

# Evaluation of the use of pulp mill effluent in eucalyptus plantation

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## ABSTRACT

The pulp industry requires a large water volume in its industrial process, which produces as a consequence a large volume of effluents. The effluent characteristics vary according to the industrial process. The land application of pulp mill effluents via fertirrigation becomes an attractive option for the final destination of this wastewater. It acts as a supplementary process to the existing treatment systems, reducing the effluent load discharged into the receiving water body. It is also a supplementary source of water and nutrients for the plants. This work presents an approach to the use of treated effluent from the pulp industry in eucalyptus fertirrigation, and discusses the implications of this practice. The effluent from the secondary treatment of the bleached kraft pulp mill was characterized and the feasibility of using this effluent in eucalyptus fertirrigation was evaluated. It was investigated the effects of eucalyptus fertirrigation in soils of potential different plantation areas using controlled environment experiments. The main results obtained after a six month period fertirrigation at effluent application

rates corresponding to applications of 0 to 8 t ha<sup>-1</sup> of sodium have outlined handling practices during effluent application, considering in this work the effects on water availability, effects on soil characteristics, and the groundwater vulnerability to contamination. The present research work allowed to progress in the feasibility study of fertirrigation with treated wastewater from the bleached kraft pulp manufacturing process in handling planted eucalyptus forests, so as to guarantee its sustainability, considering its potential of use on the present national scenario: the trend towards shortage of hydric resources, the large volume of effluents generated in pulp manufacturing, extensive eucalyptus plantation areas near the effluent generation, the great hydric and nutritional demand of plantations and the constant need of improvement of the environmental performance of the companies.

## INTRODUCTION

The pulp and paper industry is considered to be one of the most successful activities of the Brazilian industrial forest based sector.

Most raw material for the pulp manufacturing process in Brazil comes from planted eucalyptus forests. These planned forests, depending on the climatic conditions and on water and

nutrient availability in the soil, may provide productivity rates higher than 50 m<sup>3</sup>.ha<sup>-1</sup>.year<sup>-1</sup> (Barros, 2000).

However, the high growth rates of the eucalyptus species impose a high demand on the soil resources, especially water and nutrients, demanding intense handling of the soil-water-plant system. The selection of handling techniques compatible with the capacity of the place to supply water and nutrients to the plants is one of the fundamental aspects to guarantee the forest sustainability, i.e., the maintenance or the increase of the forest production, all along rotations or successive crops. The forest sustainability will primarily depend on the maintenance of the water and nutrient flows at levels suitable to meet the respective demands by the trees, observing the soil conservation and the soil, water and air quality (Barros & Comerford, 2002). This sustainability will be jeopardized if the handling techniques will lead to excess use of recharge water of the aquifers, considering the increasing shortage of hydric resources and the deterioration of their quality, which is essential both for the human life and the industrial process.

The fertirrigation of planted forests is being applied on an experimental scale, aiming to add it to the silvicultural managing techniques, in order to increase the availability (and/



or the efficiency of use) of water resources and nutrients for plantation and consequently to increase productivity.

In the last years the legal requirements concerning water management became more restrictive, so that it was necessary to investigate alternatives for its reuse and improvement of the treatment processes and final disposal of the wastewater. This resulted in an increasing interest in applying municipal and industrial wastewater in the soil, thus becoming a way of water and nutrient addition to the forest ecosystem.

The pulp industry is among those considering and using the land disposal as an alternative in the management of its residues. The pulp industry requires a large water volume in its industrial process and may demand values above 100 m<sup>3</sup> per ton of produced pulp (Carter and Gleadow, 1994). As a consequence, it produces a large volume of effluents, the characteristics of which vary according to the manufacturing process used. Thus, the land disposal of these effluents becomes an attractive option for the final destination of this wastewater. Besides working as a process additional to the existing treatment systems, reducing the effluent load discharged into the receiving water body, it is also a supplementary source of water and nutrients for the plants.

The first effluent applications in silviculturally exploited areas took place in the 50's, in North America, most of them in experimental or pilot projects (Crawford, 1958; Westnhouse, 1960). In the 80's, the effluent application was restricted to a few mills and just to areas with no forests. Several characteristics, pointed out by Nutter and Red (1985) as unique of the forest system, contributed to this scenario: (a) the fact that forests require large application areas, due to the low

nutrient assimilation by forest species; (b) the long distances between mills and planted areas, making the application economically unfeasible; and (c) the little knowledge of the biological aspects associated with the practice. However, the fact that the pulp industry has been innovating its technologies in the last years, especially by adopting cleaner technologies, which is directly reflecting in the characteristics of the final effluent, should be taken into consideration. Also reported are studies aiming to apply the effluent from pulp manufacturing to silviculture as an attractive option to reach operations that would lead to an increasing circuit closure for the forest sector (Smith et al., 1997; Thacker, 1985).

The experience accumulated on the subject indicates that the process of land disposal of the effluent must be understood, planned and managed with the same degree of attention and importance given to any other operational process. Considering the complexity of the chemical constitution of pulp mill effluents there is still the need of further information and better understanding of the behavior of their main constituents in the soil and in the vegetative covering, as well as of the limitations regarding groundwater quality, in order that safe operational conditions of land disposal systems of these effluents may be established.

In this respect, this study discusses, based on investigation by means of experiments in controlled environments (greenhouses), the implications of adopting the practice of land disposal of the effluent from the secondary treatment of the bleached pulp manufacturing process (BKME), as well as addresses ways of managing the application of these effluents by fertirrigation, besides evaluating the effects on the soil-water environment, associated with this practice.

## Land disposal of wastewater

The term land disposal has been defined as planned and controlled application of a specific residue on the soil surface, in order to reach a specific treatment degree, by means of physical, chemical and biological processes naturally occurring in the water-soil-plant system (Metcalf & Eddy, 1991; Sanks & Asano, 1976; USEPA, 1981). In the last 20 years the interest in the methods of land disposal/residue treatment has been renewed, by understanding wastewater as a source of water and nutrients and including the use of it in forest plantations (Cromer et al., 1984; Myers et al., 1995; NCASI, 1985; Urie & Red, 1986; Crites & Reed, 1986; Nutter & Red, 1985; Rezende et al., 2001).

The aim of the land disposal of residues is their final destination in amounts that will not jeopardize the environmental components (soil, water, air, organisms etc.). In the process of land disposal, use is made of the soil-plant system for degradation, assimilation and immobilization of the constituents of wastewater, as well as of the products of its transformation in the environment.

The soil normally plays an important role in wastewater disposal, acting as deposit and treating means for the different chemical wastewater constituents. In order to identify the soil effect on wastewater and conversely, it becomes important to know the physical and chemical soil characteristics before and after its application.

