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# Aquifer Contamination by Nitrogen After Sewage Sludge Fertilization

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Municipal wastewater is an alternative source of water and fertilizer nutrients to *Citrus* trees and not detrimental effects have been found neither crops nor unsaturated zone at Castellón (Spain) area (Lapeña et al. 1995, Reboll et al. 2000). On the other hand, the sludge generated from reclaimed domestic wastewater process contains, generally, considerable amounts of N and other plant nutrients. It can be applied to soils for enhanced crop production because of its fertilizer value (Navas et al. 1999, Gori et al. 2000) and this application may be one solution to their removal (Espinoza et al. 1998). Moreover, several studies have shown the efficiency of biosolids application for maintaining and improving soil structure (Navas et al. 1996, 1999, Samaras et al. 2000) and as a source of plant nutrients (Dixon et al. 1995, Gavi et al. 1997, Espinoza et al. 1998).

There is a considerable interest in nitrogen use efficiency of *Citrus* plants to order to properly manage N input in result of maximum crop productivity or quality and minimum nitrogen pollution potential. However, the high levels of chemical fertilizer being used, generate environmental problems, such as ground water contamination, anoxia of rivers or fish kills attributable to leaching of N from agricultural soils.

The purpose of this study was to assess the potential for using sewage sludge as an alternative to chemical fertilizers and to demonstrate that these agricultural practices can minimize the chemical contamination of aquifers.

## MATERIALS AND METHODS

The study was conducted during 2000 and 2001 at the experimental lysimeter station located close to a treatment plant in Castellón (Spain). The treatment was composed of a primary treatment (screening and primary sedimentation), secondary treatment (activated sludge and secondary sedimentation) and biological sedimentation. The lysimeter station was divided in 24 tanks of 1 x 1 m and 1 m deep containing about 1 $m^3$ of soil reproducing the native characteristics on the site. At the bottom of each tank was placed a drainage water leaching system. The amount of drainage water was measured volumetrically.

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Two-year-old Naveline trees (*Citrus sinensis* L. (Osbeck)) were planted in 18 lysimeters, one tree per lysimeter. All soil surfaces were maintained weed tree with herbicide and trees were sprayed periodically for pests. Trees were irrigated by flooding with 60L/lysimeter every 21 days from March to October. During the rest of the year, irrigation was variable depending on the frequency and the intensity of rainwater.

In 2000, nine lysimeter were irrigated with wastewater (WW) and another nine with groundwater (GW). In 2001 twelve trees were fertilized as is shown in table 1 and irrigated with GW or WW (three tanks/treatment). Chemical fertilization dosage (CF) mimicked the normal treatments applied by farmers in the area. Dehydrated sewage sludge dosage (SF) contained the same N concentration as chemical fertilizer (Table 1). Another six trees were not fertilized (irrigated with GW or WW) and two more tanks were maintained without trees irrigated with groundwater and fertilized with chemical fertilizer or sewage sludge.

**Table 1.** Fertilizers (g per tree) applied during 2001 season.

| | **CF lysimeters** | **SF lysimeters** |
|---|---|---|
| **March** | | |
| Ammonium sulphate | 44 | - |
| Biammonium phosphate | 44 | - |
| Potassium sulphate | 30 | - |
| Sewage sludge | - | 750 |
| **May** | | |
| Ammonium sulphate | 97 | - |
| Sevage sludge | - | 250 |
| **Juny** | | |
| Ammonium sulphate | 100 | - |
| Potassium sulphate | 30 | - |
| Sewage sludge | - | 530 |
| **July** | | |
| Ammonium nitrate | 60.8 | - |
| Sewage sludge | - | 530 |

At every irrigation period, irrigation and drainage water were analyzed. N concentration from drainage water was multiplied by the total volume obtained to calculated quantities of nitrogen leached during the experiment. Evapotranspiration (ET) for each tank was calculated by subtracting the volume of the drainage water from the total water input (rainfall plus irrigation). This allowed us to ignore soil water storage (Boman 1994).

Spring flush leaves were sampled in October and oven dried for at least 2 days at 68 °C. Total nitrogen in leaves was determined by micro-Kjeldahl method (Bremner 1965). To estimate the growth and development of the trees, height and trunk diameter were measured.

