= -4\log K_w - \log K_{eq} - 2\,(pe + pH)$$
$$= \text{constant}$$

Our data, however, showed that solution Mn was decreased by only ≈10 fold for each unit increase in pH (Fig. 3). This is probably because the composts would produce organic substances capable of dissolving solid Mn and keeping it in solution (chelation) regardless of pH. For example, catechol can dissolve solid Mn, then chelate soluble Mn as suggested as shown above.



Also, it should be noted that the ICP-measured Mn represents total soluble Mn, not Mn^{2+}.

Although Mn concentration (and activity) in the saturated paste extract responded sensitively to soil pH changes, as illustrated by the 10- to 100-fold decrease in Mn for each pH unit increase, this parameter may not be suitable for soil testing purposes because considerable time, effort, and perhaps skills are required in its determination. Thus, an attempt was made to see if the Mehlich-3

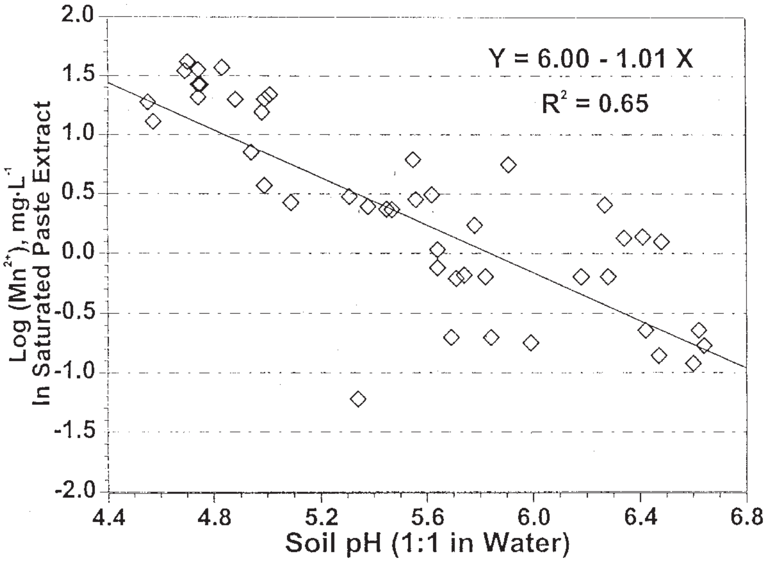


Fig. 3. Manganese concentration in the saturated paste extract of the Wahiawa Oxisol as a function of soil pH.

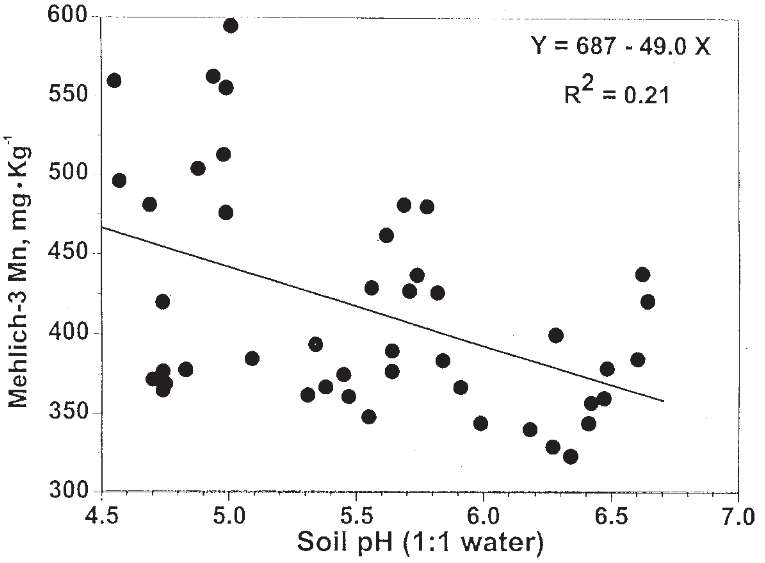


Fig. 4. Mehlich 3-extractable Mn of the Wahiawa Oxisol as a function of soil pH.

Table 2. Mehlich3 extractable Ca and Mn (mg·kg^{-1}) and soil pH of selected Hawaiian soils as affected by the addition of 2.0 g·kg^{-1} $CaCO_3$.

