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OAK RIDGE
NATIONAL
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MARTIN MARIETTA

Effects of Various Uranium Leaching Procedures on Soil: Short-Term Vegetation Growth and Physiology

Progress Report-April 1994

Nelson T. Edwards

Environmental Sciences Division
Publication No. 4289

MANAGED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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ENVIRONMENTAL SCIENCES DIVISION

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SUMMARY

This study examined the vegetation-support capacity of soils that were leached to remove uranium using various chemical leaching techniques. Short-term phytotoxicity tests were performed on 1-week-old oat and radish seedlings that were grown in nonleached soil and in soil leached with various chemicals. All soils tested were from the storage pad area at Fernald site in Ohio and were either bench-scale-leached at Oak Ridge National Laboratory (ORNL) or pilot-scale-leached at Fernald. Bench-scale leachates included 0.5 molar total carbonate solutions of sodium carbonate and sodium bicarbonate, 3.1 molar citric acid, and 0.8 molar total carbonate solution of ammonium carbonate and ammonium bicarbonate. All bench-scale-leached soils were washed with solutions of 0.05 molar calcium chloride ($CaCl_2$) followed by two additional washings with distilled water. The $CaCl_2$ washing was necessary before seeds would germinate on the soil. Some of the soils from the bench-scale leachings were amended with lime and/or fertilizer before phytotoxicity tests were performed. All pilot-scale leached soils tested were leached with sodium carbonate, but various leaching procedures were used. All tests performed on the pilot-scale leached soils were performed on unamended soil, although the leaching procedures did include thorough rinsing with water following sodium carbonate leaching. Phytotoxicity tests included whole plant gas (H_2O and CO_2) exchange, photosynthesis, dark respiration, leaf pigment concentrations, root length, root weight, stem + leaf weight, and stem height. Physiological functions and the growth of seedlings grown in soil bench-scale leached with sodium carbonate or citric acid followed by $CaCl_2$ washing and fertilization (plus lime in the case of citric acid) were equal to or greater than that for seedlings grown in nonleached soil. Physiological functions and growth rates were also comparable in seedlings grown in pilot-scale-leached soils to those in seedlings grown in nonleached soils. However, dry weight to stem height or root-length ratios were generally greater in plants grown in nonleached soils than plants grown in leached soils. This indicates that the plants grown in the nonleached soils had larger diameter stems and larger leaves or that their tissues were more dense than those of plants grown in leached soils. It also indicates that the roots of plants grown in nonleached soil were more numerous, thicker, or that root tissues were more dense than those of plants grown in leached soils. These results may indicate that the plants grown in the leached soils were less vigorous than plants grown in the nonleached soil in spite of the fact that their net growth rates (in terms of biomass accumulation and net carbon gain) were equal to or in some cases greater than growth rates of plants grown in nonleached soil.

Longer term pot studies with soils from the most promising leaching procedures should determine whether this is a major problem. Caution is advised for all of the leaching procedures in terms of damage to the soil structure. These tests were performed under ideal moisture conditions for seed germination and seedling

establishment. In the field, soils with the structure of these leached soils would require careful mulching and watering regimes. If left to natural weather conditions, the soils will either be too dry and hard at the surface or too muddy for seed germination and seedling establishment to occur.

1. INTRODUCTION

Significant volumes of soil containing elevated levels of uranium exist in the eastern United States (predominately at the Fernald site, located about 18 miles northwest of Cincinnati, Ohio, and at the Oak Ridge Y-12 Plant, Oak Ridge, Tennessee). The contamination resulted from the development of the nuclear industry in the United States requiring the mining, milling, and fabrication of a large variety of uranium products. The contaminated soil poses a collection and disposal problem of a magnitude that justifies the development of decontamination methods. Consequently, the Department of Energy (DOE) Office of Technology Development formed the Uranium in Soils Integrated Demonstration (USID) program to address the problem (ORNL-6762, 1993). The fundamental goal of the USID task group, with investigators at four DOE facilities [Argonne National Laboratory (ANL), Los Alamos National Laboratory (LANL), ORNL, and Savannah River Project (SRP)], has been the selective extraction/leaching or removal of uranium from soil faster, cheaper, and safer than what can be done using current conventional technologies. The objective is to selectively remove uranium from soil without seriously degrading the soil's physicochemical characteristics and without generating waste that is difficult to manage and/or dispose of. However, procedures developed for removing uranium from contaminated soil have involved relatively harsh chemical treatments that do affect the physicochemical properties of the soil. The questions are (1) are the changes in soil properties severe enough to destroy the soil's capacity to support and sustain vegetation growth and survival? and (2) if necessary, what amendments might be made to the leached soil to return it to a reasonable vegetation production capacity? This study examines the vegetation-support capacity of soil that had been chemically leached to remove uranium by means of a variety of chemical leaching techniques. The approach is to conduct short-term germination and phytotoxicity tests for evaluating soils after they are subjected to various leaching procedures followed by longer term (full life cycle) pot studies on successfully leached soils that show the greatest capacity to support plant growth. This report details the results from germination and short-term phytotoxicity testing of soils that underwent a variety of leaching procedures at the bench scale at ORNL and at the pilot plant at Fernald. All soils tested were from the storage pad area at Fernald.

