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EMPIRICAL MODELS TO PREDICT SOIL NITROGEN MINERALIZATION

MODELOS EMPÍRICOS PARA A PREDIÇÃO DA MINERALIZAÇÃO DO NITROGÊNIO DO SOLO

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SUMMARY

Empirical models are mathematical equations that can be fitted to experimental results. The use of these models aims to evaluate or predict observed phenomena or experimental data with the objective of helping the development of adequate soil management practices. Based on these considerations, eight mathematical models described in the literature are compared in the present work, using as experimental data the mineral N accumulated during 32 weeks of incubation in Southern Brazilian soils. To obtain mineralization values experimentally, an incubation-washing procedure with 0.01mol L^{-1} $CaCl_2$ was used. Mineral N was determined at the beginning of the incubation and in the 2^{nd}, 4^{th}, 8^{th}, 16^{th} and 32^{nd} weeks. Among the models, the best fit was obtained with the simple exponential model to describe the mineralization of organic N in the soils. The double exponential models showed quite good fit, but may be superparametrized. In addition, the hypothesis on which these models are based, i.e., the presence of two forms of organic N susceptible to mineralization, cannot be sustained in this study.

Key words: *potentially mineralizable N, mathematical models, simple and double exponential, N pools.*

RESUMO

Modelos empíricos são equações matemáticas que podem ser ajustadas a resultados experimentais. Esses modelos podem ser utilizados para avaliar ou predizer fenômenos observados ou dados experimentais e auxiliar no desenvolvimento de práticas adequadas de manejo do solo. Desse modo, o presente trabalho teve por objetivo comparar oito modelos matemáticos descritos na literatura, utilizando como dados experimentais o N mineralizado de dez solos do Rio Grande do Sul, acumulado durante 32 semanas de incubação. O N mineralizado foi obtido experimentalmente em um experimento de incubação, seguido de lixiviação com $CaCl_2$ 0,01mol L^{-1}. O N mineral foi determinado no começo do período de incubação e ao final da 2ª, 4ª, 8ª, 16ª e 32ª semanas. Entre os modelos testados, o melhor ajuste do N mineralizado foi obtido com os modelos exponenciais simples, ao passo que a obtenção desses ajustes nos modelos exponenciais duplos esteve condicionado ao aumento de parâmetros na equação. Em função dos resultados observados e das condições experimentais, conclui-se que a hipótese em que os modelos exponenciais duplos estão baseados, isto é, na presença de dois compartimentos de nitrogênio suscetíveis à mineralização, foi rejeitada.

Palavras-chave: *N potencialmente mineralizável, modelos matemáticos simples e duplo exponencial, compartimentos de N.*

INTRODUCTION

Models represent attempts of mathematical description of a natural event. In the case of N-mineralization, the main purpose of modeling is to obtain quantitative data to recommend nitrogen addition (TANJI, 1982). The degree of complexity of a model is determined by the understanding of the system to be modeled, by the availability of data and by the way the model will be applied. The main limitation of the models that include the mineralization of organic N is that

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the organic N pool is very large when compared to the inorganic N pool (CAMARGO ***et al.,*** 1997a), causing large errors when estimating the quantity of organic N available to plants (TANJI ***et al.,*** 1979).

Initially, the mathematical fittings obtained for N-mineralization as a function of time were described by simple exponential models, considering the existence of only one form of potentially mineralizable N (N_o), decomposing at a rate proportional to its concentration (STANFORD & SMITH, 1972). When this concept was introduced, several mathematical attempts to estimate N_o were proposed, using the accumulated values of mineralized N. The majority of the new models separate the organic N pool, trying to reduce the errors in the estimates of N_o. These models consider the existence of two or more fractions occurring in the process, a more stable and a less stable form, also referred to as readily and less readily mineralizable N (MOLINA ***et al.,*** 1980; INOBUSHI ***et al.,*** 1985).

Comparative studies of simple and double exponential models and other functions show that the best fit to N-mineralization in incubation studies is obtained with the double exponential function, especially for short incubation times (MOLINA ***et al.,*** 1980; DEANS ***et al.,*** 1986; CABRERA & KISSEL, 1988). However, only a simple fit for comparison is generally shown, not considering the biological response of the observed data (CAMARGO ***et al.,*** 1997b). The present work aims to compare mathematical models described in the literature, using experimental organic-N mineralization data from soils of the Rio Grande do Sul State, Brazil.