	Unamended soil			Limed soil		
Soil series/order	pH	Ca	Mn	pH	Ca	Mn
1. Halii/Oxisol	6.2	1710	4	7.1	1775	5
2. Lahaina/Oxisol	6.5	1440	350	7.8	1900	340
3. Leilehua/Ultisol	5.1	1180	11	6.3	1275	10
4. Lualualei/Vertisol	7.3	5560	225	8.0	5600	220
5. Makawili/Mollisol	7.0	1540	290	8.0	1720	290
6. Manana/Ultisol	4.6	200	4	5.5	466	4
7. Molokai/Oxisol	6.0	1500	695	7.0	1660	605
8. Paaloa/Ultisol	4.5	280	8	5.5	564	9
9. Wahiawa/Oxisol	4.4	400	550	5.5	700	520
10. Waimanalo/Mollisol	5.3	2530	150	6.3	2650	155
11. Waimea/Andisol	5.2	4200	10	6.2	4250	19

extract (Mehlich, 1984), which is a common extractant used by several soil testing labs to measure availability/toxicity of many plant nutrients including Mn, could detect changes in Mn with pH. The results were not encouraging, however. Although Mehlich-3 extractable Mn decreased slightly with increasing soil pH, its values varied greatly and independently of pH (Fig. 4). Soil pH only accounted for 21% of the variation in extractable Mn. To further confirm that the Mehlich-3 procedure did not extract Mn in proportion to the Mn solubility with respect to soil pH, a side experiment was conducted. The Mehlich-3 extractable Mn was measured in samples from 10 additional soil series from five different soil orders with and without 2.0 g·kg^{-1}

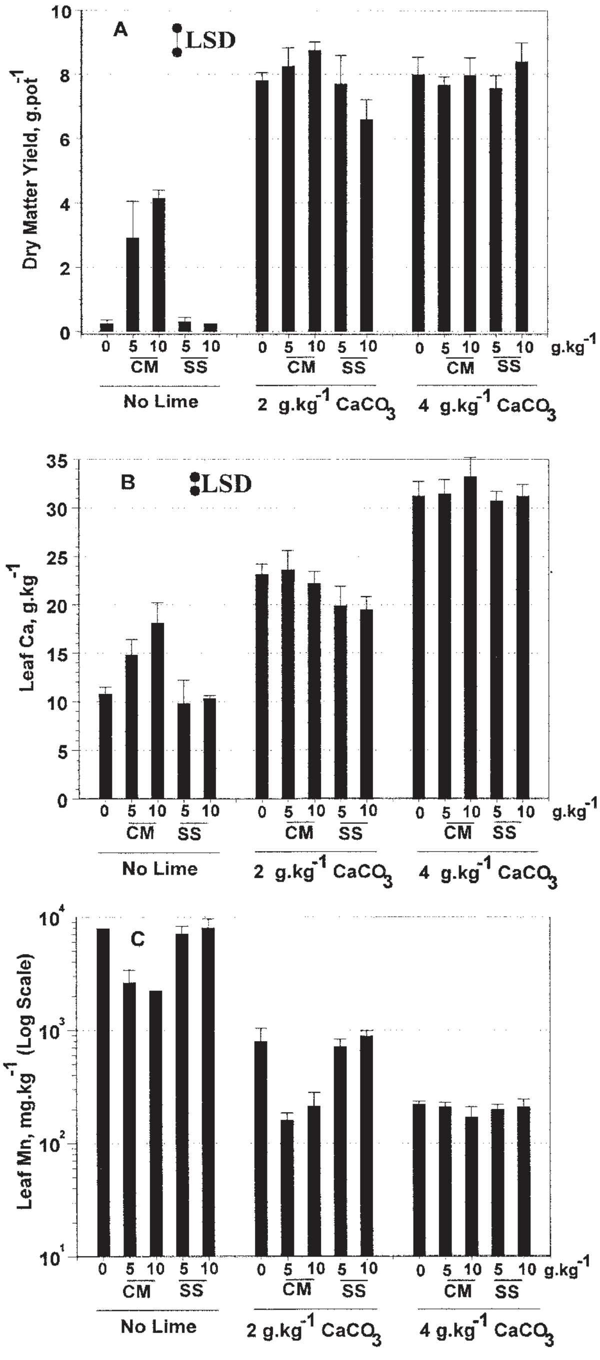


Fig. 5. Effects of lime and composts on dry-matter yield of (**A**) watermelon, (**B**) leaf Ca, and (**C**) leaf Mn. CM = chicken manure-based compost; SS = sewage sludge-based compost. Vertical T's are standard errors of the means. LSD = least significant difference.