2. METHODS AND MATERIALS

Leaching and soil preparation procedures: In the bench-scale tests at ORNL, 500-g soil samples were leached by attrition scrubbing for 5 min at 20°C with a 0.5 molar total carbonate solution of sodium carbonate/bicarbonate ($Na_2CO_3/NaHCO_3$), a 0.8 molar total carbonate solution of ammonium carbonate $(NH_4)_2CO_3/NH_4\,CO_3$), or a 3.1 molar solution of citric acid. Soils were then either air dried and pulverized or washed five times with 1.5 l of 0.05 molar $CaCl_2$ solution followed by two additional washings with distilled water before drying and pulverization. A complete description of bench-scale leaching procedures is discussed in ORNL-6762 (1993).

All of the soils were either tested with no further treatment after leaching or were fertilized with agricultural lime or fertilizer containing 10% nitrogen, 10% phosphorus, and 10% potassium. Lime was added at rates equivalent to 8 tons per acre and fertilizer was added at rates equivalent to 1000 lb per acre.

Three samples of storage pad soil were also tested after leaching at the Fernald pilot plant. All three samples were leached with sodium carbonate. Sample T4S50 was particle-size fractionated and attrition scrubbed with water. Sample T16S50 was not fractionated, and a polymer was used before attrition scrubbing with water. Sample T17S50 was particle-size fractionated and attrition scrubbed with water at 40°C. Additional tests are in progress.

Seed germination: Seed germination tests and a series of physiological and growth measurements were performed for each set of plants growing in leached soils and in a single nonleached control soil sample, as indicators of phytotoxicity. Oat and radish plants were used in all of the tests. All tests were performed in a growth chamber set to control temperature at 18°C during 10 h dark periods and at 24°C during 14 h light periods.

Maximum potential germination percentages were determined by placing 100 seeds of each species between single layers of moistened blotter paper within a small covered plastic dish. The germination dishes were left in the growth chamber but in a darkened container for 72 h. Germinated seeds were then counted. This procedure was repeated with each soil tested, except that a 5-mm moist layer of each treated soil replaced the bottom layer of blotter paper.

Physiological processes: Physiological measurements were performed on 1-week-old seedlings of each species. Plants were grown in small glass tubes (10 cm tall, 5 cm wide, and 0.5 cm thick). Each tube contained 20 g (dry weight) of soil. The glass tubes were open on each end. A 1-cm-thick layer of absorbent cotton placed in the bottom of each tube held the soil in place and also provided a wick to transport water from an external water reservoir into the soil. Eight glass tubes were used for each species per treatment. Germinated seeds of each species were planted slightly beneath the surface of the soil in each tube. Three radish or two oat seedlings were established in each tube. The seedlings were allowed to grow under the growth chamber conditions described above.

Physiological measurements included whole plant gas exchange (net CO_2 gain and dark respiration), photosynthesis and transpiration, and plant pigment concentrations. Gas exchange measurements were performed with a Li-Cor infrared gas analyzer (Li-Cor, Lincoln, Nebraska). For whole plant gas exchange, a single glass tube with plants was placed inside a 1-l Li-Cor gas exchange chamber. To correct for soil respiration, a single glass tube containing soil but without plants was placed in the chamber. Photosynthetic and transpiration rates were measured by placing a measured area of intact leaf tissue inside the Li-Cor 1-l chamber. The photosynthesis and transpiration measurements were performed only on the oat seedlings. Plant pigment concentrations (chlorophyll a, chlorophyll b, and carotenes) were determined for plants grown in bench-scale sodium carbonate-leached soil with fertilizer added vs nonleached controls. Plant pigments were removed from the plant leaves by soaking overnight in ethyl alcohol. Absorbence values of the extracts were determined at wavelengths of 665, 649, and 470 nm, and the pigment concentrations were calculated (Lichtenthaler and Wellburn, 1983).

Growth measurements: Net growth was determined by measuring the root length and dry weight and the stem height and dry weight of each plant. At the end of a 1-week growth period, seedlings were removed from the soil; the soil was rinsed from the roots with water; and the stem height and root length of each seedling were measured. Roots and stems were separated, dried at 75°C, and weighed. For the bench-scale treatments, it was necessary to ash the roots for determining ash-free dry weights, because of the difficulty of removing soil from the roots. In the pilot-scale study, the roots were allowed to soak in water for 2 h before a final rinsing. In this way, all of the soil was removed from the roots, thus eliminating the necessity to ash them.

Treatment designations and data analyses: Treatment designations throughout the text and figures are defined below:

Washed - Leached soil was washed with $CaCl_2$, as described above, with no lime or fertilizer.
Not washed or unwashed - Soil received no further treatment after leaching.
Limed - Lime was added to soil after leaching and washing.
Sodium carbonate (fertilized) - Fertilizer was added after $CaCl_2$ washing.
Citric acid (fertilized) - Fertilizer was added after $CaCl_2$ washing *and* liming.

Gas exchange data are expressed as means of eight measurements. Rooting depth, stem height, and weight data are expressed as means of 24 measurements for radish seedlings and 16 measurements for oat seedlings. All error bars in the figures represent 1 standard error.

3. RESULTS AND DISCUSSION

3.1 BENCH-SCALE STUDIES

Seed germination: Germination percentages averaged 80% and 60% for radish and oats, respectively, on nonleached soil. These rates were near the maximum rates that were determined with absorbent paper. No germination occurred on unwashed sodium carbonate-leached soil, presumably due to high sodium concentrations caused by the leaching process. After washing the sodium carbonate-leached soil with $CaCl_2$, germination rates were comparable to rates in the nonleached control soil. Germination percentages in unwashed ammonium carbonate-leached soil were less than 20% for both radish and oats. Percentages in citric acid-leached soil after washing with $CaCl_2$ were 50% for radish and 40% for oats. Results from the germination tests are illustrated in Fig. 1. Note that the tests were conducted under ideal moisture conditions. Due to damage to the soil structure and the high salt concentrations that result from the various leaching processes, the surface soil dries rapidly and is resistant to re-wetting. The soil dries to a concrete-like crust at the surface; water added to the surface is not readily absorbed by the soil. Moisture movement from a water reservoir beneath the soil moves very slowly up through the soil by capillary action. Evaporative losses exceed this capillary movement, even in the relatively mild environmental conditions of a growth chamber. Therefore, establishment of vegetation in the field would require a very careful watering regime and/or heavy mulching. Also, in the field, rainfall would tend to runoff rather than soak into the soil after the soil drys.