Vegetation is another important component of the system, acting as an extractor of a large part of the macro and micronutrients available, avoiding their accumulation and the consequent salinization of the environment and contamination of surface and groundwater. It also permits to improve some soil characteristics such as its structure, providing an increase of water



infiltration and percolation. According to Feigin et al. (1991), the vegetal species, when being selected, must meet at least one of the following targets: water consumption, contaminant removal, salt tolerance, biomass production, quick growth and ability to fix atmospheric nitrogen.

The evaluation of the microbial and biochemical characteristics of wastewater i.e. of the effluent, allows to check whether they embody the conditions specified by the standards and criteria of public health, taking into consideration the type of culture, the soil, the way of application in the soil. The knowledge of the effluent chemical characteristics permits to determine the application rate and the means by which the negative impacts of it on the area destined for its disposal may be minimized.

The land disposal regulation principles are related to the maintenance and improvement of the soil-plant system quality at the minimum risk to the human health and to other environmental receptors.

## MATERIALS AND METHODS

The experiment with the effluent from the secondary treatment of the bleached kraft pulping process (BKME) in the soil was conducted in a greenhouse of the Soil Department of the Federal University of Viçosa, UFV, and had the assistance of the Laboratories of Water Quality and Chemical Analysis of Residues of the Department of Agricultural Engineering/UFV, Pulp and Paper Laboratory of the Department of Forest Engineering/UFV and Laboratory of Soil Physics and of Forest Soils of the Soil Department/UFV.

The experiment comprised 90 experimental units, consisting of columns containing samples of material of three types of soils and eucalyptus plants, submitted to five different fertirrigation rates with treated effluent from the kraft pulping

process, in daily dripping applications (0; 114.6; 286.5; 515.7 and 1146 mm year<sup>-1</sup>), corresponding to application loads of 0; 0.85; 2.12; 3.80 and 8.50 t ha<sup>-1</sup>year<sup>-1</sup> of Na<sup>+</sup>. Under a second condition, additional to the fertirrigation with effluent, application rates totaling an average yearly precipitation of 1300 mm were applied at 45 experimental units from the ninetieth day of culture.

The columns were filled up with clayey typic kandiodults, sandy typic kandiodults and typic quartzipsammments from representative eucalyptus plantation areas, previously air-dried, crumbled and duly homogenized and screened in 4 mm mesh screens, accommodating layer by layer, so as to repeat positions identical to those found on the field.

The wastewater used in the experiment consisted of the effluent from the secondary treatment system of the bleached kraft pulp manufacturing process, consisting of a system of facultative aerated lagoons in series, followed by a decantation lagoon.

The study also encompassed the use of a genetic material, coming from a clonal hybrid of *Eucalyptus grandis* x *E. Urophyla*. Along 180 days of culture of the eucalyptus plants, following was monitored: the ambient temperature, the evapotranspired water application rates, the application rates of simulated rain, the physical and chemical characteristics of the water drained from the columns, the visual symptoms of deficiency or toxicity in the plants, as well as the morphological characteristics (size, stem diameter, coloring) of the plants. After this period, the analyses of the physical and chemical characteristics of the soils of the experimental units have been carried out at samples collected in five depths in the columns, as well the chemical analysis of the foliar material of the eucalyptus plants. To determine most attributes, the

methodology described in EMBRAPA (1997) and APHA (1995) was adopted.

In the statistic analyses, the normality of the data and the homogeneity of variances between both experiments were verified and it was decided for the joint analysis of both experiments. The data were interpreted by variance and regression analysis.

## RESULTS AND DISCUSSION

### Evaluation of the quality of the treated effluent from the kraft pulping process (BKME) as water for agricultural utilization

The information found in the literature about quality of water for agricultural utilization refers to its use as irrigation water. In this technique the application rates are always much larger than in case of use as fertirrigation. However, the found information may be useful to predict the problems that may occur with the soil and plants.

Some relevant factors for irrigation water evaluation comprise: health risks, salinity, sodicity, the specific toxicity of ions, the concentration of nutrients (N, P) and the bicarbonates, the pH, the residual chlorine and the possibility of alteration of the soil permeability.

As to the irrigation water restriction concerning health risks coming from its microbiological quality, the criteria adopted by the U.S. Environmental Protection Agency, USEPA (1981), and those recommended by the World Health Organization (WHO, 1989) are followed. It is stressed that microbiological standards of quality of wastewater similar to the water potability standards are recommended by USEPA for the unrestricted irrigation or irrigation by spraying in any situation, while the criteria established by WHO for unrestricted irrigation are stricter as to the presence of helminth eggs, but flexible as to the



bacteriological quality, and omissive with regard to viruses and protozoans.

Directives to interpret the quality of water for irrigation purposes, formulated foreseeing the potential risk to the production of the culture associated with irrigation for a long period, are presented by Ayers and Westcot (1991). A classification of the irrigation water quality in terms of risk of salinity, normally measured either by the total dissolved solids (TDS) or the electrical conductivity, was worked out by McNeal (1981).

Considering the BKME characteristics presented in Table 1 and the usual criteria of quality of irrigation water (Ayers & Westcot, 1991; USEPA, 1981; UNEP, 1996; USEPA, 1992; WHO, 1989), it can be inferred as to the quality of this effluent, in terms of its fertilizing potential and restrictions of use:

- the treated effluent from the kraft process has intense color, reaching values higher than 1000 mg.L<sup>-1</sup> of Pt. However, this is no limiting factor for its use in fertirrigation, since, according to Juwarker & Subrehmany (1986), the color is mainly due to the presence of lignin derivatives, which does not result in toxic effects to the plants, when present in the soil; and the latter has been showing capacity to remove it. The high color intensity of the effluents has been a further factor of incentive to consider the application in the soil, thus avoiding to cause esthetic problems to the receiving water bodies.

- the high electric conductivity values found in the effluent (2.5 to 4.5 dS. m<sup>-1</sup>) indicate a high concentration of ions in solution. Considering the guide of USEPA (1981) for irrigation water salinity, the BKME fits as water that should be used for tolerant cultures in permeable soils, requiring careful handling. According to Ayers and Westcot (1991), BKME is classified as water with severe

restriction of the soil, and this in case it does not occur any removal of the salts accumulated in this environment.

- the pH may be the limiting factor to the effluent application in the soil, since high acidity conditions may cause toxicity problems, as making some metals available, while alkalinity may make unavailable a large part of the micronutrients essential for the plants. According to the pH values of BKME, between 6.9 and 8.2, the effluent is characterized as slightly basic, possibly due to the presence of alkalinity, fitting into the pH range which is considered to be optimal for use in agriculture, thus permitting, according to Gheyi et al. (1997), to predict the adverse effects related to the nutrient availability.

- the nitrogen concentrations present in BKME indicate that the sources of nitrogen readily available for the plants are small, so that there is a need of nutritional supplementation with another source of N in case of using the effluent for fertirrigation of agricultural cultures. The results indicate that if on the one hand the effluent does not represent the possibility of replacing nitrogenized fertilizers in the eucalyptus cultures, on the other hand they indicate little risk of groundwater contamination with nitrate. The nitrate lixiviation as a result of the application of the effluent from the pulp industry in the soil, due to the low nitrogen concentrations in the effluent, is less worrying, when compared to the risk of application of domestic sewage.

