Fertilizer Research **26**: 319–325, 1990.
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A derivation of the Pierre-Sluijsmans equation used in the Netherlands to estimate the acidifying effect of fertilizers applied to agricultural soils

K. Harmsen, H. Loman & J.J. Neeteson
Institute for Soil Fertility Research (IB), P.O. Box 30003, NL 9750 RA Haren, The Netherlands

Key words: Leaching, denitrification, nitrogen uptake, mineralization, immobilization, ionic balance, soil acidification

Abstract

The acidifying effect of fertilizers applied to agricultural soils can be estimated from their chemical composition and a quantification of the nitrogen cycle in the agricultural system under consideration. In The Netherlands, the acidifying effect of fertilizers is estimated from an ionic-balance equation, referred to as the Pierre-Sluijsmans equation. This equation estimates the amounts of lime required to neutralize the acidifying effect of fertilizers applied to agricultural soils. In the present paper this ionic-balance equation is derived from chemical considerations and its theoretical background is discussed. Particular attention is paid to the acidifying effect of the nitrogen component of fertilizers applied to agricultural soils.

Introduction

In natural soils acidification occurs as a result of physical, chemical and biological processes. For example, dissolution and dissociation of CO_2 in rainwater, adsorption of H^+ and leaching of cations from soils, uptake of cations by plants, and nitrification of ammonium may all contribute to acidification of the soil [9].

In agricultural soils, acidification may also occur as a result of fertilization. The nitrification of fertilizer-applied ammonium-nitrogen results in the production of protons in the soil, whereas the leaching of fertilizer-derived anions from the soil further adds to the accumulation of protons in the rooting zone of the soil. Nitrogen fertilizers applied to agricultural soils would have their maximum acidifying effect if the added ammonium-nitrogen would all be nitrified, and all nitrate-nitrogen would subsequently be leached from the rooting zone of the soil, with cations other than protons as co-ions. Under field conditions, however, not all ammonium-nitrogen is nitrified, and nitrate-nitrogen may be involved in processes other than leaching, such as uptake by crops, mineralization/immobilization turnover, or denitrification. In some of these processes protons are consumed and therefore only part of the fertilizer-applied nitrogen would have an actual acidifying effect.

In The Netherlands, an ionic balance equation is used to estimate the effect of a fertilizer on the acid-neutralizing capacity ('lime status') of a fertilized soil relative to an unfertilized soil. This ionic-balance equation was originally proposed by Pierre [4, 5, 6] and further developed and extensively tested under Dutch conditions by Sluijsmans [7, 8]. Therefore, in The Netherlands, this equation is commonly referred to as the 'Pierre-Sluijsmans' equation [2], a convention that will be followed in the present treatment.

Although the Pierre-Sluijsmans equation is widely used in The Netherlands [1], it seems that its theoretical background has not been discussed in the literature. Recently there has been a renewed interest in the Pierre-Sluijsmans equation, as it may also be applied to estimating the acidifying effect of atmospheric deposition on agricultural as well as forest soils [3]. The objective of this paper is to present a derivation of the Pierre-Sluijsmans equation, as given by Sluijsmans [8], and to discuss its theoretical background. This may foster its proper use in estimating the acidifying effects of fertilizers as well as atmospheric deposition in soils in The Netherlands.

Theory

The amount of lime that is needed to neutralize the acidifying effect of fertilizers applied to the soil may be estimated from the Pierre-Sluijsmans equation [8]:

$$\begin{aligned}E = &-1.0\,\mathrm{CaO} - 1.4\,\mathrm{MgO} - 0.6\,\mathrm{K_2O}\\ &- 0.9\,\mathrm{Na_2O} + 0.4\,\mathrm{P_2O_5}\\ &+ 0.7\,\mathrm{SO_3} + 0.8\,\mathrm{Cl} + \mathrm{nN}\end{aligned} \tag{1}$$

where E denotes the effect of 100 kg of fertilizer on the lime requirement of the soil (kg CaO per 100 kg of fertilizer) and the components of the fertilizers are expressed as percentages (w/w). The coefficient n (dimensionless) is a measure for the acidifying effect of the nitrogen contained in the fertilizers. Values for n are theoretically in the range of 0 to 2. Equation (1) has first been reported by Pierre [4, 5, 6], who used experimentally determined values for n, without explicitly relating these values to the nitrogen flows in the soil-crop systems studied. Sluijsmans [8] explained the coefficient n in terms of the nitrogen dynamics in agricultural soils and showed how changes in the ionic balance upon nitrogen uptake affect the acidifying effect of the fertilizers involved.

Equation (1) may be written in the alternative form:

$$E = \Sigma'[\mathrm{an}] - \Sigma'[\mathrm{cat}] + \alpha\Sigma[\mathrm{N}] \tag{2}$$

where square brackets denote amounts of ionic charge [$\mathrm{kmol_c\,ha^{-1}\,yr^{-1}}$), and where:

$$\Sigma'[\mathrm{an}] = [\mathrm{Cl^-} + \mathrm{H_2PO_4^-} + \mathrm{SO_4^{2-}}] \tag{3}$$

and:

$$\Sigma'[\mathrm{cat}] = [\mathrm{Na^+} + \mathrm{K^+} + \mathrm{Ca^{2+}} + \mathrm{Mg^{2+}}] \tag{4}$$

and:

$$\Sigma[\mathrm{N}] = [\mathrm{NO_3^-} + \mathrm{NH_4^+}] \tag{5}$$

The coefficient α (dimensionless) is a measure for the acidifying effect of the nitrogen in the fertilizers; its values are in the range of 0 to 1. Hydrogen ions are not included in the sum of the cations (eq. 4), because any H^+ present in the fertilizers will be included in the estimate for the acidifying effect (eq. 2), as it is assumed that the excess anionic charge is neutralized by an equivalent amount of hydrogen ions. Any excess positive charge is assumed to be neutralized by $\mathrm{HCO_3^-}$ (or $\mathrm{OH^-}$) ions.