Physiological processes: Both radishes and oats grown in sodium carbonate-leached soil that had been washed with $CaCl_2$ and fertilized exhibited a greater net carbon gain rate just prior to harvest than did plants grown in any of the other soils, including the nonleached control soil (Fig. 2). Radishes grown in citric acid-leached soil that had been washed, limed, and fertilized had a greater net carbon gain rate than oats grown in any of the other soils, except for those grown in washed and fertilized sodium carbonate-leached soil. The lowest net carbon gain rate was observed in plants grown in sodium carbonate soil that had only been washed and in citric acid-leached soil that was only washed or washed and limed. Net carbon gain could not be determined for any of the soils that were not washed, because no growth occurred. The growth data presented below supports the net carbon gain results, except that the growth data generally indicate better growing conditions in the nonfertilized and nonlimed soils than do the net carbon gain data. However, much of the growth of young seedlings comes from nourishment stored in the seeds and therefore may not be as indicative of longer-term growth as the gas exchange data. The gas exchange data were collected after depletion of most of the stored food supply.

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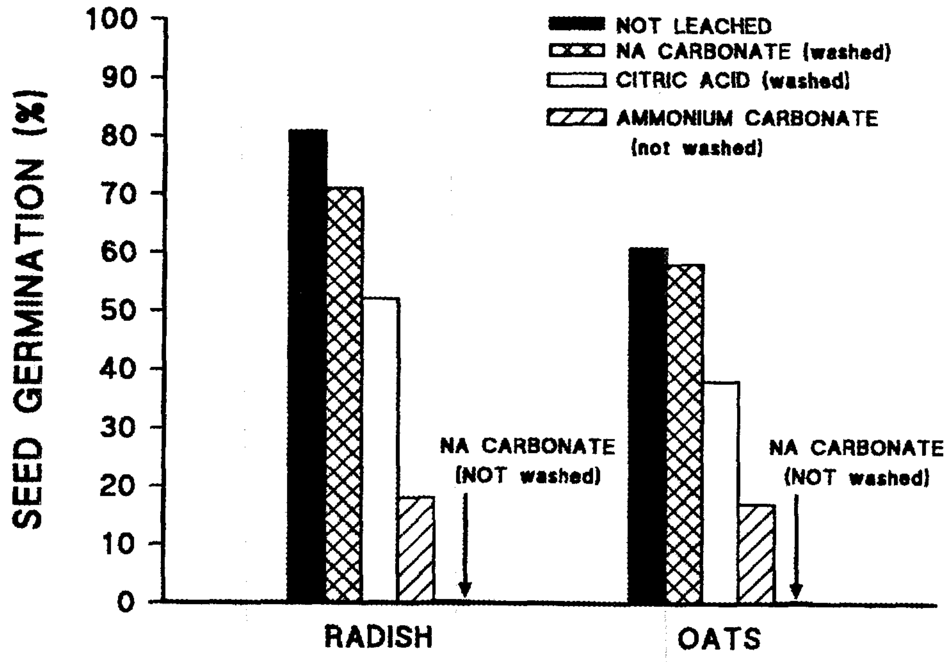

Fig. 1. Germination percentages of radish and oat seeds in nonleached control soil and in washed and unwashed soil leached with sodium carbonate, citric acid, and ammonium carbonate.

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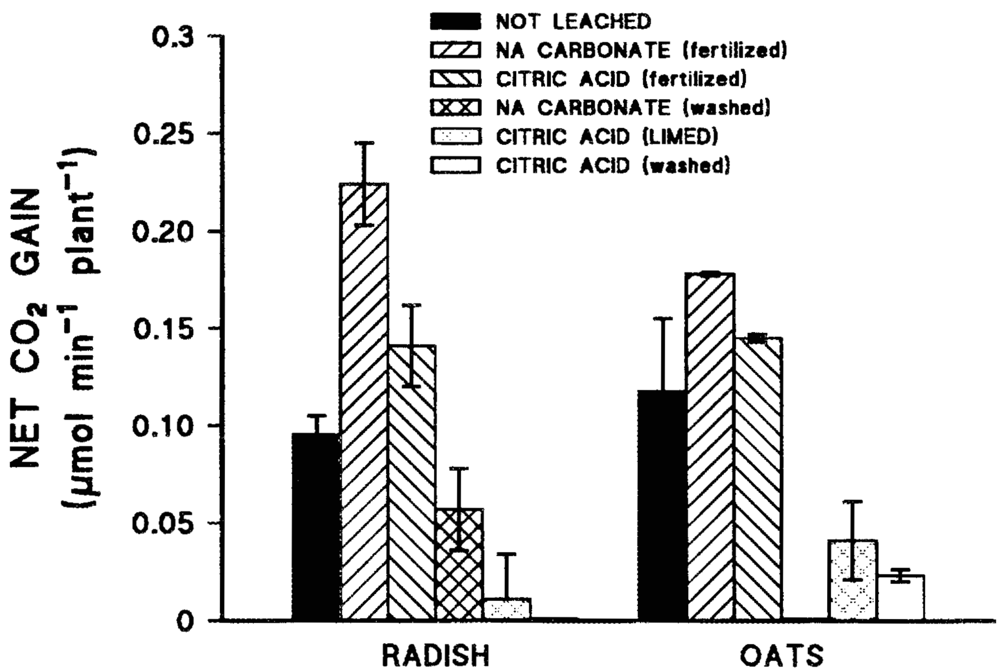

Fig. 2. Net carbon dioxide gain in radish and oat seedlings growing in nonleached soil and soils that had been leached and treated using various techniques.