MATERIAL AND METHODS

Theoretical background: STANFORD & SMITH (1972) proposed a biological method to study N-mineralization from organic matter incubated at constant temperature and moisture. The mineralization potential for soil nitrogen (N_o) was defined as the fraction of the N pool considered susceptible to mineralization, as long as the process follows a first order kinetic. They suggested that the mineralization potential, N_o, and the mineralization constant, k, could be used to predict the soil nitrogen availability to plants in a time period, considering that mineralized N is a linear function of the square root of time. This exponential model is based on the assumption that there is only one form of potentially mineralizable nitrogen, decomposed at a rate proportional to its concentration (Table 1). This form is assumed to be of a discrete size (N_o) and its magnitude approaches asymptotically that of mineralized N (N_m) accumulated through time.

Ever since STANFORD & SMITH (1972) introduced the concept of potentially mineralizable N (N_o), different mathematical approaches have been used to estimate its value using mineralized N data. From the simple exponential model (Table 1) a physical meaning for k (specific decomposition rate) is obtained, and k is not linear with time (DEANS ***et al. ,*** 1986). When it is difficult to obtain a non-linear regression, N_o can be obtained by the hyperbolic equation described by JUMA ***et al.*** (1984). According to MOLINA ***et al.*** (1980), neither the forms obtained by a non-linear nor those by linear methods give satisfying results for N_o or k.

A quite popular model is the double exponential described by MOLINA ***et al.*** (1980) and INOBUSHI ***et al.*** (1985). Both authors consider the existence of two fractions within the process, one being a less stable form, and the other one more resistant. These models are mathematically and conceptually identical, but with some differences in interpretation of their parameters. MOLINA ***et al.*** (1980) described this behavior by the equation in table 1, where S and (1-S) represent, respectively, the less stable and the resistant fractions of organic-N, decomposing at specific rates h and k. The model described by INOBUSHI ***et al.*** (1985) was used to fit N-mineralization data for submerged soils. These authors proposed a double exponential equation (Table 1), where N_{oq} and N_{os} are described by first order kinetic reactions, with readily and less-readily mineralizable N, decomposed at specific rates of kq and ks, respectively. According to DEANS ***et al.*** (1986) the simple exponential equations exhibit a systematic subestimation of N_o and a superestimation of the mineralization rate k. These authors observed that the double exponential models gave better estimates of k with lower mean squares, which supports the hypothesis that at least two organic N pools contribute to mineralized N.

Notwithstanding the relevance of these studies, TALPAZ ***et al.*** (1981) proposed a non-linear fitting, considering only one mineralizable N-pool, whereas SIERRA (1990) considered the simple and double exponential models only as approaches to the mineralization that happens within the soil organic matter. This author showed that the mineralization rate diminishes continuously, and that it is impossible to recognize mineralized N pools as proposed for exponential models, due to the non significance of the pool size (mineralized N). From a practical point of view, the number of pools to be included in the model depends on the fit and the desired precision.

Table 1 - Empirical models for estimating organic N mineralization.

Model	Equation	Description	Reference
a) Simple Exponential	$N_m=N_o(1-\exp^{-kt})$	Considers that N-mineralization is described by only one pool, N_o, which defines the potentially mineralizable N as a function of t and a rate, k.	STANFORD & SMITH (1972)
b) Simple Exponential	$N_m=N_o(1-\exp^{-kt^b})$	Similar to that described by STANFORD & SMITH (1972), introducing a time exponent (b).	MARION ***et al.*** (1981)
c) Simple Exponential	$N_m=N_1+N_2-(N_2 \exp^{-k_2 t})$	This model considers the presence of an initial flush of mineralization (N_1) and of the active fraction (N_2) as a function of t and a rate k_2.	JONES (1984)
d) Simple Exponential	$N_m=N_1(1-\exp^{-k_1 t})+k_o t$	The model describes two pools, k_o being the mineralization rate of the most stable pool.	CABRERA (1993)
e) Double Exponential	$N_m=N_oS(1-\exp^{-ht})+N_o(1-S)(1-\exp^{-kt})$	Considers two pools, where S and (1-S) represent the labile fraction and recalcitrant fraction of N_o, decomposing at a specific rate of h and k, respectively.	MOLINA ***et al.*** (1980)
f) Double Exponential	$N_m=N_{oq}(1-\exp^{-kqt})+N_{os}(1-\exp^{-kst})$	The first term describes the readily mineralizable and the second the less readily mineralizable N, decomposing at a specific rate of k_q and k_s, respectively.	INOBUSHI ***et al.*** (1985)
g) Hyperbolic	$N_m=N_o t/(t^{½}+t)$	Gives an estimate of the half-life of mineralization ($t^{½}$).	JUMA ***et al.*** (1984)
h) Parabolic	$N_m=At^b$	Used by STANFORD & SMITH to give a pre-estimate of N_o by a non-linear model.	BROADBENT (1986)

