

## Decline of *Citrus* Due to Presence of Landfill Gases in the Soil

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Over a period of years, the decline of citrus has been noted near sites located on or near landfill areas where aerobic or anaerobic decomposition is taking place. The symptoms of these declines in vigor are: growth reduction, leaf abscission, wilting, dieback, and eventual death of the tree. Similar declines have occurred in peach (*Prunus persica* Batsch.) orchards near landfills (Flowers 1977), as well as in woody species of ornamentals (Costa 1971).

A number of compounds have been associated with the aerobic digestion of organic matter, and as the amount of oxygen decreases, the products of various organisms can change (Bird and Lynch 1974). The aerobic products produced can include as the major constituents carbon dioxide,  $H_2O$ , and organic acids, notably acetic acid (Farquhar 1968). Other compounds can include simple sugars and amino acids (Flowers 1977).

One hydrocarbon found in both anaerobic (Smith 1972) and aerobic soil conditions (Cornforth 1975) is ethylene. This compound can be formed from a number of organisms using methionine and glucose and other compounds as substrates (Bird and Lynch 1974; Ilag 1968; Lynch 1972). Ethylene is an active hormone in *Citrus* and can affect plant metabolism by changing membrane permeability of cells (Abeles 1972), leaf and fruit abscission (Cooper 1969), reduction in stem elongation and leaf chlorosis. The addition of ethylene to the soil by injection can cause a variety of symptoms, including tree defoliation, dieback, loss of young fruit, and delay in growth or death (Wilson 1966). It is at present the only olefin with hormonal properties (Abeles 1972).

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<sup>1</sup>Mention of a chemical or proprietary product does not constitute a recommendation for use by the U.S. Department of Agriculture to the exclusion of other products that may also be suitable.

Although methane is the major product of anaerobic digestion, it, in itself, has not been found to be toxic to trees (Flowers 1977). Under low oxygen conditions citrus will have reduced growth, but only at oxygen levels <1.5%. Girtton (1927) established that high levels of CO<sub>2</sub> (<37.2%) were necessary to arrest root growth in sour orange (Citrus aurantium L.), but in neither case is there death of tissue.

The low oxygen created by anaerobic conditions in landfills is considered by some workers to be like that of flooding for extended periods of time (Gilman 1981).

Disturbed uptake can be a factor in the disposition of minerals due to low oxygen potentials. Zinc is found to accumulate in the roots of citrus subjected to low oxygen potentials either by high moisture or low oxygen content of the root media (Labanauskas 1966). The disturbance of Zn movement in citrus blight by some unknown stress will cause accumulation of this element in xylem and phloem tissue of Citrus. This type of accumulation does not occur with other known stresses.

These studies were designed to determine the soil gas near landfills where citrus tree decline occurs and whether the environmental stresses imposed in the areas studied could alter Zn accumulation in wood tissue.

#### MATERIALS AND METHODS

The trees selected for these experiments were adjacent to two landfill sites located in two different areas in Florida. The Lake County site consisted of 'Valencia' (Citrus sinensis (L.) Osb.) orange trees on 40-year-old rough lemon (C. limon (L.) Burm. f.) rootstock, and a site located in Highlands County with 21-year-old trees with the same rootstock/scion combination. Both sites are located in areas having the same soil type, namely deep Astatula (hyperthermic, uncoated; typic quartzipsamments), which is extremely porous sand with <1% organic matter. Both sites have an underlying horizon of reddish sand which is high in iron content.

Each site is a deeply excavated area that contains household refuse which is the source of substrate for the production of volatiles. The landfills were approximately 25 m deep in both sampling sites.

Gas sampling tubes were placed in the soil where obvious signs of tree decline (wilting, leaf abscission) were observed, as well as in an area where visual symptoms of decline were absent.

Gas sampling tubes were located 0 (on top of landfill), 30, 60, and 90 m from the approximate border of the landfill in Highlands County, and 18, 30, 45, and 90 m from the landfill in

Lake County. At each of these positions, two tubes were placed parallel to the landfill perimeter and 15 m apart.