Photosynthetic rates in oats were greater in plants grown in washed and fertilized, sodium carbonate-leached soil than in either washed, limed, and fertilized citric acid-leached soil or in nonleached soil (Fig. 3). There were no differences in photosynthetic rates in oats grown in nonleached soil and citric acid-leached soil that had received lime and fertilizer. Transpiration rates were generally greater in the oats having the greatest photosynthetic rates. However, transpiration rates in oats grown in the limed and fertilized citric acid-leached soil were very low relative to photosynthesis rates resulting in a relatively high water-use efficiency (Fig. 3). This is a desirable trait in plants, especially those growing in areas where drought is a problem. However, these plants were grown in water-saturated soil, and any advantage in terms of water use efficiency would not have been manifested in terms of growth or survival.

Growth: Dry matter accumulation in both the oat and the radish seedlings grown in washed, limed, and fertilized citric acid-leached soil was significantly greater than that of seedlings grown in any of the other soils including the nonleached control soil (Fig. 4). Dry matter accumulation in radishes was poorest in sodium carbonate and citric acid-leached soils that were treated only by washing, with essentially no growth occurring in the washed, citric acid-leached soil. Similar results were recorded for oat seedlings except that the lowest dry matter accumulation occurred in the limed or washed, citric acid-leached soil. However, while dry matter accumulation in radish plants was near zero in washed, citric acid-leached soil, oat seedlings grown in washed citric acid-leached soil exhibited dry matter accumulation that was not significantly less than that of oat seedlings grown in limed, citric acid-leached soil.

Except for the radishes grown in washed citric acid-leached soil (where no growth occurred), stem heights and rooting depths were as great or greater in the leached soils than in the nonleached control soil (Fig. 5). In both radish and oat seedlings, stem height and rooting depth were greatest in the sodium carbonate-leached soil regardless of treatment, except that no growth occurred in the unwashed sodium carbonate soil. Rooting depth and stem height were less in plants grown in the nonleached control soil than in any of the leached soils, whereas the dry weights of the plants grown in nonleached soil were near equal to or in some cases greater than dry weights of the plants grown in leached soil. Figures 6 (a, b, and c) illustrate the relationships between stem height and weight or root length and weight for the various treatments including both bench-scale and pilot-scale treatments. Weight/stem height ratios were greater in both radish and oat plants grown in nonleached soil and fertilized, citric acid leached soil than in plants grown using any of the other treatments. The lowest (stem + leaf weight/stem height) ratios were observed in plants grown in pilot-scale leached soil. Root weight/root length ratios of plants grown in nonleached soil were equal to or greater than ratios for plants grown in any of the treated soils, except for oats grown in pilot-scale treated soils. Root weight/root length ratios of oats grown in pilot-scale leached soil were greater than for oats grown in any of the other soils

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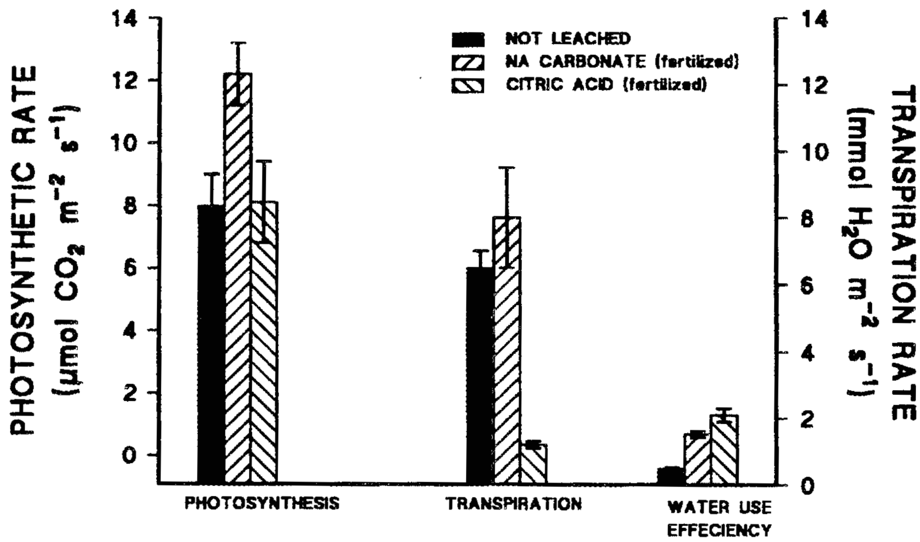

Fig. 3. Photosynthetic and transpiration rates in leaves of oat seedlings growing in nonleached soil; washed and fertilized sodium carbonate-leached soil; and washed, limed, and fertilized citric acid-leached soil. Water-use efficiency (photosynthesis/transpiration) is a ratio and may be read from either scale.