Experimental data: Eight mathematical models (Table 1) were compared using experimental data from the average of ten representative soils from Rio Grande do Sul, Brazil, comprising a wide range of organic matter (17 - 56g kg^{-1}), nitrogen (0.9 - 3.6g kg^{-1}), pH (4.0 - 6.4) and clay content (180 - 550g dm^{-3}), described by CAMARGO ***et al.*** (1997b). Initially, these soils were incubated for four years in micro-plots (30L) with drainage and exposed at field conditions. This procedure was used to minimize sampling and handling effects at the beginning of incubation. One sample was colleted to determine soil moisture and another one with natural moisture to be used, immediately, in the procedures of incubation described below. This method of incubation described by STANFORD & SMITH (1972) was used. The method consist of using 25g of each soil mixed with 25g of washed sand in leaching tubes of 100mL. These tubes were washed first with 0.01mol L^{-1} $CaCl_2$, and then a solution without N (0.002mol L^{-1} $CaSO_4.2H_2O$, 0.002 mol L^{-1} $MgSO_4$, 0.005mol L^{-1} $Ca(H_2PO_4)_2$. H_20 and 0.00025mol ℓ^{-1} K_2SO_4) was added and the water excess removed by suction at 80 kPa. The tubes were closed to maintain aerobic conditions and incubated at 35°C. At the beginning, and after 2, 4, 8, 16 and 32 weeks, the soils were washed using the same procedure, according to STANFORD & SMITH (1972). NH_4^+ (distillation with MgO) and NO_3^- + NO_2^- (distillation with devarda alloy) were determined in the wash water.

The fitting of the mathematical models was done using SAS-ProcNLIN (SAS Institute, 1990), an iterative method using MARQUARDT (1963) algorithm, obtaining convergence when the decrease in residual squares in relation to that of the previous iteration become negligible. The method depends on initial values for the parameters, important to reduce the number of iterations and to avoid non-convergence problems.

The initial estimates were based on the behavior of the results in relation to those observed in the literature and afterwards determined by the least number of iterations for convergence, least standard error and coefficient of variation, by the variation inflation factor (VIF) and by the least dependence between parameters (1/(1-VIF)), given by the SIGMASTAT (1994) software, using the MARQUARDT (1963) algorithm for non-linear models. To identify the most convenient model, the mathematical properties of the functions were considered (from a biological point of view) as well as the determination coefficient (R^2), the adjusted coefficient of determination (adjusted R^2), the

residual mean of squares (RMS), the residual standard deviation (RSD), the distribution of residues and the asymptotic correlation matrix for the parameter estimates.

RESULTS AND DISCUSSION

Due to the similarity of the R^2 values (Table 2) calculated from the 8 models, it was not possible to determine the best model, using this index. Eliminating the effect of the number of parameters by the adjusted R^2, the fitted models showed differences, with a slight superiority of the simple exponential ones. This behavior could be expected, given the similarity of the R^2 and the higher number of parameters of the double exponential models. Among the models studied, that of JONES (1984) is outstanding by its fitting capacity and description of observed data. With one parameter less than the models of MOLINA ***et al.*** (1980) and INOBUSHI ***et al.*** (1985), this model showed highest adjusted R^2 value, showing that the use of superparametrized models, like the double exponential, is not essential for obtaining an adequate fit.

Comparing the models by residual mean squares and residual standard deviation, the superiority of JONES' (1984) model is clearly seen (Table 2). The fit of the double exponential models (MOLINA ***et al.,*** 1980; INOBUSHI ***et al.,*** 1985) was of lower quality, being superior only to the model described by BROADBENT (1986), that, in the context of the present study, was the most inadequate. As far as the distribution of residues is concerned, the models were alike; however, JONES (1984) model showed a distribution of residues that suggests better fit. Examining the asymptotic correlation matrix of the parameter estimates, high positive and negative correlations are observed for the double exponential models (MOLINA ***et al.,*** 1980; INOBUSHI ***et al.,*** 1985), indicating that they are superparametrized for the used data set. Models with less parameters show low correlations, especially that of JONES (1984).