The steel tubes were 5.1 cm in diameter and 3.05 m deep. Each tube was sealed with concrete which was placed 3 m around its circumference to prevent any ambient contamination of the gases in the tube.

Sampling was accomplished using either an evacuated metal or glass container connected to the soil tube by 4 mm I.D. polypropylene tubing. The metal sampling container had an internal volume of 12.46 L, the glass sampling bulb 250 mL, and the soil tube 25.20 L. The metal and glass sampling containers were equipped with rubber septa.

The soil gases, carbon dioxide ( $\text{CO}_2$ ), oxygen ( $\text{O}_2$ ), carbon monoxide ( $\text{CO}$ ), and methane ( $\text{CH}_4$ ), were analyzed using a Shamadzu Model 7AG gas chromatograph using 120-140 mesh carbosieve B in a stainless steel 3.17 mm O.D. X 1.82 m column, helium with a flow rate of 40 mL/minute and a thermoconductivity detector that was programmed to rise from 50°C to 130°C at a rate of 30-32°C/minute.

Due to the high concentrations of methane found in soil gases near landfills, a method was devised to separate  $\text{CH}_4$  from ethylene ( $\text{C}_2\text{H}_4$ ) using an exclusion method based on the trapping of olefins by a mercuric perchlorate solution. The system described by Young (1952) was modified to allow the injection of a quantity of landfill gas (50 mL) into a stream of nitrogen which had first passed through a 0.25 M mercuric perchlorate solution in a gas washing apparatus, and then passed into a gas bubbler (c porosity) which contained 10 mL of 0.25 M mercuric perchlorate - 4 M perchloric acid (at 0°C). The  $\text{N}_2$  was allowed to flow at a rate of 50 mL/minute for 10 minutes. A portion (2 mL) of the reacted solution was transferred to a serum vial (10 mL) fitted with a miniert valve (Supelco, Bellefonte, Pennsylvania). These valves are constructed of Teflon<sup>TM</sup> and have a compression mechanism which allows a gastight seal and are equipped with a septum and a sliding locking valve. The  $\text{C}_2\text{H}_4$  releasing system (2 mL 4 M HCL) was added to the  $\text{Hg}(\text{ClO}_4)_2$  by injection through the septum and mixed. After 10 minutes of equilibration of the released gas, a 1-mL sample was removed by a gastight syringe and injected into a gas chromatograph using the conditions described earlier (Evensen 1981).

The trapping system was found to be 90-95% efficient at 0°C for  $\text{C}_2\text{H}_4$ . Care was taken to equalize pressures inside the vial by the removal of an appropriate gas volume to compensate for the addition of liquids. Authentic gas standards were used to compare retention times with the unknowns for identification.

Table 1. Soil gas concentrations under 'Valencia' orange trees (Lake County site).

Month	DLF <sup>1</sup> (m)	CO <sub>2</sub> (%)	O <sub>2</sub> (%)	CH <sub>4</sub> (%)	C <sub>2</sub> H <sub>4</sub> (ppm)	TD <sup>2</sup>
Sep	18.3	49.5 ± 3.1	1.1 ± 0.05	45.1 ± 1.5	11.0 ± 1.1	+
Nov		45.0 ± 4.2	4.0 ± 0.1	42.0 ± 2.1	12.4 ± 1.7	+
Sep	30.4	17.6 ± 2.8	4.0 ± 0.6	20.1 ± 3.1	12.4 ± 2.6	+
Nov		17.2 ± 3.1	3.0 ± 0.2	19.0 ± 2.5	21.1 ± 1.4	+
Sep	45.0	4.1 ± 0.5	18.8 ± 1.1	0.1 ± 0.01	2.0 ± 0.8	-
Nov		8.1 ± 0.8	17.0 ± 1.3	0.6 ± 0.02	1.0 ± 0.2	-
Sep	95.0	0.1 ± 0.05	21.0 ± 2.4	2.0 ± 0.5 (ppm)	-	-
Nov		0.1 ± 0.05	20.8 ± 4.1	2.5 ± 0.1 (ppm)	-	-

<sup>1</sup>Distance from landfill.