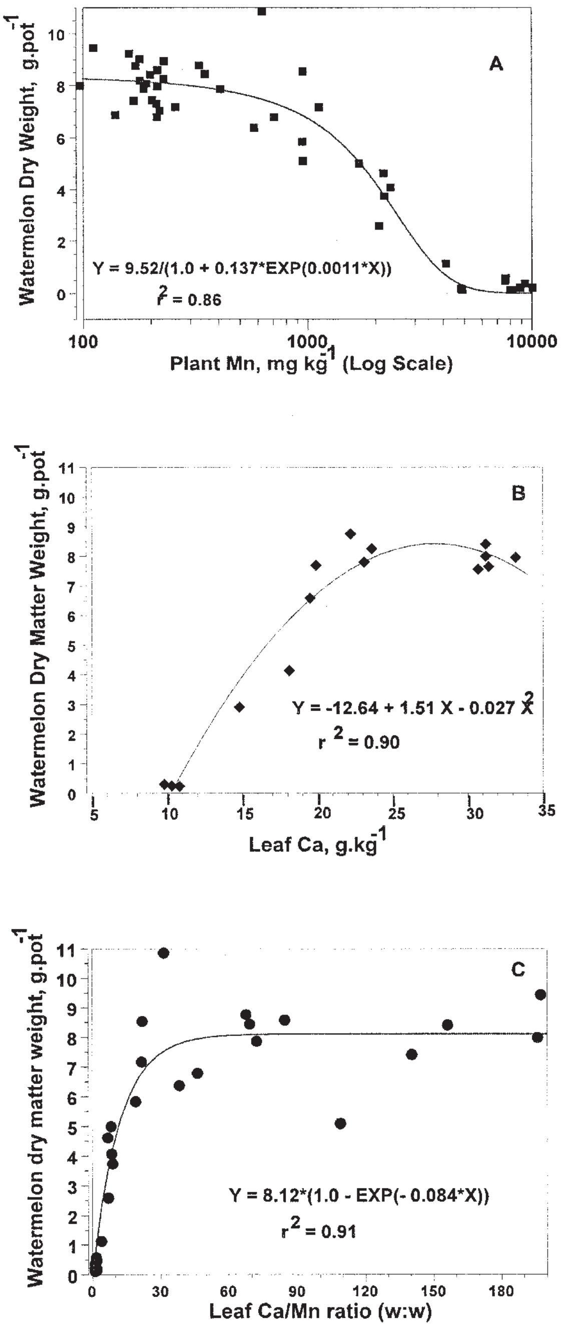


Fig. 6. Responses of watermelon, as measured by dry-matter weight, to (**A**) leaf Mn, (**B**) leaf Ca, and (**C**) leaf Ca/Mn ratio.

$CaCO_3$ and 2 weeks of incubation in moist/dry conditions. The data (Table 2) indicated that the Mehlich-3 solution could extract different amounts of Mn from different soils (suggesting the solution is perhaps sensitive to different Mn minerals), but it failed to extract different quantities of Mn from the same soil having different pHs.

Watermelon dry biomass and Ca, Mn concentrations in leaves as affected by lime and compost amendments. As expected, liming improved watermelon growth significantly: from <0.5 g per pot in the unamended control to ≈8.0 g per pot in the 2.0 $g \cdot kg^{-1}$ $CaCO_3$ treatment (Fig. 5A). Liming above this rate (i.e., at 4.0 $g \cdot kg^{-1}$ $CaCO_3$), however, did not increase the dry biomass. If leaf area and/or biomass is an early requirement for fruit weight and quality (the saleable product) then liming at ≈2.0 $g \cdot kg^{-1}$ $CaCO_3$ (or ≈4 tons per hectare $CaCO_3$ at 15 cm depth) would be more economical.

The effect of composts on watermelon growth was most striking in the no-lime treatments, in which the CM compost increased dry weight nearly 10 fold, whereas the SS compost slightly decreased growth relative to the control (Fig. 5A). With lime, the effect of composts on watermelon growth was not significant, except for the 10 $g \cdot kg^{-1}$ SS compost at 2 $g \cdot kg^{-1}$ $CaCO_3$, which significantly reduced growth as compared to growth in the other limed treatments (Fig. 5A). The cause of differential growth might have been related to the levels of Ca and/or Mn in the leaf tissue as discussed below.

Leaf Ca, at flowering, was ≈10 $g \cdot kg^{-1}$ in the unamended control, and no lime, but amended with the SS compost (Fig. 5B). In contrast, the 2 $g \cdot kg^{-1}$ $CaCO_3$ treatment raised leaf Ca to ≈22 $g \cdot kg^{-1}$ and the 4 $g \cdot kg^{-1}$ $CaCO_3$ treatment to ≈32 $g \cdot kg^{-1}$ (Fig. 5B).

Based on the Ca sufficiency range of 17–30 $g \cdot kg^{-1}$ for watermelon sampled at the early flowering–small fruit stage (Jones et al.,1991), it is likely that watermelon plants in the no lime treatments, except the one receiving 10 $g \cdot kg^{-1}$ CM compost, were Ca deficient. In contrast, plants in the limed treatments were all sufficient in Ca based on that same range (Jones et al., 1991).