Considering the mathematical properties of the functions, based on the graphical representation of the fitted models, using the simple exponential as reference (Figure 1), it can be seen that, as they turn into typical zero order kinetic reactions, a slight subestimation of mineralized N occurs for the last point of measurement (32 weeks). JONES (1984) model fits this point better (Figure 1), including the concept of initial flux of mineralization and active nitrogen fraction.

The parameter added to the model of STANFORD & SMITH (1972) by MARION ***et al.*** (1981), a time exponent, improved the fitting (Figure 1). STANFORD & SMITH (1972) observed that the model, made up of two parameters, does not describe adequately the initial nitrogen mineralization rate. These authors corrected their estimates adding N mineralized during the first two weeks of incubation to the potentially mineralizable N, obtained by the simple exponential function (containing two parameters), on the mineralized N after 30 weeks of incubation. The parameter (time exponent) added by MARION ***et al.*** (1981) to the basic mineralization equation described by STANFORD & SMITH (1972) has the same objective and is a simpler way. Recently, CABRERA (1993) used a simple exponential model,

Table 2 - Non-linear coefficients obtained by adjusting accumulated mineralization of organic N (average of ten soils and their replicates), incubated for 32 weeks in laboratory.

Model	a	R	R^2	R^2 adjusted	RDF	RSS	RMS	RSD	CV%	F
a) STANFORD & SMITH (1972)	2	0,992	0,984	0,973	4	223,85	56,0	7,48	7,51	-
b) MARION ***et al.,*** (1981)	3	0,994	0,989	0,972	3	155,4	51,8	7,2	7,23	1,32nsF10(1,3)=5,5
c) JONES (1984)	3	0,998	0,996	0,990	3	55,4	18,5	4,3	4,32	9,10*F10(1,3)=5,5
d) CABRERA (1993)	3	0,995	0,990	0,975	3	137,4	45,8	6,77	6,80	1,89nsF10(1,3)=5,5
e) MOLINA ***et al.,*** (1980)	4	0,995	0,990	0,951	2	136,9	68,5	8,28	8,32	0,63nsF10(2,2)=9,0
f) INOBUSHI ***et al.,*** (1985)	4	0,995	0,990	0,951	2	136,9	68,5	8,28	8,32	0,63nsF10(2,2)=9,0
g) JUMA ***et al.,*** (1984)	2	0,994	0,989	0,982	4	154,2	38,5	6,2	6,23	1,45nsF10(4,4)=4,1
h) BROADBENT (1986)	2	0,976	0,958	0,930	4	584,7	146,2	12,09	12,15	2,61nsF10(4,4)=4,1

a = Number of parameters; F = comparison with STANFORD & SMITH (1972); RDF = Residual degrees of freedom; RSS = Residual sum of squares; RMS = Residual mean of squares; RSD = Residual standard deviation.

