Outlines of a computer simulation model on residual and added nitrogen changes and transport in soils[1]

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Introduction

The nitrate content of underground and surface waters is often related to agrotechnical practices. *Amberger* (1972) summarized the situation in view of research projects carried out in Western Europe. *Taylor* et al. (1971) measurements show that nutrient losses from farmland were significantly greater than those from woodland. *Viets* (1971) and *Viets* and *Hageman* (1971) analysed the possible contribution of fertilizer use to water pollution. Since available literature data on specific contributions of fertilizers and manures to nitrate pollution are inconclusive, an attempt was made, following *de Wit* and coworkers (1972), to contribute to the study of the problem through model simulation of nitrogen changes in the soil.

The aim of the work presented here is to describe the main nitrogen processes in the soil which were the basis for computer programming. Most of the material is based on literature findings, although assumptions and simplifications had to be made. Thus, many improvements over the presented material could be made in the future. A complete description of the model is given in a mimeographed report.[2]

The programming of the model was based on the Continuous System Modeling Program (CSMP) developed by IBM and used by *de Wit* and coworkers for simulating ecological processes and transport phenomena in soils (*de Wit* and *van Keulen*, 1972).

Mineralization and Immobilization of Nitrogen

We assumed that the rate of mineralization of organic nitrogen is influenced by the composition of the organic matter, availability of energy sources, soil moisture,

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pH, and temperature. Further, it was assumed that this process is different in the residual soil organic matter and in the freshly added plant residues and manures. Parallel to mineralization, immobilization of inorganic ammonia and nitrate was assumed. Here the ratio of available organic carbon to available nitrogen determines which of the two processes predominates. Reactions were assumed to occur in situ in the upper soil layer and ammonium [NH$_4$] was taken as the product of mineralization of both forms of organic materials. The diversity of microorganisms responsible for these reactions rules out an exact kinetic description. The best approximation seemed to be first-order kinetics.

The equation describing the mineralization of residual soil organic matter was based on *Stanford* and *Smith's* (1972) paper:

$$\frac{dN}{dt} = -kN \tag{1}$$

where N = concentration of potentially mineralizable nitrogen, t = time, and k = constant.

The potentially mineralizable nitrogen in equation (1) was found as follows:

$$N = No - [NH_4] \tag{2}$$

where N$_0$ = initial concentration of mineralizable nitrogen and [NH$_4$] = nitrogen already mineralized into ammonium. By derivation of equation (2) we obtain

$$\frac{dN}{dt} = - \frac{d[NH_4]}{dt} \tag{3}$$

and

$$\frac{d[NH_4]}{dt} = k(No - [NH_4]) \tag{4}$$

To solve equation (4) numerical values for k and No are needed. Measurements of rates of mineralization and of the potentially mineralizable nitrogen are scarce. For our simulation these values were calculated from *Stanford* and *Smith's* (1972) data. A mean value for k (k = 0.0832/day) was calculated, while N$_0$ correlated reasonably well (r = 0.85) with the percentage of total soil nitrogen (N$_{tot}$) and thus a regression was calculated (N$_0$ = 0.0053 + 0.113 N$_{tot}$). The last was checked with good agreement against *Allison* and *Sterling's* (1949) data. Obviously, these calculations imply·the assumption of a uniform character of soil organic matter.

The freshly added organic materials were classified by the ratio of the most common carbohydrates in them, namely lignin, cellulose and sugar, assuming that the energy for the mineralization process is provided by decomposition of these carbohydrates.

The rate of decomposition of lignin is given by:

$$\frac{dL}{dt} = -k_L L \tag{5}$$

where L = concentration of lignin and k_L = specific constant. The value of (k_L = 0.0019/day) was calculated according to *Alexander* (1961), who states that about 1/3 decomposes in 6 months and about 1/2 in a year.

There is evidence of an inhibitory effect of the lignin on the decomposition of other fractions (*Ghildyal* and *Gupta*, 1969; *Norman*, 1929). *Norman* (1929) showed that the percentage ratio of the relatively easily decomposable fractions and lignin is a good index of decomposability of plant residues. The correlation coefficient was found to be $r = -0.88$. Accordingly, a regression equation, relating *Norman's* index (y) to the percentage of lignin (x) was calculated and a lignin inhibition factor (f) obtained.

$$f = \frac{y}{9.27} = \frac{9.27 - 0.26x}{9.27} \tag{6}$$

The decomposition rates of added organic materials were multipled in the simulation program by this factor.