Nitrate, cations, chloride, boron, phosphate and organic matter were determined from irrigation water and nitrate from drainage water. Cations were analyzed by atomic absorption spectrophotometry (Standard Methods 1989). Boron was analyzed following the azomethine-H method (Lopez et al. 1993). Chloride and nitrate were determined using a segmented flow colorimeter. UV-vis spectrophotometry was used to analyzed phosphate and organic matter was measured by the volumetric method (Norwtiz and Keliher 1985).

Soil and sewage sludge samples were dried at room temperature for 1 week, ground and sieved through a 2 mm nylon mesh before analysis. Correction for dry mass was obtained from a separate portion by drying at 105 °C for 24 h. Total nitrogen was analyzed by micro Kjeldhal method (Bremner 1965). Calcium, potassium, magnesium, phosphorus and heavy metals were determined by ICP-Inductively Coupled Plasma spectrophotometry. For mercury atomic absorption spectrophotometry was used and organic matter was measured by a volumetric method (Standard Methods 1989).

All data were tested using the Statgraphics software support. Means were expressed with their SE. They were compared by an LSD test. Differences were taken into account only when they were significant at the 5% level.

## RESULTS AND DISCUSSION

No limitations were found to use reclaimed wastewater from the treatment plant of Castellón to young *Citrus* trees irrigation as previously studies (Lapeña et al. 1995, Reboll et al. 2000). Water irrigation analyses are shown in table 2.

**Table 2.** Chemical analysis of groundwater (GW) and wastewater (WW).

| | 2000 | | | 2001 | | |
|---|---|---|---|---|---|---|
| **($mg\ L^{-1}$)** | **GW** | **WW** | | **GW** | **WW** | |
| **N** | 11.22±0.12 | 69.91±15.34 | * | 5.99±0.21 | 26.74±4.71 | * |
| **$Cl^-$** | 41.7±1.7 | 230.0±25.5 | * | 47.3±2.4 | 309.5±30.7 | * |
| **B** | 0.02±0.03 | 0.62±0.05 | * | 0.1±0.10 | 1.1±0.10 | * |
| **$Na^+$** | 34.5±2.6 | 264.7±38.1 | * | 33.2±5.1 | 214.3±15.9 | * |
| **$K^+$** | 2.6±0.5 | 18.4±0.6 | * | 3.1±2.4 | 18.0±1.0 | * |
| **$PO_4^{3-}$** | 3.8±0.6 | 12.2±1.6 | * | 4.2±0.8 | 11.5±0.9 | * |
| **Organic mater** | 9.4±1.5 | 60.2±6.5 | * | 10.5±1.3 | 59.6±5.9 | * |

n =10 (2000), n=7 (2001). * Significant at 5% level (LSD).

The sewage sludge characteristics are given in table 3. This sludge was slightly basic with very high organic matter, nitrogen content and moderate salinity. The concentration of trace metals showed values below the maximun allowable concentrations recommended by the European Union (86/278/EEC) and the Spanish Ministry of Agriculture (1310/1990) for agricultural land application of biosolids (Table 3).

**Table 3.** Characteristics of the sewage sludge of Castellon water treatment plant during 2001.

| | Average | Sd | Standard limits |
|---|---|---|---|
| pH ($H_2O$) | 7.60 | 0.50 | |
| Ashes (%) | 18.87 | 5.80 | |
| Moisture (%) | 81.13 | 3.50 | |
| Organic matter (%) | 55.00 | 6.52 | |
| Nitrogen (%) | 3.96 | 0.50 | |
| Phosphorus (%) | 2.72 | 0.12 | |
| Potassium (%) | 0.14 | 0.05 | |
| Magnesium (%) | 0.29 | 0.08 | |
| Calcium (%) | 4.82 | 0.94 | |
| Iron (mg/kg) | 37440.00 | 4327.00 | |
| Copper (mg/kg) | 143.00 | 20.00 | 1000-1750 |
| Zinc (mg/kg) | 1604.00 | 325.70 | 2500-4000 |
| Cadmium (mg/kg) | < 2 | - | 20-40 |
| Lead (mg/kg) | 380.00 | 90.50 | 750-1200 |
| Chromium (mg/kg) | 87.70 | 20.15 | 1000-1500 |
| Nickel (mg/kg) | 18.54 | 5.70 | 300-400 |
| Mercury (mg/kg) | < 0.3 | - | 16-25 |

In the first year of the experiment the irrigation water was the only source of N applied (Table 2). Leaf and soil N content were constant and no significant differences were observed between samples from trees irrigated with to different water qualities (GW and WW) (Table 4).