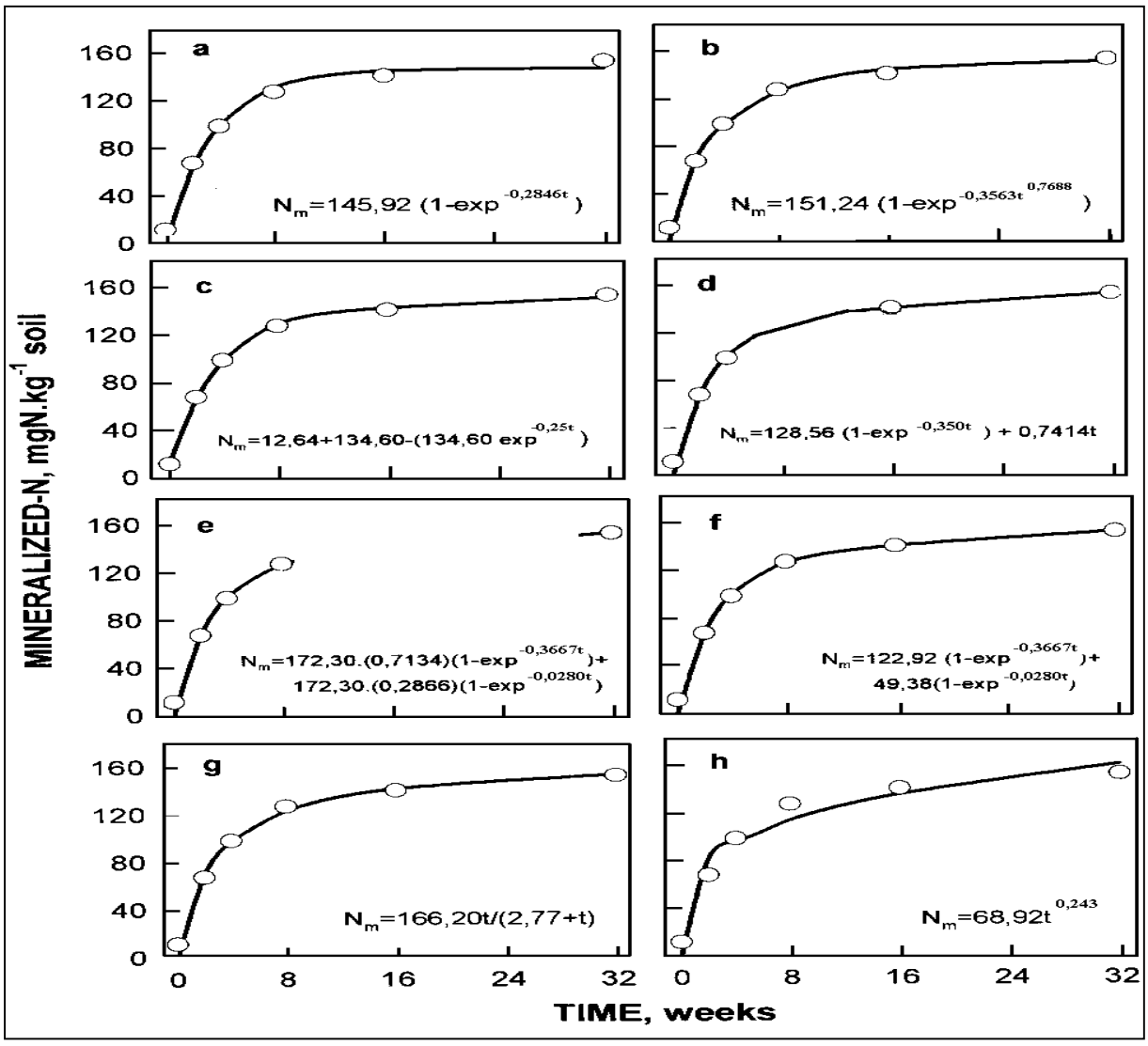

Figura 1 – Comparison of impirical models for the estimating of potentially mineralizable N. Letters correspond to the models described in tables 1 and 2.

including the mineralization rate of the most stable nitrogen pool. Therefore, the application of this model is based on the existence of two pools of organic N being mineralized.

STANFORD & SMITH (1972) verified for 39 soils that the variation of N-mineralization rate was very small (k=0.054 ± 0.009. week^{-1}), suggesting that the main source of mineralized N was the same for all soils. A reevaluation of the experimental data of these authors by MOLINA ***et al.*** (1980) revealed that a better fit of the simple exponential model depends on the presence of various organic N pools, each one with its own mineralization rate. Following this principle, INOBUSHI ***et al.*** (1985) assumed the existence of two nitrogen pools in the mineralization process in 30 flooded soils from Japan. The application of the double exponential model described by these authors (MOLINA ***et al.*** 1980; INOBUSHI ***et al.*** 1985) to the results of the present paper showed good fit to the data observed after the second week (Figure 1).

The first data point was not well fitted by the models, with a deviation of 11.55mg N kg^{-1} of soil. The only model that estimated zero time well was the simple exponential model described by JONES (1984), representing an important factor for the mathematical estimation of residual nitrogen present at the beginning of mineralization. This result was expected because the other authors did not generally use time zero in their adjustments because at this time the mineralization had not begun. The hyperbolic function (JUMA ***et al.,*** 1984) did not

describe the data as well as the double exponential models, mainly for data at week eight (Figure 1).