Toxic Mn levels in watermelon seem to be 1000 to 1500 $mg \cdot kg^{-1}$ (depending on variety). For example, Elamin and Wilcox (1986) reported a toxic level of 1300 $mg \cdot kg^{-1}$ for 'Sugar Baby'. However, Jones et al. (1991) listed as "high" when leaf Mn exceeds 250 $mg \cdot kg^{-1}$ for watermelon (no variety mentioned). Based on such information, Mn toxicity likely contributed to the poor growth of watermelon in the unlimed treatments and in the low lime treatments receiving the SS compost (Fig. 5C). In fact, in these treatments we observed blackish brown spots on lower leaf surface, and necrotic lesions and chlorosis of leaf edges and tips. These visual symptoms are a strong indication of Mn toxicity (Hue et al., 1998; Simon et al., 1986). According to Wissemeier and Horst (1991), the brown color of these spots derives from oxidized polyphenols, not from MnO_2, although oxidized Mn concentration at the spots may be high as well.

By plotting dry matter from all the treatments as a function of leaf Mn (Fig. 6A), leaf Ca (Fig. 6B), and leaf Ca/Mn ratio (Fig. 6C), our results show the critical values for watermelon 'Crimson Sweet' tentatively (based on regression models) were: 1) Mn levels: 1000–1200 $mg \cdot kg^{-1}$ in leaf for a 10% reduction in dry weight; 2) Ca levels: 24–26 $g \cdot kg^{-1}$ for maximum growth; 3) a leaf Ca/Mn ratio ($g \cdot g^{-1}$) >25 is the best predictor of good growth if both Ca deficiency and Mn toxicity exist. According to Marschner (1995), a balance between Ca and Mn is required for normal plant growth. This is because high Mn increases activities of polyphenoloxidase enzymes, especially indolacetic acid (IAA) oxidase. This results in increased degradation of IAA and reduced basipetal transport of IAA, which is required to counter balance the acropetal transport of Ca. Thus, Ca deficiency, such as crinkle leaf in cotton (*Gossypium hirsutum*) is an indirect result of Mn toxicity.

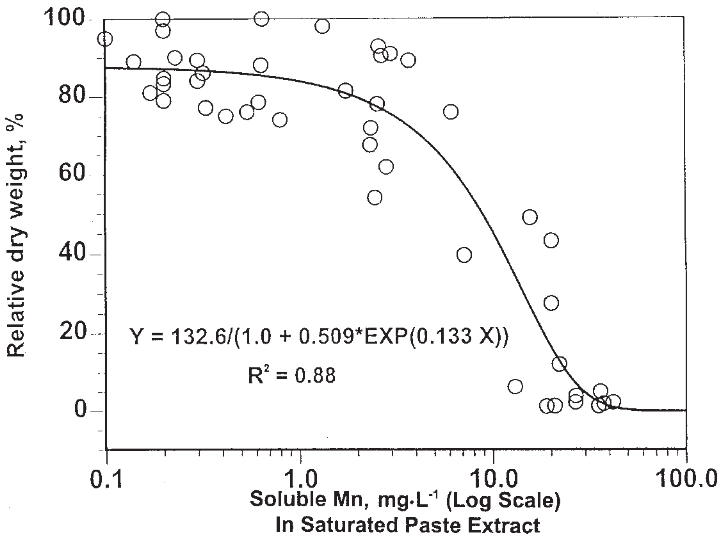


Fig. 7. Relationship between watermelon dry weight and Mn concentration in the saturated paste extract of the Wahiawa Oxisol.

A critical level of ≈2.0 $mg \cdot L^{-1}$ Mn was obtained by regressing relative dry weight against soil-solution Mn (as extracted from the saturated paste), above which 10% growth reduction would be expected based on the regression (Fig. 7).

Conclusions

The effects of lime and organic amendments on the availability/toxicity of soil Mn were studied in a greenhouse experiment, using watermelon as a test crop. The soil, an acidic (pH 4.4) Oxisol with 15$g \cdot kg^{-1}$ total Mn, was amended with 2 composts at 3 application rates and with lime ($CaCO_3$) at 3 rates, in the treatments factorially arranged. The results showed that: 1) adequate lime to raise soil pH to 5.7 or above, resulting in soluble (paste extracted) Mn <2.0 $mg \cdot L^{-1}$, is essential for normal watermelon growth; 2) Mehlich3 extracting solution is not sensitive to soluble/available Mn in a given soil at different pH levels; 3) reduced growth is expected if leaf Mn exceeds 1000 $mg \cdot kg^{-1}$ in watermelon; and 4) leaf Ca/Mn ratio should be used in predicting Mn toxicity.

Although our results were preliminary, it is recommended that when watermelons are grown on high-Mn soils, lime be applied enough to attain pH >5.7, so Mn in the soil can be reduced. The use of composts on such soils is more complex: compost properties must be identified. This is because some are beneficial while others may be detrimental to the plant yield.

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