The equation for the rate of decomposition of cellulose was assumed to be:

$$\frac{dC}{dt} = -k_C C \tag{7}$$

where C = concentration of cellulose and k_C = reaction constant. The last one was calculated from *Anderson's* (1926) data. He found that the rate of decomposition is influenced by the ratio of cellulose to mineral nitrogen added. The following equation gives an approximate expression of this reaction:

$$k_C = 0.04646 - 0.00026 \frac{C_o}{N} \text{ day}^{-1} \tag{8}$$

where C_0 = initial concentration of cellulose and N = mineral nitrogen concentration.

The rate of decomposition of sugars was based on *Winsor* and *Pollard's* (1956) work, who found that the inorganic nitrogen immobilization at different levels of sucrose terminates after about two days. Accordingly,

$$\frac{dS}{dt} = -k_S S \tag{9}$$

where S = concentration of sugars and $k_S = 1.15$/day.

The rates of mineralization of added organic nitrogen were assumed to be related to availability of energy sources. The energy is supplied by decomposition of carbohydrates. Thus the rate of nitrogen mineralization is closely linked to rates of decomposition of the carbohydrates present in the material.

In addition to the above rates of decomposition of carbohydrates, influencing nitrogen mineralization, another consideration was taken into account. There is a higher efficiency of carbon utilization, as energy source for mineralization of

nitrogen, for sugars than for cellulose or lignin. Accordingly, the ratio between carbohydrate consumed and nitrogen mineralized was expressed by the following proportionality factors. For lignin $m_L = 1/50$, for cellulose $m_C = 1/35$ (both according to *Alexander*, 1961) and for sugars $m_S = 1/25$ (according to *Winsor* and *Pollard*, 1956).

Following previous assumptions, again assuming first order kinetics and by integration of equations (5), (7) or (9), the concentration of decomposed material of any carbohydrate D at time t, is

$$D = D_0 (1-e^{-k_D t}) \tag{10}$$

where D_0 = concentration of the carbohydrate at t = 0 and k_D = reaction constant.

The concentration of ammonium nitrogen [NH₄] at time t mineralized from compounds linked to the carbohydrate, assuming for the time being that energy supply is the only rate influencing factor, is then:

$$[NH_4] = D_0 \, m_D \, N_f \, (1-e^{-k_D t}) \tag{11}$$

where N_f = fraction of total organic nitrogen involved in the reaction at t = 0 and m_D proportionality factor for any carbohydrate.
Differentiation yields:

$$\frac{d[NH_4]}{dt} = D_0 m_D N_f k_D e^{-k_D t} \tag{12}$$

and from equation (11):

$$e^{-k_D t} = 1 - \frac{[NH_4]}{D_0 m_D N_f} \tag{13}$$

Further by substituting equation (13) into (12):

$$\frac{d[NH_4]}{dt} = k_D \, (D_0 N_f m_D - [NH_4]) \tag{14}$$

Concentration of [NH₄] obtained by mineralization of freshly added organic materials at time t is calculated by integration of the rate of mineralization expressed in equation (14). Thus

$$[NH_4] = \int_o^t \frac{d[NH_4]dt}{dt} \tag{15}$$

Finally, it must be ascertained whether the nitrogen present undergoes a process of immobilization or mineralization. The dominant process is dictated by the C : N ratios in the material added (C_{OM}/N_{OM}) and in the soil (C_S/N_S). It is generally accepted that the ratio at equilibrium in the residual soil organic matter is $C_S/N_S \simeq 10$. According to *Alexander* (1961), 1.7 % N in added organic material is the limiting value for the release or fixation of mineral nitrogen in it. The average C

content in plant organic materials is 40 %. Hence the corresponding critical $C:N$ ratio is

$$\frac{C_{OM}}{N_{OM}} = \frac{40}{1.7}.$$

The equilibrium ratios for mineralization and immobilization are thus defined for the residual soil organic matter and for the added organic material as:

$$\frac{C_S}{N_S} = 10 \quad \text{and} \quad \frac{C_{OM}}{N_{OM}} = 23$$

It may be assumed that both processes are in equilibrium in the soil after addition of organic materials, thus

$$\frac{C_{OM}}{N_{OM}} = \frac{C_S}{N_S} \cdot 2.3$$

In order to adapt the above equation for cases in which the nitrogen percentage in the added material is not 1.7 %, a certain amount N_x must be added or subtracted. At the same time, the added mineral nitrogen (N_{AM}), which may affect the amount of nitrogen in the soil, must be included with the soil nitrogen (N_S). Thus we obtain:

$$\frac{C_{OM}}{N_{OM} \pm N_x} = \frac{C_S}{N_S + N_{AM}} \cdot 2.3$$

The amount of nitrogen producing disequilibrium, i. e., a trigger of mineralization or immobilization is thus obtainable as:

$$\pm N_x = N_{OM} - (N_S + N_{AM}) \cdot \frac{C_{OM}}{C_S \cdot 2.3} \tag{16}$$

Mineralization or immobilization of nitrogen sets in according to whether the N_x value is negative or positive.