**Table 4.** Evapotranspiration (ET), soil and leaf N content of *Citrus* trees in lysimeters irrigated with groundwater (GW) or wastewater (WW) during 2000.

| Irrigation | ET (mm) | Soil N (%) | Leaf N (%) |
|---|---|---|---|
| GW | 643.6±15.3 | 0.14±0.01 | 2.35±0.16 |
| WW | 655.1±17.8 | 0.15±0.02 | 2.36±0.26 |

n=3. Means within columns are not significantly different at $p \leq 0.05$.

As it is shown in table 5 evapotranspiration values raised a high availability of water during 2001 that would be helping the absorption by roots. The percentage of N leached in relation to the N applied was higher in CF than in SF tanks and mainly in those irrigated with groundwater (Table 5). Differences found in lysimeters fertilized with sewage sludge may be related to the organic matter present in this biosolid and the slower mineralization and less leaching capacity (Harding 1985, Samaras 2000). This fact may be confirmed by the results obtained in the no-tree tanks where the relation between N leached and N applied was 70.4% to CF tank and 38.5% to SF tank. Moreover, the percentage of N leached in CF no tree-tank shown that the losses of N attributable to volatilization or denitrification not have exceeded from 40% of the N applied as has been reported previously by Stevenson (1986).

**Table 5.** Evapotranspiration, N applied and N leached in lysimeters with trees irrigated with groundwater (GW) or wastewater (WW) during 2001.

| Irrigation | Fertilizers | ET (mm) | N applied (g) | N leached (g) | N leached/ N applied (%) |
|---|---|---|---|---|---|
| **GW** | **CF** | 526.5±20.2b | 84.2 | 7.98±2.11a | 9.47 |
| **GW** | **SF** | 572.9±19.7a | 83.7 | 3.50±0.93b | 4.03 |
| **GW** | **NF** | 578.7±10.7a | 2.52 | 0.16±0.08c | 6.34 |
| **WW** | **CF** | 583.7±22.8a | 92.9 | 4.94±1.08ab | 5.31 |
| **WW** | **SF** | 586.8±19.3a | 92.4 | 3.68±0.75b | 3.98 |
| **WW** | **NF** | 575.9±11.8a | 11.23 | 0.70±0.10bc | 6.23 |

Evapotranspiration (ET). Chemical fertilization (CF). Sewage sludge fertilization (SF). Non fertilized (NF). Means within columns followed by different letters are significantly different at $p \leq 0.05$.

The N uptake efficiencies, in tanks with tree, can be calculated from the difference between the N leaching losses and that from the no-tree tank. The percentage of N availability to CF trees was between 60.9-65% and between 34-34.5% to SF trees when groundwater or wastewater was used respectively.

The soil N values found in fertilized lysimeters were in the optimal range in soils to *Citrus* crop (Legaz et al. 1995) (Fig 1b). The higher N applied to the lysimeters irrigated with wastewater had not produced an increase of soil N concentration in relation with the lysimeters irrigated with groundwater. Previous work carried out with young *Citrus* trees from field conditions, shown that the lythology of the soil (clays and sands) have an influence on the levels of soil N (below 0.2 %) in the first years of irrigation with wastewater (Reboll et al. 2000). This result, and the lower amounts of leached nitrogen found in sludge fertilized lysimeters, support the beneficial effects of organic matter on nitrogen contamination control as well in unsaturated zone as in the aquifer.

Levels of N in leaf tissue of fertilized trees were in the optimal range (2.2-2.8 %) to three years-old Naveline trees (Embleton 1973) (Fig. 1a). A deficiency was observed in the non fertilized trees after two years. In Citrus plants it was found, previously, that nitrate uptake rate follows a linear relationship with the external nitrate concentration (Cerezo et al. 2000).

Levels of N in leaf tissue were in relation with soil N (Fig. 1c). The higher soil N contents in CF tanks were reflected in the leaf N concentration, but the use of sewage sludge produced an optimal balance between leaf and soil N levels in both wastewater and groundwater treatments (Fig 1c).