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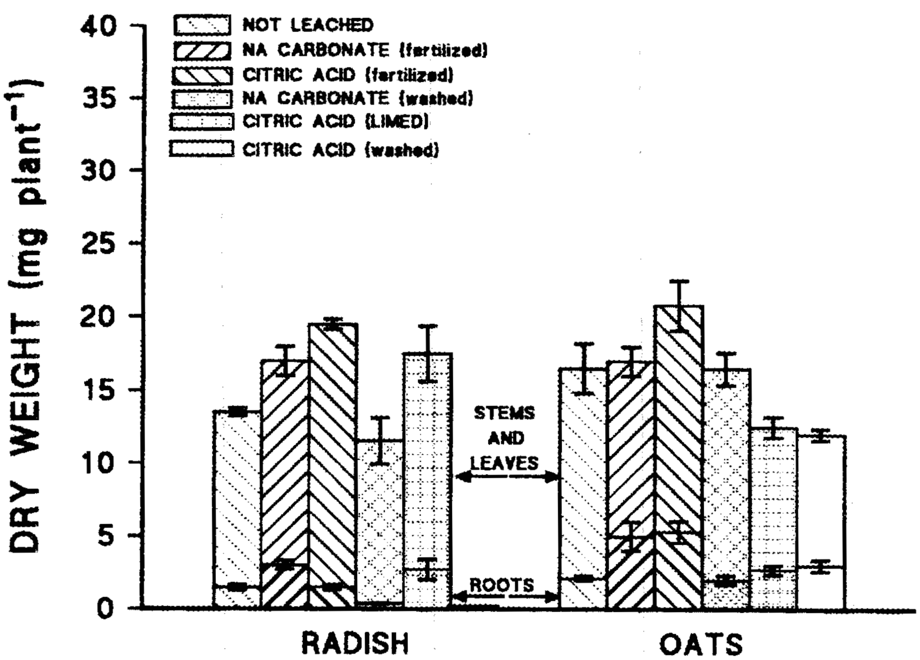

Fig. 4. Dry weights of 1-week-old radish and oat seedlings grown in nonleached soils and in soils treated by various leaching, washing, and fertilization regimes.

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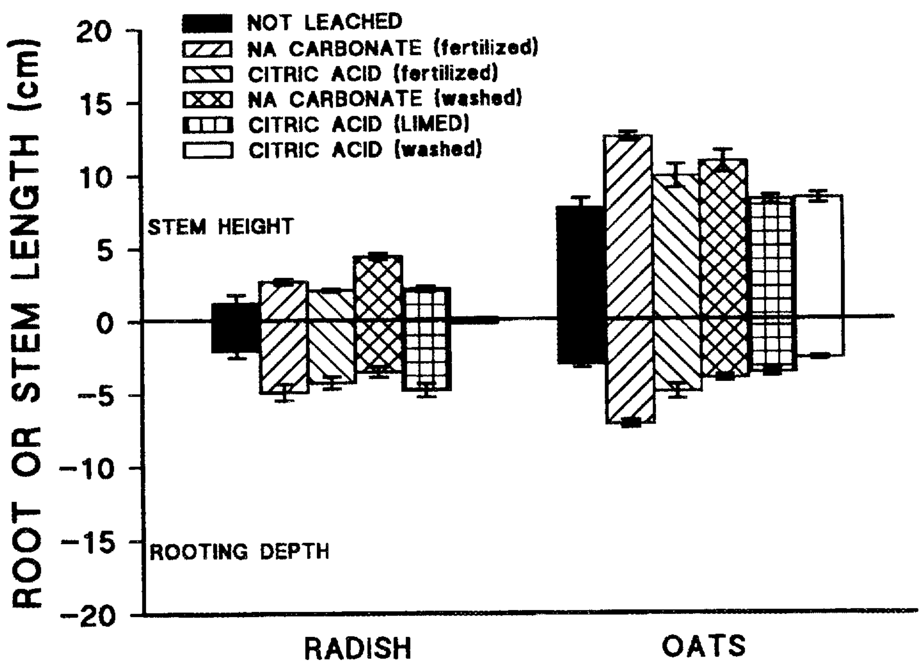

Fig. 5. Rooting depths and stem heights of 1-week-old radish and oat seedlings grown in nonleached soil and in soils that had been treated by various leaching, washing, and fertilization regimes.

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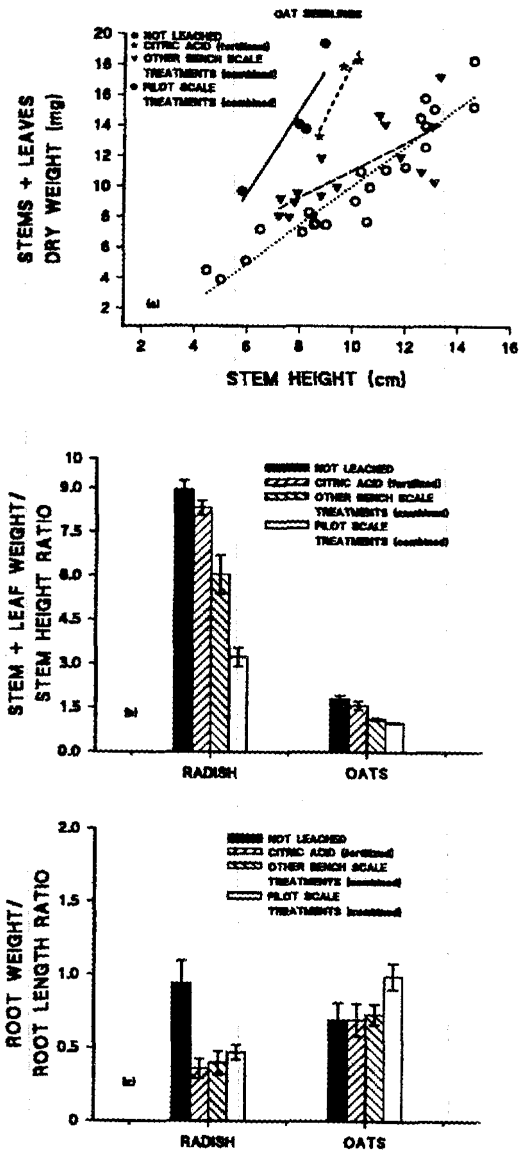