Parallel to the mineralization or immobilization process, there is one of humification, in which the nitrogen which is not released or which is immobilized ($N_{OM} - N_x$, or $N_{OM} + N_x$), is converted into soil organic nitrogen and treated as such.

The influence of environmental conditions such as soil pH, temperature, and moisture on nitrogen mineralization was expressed in the simulation by functions in the form of an optimum curve relating a factor to the environmental parameter. The numerical values of the factors ranged from 0.0 at a level of the environmental parameter where the mineralization is arrested by adverse conditions, to 1.0 at the optimum, and then again to 0.0 at a level of the parameter where the mineralization would stop.

It was assumed according to *Russell* (1963) and *Alexander* (1961) that mineralization may set in at 0 °C, increasing in the 5 °C–30 °C interval and being at an optimum in the range of 30 °C – 60 °C, above which it drops to nil at 75 °C. The temperature of a soil layer for this and subsequent processes was found from the ratio of its volumetric heat content and volumetric heat capacity. The heat content was found by integration from the net heat flux in the soil layer, composed of the heat flowing with water flow, transpiration current and conduction. The heat conductivity of a soil varies with its moisture content (*de Vries* 1963). The heat capacity of the soil layer was calculated as the product of the heat capacity of soil minerals (0.48, that of silicate minerals) and their relative content, plus the heat capacity of water in the same volume of soil layer (*de Wit* and *van Keulen*, 1972).

It seems that nitrogen mineralization is partial at pH as low as 4 and has a broad optimal interval at pH 6–9 (*Harmsen*, 1955 and *Winsor* 1958).

The process apparently takes place under extreme moisture conditions, although the optimal interval for ammonification seems to be between 50–70 % of the water holding capacity (*Miller* and *Johnson*, 1964; *Alexander*, 1961; *Black*, 1968; *Kononova*, 1966). Soil moisture in a soil layer was evaluated from water inputs by irrigation or rain, water flow through the soil profile and water losses by evapotranspiration. The conceptual background and programming of heat and water flow in the soil were adapted mainly from *de Wit* and *van Keulen* (1972).

Oxidation of Ammonium into Nitrate

Similar to the reactions in the previous section, those described here are assumed to occur in situ in the uniform top soil layer. Nitrite accumulation was not included in the program, neither was accumulation of H^+ ions.

The nitrification rate equation was adopted from *Lees* and *Quastel* (1946) as follows:

$$10 \text{ g} \frac{y}{A-y} = K(t-t_1) \tag{17}$$

where $y = NO_3 - N$ produced; A = asymptotic value of y (80 % of original ammonium concentration); t = time; t_1 = time for $y = A/2$; K = constant. Differentiation yields:

$$\frac{dy}{dt} = 2.303 \frac{K}{A} (A-y)y \tag{18}$$

or denoting $2.303 \frac{K}{A} = R$,

$$\frac{dy}{dt} = R(A-y)y \tag{19}$$

It seemed justified to assume that the microbial population active in the process is proportional to the amount of ammonia consumed. Then, y represents also the microbial population and the equation may be interpreted as stating that the rate of nitrate production is proportional to the available ammonium present times the amount of nitrifiers present. Equation (19) was slightly modified to include both the nitrifiers initially present (x_i) and those produced during the process (y), namely

$$\frac{dy}{dt} = R(A-y)(x_i + y) \tag{20}$$

The constants $R = 4.2 \cdot 10^{-4}$ and $x_i = 88.4$ were determined from *Lees* and *Quastel's* work by least-squares fitting.

Correction factors accounting for influences of environmental conditions were presented in a similar way to those in the previous section.

The influence of soil pH on rate of nitrification was reproduced mainly from *Alexander* (1961) and *Kundler* (1970). The factor has a zero value at pH 4.0 and 9.5, rises steeply to a value of 1.0 at pH 6.6, and decreases from that value again steeply at pH 8.4. Possible local, temporary influences of added fertilizers on the pH were not taken into account in this simulation.