<sup>2</sup>Trees declined.

Wood tissue was taken from 10 trees of the same age at the Lake County and 18 trees at the Highlands County sites. The wood tissue was collected and prepared according to a procedure used by Wutscher and Hardesty (1979).

Zinc in wood tissue was analyzed with a Jarrell-Ash Plasma Atom Company 750 (Jones 1977).

## RESULTS AND DISCUSSION

Anaerobic conditions near landfill sites are judged by the results of the gas analyses from each site. The O<sub>2</sub> content ranged from 21% at a distance of 95 m from the apparent border of the landfill to 1% at a distance of 18 m at the Lake County site. A decreasing O<sub>2</sub> concentration gradient in the soil corresponded with an increasing amount of decline as distance to the landfill decreased. A similar gradient exists at the Highlands County site, with the O<sub>2</sub> at the lowest levels on or near the landfill (Table 2). The value for 0 in m in Table 2 (directly on landfill) produced a higher O<sub>2</sub> value than the nearest position near the landfill. This is primarily due to the dense material used to cap the landfill after filling is completed. This sampling area was approximately 12 m from the first sampling (18 M) area, and the effects of gravity on the heavier gases are evident.

Methane is produced in the soil near the landfill sites and decreases as the distance from the site is increased. The levels range from 45% at the 18 m area in the Lake County site

Table 2. Soil gas concentrations under 'Valencia' orange trees (Highlands County site).

Month	DFL <sup>1</sup> (m)	CO <sub>2</sub> (%)	O <sub>2</sub> (%)	CH <sub>4</sub> (%)	C <sub>2</sub> H <sub>4</sub> (ppm)	TD <sup>2</sup>
Jan	0.0	17.2 ± 1.2	7.2 ± 0.8	28.1 ± 2.4	55.0 ± 2.1	-
Feb		22.1 ± 0.5	10.4 ± 0.5	24.0 ± 5.1	22.1 ± 1.3	-
Jan	30.0	46.6 ± 1.2	2.2 ± 0.2	60.5 ± 2.2	68.0 ± 1.1	+
Feb		40.2 ± 0.5	0.8 ± 0.1	56.2 ± 1.2	74.0 ± 1.3	+
Jan	60.0	28.9 ± 1.7	13.4 ± 0.2	11.4 ± 3.1	31.0 ± 1.1	+
Feb		42.8 ± 2.1	14.0 ± 2.1	27.5 ± 2.1	32.2 ± 1.4	+
Jan	90.0	11.2 ± 1.2	16.9 ± 1.8	10.6 ± 1.1	3.0 ± 0.2	-
Feb		12.1 ± 0.8	15.8 ± 1.4	10.1 ± 0.2	-	-

<sup>1</sup>Distance from landfill.

<sup>2</sup>Trees declined.

to 2.5 ppm 95 m from the landfill boundary. The levels of gas dropped slightly during the two sampling dates, but the change was not significant.

Similar data were collected at the Highlands County site, with concentrations ranging from 60.5% to 10.6% methane at the first collection date and 56.2% to 10.1% on the second collection date. The presence of 10% methane in the samples has no visible influence on the decline of these trees at the 95 m area.

Ethylene was present in these anaerobic conditions in Highlands and Lake county areas and ranged from 2 to 11 ppm on the first collection date to 1 to 21.1 ppm on the later date at the Lake County site (Table 1). The levels of ethylene were 21.1 to 90 ppm higher in the Highlands County site (Table 2) with the highest value occurring at the sampling site nearest the landfill.

Carbon dioxide levels were highest in the areas nearest the landfill sites with values ranging from 11.2 to 46.6%, respectively, at the 90 m and 30 m from the Highlands County site (Table 2). At the Lake County site the CO<sub>2</sub> levels ranged from 0.1 to 49.5% at the 95 m and 18 m sites, respectively (Table 1).