Fig. 6. (a) Mean stem + leaf weights plotted against mean stem heights for oat seedlings grown in nonleached soil and in leached soils. (b) Mean stem + leaf weight/stem height ratios for oats and radishes grown in nonleached soil and leached soils. (c) Mean root weight/root length ratios for radish and oat seedling in nonleached soil and in leached soils.

including the nonleached soil. These data indicate a more branched rooting system and possibly larger (diameter) stems and leaves or higher density tissue in the plants from the nonleached soil and from the fertilized, citric acid-leached soil. Either or both conditions are indicative of vigorous vegetation with better survival capabilities than for plants with lower weight to stem height and root length ratios. For example, a more branched rooting system as opposed to a poorly branched rooting system would be an advantage for root exploration of the soil for moisture and nutrients.

Although no fertilizer tests were performed with the nonleached control soil, these results indicate the probability of a nutrient deficiency in that soil. This is supported by the pigment concentration data that showed lower chlorophyll a concentrations in leaves of radishes grown in nonleached soil than in leaves of radishes grown in fertilized, sodium carbonate-leached soil (Fig. 7). Such differences were not observed in oats, perhaps because oat seeds are larger than radish seeds with a potentially greater supply of nutrients, such as nitrogen, that are essential for chlorophyll production in plants. The fact that growth was greater in plants growing in some of the washed, leached soils without fertilizer additions (e.g., oats in washed, citric acid-leached soil and both oat and radish seedlings in washed, sodium carbonate and citric acid-leached soils) than in the nonleached control soil indicates that some previously unavailable nutrients may have been made available for plant uptake by the leaching and washing procedures.

3.2 PILOT-SCALE STUDIES

Seed germination: Radish seed germination was 85%, 84%, and 80% in the T4S50, T16S50, and T17S50 leached soils. These percentages were not different from the percentages in the nonleached control soil (82%). Oat seed germination averaged about 62% in an earlier test of nonleached control soil, as compared with a range of 44% to 53% in the three pilot-scale leached soils. However, the lower oat seed germination percentages in the leached soils were about equal to concurrently run germination tests with absorbent paper. Therefore, the lower germination percentages were due to decreased natural viability of the oat seeds over time. No nonleached soil was available at the time these tests were performed.

Physiological responses: Net CO_2 gain in nonleached soil and in the T17S50 soil was greater with both radish and oat plants than in the other pilot-scale leached soils tested (Fig. 8). The lowest whole plant CO_2 gain rates, for both radishes and oats, were in T4S50 soil. However, the greatest rate of photosynthesis was measured in oats grown in the T4S50 soil, indicating relatively high rates of respiration in the roots and stems (Fig. 9). Transpiration rates were greatest and water-use efficiencies were lowest in oat plants grown in nonleached and T17S50 soils. Water-use efficiencies were about twice as high in oats grown in T4S50 and

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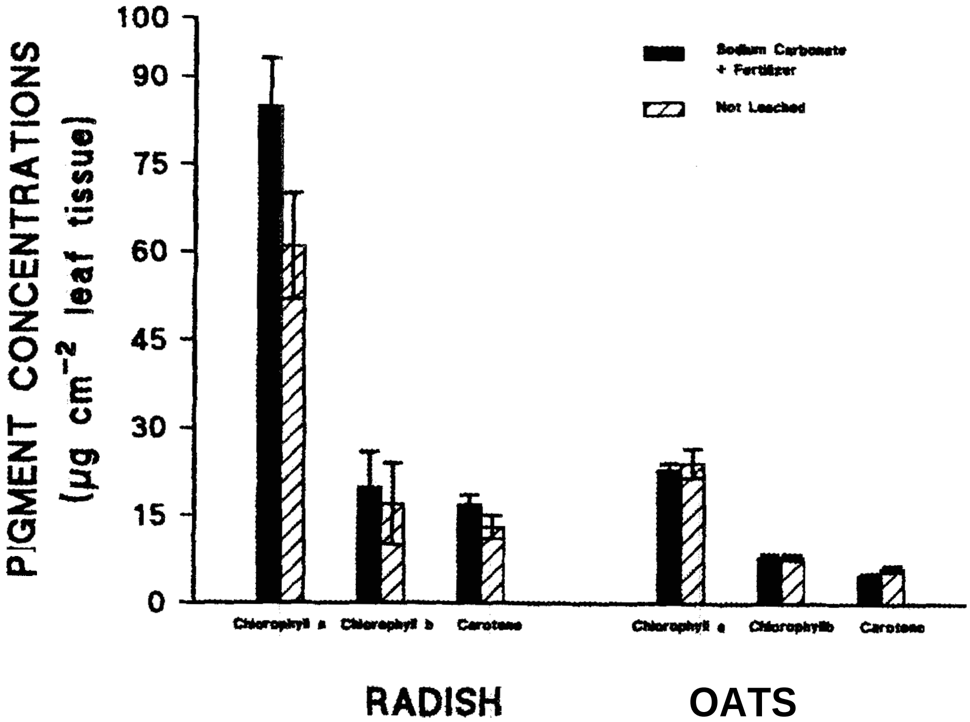

Fig. 7. Pigment concentrations in leaves of radish and oat seedlings after 1 week of growth in nonleached soil and in fertilized sodium carbonate-leached soil.