- the potassium, calcium, magnesium and sulfur concentrations, comparatively to inorganic nitrogen and phosphorus, can be considered to be relatively high, making feasible the BKME utilization to supply these nutrients as substituent of commercial fertilizers. The BKME would be able to supply the total demand of potassium and calcium for most cultures, but just partially the

phosphorus demand in the treatments with higher application rates. In practice, if manuring supplementation is made, it would be possible to totally dispense with the potassic fertilizer and to reduce the phosphorus dose. The sulfur and phosphorus fertilizer for lower rates would be still necessary. These data reveal that the BKME application may be enough to meet the demand of most cultures, and on the other hand the need of suitable handling, comparing the hydric and the nutrient balance.

- with regard to the micronutrients, the BKME presents, in general, low concentrations, and is able to supply the demands of Cu, Fe, B, Mn and Mo (these ones in excess for higher rates), a supplementation with Zn being required for most cultures. Nutrients in excess may jeopardize the culture productivity and quality, as well as result in environmental problems, mainly by lixiviation and groundwater contamination. The values of iron in the effluent are within the quality limits of irrigation water according to UNEP (1996), but indicate a moderate risk of dripper plugging, in case the effluent is applied in the soil by using this method. This is not observed for the concentration of manganese.

- the sodium concentration in the effluent may be found ranging from 450 to 1000 mg.L<sup>-1</sup>, with average values around 750 mg.L<sup>-1</sup>. These high sodium concentrations are undoubtedly the most limiting factor for using this water for agricultural fertirrigation. Concentrations higher than 70 mg.L<sup>-1</sup> present already a slight to moderate degree of restriction for use in irrigation by spraying, and may present toxicity problems.

- the high value of the Sodium Adsorption Ratio (SAR around 26), as a result of the high concentration of Na and of the relatively low concentration of Ca and Mg, involves risks such as of clay dispersion, of soil pore sealing, with a consequent



reduction of the rate of water infiltration into the soil and higher susceptibility to erosion, as a result of the intensification of the superficial flow off. These risks are not very high, since the electric conductivity is also high. Under the point of view of effects on the soil permeability, the BKME presents a slight to moderate degree of restriction for being used as irrigation water.

Irrigation water with SAR between 4 and 9 are considered to be of low risk with regard to clay dispersion, and consequently, to the soil permeability. According to the literature, some combined effluents from pulp mills have SAR in this range, and are thus potentially suitable for fertirrigation (Cromer et al., 1984). However, the low electric conductivities of some sectorial effluents of pulp mills, such as those coming from pulp cooking and washing, combined with the relatively high SAR, may cause physico-chemical problems to the soil, destroying its structure and consequently its macroporosity. For utilization of this wastewater, mixtures must be carried out, or the solution of calcium and magnesium must be provided in order to reduce the SAR value.

- the chloride ion, as counterion of the sodium ion in the salt formation, is also present in high concentrations in the effluent of the secondary treatment. The chloride concentrations in the pulp industry effluent range from 120 to 350 mg.L<sup>-1</sup>, which may jeopardize the use of the effluent for fertirrigation, since such concentrations may cause severe damages to the plant leaves in case of effluent application by spraying.

- the aluminium concentration in BKME presents values lower than 12 mg L<sup>-1</sup>. Aluminium is a toxic element for the plants, with recommended maximum limits of 5 and 20 mg.L<sup>-1</sup>, for continuous irrigation and for

periods of up to 20 years, respectively, according to UNEP (1996). Notwithstanding, the high eucalyptus tolerance to aluminium should be highlighted (Neves et al., 1982).

- the low heavy metal concentrations in the treated effluents present values below the permissible level for being discharged in receiving water bodies, as established in CONAMA Resolution 20/86 (CONAMA, 1986). Cadmium is one of the heavy metals presenting higher environmental risk when disposed of in the soil, in view of its higher mobility and its high toxicity, even in very low concentrations. The cadmium concentration values in BKME are low, lower than 0.020 mg.L<sup>-1</sup>.

- the concentration of chlorinated organic material in BKME, measured by the AOX (adsorbable organic halogen) concentration, was around 4.0 mg.L<sup>-1</sup> of chloride. The effluent applied to the soil, even after being submitted to the secondary treatment, may still contain a significant concentration of organic compounds of high molecular weight, of high toxicity for the biota. Due to their high molecular weight, these compounds remaining in the effluent after the secondary treatment are believed to be physically retained and degraded in the first layers of the soil, as discussed by Kookana and Rogers (1995).

- the concentration of suspended solids, of about 60 mg.L<sup>-1</sup>, indicates a moderate dripper plugging risk (Ayers and Westcot, 1991), in case this is the method chosen for effluent application. In nontreated effluents, the solids in suspension predominantly consist of fibers and for this reason may provide clogging of piping and hydraulic emitters (Adin & Sacks, 1991 and Ravacha et al., 1995). The suspended solids present in the final effluent are of biological nature, consisting essentially of biological flocks. Thus, as far as the suspended solids parameter is concerned, it is the

magnitude of concentration of these biological solids in the effluent that will or will not make feasible its utilization in fertirrigation with applications by dripping. In principle, the suspended solids also contribute to problems of soil surface colmation (Braile, 1979).

- the presence of fecal coliforms is verified in countings lower than 1000 FC/100 mL, the value of the bacteriological standard of WHO for unrestricted irrigation. In spite of occurring domestic sewage contribution to the final mill effluent generation, this is a little significant portion of the total volume. In terms of helminth eggs, the detention time in the stabilizing lagoons is fully sufficient for their total removal to take place. As to the quality criteria recommended by USEPA, the values of which are similar to those of potable water, it is expected that with BKME disinfection it will be possible to reach them in microbiological terms.

## METHOD AND APPLICATION RATE DEFINITION

The method and the wastewater application rate must be consistent with the soil-plant system characteristics.

According to Metcalf & Eddy (1991), there are three basic methods of wastewater application in the soil: quick infiltration (inundation or infiltration-percolation methods), superficial flow off and slow infiltration or irrigation, among other natural processes, such as wetlands, subsurface application and aquiculture.

Irrigation corresponds to the most used process, where the effluent is applied by spraying, in a localized way or by surface. In general, the project of application systems by irrigation is controlled by the hydraulic application rate i.e. by the volume of wastewater applied by unit of area for a certain period of time. In case of irrigation of agricultural cultures, the systems are projected according to the water



required for the plant production, the hydraulic application rate is the water amount to be used by the plant under unrestricted conditions, and they must be compatible with the agricultural practices, the soil and the water characteristics, the climatic conditions, the type of irrigated culture, the irrigation technique, as well as with the risks of soil salinization and groundwater contamination and the flow of wastewater to be disposed of in the soil (Ghassemi, 1995; Overcash & Pal, 1979).