Comparisons of the zinc levels in the first 2 cm of xylem (wood tissue) in the areas of aerobic and anaerobic conditions show no relationship between the levels of O<sub>2</sub> found in the soil and the occurrence of Zn in the wood (Fig. 1). In fact, the highest

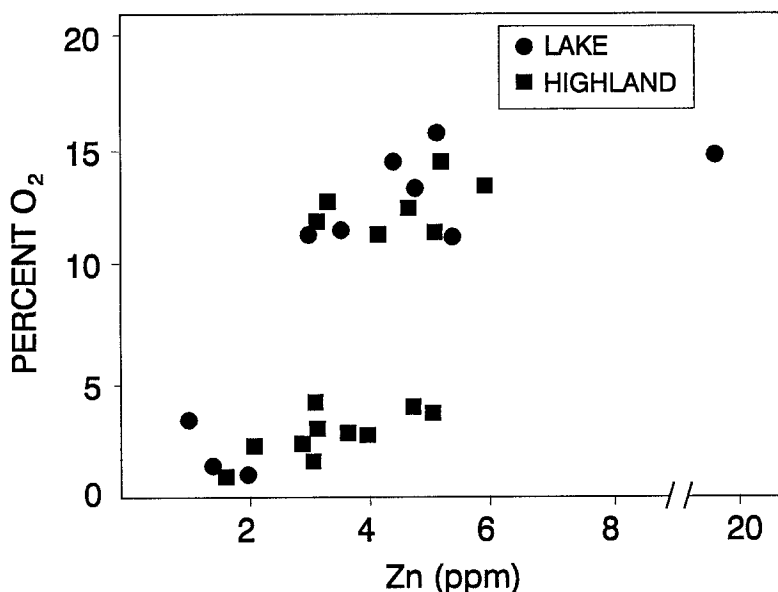


Figure 1. Values for zinc from wood samples taken from the highlands and Lake County sites. No significant difference was found when O<sub>2</sub> content of the soil and Zn content were compared.

level of Zn found was in a tree in an area with high O<sub>2</sub> (20.8%) content. The Zn level was 19.4 ppm in the trunk wood.

Although the soil gas environment created by the influence of the landfills in this study are detrimental to the growth and development of Citrus, the decline of citrus trees cannot be attributed solely to the presence of anaerobic conditions, since the changes in O<sub>2</sub> and CO<sub>2</sub> content of the soil were gradual with increased O<sub>2</sub> levels as the distance increased from the landfill, yet citrus trees were still in decline (Table 2). Growth of citrus can be maintained at 14% O<sub>2</sub>, but the decline of the trees at the landfill sites was apparent from leaf loss and twig dieback. The level of ethylene in this area was found to reach a level of 27.5 ppm which is high for physiological activity in citrus. Sour orange (C. aurantium L.), and 'Mexican' lime (C. aurantifolia (Christm.)) seedlings will die back if aqueous solutions of 10-25 ppm ethylene are present (Bausher unpublished). Ethylene has been mentioned as a possible cause of vegetation death in landfills, but without published record (Flowers 1977).

The existence of ethylene can be attributed to the possible lack of  $O_2$  which reduces its ability to be oxidized to  $CO_2$  (Cornforth 1975); however, the large level of substrates found in landfills can increase the production of ethylene above that required by aerobic organisms for use as substrate for the formation of  $CO_2$ . Ethylene can also be formed by chemical breakdown of rubber and plastic wastes due to chemical and bacterial action (Bird and Lynch 1974). The accumulation of Zn found in citrus blight was not found in these sampling sites. The presence of elevated Zn is not correlated with the presence of external anaerobic conditions in the root systems of Citrus. The uptake of mineral elements under reduced  $O_2$  potentials can increase the uptake of Zn in roots of Citrus sinensis (L.) Osb. due to the presence of periodically flooded soil conditions (Labanauskas 1966) or imposed low  $O_2$  levels (Labanauskas 1965). The changes in Zn or its accumulation in citrus blighted trees seems to be specific for this disease and not predicated on a general stress reaction such as low  $O_2$  levels caused by periodic high water tables or other anaerobic sources. Also, the conditions of high soil ethylene did not seem to affect the Zn content of the wood.

The production of gases by organic substrates makes landfill areas a poor location for planting citrus or other ethylene-sensitive plant systems.

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