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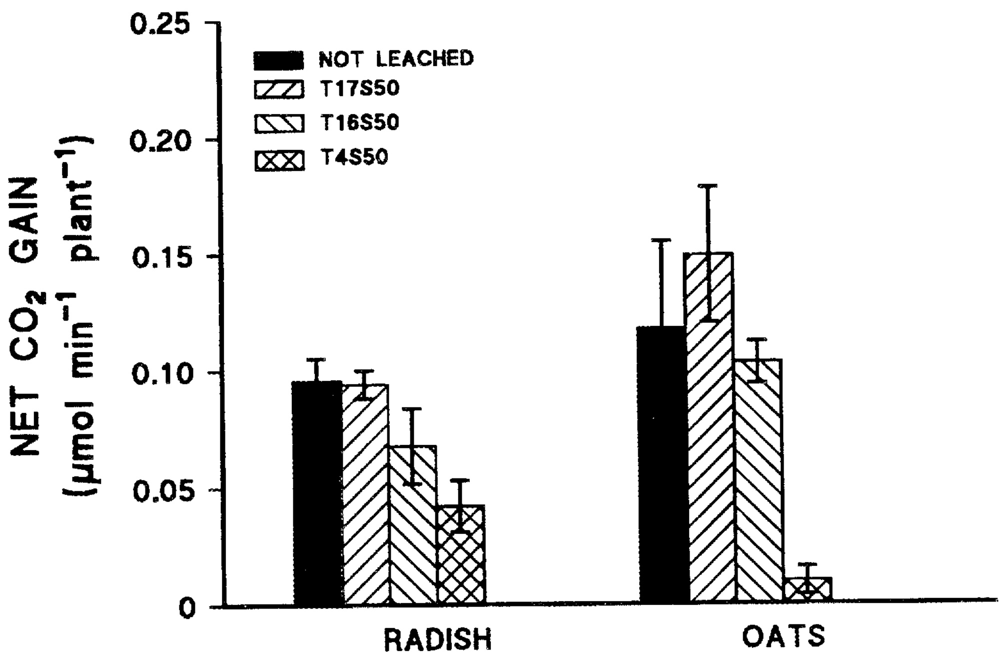

Fig. 8. Net carbon dioxide gain in 1-week-old oat and radish seedlings growing in nonleached soil and in storage pad soil having undergone three sodium carbonate leaching procedures at the pilot scale.

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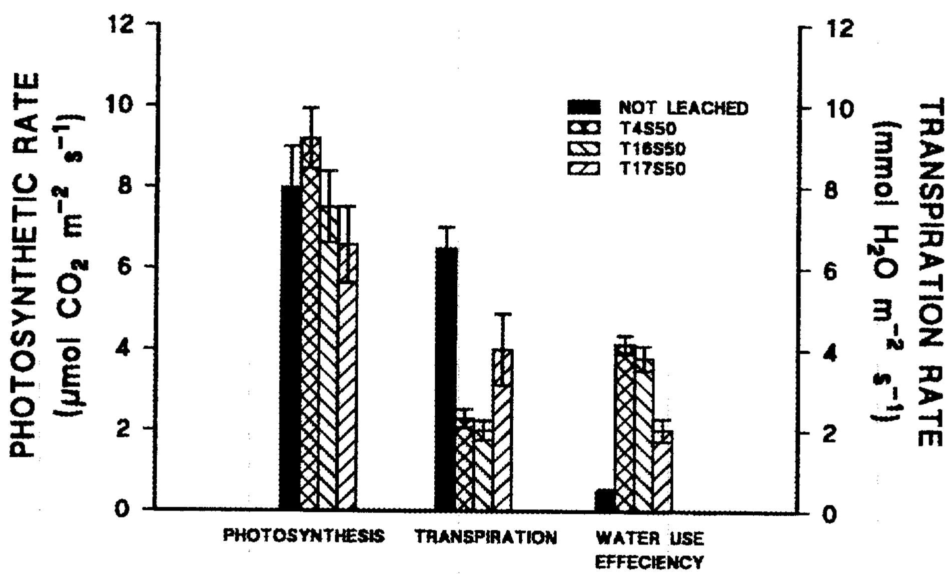

Fig. 9. Photosynthetic and transpiration rates in leaves of 1-week-old oat seedlings growing in nonleached soil and in storage pad soil having undergone three sodium carbonate leaching procedures at the pilot scale. Water-use efficiency (photosynthesis/transpiration) is a ratio and may be read from either scale.

T16S50 soils than water-use efficiencies of oats grown in any of the other soils including nonleached and bench-scale leached soils.

Growth: Net CO_2 gain results generally supported the dry matter accumulation data. However, contrary to indications of a low net CO_2 gain rate in plants grown in the T4S50 soil, the actual accumulated dry weight at harvest was greater in radishes grown in this soil than in any of the other pilot-scale leached soils (Fig. 10). Dry weight accumulation in oats was about equal among plants grown in nonleached soil and in the pilot-scale leached soils, including the T4S50 soil. These seemingly contradictory results with plants grown in the T4S50 soil could be explained if nighttime respiratory carbon losses were unusually low. However, we did not measure gas exchange at night; therefore, we have no evidence to support this theory.