The use of the term fertirrigation with wastewater is recommended in replacement for that of slow infiltration or irrigation, while it should be understood that using wastewater as water source to meet the hydric requirements of the culture may jeopardize the soil and groundwater quality, as well as the plant development (Matos, 2003; Matos et al., 2003). Thus, the reference for defining the application rates no longer consists of exclusively hydraulic criteria or calculations based on the evapotranspiration of the culture, and the application rate is calculated, instead of it, based on the constituent present (limiting parameter), proportionally, to the greatest extent, considering the capacity of the soil and the cultures to assimilate the residue applied. In this case, the nutrients, such as nitrogen, phosphorus, potassium, or even toxic elements, which become limiting factors, such as sodium, chlorine, BOD and heavy metals, may be considered. In some cases, the control factor is the organic load rate, as in case of very concentrated wastewater. In some industrial wastewaters, other factors should be taken into consideration, such as salt load or metal load. However, in general, for wastewater with BOD values equal to or lower than 1000 mg.L<sup>-1</sup>, the application rates can be defined based on the hydraulic application rate (Rowe & Abdel-Magid, 1995).

The fertirrigation can be carried out using application techniques identical to those used for conventional irrigation (spraying, either localized or by surface), but in case ions or substances that may cause problems if applied on the plant canopy are in solution, the localized application (microspraying or dripping) is recommended (Costa e Brito, 1994).

As far as localized irrigation is concerned, it is stressed however that the water quality should be taken into consideration under the point of view of possible emitter clogging, caused by factors of physical, chemical and/or biological nature (Leon & Cavallini, 1999). The system plugging may occur due to a factor or combination of factors, as follows: (a) presence of high concentrations of suspended matter (both organic and inorganic material); (b) precipitation of calcium and magnesium compounds, as well as dissolved fertilizers (phosphate, ammonia, iron, zinc, copper and manganese) and heavy metals in the water; and (c) presence and growth of algae, bacteria and other organisms in the water. No study was yet carried out about which of these factors and their combinations might cause the greatest plugging problem (Sandri et al., 2001; Leon & Cavallini, 1999; Metcalf & Eddy, 1999; Oron et al., 1980).

In case of effluents from the kraft pulping process, the element sodium becomes the limiting factor, indicating the use of the localized effluent application technique (dripping).

#### **Effects of BKME application on the physical and chemical soil characteristics**

The clayey typic kandiodults and sandy typic kandiodults (PA<sub>d1</sub> and PA<sub>d2</sub> soils) used in the experiment, have characteristically a more sandy superficial horizon (A) and the subsurface horizon (B) with yellowish coloring and medium to clayey texture, which conditions a significant

reduction of permeability in depth, have natural low fertility and low capacity of cation exchange and are poor in iron and in exchangeable bases.

The typic quartzipsamments (RQo) comprise sandy soils, with granulometric distribution fitting into the textural classes of sand and fine sand (% of sand - % of clay > 70%) and a slight increase in the clay percentage as the profile depth increases. These are excessively drained soils, with quick permeability all along the profile, porous, little susceptible to erosion, predominantly acid, with low base saturation. They present low organic material and nutrient contents, which decrease as depth increases.

In general it turns out that the water availability classes of the soils are predominantly low, which may be due to the low clay activity and mainly to the relatively high macro/micropore ratio present in the studied soils. Table 2 shows the main chemical characteristics of the studied soils.

The application of effluent to the soil may cause undesirable effects on its chemical characteristics (Fuller & Warrick, 1985). A great concern is related to salt accumulation, as a result of long time application. Adisesha et al. (1997) did not find any significant changes in the characteristics of the soil submitted to fertirrigation with pulp and paper industry effluent for a period of time shorter than 3 years. On the other hand, several studies (Cromer et al., 1984; Hansen et al., 1980; Aw, 1994; Hayman & Smith, 1979) addressing chemical effects of the application of pulp and paper industry effluent to the soil indicated high concentrations of Na<sup>+</sup>, Cl<sup>-</sup>, K<sub>2</sub>O, Ca<sup>2+</sup> and K<sup>+</sup>, besides high values of electric conductivity and sodium adsorption ratio (SAR), as well as a low concentration of Mg<sup>12</sup> and other essential nutrients (Aw et al., 1993; Aw, 1994). Irrigation with nondiluted pulp and paper industry



**Table 1: Characterization of the effluent from the secondary treatment of kraft bleached pulp mill**

Parameters	Unit	Average Value <sup>1</sup>
Color	mgL <sup>-1</sup>	1369
PH	-	7.58
Total solids (TS)	mgL <sup>-1</sup>	2446
Total dissolved solids (TDS)	mgL <sup>-1</sup>	1789
Solids in suspension (SS)	mgL <sup>-1</sup>	60.0
Sedimentable solids	mL.L <sup>-1</sup>	0.25
DQO <sub>3</sub> /	mgL <sup>-1</sup>	375
BOD <sub>4</sub> /	mgL <sup>-1</sup>	68
E.C.5/	dSm <sup>-1</sup>	3.4
Sodium (Na)	mgL <sup>-1</sup>	742.48
Calcium (Ca)	mgL <sup>-1</sup>	49.44
Magnesium (Mg)	mgL <sup>-1</sup>	7,6,86
SAR <sub>6</sub> /	-	26.2
Kjedhal nitrogen	mgL <sup>-1</sup>	6.8
Ammonium	mgL <sup>-1</sup>	1.69
Nitrate	mgL <sup>-1</sup>	0.00
Phosphorus (P)	mgL <sup>-1</sup>	1.074
Potassium (K)	mgL <sup>-1</sup>	27.357
Sulfur (S)	mgL <sup>-1</sup>	116.65
Copper (Cu)	mgL <sup>-1</sup>	0.010
Iron (Fe)	mgL <sup>-1</sup>	0.574
Zinc (Zn)	mgL <sup>-1</sup>	0.048
Manganese (Mn)	mgL <sup>-1</sup>	0.305
Aluminium (Al)	mgL <sup>-1</sup>	3.45
Boron (B)	mgL <sup>-1</sup>	0.170
Cadmium (Cd)	mgL <sup>-1</sup>	0.001
Cobalt (Co)	mgL <sup>-1</sup>	0.005
Chromium (Cr)	mgL <sup>-1</sup>	0.135
Mercury (Hg)	mgL <sup>-1</sup>	0.016
Molybdenum (Mo)	mgL <sup>-1</sup>	0.029
Nickel (Ni)	mgL <sup>-1</sup>	0.016
Lead (Pb)	mgL <sup>-1</sup>	0.027
Silicon (Si)	mgL <sup>-1</sup>	5.001
Vanadium (V)	mgL <sup>-1</sup>	0.009
Chloride (Cl-)	mgL <sup>-1</sup>	493
Sulphate (SO <sub>4</sub> -2)	mgL <sup>-1</sup>	426.12
Carbonate	mgL <sup>-1</sup>	0
Bicarbonate	molcL <sup>-1</sup>	8.3
AOX <sub>2</sub>	mgL <sup>-1</sup> of Cl <sup>-</sup>	4.59
Total Coliform	NMP/100mL	<5000
Fecal Coliform	NMP/100mL	<1000

1/Average values from monthly determinations, during experiment monitoring. 2/ Adsorbable organic halogens. 3/Chemical oxygen demand. 4/Biochemical oxygen demand. 5/Electric conductivity. 6/Sodium adsorption ratio given by  $SAR = Na^+ / (Ca^{+2} + Mg^{+2})^{-1/2}$ , where the contents of Na<sup>+</sup>, Ca<sup>+2</sup> and Mg<sup>+2</sup> are expressed in cmolc.L<sup>-1</sup>

effluent may also cause an increase in the pH values of the soil and in the contents of organic carbon, organic matter and available nutrients (Johnson & Ryder, 1988; Kannan & Oblisami, 1990a, 1990b; Juwarkar & Subrahmanyam, 1986).