The change of nitrification rate with soil temperature was calculated according to *Black* (1968), *Sabey* et al. (1956, 1959), *Parker* and *Larson* (1962), and *Frederick* (1956). Zero values were assigned to the temperature factor for any temperature below 3 °C and above 50 °C. It increased to a value of 1.0 for temperatures of 24 °C to 32 °C.

Soil moisture influence on rate of nitrification was deduced from *Reichman* et al. (1966), *Miller* et al. (1964), *Parker* and *Larson* (1962), *Justice* and *Smith* (1962), and *Black* (1968). Nitrification was assumed to proceed in the moisture range equivalent to suctions of 0.01 to 22.0 bars with an optimum between 0.3 and 1.0 bars.

The presentation of influence of oxygen concentration in the soil atmosphere on nitrification was based on *Amer's* work (*Black*, 1968). The factor gradually increases from zero at zero oxygen concentration to its optimal value at 16 %.

Denitrification of Nitrates

The denitrification of nitrates [NO_3] was also treated as a first-order reaction, with the following factors influencing its rate taken into account: soil temperature (k_t), pH (k_H), available organic matter (k_{om}) and oxygen including moisture (ko). Accordingly

$$-\frac{d[NO_3]}{dt} = k\,[NO_3] \tag{21}$$

where $k = k_t \cdot k_H \cdot k_{om} \cdot k_o$.

The temperature dependent rate constant k_t was taken from *Avnimelech's* (1971) work and was found in agreement with results obtained by some other authors (*Bremner* and *Shaw*, 1958; *Mahendrappa* and *Smith*, 1967). The factor relating denitrification rates to soil pH (k_H) was based mainly on *Valera* and *Alexander's* (1959) paper, again in good agreement with data by other authors (*Bremner* and *Shaw*, 1958). The values for this factor have a value of 1.0 between pH 6.2 and 8.0 and decrease at both sides to zero at pH 4.0 and 10.0. The factor k_{om}, representing the availability of organic carbon equals 1 at an abundant supply of available organic carbon. If, however, this is limited, the factor is smaller than 1 and proportional to the sum of rates of decomposition of soil organic carbon and of the carbohydrates in added organic material. Calculation of those was presented in a previous section.

The oxygen concentration in soil water, either entirely inhibits denitrification or does not influence it at all (*Greenwood* 1963). Thus the factor ko due to oxygen concentration in soil water is either 0 or 1. The limiting concentration was taken as 0.00648 cm^3 0$_2$/100 ml water (*Greenwood* 1963).

The oxygen concentration in soil water of any layer was found by integration of the total flux of oxygen in it. It was assumed that oxygen diffuses simultaneously in the air and water of the soil and that there is an instantaneous equilibrium between the gaseous and dissolved oxygen at any given depth. Further, it was assumed that part of the oxygen is consumed by ammonia and organic carbon oxidation.

Vertical flow of oxygen through soil air may be represented by *Fick's* equation:

$$\frac{\partial C_a}{\partial t} = \frac{\partial}{\partial z}\left(D_a\,\frac{\partial C_a}{\partial z}\right) = D_a\,\frac{\partial^2 C_a}{\partial z^2} + \frac{\partial C_a}{\partial z}\,\frac{\partial D_a}{\partial z} \tag{22}$$

Where C_a = oxygen concentration
  D_a = diffusion coefficient in soil air
  z = depth
  t = time

Assuming D_a to be depth – invariant, the above reduces to:

$$\frac{\partial C_a}{\partial t} = D_a\,\frac{\partial^2 C_a}{\partial z^2} \tag{23}$$

Accounting for changes per unit soil volume (V_a) and for oxygen dissolving in the soil water per unit time ($\partial s/\partial t$), we obtain:

$$V_a\,\frac{\partial C_a}{\partial t} = D_a V_a\,\frac{\partial^2 C_a}{\partial z^2} - \frac{\partial s}{\partial t} \tag{24}$$

A similar reasoning yields the equation for the dissolved oxygen:

$$V_w \frac{\partial C_w}{\partial t} = D_w V_w \frac{\partial^2 C_w}{\partial z^2} + \frac{\partial s}{\partial t} - MV_w \tag{25}$$

where: V_w = volume fraction of soil water

   C_w = oxygen concentration in soil water,

   D_w = diffusion coefficient in soil water, and

   M = amount of oxygen consumed by oxidation of ammonia and organic carbon per volume of soil water and per unit time („oxygen sink").

In view of the instantaneous equilibrium referred to earlier, we have:

$$C_a = K C_w \tag{26}$$

where K = distribution constant of oxygen between air and water.