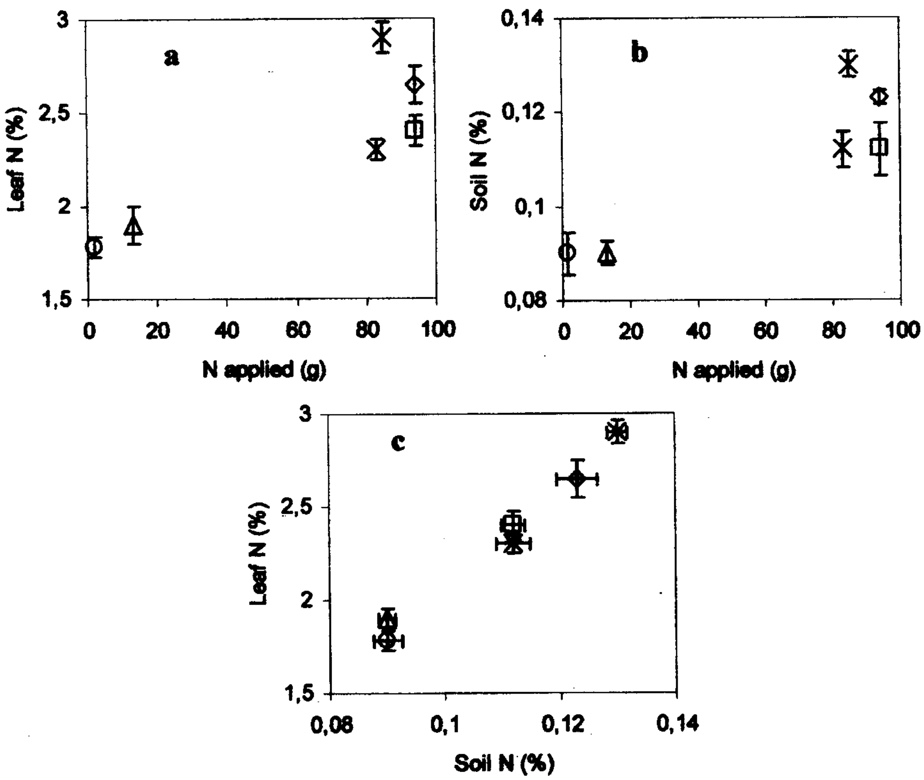


**Figure 1.** Relation between N applied, soil N and leaf N of *Citrus* trees in lysimeters irrigated with groundwater (GW) and fertilized: CF (x), SF (•), NF (o), or wastewater (WW) and fertilized: CF (◊), SF (□), NF (Δ), during 2001 (n=3).

Nutrients applied with the irrigation water on the non fertilized trees were not enough for an optimal range of development. There were no significant differences in plants fertilized with chemical compounds or sewage sludge in respect of the height or trunk diameter (Table 6). During the experiment a satisfactory increase in the height was observed.

The use of sewage sludge from the treatment plant of Castellón was suitable to fertilize 3 year-old citrus trees. N availability was adequate for root uptake and for satisfactory leaf N levels and growth. The use, under control, of sewage sludge to fertilize with wastewater or groundwater irrigation have not produced an increase of N leached or soil N contents. Thus, it can be a solution to their disposal avoiding soil and water pollution effects.

**Table 6.** Growth and development of trees irrigated with groundwater (GW) or wastewater (WW) and amended with chemical fertilization (CF), sewage sludge fertilization (SF) or non fertilized (NF), during 2001.

| Irrigation | Fertilizers | Height | | | Trunk diameter | | |
|---|---|---|---|---|---|---|---|
| | | Initial | Final | Δ | Initial | Final | Δ |
| GW | CF | 70.0±2.3 | 82.0±3.9 | 12.0a | 1.7±0.1 | 2.5±0.2 | 0.8a |
| GW | SF | 63.3±1.8 | 71.2±1.3 | 7.9a | 1.8±0.2 | 2.5±0.1 | 0.7a |
| GW | NF | 66.7±2.6 | 69.7±1.5 | 3.4b | 1.8±0.1 | 1.9±0.1 | 0.1b |
| WW | CF | 62.2±3.2 | 70.5±2.2 | 8.8a | 1.6±0.2 | 2.3±0.2 | 0.7a |
| WW | SF | 68.2±2.9 | 76.3±3.1 | 8.1a | 1.8±0.2 | 2.4±0.1 | 0.6a |
| WW | NF | 62.2±1.3 | 65.3±1.6 | 3.1b | 1.7±0.1 | 1.8±0.2 | 0.1b |

Means within columns followed by different letters are significantly different at $p \leq 0.05$ (n=3).

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