In the present study, the electric conductivity in the saturation extract and the concentrations of available chloride and exchangeable sodium in the soil (Figures 1 and 2) showed similar behavior at all BKME rates applied and in all soils evaluated. High values of EC<sub>se</sub> and high concentrations of exchangeable Na and chloride have been obtained in the superficial layers of the column soils. The reason for the electric conductivity values to follow the variations of Na<sup>+</sup> and Cl<sup>-</sup> is the higher contribution of these ions to the EC of the soil saturation extract than of other ions also present, due to the fact that they are more concentrated in the BKME. In the soil columns that received the highest application rates of BKME, as well as of sodium, there were higher increases in the electric conductivity of the soil saturation extract (Figure 1). Considering the soils that did not receive any simulated rain, a more significant increase in the electric conductivity or salinity of the superficial layers (0-40cm) of the soils of medium/clayey (PA<sub>d1</sub>) and sandy/medium (PA<sub>d2</sub>) texture can be verified. A less expressive effect was observed in the sandy texture soil (RQ<sub>o</sub>).

As reported by Johnson and Ryder (1988), the increase in the sodium concentration in the soil surprisingly seems to be independent of the effluent application rates. This increase in Na content may cause the soil clay dispersion, leading to a poor infiltration and aeration of this environment. This also suggests that fertirrigation with pulp industry effluents may be a potential danger for the cultures.



**Table 2: Chemical characteristics<sup>1/</sup> of the samples of the soils used in the experimental columns**

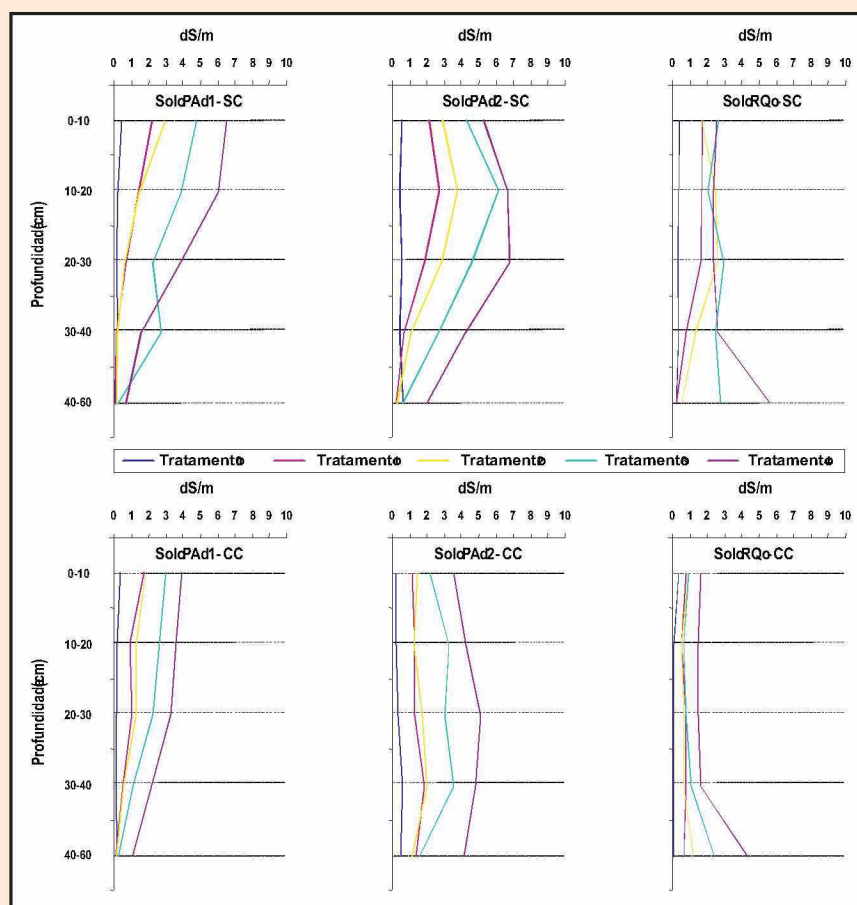
Soil <sup>2</sup>	Depth	pH H <sub>2</sub> O	P	K	Na	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Al <sup>3+</sup>	CTC (t)	CTC (T)	V
				mg.dm <sup>-3</sup>				cmolc.dm <sup>-3</sup>			%
PAd1	0-10cm	4,67	19,06	16	10	0,48	0,19	0,72	1,47	6,23	12,0
	10-60cm	4,79	3,20	10	11	0,71	0,20	0,72	1,71	5,68	17,4
	60-100cm	4,59	0,56	9	7	0,15	0,06	0,96	1,22	3,43	7,6
PAd2	0-10cm	4,79	4,86	9	10,0	0,20	0,11	0,36	0,73	3,01	12,3
	10-60cm	4,90	0,86	4	6,0	0,27	0,16	0,24	0,71	2,78	16,9
	60-100cm	4,88	0,47	8	6,0	0,41	0,11	0,36	0,93	3,01	18,9
RQo	0-30cm	5,51	4,75	6	4,0	0,65	0,23	0,00	0,92	2,7	34,1
	30-60cm	5,59	0,78	1	2,0	0,18	0,04	0,00	0,23	0,56	41,1
	60-100cm	5,50	1,12	1	2,0	0,17	0,05	0,00	0,23	0,82	28

1/ pH in water - ratio 1:2.5; P - K - Na - Extractor Mehlich 1; Ca - Mg - Al - Extractor KCl 1 mol.L<sup>-1</sup>; CTC(t) - Effective cation exchange capacity; CTC (T) - Cation exchange capacity at pH 7.0; V - base saturation index. 2/ PAd1-Dystrophic Yellow Clayey Typic Kandiodult A moderate flat relief, medium/clayey texture; PAd2- Dystrophic Yellow Sandy Typic Kandiodult A moderate flat relief, sandy/medium texture; RQo - Orthic Typic Quartzipsamment, sandy texture.