Substituting equation (26) into (24), adding equation (25) and rearranging, we obtain an equation describing the flux of oxygen in soil water:

$$\frac{\partial C_w}{\partial t} = \frac{V_w D_w + K V_a D_a}{K V_a + V_w} \cdot \frac{\partial^2 C_w}{\partial z^2} - \frac{MV_w}{K V_a + V_w} \tag{27}$$

The boundary and initial conditions are as follows: At the atmosphere-soil interface ($z = 0$) at $t = 0$, the oxygen concentration is equal to the atmosphere:

$$C_a = C_{atm} \tag{28}$$

and

$$C_w = C_{atm}/K \tag{29}$$

For the intermediate soil layers, a linear decrease with depth was assumed; while for the last layer, the soil was assumed saturated.

The consumption factor M, is governed by two processes, organic carbon and ammonia oxidation. In the carbohydrate process, the rate of oxygen (O_2) consumption is proportional to the rate of organic carbon (C) oxidation, represented by the reaction: $CH_3COOH + 2O_2 \rightarrow 2CO_2 + 2H_2O$:

$$\frac{dO_2}{dt} = f_C \frac{dC}{dt} \tag{30}$$

where f_C = proportionality factor for the carbohydrate process.

Since 24 units of organic carbon are oxidized by 64 units of oxygen, the proportionality factor equals $f_C = 24/64$. A 80 % efficiency for the reaction was assumed.

In the nitrification process ($NH_4^+ + 2O_2 + OH^- \rightarrow NO_3^- + H^+ + 2H_2O$), the proportionality is given by:

$$\frac{dO_2}{dt} = f_N \frac{dNH_4}{dt}$$

f_N = proportionality for the nitrification process.
Here 14 g nitrogen consume 64 g of oxygen.
and thus $f_N = 14/64$

$$\text{Finally } M = f_C \frac{dC}{dt} + f_N \frac{dNH_4}{dt} \tag{32}$$

As in previous cases, the organic carbon is represented by carbohydrates having different rates of decomposition. The influence of moisture is taken care of by the assumption that soil pores may be occupied by air or water, with one gradually expelling the other.

The nitrate-nitrogen formed in the top soil layer, or applied to it, was assumed to undergo additional changes. The nitrates are transported from the top layer into other soil layers by diffusion and by flow. The rate of the latter is a function of the water flow. This part of the simulation was programmed according to *de Wit* and *van Keulen* (1972). The nitrate transport with water may lead finally to losses with drainage water. Additional nitrate losses are assumed to be due to nitrogen uptake by plants. Nitrate uptake was calculated as a product of the amount of water taken up by roots from a certain layer and the concentration of nitrates in it. The uptake was limited, however, to a predetermined maximum concentration (18 mg/100 ml).

The rate of water uptake by roots was assumed to be proportional to the transpiration rates and the water content of soil layers and distributed between the layers according to root activity. Again, a limiting value was taken so that water uptake would stop from any layer having a moisture content equivalent to a suction of 20 atm.

Summary

A computer model simulating nitrogen changes and transport in soils was developed. The theoretical background and the assumptions needed for development of the program are presented. The rates of changes of organic nitrogen into ammonium, oxidation of ammonium into nitrate, and denitrification of nitrate are presented as first-order kinetics reactions. The sources of rate constants and of some parameters are shown and the influences of environmental factors on the rate of reactions are described. The nitrate so formed or applied moves through the soil profile and may be lost from it with drainage water, by plant uptake, immobilization, or by denitrification.

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Grundzüge eines Computersimulationsmodelles über die Veränderungen und den Transport von Boden- u. Düngerstickstoff

Von *J. Hagin, A. Amberger, G. Kruh* und *E. Segall*

Es wurde ein Computermodell entwickelt über die Veränderungen und den Transport von Stickstoff in Böden. Der theoretische Hintergrund und die notwendigen Annahmen für die Aufstellung eines solchen Programmes werden dargelegt. Die Umsetzungen von org. Stickstoff in Ammonium bzw. Nitrat sowie die Denitrifikation von Nitrat werden dargestellt als

eine Reaktionsgleichung ersten Grades. Die Reaktionskonstanten und einige Parameter werden aufgezeigt und die Einflußgrößen der Umweltfaktoren auf die Reaktionsraten beschrieben. Das gebildete oder zugegebene Nitrat bewegt sich durch das Bodenprofil und kann verlorengehen durch Auswaschung, Aufnahme von Pflanzen bzw. immobilisiert oder denitrifiziert werden.

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