STANFORD & SMITH (1972) observed that most of their results fitted well to the parabolic equation (Table 1). However, they did not give any more attention to this model, used only as a mean of pre-estimating N_o for the simple exponential model. It is a fact that the parabolic model fitted the data better than the simple exponential model for some soils, not for all of them. BROADBENT (1986), based on the hypothesis that a better estimate of mineralized N could be obtained using the parabolic equation, used values for the time exponent different from 0.5. A comparison between the parabolic model and the exponential model described by STANFORD & SMITH (1972) (Figure 1) showed a better fit with the late model. In their original paper, STANFORD & SMITH (1972) estimated k and N_o by a non-linear fitting of mineralized N as a function of incubation time. This fit is worse than the non-linear model when estimating the mineralization parameters and for the fitting of exponential data (SMITH ***et al.,*** 1980). As in the present paper, the treatment given to the exponential model is non-linear; hence the greate difference between the models can be justified.

Some other differences among the parameters estimated by the eight models can be observed (Figure 1). As far as the potentially mineralizable N (N_o = 145.92 mg kg^{-1}) in the model of STANFORD & SMITH (1972) is concerned, some models subestimated its value, such as those described by JONES (92.2% of N_o), CABRERA (88.1% of N_o) and BROADBENT (47.2% of N_o), while other superestimated it, like those of MARION ***et al.*** (103.6% of N_o), JUMA ***et al.*** (113.9% of N_o), INOBUSHI ***et al.*** (118.1% do N_o), and MOLINA ***et al.*** (118.1% of N_o).

Nitrogen mineralization rates obtained in this work were higher than those cited in the literature. This may be due to the nature of the process, with intense mineralization in the first weeks followed by a relatively long period of stabilization when compared to the mineralization periods. The occurrence of these high initial rates may be associated with an increase of microbial activity, induced by the handling and preparing of the samples. This is a recognized fact, and some authors even neglect the zero time mineralized N for this reason (STANFORD & SMITH, 1972), while other (MATAR ***et al.,*** 1991) start their calculations of accumulated mineralized N from the 4th week. Applying these methods to the data of the present paper, mineralization rates decrease from an average of 0.3196 $week^{-1}$, to 0.2530 $week^{-1}$, if considered from the 2nd week, and to 0.129 $week^{-1}$, if considered from the 4th week (CAMARGO ***et al.,*** 1997b).

The comparison of the models reveals that the simple exponential model described well the mineralization of organic N in soils from Southern Brazil. The active fraction (N_2) and the initial flux (N_1) parameters that compose the model of JONES (1984) can represent the residual mineral N (N_1) that, together with the potentially mineralizable fraction (N_2) and could be used as a simplification for N fertilizer recommendation. Considering that the models studied in this paper are linked to that one of STANFORD & SMITH (1972), but including a higher number of parameters, the only model that presented a significant increase 10% of the explanation variability for mineralized N (F value; Table 2), in relation to the model of STANFORD & SMITH (1972) was the model of JONES (1984).

The double exponential models fit the data satisfactorily, however, only a little improvement was observed when compared to the simple exponential ones, indicating that the superparametrization of these models is not justified when few points are used. In this case, the use of models with more than tree parameters did not fit very well because the low number of degrees of freedom. Nevertheless, it is possible to conclude that during the growing period, or during the incubation period studied (32 weeks), there was no contribution from the recalcitrant fraction to the mineralized organic N balance. This fact is confirmed by data obtained from 26 soils by KEENEY & BREMNER (1966), who divided organic N into that found before and after a year of incubation. The differences between the divided organic forms in this period were not significant, as well as the recovery of exchangeable ammonium, showing that such a situation does not require a higher number of parameters for a better fit.

The unstable or easily mineralizable fraction is mineralized at rates up to 10% per year (HEBERT, 1982). In the present study, a rate of about 7% was observed, characterizing the absence of nitrogen originating from the recalcitrant fraction. The double exponential models give a better fit than the simple exponential models, which is expected given the elevated number of parameters. However, those models present indications of superparametrization and the hypothesis that these models characterize the presence of two forms of organic N susceptible to mineralization can not be sustained or is sometimes impossible (DENDOOVEN ***et al.,*** 1997). To test the hypothesis that there are two organic N pools a 1-2 year-

period of incubation is need. In this case, it is possible that the double exponential models would show some advantages over the single exponential models. Results from DOU ***et al.*** (1996) study show that the pool size and mineralization rate parameters in the different models are merely mathematically defined quantities and do not represent any rigorously-defined pool size of potentially-mineralizable N and their mineralization rate constants in the soils.

CONCLUSIONS

Based on the results obtained in this study and on the numerical and graphical analysis of the models, is possible to conclude that the simple exponential model proposed by JONES (1984) is the one that best describes the mineralization process and the potential of organic N to be released to the environment and to become available for plants.

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