To avoid the risk of sodium predominance in the soil, the effluent application must be offset with other sources of Ca<sup>1/2</sup> and Mg<sup>1/2</sup>, which combined with high pluviometric indices may promote the lixiviation of Na<sup>1</sup> and the reestablishment of the cation balance. Thus, no reduction in the soil permeability should be observed (Johnson & Ryder, 1988).

The rain simulation in the experiment provided a reduction in the electric conductivity values in the saturation extract of samples taken on the whole soil profile. This demonstrates that in rainier periods, such as those occurring in the coastal region, the risk of soil salinization, as a result of BKME application for eucalyptus fertirrigation, becomes low. The exchangeable sodium lixiviation is suspected to be mainly responsible for these results and it can be expected that the use of methods making possible to remove sodium from the profile will also control the electric conductivity in the soil.

Evaluating the exchangeable sodium concentrations, it can be verified that there was a significant variation in the concentrations of this cation in the whole profile of the soil



**Figure 1: Electric conductivity evolution in five depths of the studied soils - PAd1, PAd2 and RQo -, evaluated 180 days after planting eucalyptus scions and applying BKME, corresponding Treatm. 0, Treatm. 1, Treatm. 2, Treatm. 3 and Treatm. 4 to the application rates of 0mm, 114.6mm, 286.5mm, 515.7mm and 1146mm, considering two conditions for conducting the experiment: without rain simulation and with rain simulation (average values from three repetitions)**

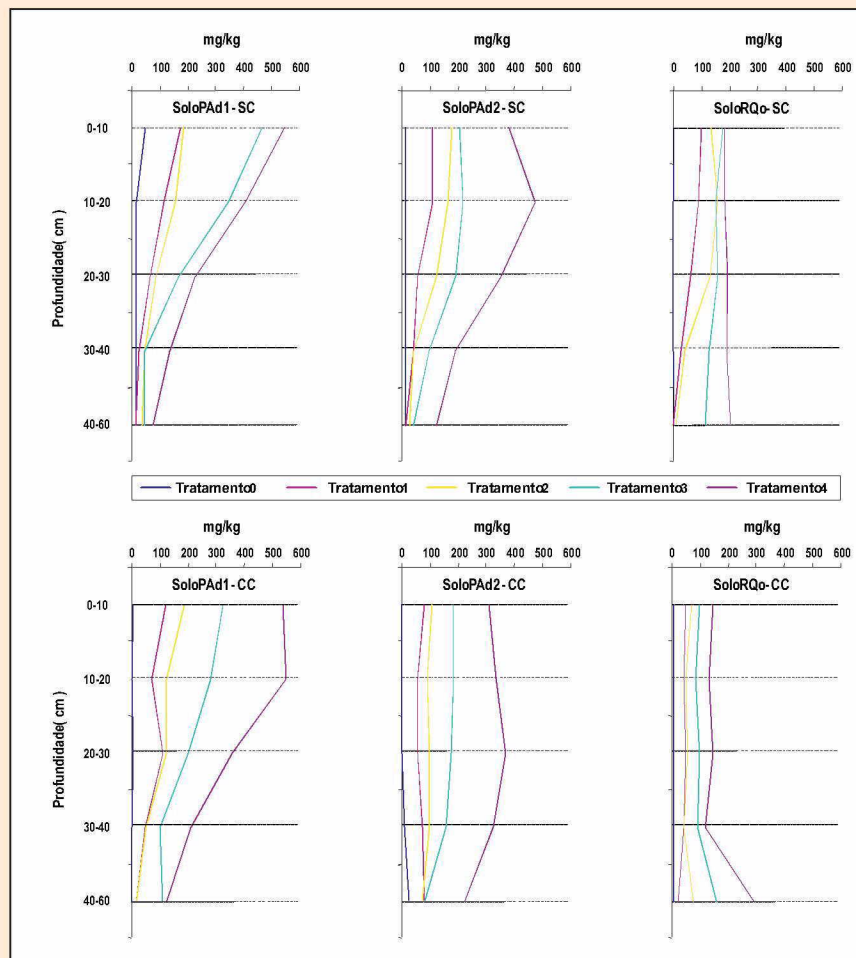


in the columns where the effluent was applied (Figure 2). The regression equations adjusted as a function of BKME application rates for different depths show different behaviors of the studied soils as far as sodium is concerned. The PAD1 soil showed a tendency to sodium variation adjusted to the quadratic function, while the PAD2 and RQo soils presented a linear increase in the sodium concentrations with BKME application rates along each depth. The results of exchangeable Na concentration, obtained from the collected samples, confirm that this is the most limiting factor for land disposal of secondary treatment effluent, since there is a clear concentration of this cation in the profile. However, considering how easy it is also to remove the Na from the profile, it is believed that the disposal of BKME in sandy soils, or with artificial drainage systems implanted, may be sufficient to provide better control of the soil sodification.

In the typic quartzipsamment, RQo, the sodium concentrations have been kept at lower levels than those found in the clayey typic kandiuults and sandy typic kandiuults PAD1 and PAD2 for all BKME application rates, with no variation along the soil profile for the treatments with higher application rates (515.7 and 1146 mm). In the rain simulation experiment a tendency to lixiviation of this cation was verified, since there was an increase in its concentration in the deeper layers (Figure 2).

The easy drainage of RQo soil made it possible to avoid severe effects of salinization and permeability reduction, as a result of the effluent application, as reported in other studies about sandy texture soils (Cromer et al., 1984; Johnson & Ryder, 1988; Juwarkar & Subrahmanyam, 1986).

Although BKME shows high sodium concentrations (480 to 990 mg.L<sup>-1</sup>) and relatively low ones of calcium (28 to 110 mg.L<sup>-1</sup>) and



**Figure 2: Sodium content evolution in five depths of the studied soils - PAD1, PAD2 and RQo -, evaluated 180 days after planting eucalyptus scions and applying BKME, corresponding Treatm. 0, Treatm. 1, Treatm. 2, Treatm. 3 and Treatm. 4 to the application rates of 0mm, 114.6mm, 286.5mm, 515.7mm and 1146mm, considering two conditions for conducting the experiment: without rain simulation and with rain simulation (average values from three repetitions).**

magnesium (4 to 11 mg.L<sup>-1</sup>), which imparts a high sodium adsorption ratio (SAR) to it, the high electric conductivities minimized in a certain way the dispersive effects of clay, provided by applying this effluent. The BKME application did not cause any significant alteration in the percentage of dispersed clay of PAD2 and RQo soils of the columns at the different BKME application rates, nor as a result of the fact that the soils received or did not receive simulated rains. The sandy texture soil, RQo, due to its low clay content, was the soil that showed the lowest dispersed clay values at any sodium application rate. This confirms

that observed by Johnson & Ryder (1988) and Cromer et al. (1984), i.e., coarse texture soils did not present any severe deterioration effects and any permeability loss as a result of applying effluents having a high percentage of exchangeable sodium. Variations in the dispersed clay percentage were observed in the medium/clayey texture soil, PAD1, in the subsurface layers (20-30 cm).