Stem height and rooting depth were least in the nonleached soils, as observed in the bench-scale test (Fig. 11). Also, as in the bench-scale test, the rooting depths and stem heights were less than might be indicated by the comparison of biomass accumulation among the various tests.

4. CONCLUSIONS

Although all tests have not been completed (e.g., long-term growth studies are just beginning), several tentative conclusions can be drawn from the results presented: (1) seed germination results indicate that bench-scale leached soils require additional washing to bring germination percentages up to acceptable levels; (2) plants will grow in soil leached at the bench scale with sodium carbonate or citric acid only if they have been "washed," and fertilization is necessary for growth rates to approach rates in unleached soil; (3) plant growth rates (dry mass production) in pilot-scale leached soils that have been tested to date (T17S50, T16S50, and T4S50) are equal to or greater than plant growth rates in unleached soil; (4) stem and leaf tissue density measurements indicate that plants grown in unleached soil or fertilized,citric acid-leached soil are structurally stronger than plants grown in any of the other leached soils that were tested; and (5) all of the leaching procedures damage the soil structure to the extent that seedling establishment is extremely difficult, requiring heavy mulching and regular water misting.

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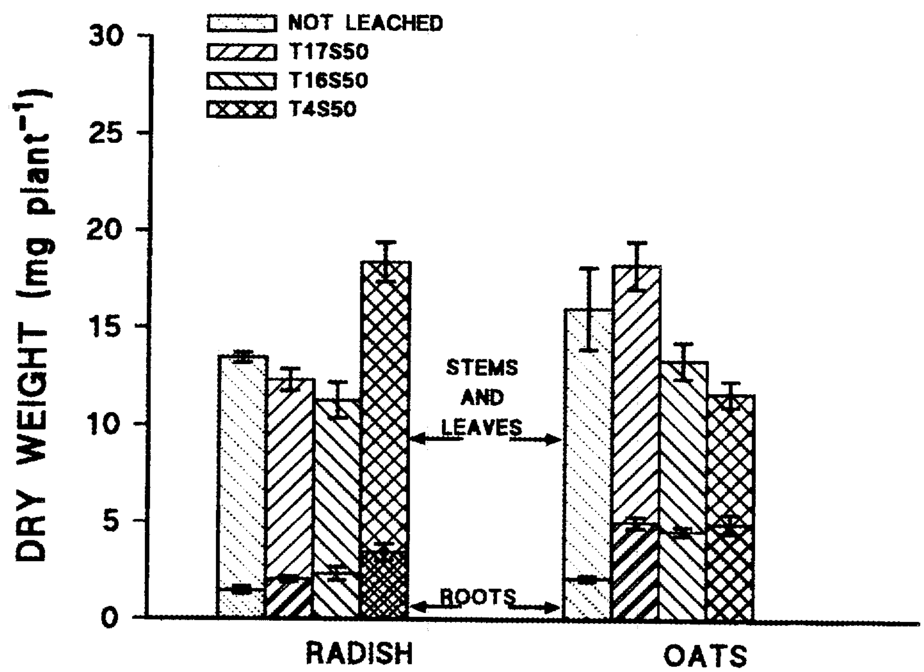

Fig. 10. Dry weights of 1-week-old radish and oat seedlings grown in nonleached soil and in storage pad soil having undergone three sodium carbonate leaching procedures at the pilot scale.

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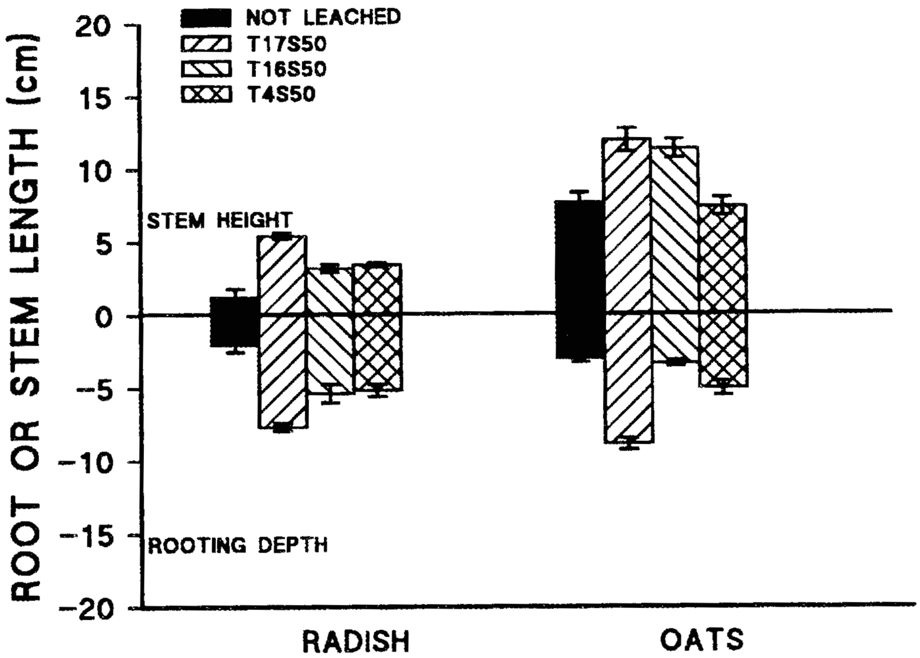

Fig. 11. Rooting depths and stem heights of 1-week-old radish and oat seedlings growing in nonleached soil and in storage pad soil having undergone three sodium carbonate leaching procedures at the pilot scale.

5. ACKNOWLEDGEMENTS

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