As to the alteration in the concentration of heavy metals in the soil profile, it turned out that Cd, Cr, Pb and Ni did not come to reach the last layer in the soil columns that received the highest application rate,



which made impossible their presence in solution in the percolated water. The alteration in the concentrations of Cu, Cr, Pb and Ni in the soils submitted to the different application rates was insignificant and low concentrations, comprised in the range of nondetectable to 2 g.dm<sup>-3</sup>, have been obtained. According to the data obtained and the maximum application loads of heavy metals suggested by the European Economic Community, cited by Loehr (1984), Simon and Tedesco (1993) and adopted in the State of Paraná (SANEPAR, 1997) for application to the soil, the highest BKME application rate did not provide the incorporation of amounts of Cd, Cr, Ni and Pb higher than the maximum values proposed. The maximum limits of metal concentration in the soil, adopted by the State of Paraná by an adaptation of the Spanish legislation, expressed in mg.kg<sup>-1</sup>, are: 1.0; 50; 100 and 30, for Cd, Pb, Cr and Ni, in soils of pH lower than 7.

With regard to mobility, the analyzed metals showed little movable, even with simulated rain application, reflecting their great affinity with the solid phase, even in sandy soils.

### **Salt lixiviation and possible groundwater contamination risks**

The environmental risks in disposing of bleached pulp effluents in water courses are being well documented (Dell et al., 1996). A few articles refer to the risk evaluation at effluent application to the soil. Most articles address the land disposal of the sludge coming from the effluent treatment (Vasconcelos e Cabral, 1993; Keenan et al., 1990).

Since certain chemical substances, oils, solvents or other dangerous and toxic substances have been eliminated from the industrial process, the secondary effluent from the pulp industry can be considered to be of good quality, at least equal to that supplied by the secondary treatment of

domestic sewage. However, when it is disposed of in the soil, the salt mobility in that environment may be a potential pollution problem for the groundwater.

The simulation of the pluviometric precipitation was employed in this experiment in order to evaluate the capacity of natural rains to lixivate the salts retained in the soil as a result of BKME application. Analyzing the quality of the samples of the percolated liquid, collected in the drains of the soil columns, the risk that the practice of fertirrigation with BKME may involve in terms of groundwater contamination was evaluated.

The clayey typic kandiuults of medium/clayey (PA1) and sandy/medium (PA2) texture showed higher water retention capacity and did not present any percolate resulting both from BKME application and from simulated rain application. The sandy texture soil (RQo) showed vulnerability to groundwater contamination, in spite of the fact that the physical and chemical characteristics of percolated water indicated that there was an attenuation of some parameters of the applied effluent, such as BOD reduction by 50%, and lower micronutrient and nitrate concentrations. An expressive gain in the percolate color, as well as an increase in the concentrations of elements as potassium, sodium and chloride at the highest application rate, due to the high concentrations of these ions in the effluent and also due to the sandy nature of the soil itself, were observed for this soil, resulting in a high lixiviation rate, corresponding to the BKME rate of 1146 mm i.e. 8 t.ha.<sup>-1</sup> of sodium.

In studies effected by Johnson & Ryder (1988) and Cromer et al. (1984), increases in the concentration of some contaminants in groundwater have been also observed. Initially, higher concentrations were found in the upper layers of the aquifer, showing a trend toward reduction as depth increased. Applications for extended periods (periods corresponding to 3 years) in

areas of effluent disposal provided a great increase in sodium and chloride concentrations in all depths, indicating a uniform distribution of the solution.

In the conducted experiments it became evident that as the application rate increased there was a proportional increase in the amount of movable ions in the lixivate. On the other hand, in case the ionic retention capacity is exceeded, even the nutrients considered to be of low mobility may reach the groundwater. Sandy soils provide higher infiltration rates and lower plugging capacity to the incorporated ions, and are for these reasons those involving more risks to the groundwater quality.

The groundwater contamination potential depends, among other factors, on the groundwater depth and on the physico-hydric characteristics of the porous environment. The pollutant, when being incorporated to the groundwater of aquifers of granular nature, disperses and spreads in a generalized way along with the water and may extend for long distances, encompassing large areas. In case the incorporated quantities are low, the dilution of the pollutants in the groundwater may result in permissible concentrations, of low toxicity for human beings and animals. Depending on the physical and chemical nature of the pollutant, some may be adsorbed by the solid material of the aquifer, becoming adsorbed to the clays (Matos, 2001).

Although the monitored soil depth (55 cm) is relatively small, the results found can be used to analyze the groundwater vulnerability, in case different rates of BKME are applied to the sandy soil (RQo). The great lixiviation potential in this soil, maximized by the low interaction of the sand fraction with ions present in the water of the soil, indicates that the fertirrigation parceling is recommendable in case an application of higher BKME rates is required.

Comparing the concentration values obtained in the percolate to those established by CONAMA Resolution



No. 20, dated 06/18/86 (CONAMA, 1986), for water body Class 1, to the potability standards established by GOVERNMENTAL DECREE No. 518, of 2004, from the Ministry of Health (FUNASA, 2004), and to typical values found in groundwater (Mathess & Harzey, 1982; Szikszay, 1993), it turns out that they are similar or overcome the referential values (Table 3).

In general, it can be observed that there was a gradual increase in the electric conductivity values and in the sodium concentrations in the percolate along the period of time of BKME application, having the higher application rates provided more significant increases in the values of these parameters, when compared to those obtained in the percolate of the witness columns.

The high sodium concentrations in the percolate indicate that the sandy soil has enough permeability to permit a quick sodium lixiviation in the profile. A tendency of the sodium concentrations in the percolate of the columns that received the BKME application rate of 1146 mm towards stabilization at a value below the affluent concentration can be also verified. The dilution of the soil solution with the application of simulated rains provided a continuous sodium removal by lixiviation, indicating that the local pluviosity may be sufficient to promote a suitable lixiviation of the sodium of the soil profile.

In fertirrigation, the nitrogen is the element which is most subject to lixiviation in the soil. However, while the nitrogen is in its form of cation ammonium ( $\text{NH}_4^+$ ), the possibility of its loss by lixiviation is low. Under normal cultivated soil conditions, the ammonium is nitrate oxidized ( $\text{NO}_3^-$ ), negative load ion, which moves more freely with the soil water. Lixiviation may occur if nitrate is present in large quantities in the soil prior to plantation, which is not the case, or when the culture is not using this nutrient, or else when the irrigation or the rain exceeds the soil retention capacity and the

moisture required by the culture.

Potassium showed in the soil a very similar behavior to that of sodium i.e. it accumulated to a certain extent, but became susceptible to strong removal, showing a tendency to have its concentration in the column percolate stabilized with the time of application of BKME. The high potassium concentrations in the column percolate indicate that the removal of this nutrient from the sandy texture soil profile is relatively high, as it is the case of the evaluated RQo, especially in the rainy period.

The phosphorus mobility in the soil is small because it is strongly adsorbed to the argillo-humic complex of the soil and does not involve a serious danger in terms of groundwater contamination. However, in sandy soils, considering its high macroporosity and low retention capacity of this nutrient, this panorama may change and there may be risks of groundwater contamination. The average value of concentration of P found in BKME was  $1.75 \text{ mg.L}^{-1}$ , while in the percolate it varied from  $0.111$  to  $5.17 \text{ mg.L}^{-1}$  for the lowest (114.6 mm) and the highest (1146 mm) application rate, respectively, suggesting that there was a movement of P along the profile. These values are within the typical range for groundwater.

#### **Suitable application handling**

In many disposal techniques, resting periods from wastewater application, as seasonal applications, are usual, allowing a time for organic matter degradation by the microorganisms, as well as for water drainage so as to reestablish aerobic conditions. These resting periods depend on the method of disposal, on the soil, on the culture and on handling considerations. They usually result in relatively great variations in the application rates and consequently require larger storage and wastewater application areas.

The results of this study indicated that occasional discharges of the salts

accumulated in the radicular zone would be required to minimize their effects on water availability and toxicity for the plants. The rates of simulated rain applied turned out to be sufficient to allow lixiviation of sodium and chloride of the profile of the studied soils. Thus, the intermittent effluent application, intercalated by periods with application of nonsaline water (rain or irrigation) should make possible to remove a large amount of salts, especially in the more sandy soils.

The incorporation of organic correctives, including silt from pulp industries and bovine dung, into the soil of the effluent application areas, is also a handling option. According to Vasconcelos e Cabral (1993) and U.S.EPA (1981), this practice made possible to maintain productivity during fertirrigation with effluent due to the supply of unavailable nutrients to the cultures and the contribution to the soil structure improvement, increasing infiltration rates and the sodium lixiviation potential.

#### **CONCLUSIONS AND CONSIDERATIONS**

The present work permitted to progress in the feasibility study of using fertirrigation with treated wastewater from the bleached pulp kraft manufacturing process in the management of planted eucalyptus forests, so as to guarantee its sustainability, considering its potential of utilization on the present national scenario: tendency to the shortage of hydric resources, large volume of effluents generated in pulp manufacturing, extensive eucalyptus plantation areas near the effluent generation sources, high hydric and nutritional demand of the plantations and constant need of improvement of the environmental performance of the companies.

The information obtained in this study provide resources for a better understanding of BKME behavior, when applied to eucalyptus plantations in



**Table 3 - Characteristics of the percolate of the columns of RQo soil submitted to different application rates of the secondary effluent from the bleached pulp manufacturing process (BKME), with rain simulation<sup>1</sup>.**

Parameters	Period of application (days) <sup>2/</sup> Treatments <sup>3/</sup>															
	114,6 mm				286 mm				515,7 mm				1146 mm			
	90	120	150	180	90	120	150	180	90	120	150	180	90	120	150	180
pH	5,1	6,38	5,05	6,07	6,08	4,45	5,58	6,02	5,54	4,06	5,84	5,84	4,97	4,24	6,66	6,19
Color mg.L	-	-	-	-	-	123	136	140	90	192	240	259	530	1295	2556	3120
CE dS. m <sup>-1</sup>	0,45	0,45	1,07	1,07	0,43	0,81	1,57	1,26	0,87	0,87	2,50	2,38	1,44	2,19	1,73	1,90
Na <sup>+</sup> mg.L <sup>-1</sup>	16	27	120	2714/	26	70	220	5044/	72	193	522	14104	272	357	397	365 4/
Ca <sup>+2</sup> mg.L <sup>-1</sup>	10,23	-	-	35,09	49,67	-	-	47,15	55,74	-	-	42,77	35,93	-	-	4,5
Mg <sup>+2</sup> mg.L <sup>-1</sup>	7,03	-	-	16,31	28,44	-	-	17,71	22,31	-	-	23,16	12,38	-	-	0,392
Cl <sup>-</sup> mg.L <sup>-1</sup>	56	136	227	187	164	-	186	448	216	-	495	1095	685	667	894	1293
N mg.L <sup>-1</sup>	-	-	31,08	15,84	35,84	23,94	21,95	-	47,32	20,19	11,27	10,29	50,82	27,02	15,50	11,06
P mg.L <sup>-1</sup>	0,165	-	0,184	0,25	0,111	-	0,175	0,526	-	0,118	0,195	0,200	-	4,27	6,6	5,17
K mg.L <sup>-1</sup>	15,69	-	25	85	19	-	46	-	-	50	165	190	-	150	207	200
Cu µg.L <sup>-1</sup>	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Fe µg.L <sup>-1</sup>	1,70	-	-	12,08	1,109	6,18	-	12,30	5,32	-	-	5,197	1,15	2,62	-	2,298
Zn µg.L <sup>-1</sup>	0,027	-	-	0,033	0,086	-	-	0,050	0,016	-	-	0,083	0,039	0,070	-	0,026
Mn µg.L <sup>-1</sup>	6,39	-	-	4,49	2,908	-	-	6,83	2,746	-	-	5,547	2,486	-	-	0,331
CO mg.L <sup>-1</sup>	80,06	48,5	62,3	32,85	131,9	33,03	55,7	71,5	101,3	82,7	130,9	152,5	155	-	-	-
DBO mg.L <sup>-1</sup>	30,9	31,14	37,24	24,57	36,66	-	-	32,14	32,77	28,9	-	24,21	30,85	34,67	31,11	31,37
AOX mgCl.L <sup>-1</sup>	-	14,55	4,23	4,23	-	-	-	-	-	-	-	-	-	3,4	4,0	6,0

1/ average values monitored along the experiment. 2/ days counted from the beginning of BKME application. 3/ Applied rates, corresponding to 0, 0.85; 2.1; 3.8 and 8.45 t.ha<sup>-1</sup> of Na.

clayey and sandy typic kandudults and typic quartzipsamments, contributing to establish safe handling conditions in accordance with the assimilation capacity of the soil-plant system.

The study showed that characteristics of each soil should be considered in defining application rates and that the intermittent effluent application, intercalated by periods with application of nonsaline water (rain or irrigation) should make possible to remove a large amount of salts, especially in the more sandy soils.

In general, the relatively good behavior of the soil-plant system, when submitted to different effluent application rates, confirms the possibility of the agricultural utilization of BKME.

Future studies should verify the influence of fertirrigation application with BKME in the long term on the eucalyptus growth indices, on the woody tissue quality and on the pulp yield.

Another study to be considered

would concern the variation in the microbial population of the soil exposed to the treated pulp and paper effluent, as in its enzymatic activities, considering that the microorganisms of the soil have a vital participation in the degradation of organic material incorporated, constituting an important source of enzymes for the environment, and that these organisms are the first ones to come across the environmental changes caused by the disposal of effluents.

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