Reference state and fertilizer application

The objective of equation (2) is to estimate the acidifying effect of fertilizers, relative to a reference state in which no nitrogen is added to the soil. In the reference state, mineral nitrogen may be derived from atmospheric deposition of nitrogen compounds, from fixation of atmospheric N_2 by microorganisms in the soil, or from net mineralization of nitrogen contained in crop residues, roots, microbial biomass or organic matter. Assuming that all ammonium-nitrogen in the soil is nitrified during the growing season and that the $\mathrm{NO_3}$-N in the soil in the reference state N_s^0) is either taken up by the crop (N_u^0), denitrified (N_d^0) or leached down to below the rooting zone (N_l^0), it follows that in the reference state:

$$[N_u^0 + N_d^0 + N_l^0] = [N_s^0] \tag{6}$$

Dividing both sides of equation (6) by $[N_s^0]$ yields:

$$f_u^0 + f_d^0 + f_l^0 = 1 \tag{7}$$

where the amounts of nitrate-nitrogen

($\mathrm{kmol_c\,ha^{-1}\,yr^{-1}}$) involved in crop uptake, denitrification and leaching, are expressed as fractions of the initial amount of NO_3-N in the reference soil. In order to estimate the acidifying effect of the nitrate-nitrogen in a reference soil, a coefficient α^0 may be defined according to:

$$\alpha^0 \equiv \alpha_u^0 f_u^0 + \alpha_d^0 f_d^0 + \alpha_1^0 f_1^0 \qquad (8)$$

where the coefficients α_u^0, α_d^0 and α_1^0 are measures for the acidifying effect of the nitrate fractions involved. The acidifying effect of the NO_3-N in a reference soil is thus given by $\alpha^0[N_s^0]$ ($\mathrm{kmol_c\,ha^{-1}\,yr^{-1}}$).

When fertilizer nitrogen is applied to a soil, the amount of soil mineral-nitrogen increases. In the present treatment it is assumed that all applied nitrogen (N_f) mixes thoroughly with the initial soil mineral nitrogen (N_s^0) and that all added ammonium-nitrogen is nitrified in the soil. Furthermore, it is assumed that no net immobilization of fertilizer nitrogen occurs.

The added nitrate-nitrogen in the mineral-nitrogen pool of the soil can thus either be taken up by the crop (ΔN_u), be denitrified (ΔN_d), or be leached down to below the rooting depth of the crop (ΔN_1), where:

$$[\Delta N_u] = [N_u - N_u^0] \qquad (9a)$$

$$[\Delta N_d] = [N_d - N_d^0] \qquad (9b)$$

$$[\Delta N_1] = [N_1 - N_1^0] \qquad (9c)$$

and where $[N_u]$, $[N_d]$ and $[N_1]$ refer to the total amounts of nitrate-nitrogen ($\mathrm{kmol_c\,ha^{-1}\,yr^{-1}}$) involved in these fluxes in the fertilized soil. Assuming that no added nitrate-nitrogen is stored in the mineral-nitrogen pool of the soil, it follows that:

$$[\Delta N_u + \Delta N_d + \Delta N_1] = [N_f] \qquad (10)$$

Dividing both sides of equation (10) by $[N_f]$, it follows that:

$$f_u + f_d + f_1 = 1 \qquad (11)$$

where the amounts of fertilizer-derived nitrate-nitrogen that are involved in crop uptake, denitrification and leaching, are expressed as fractions of the amount of fertilizer nitrogen applied to the soil. The coefficient α in equation (2) is now defined as:

$$\alpha \equiv \alpha_u f_u + \alpha_d f_d + \alpha_1 f_1 \qquad (12)$$

where the coefficients α_u, α_d and α_1 are measures for the acidifying effect of each of the nitrogen fractions involved. The acidifying effect of the added nitrogen thus equals $\alpha[N_f]$ ($\mathrm{kmol_c\,ha^{-1}\,yr^{-1}}$), whereas the acidifying effect of all fertilizers applied is obtained form equations (2) and (12). Equation (2) thus estimates the additional acidifying effect of fertilizers added to the soil, relative to a reference state in which no fertilizers are applied.

Derivation of the Pierre-Sluijsmans equation

The Pierre-Sluijsmans equation (eq. 2) can be derived from the ionic balance of the fertilizers applied, and a quantification of the nitrogen fluxes and rates of uptake in soil-crop systems.

From the condition of electroneutrality it follows that the chemical composition of fertilizers should satisfy the equality:

$$\Sigma[\mathrm{an}] = \Sigma[\mathrm{cat}] \qquad (13)$$

or:

$$\Sigma'[\mathrm{an}] + [NO_3] = \Sigma'[\mathrm{cat}] + [H] + [NH_4] \qquad (14)$$

where ionic charge numbers have been omitted for ease of notation and where $\Sigma'[\mathrm{an}]$ refers to all anions other than NO_3^- (cf. eq. 3) and $\Sigma'[\mathrm{cat}]$ refers to all cations other than H^+ and NH_4^+ (cf eq. 4). Equation (14) can be written as:

$$\Sigma'[\mathrm{an}] - \Sigma'[\mathrm{cat}] + [NO_3] - [NH_4] = [H] \qquad (15)$$

Now assume that all ammonium-nitrogen is nitrified according to:

$$NH_4^+ + 2O_2 \rightarrow 2H^+ + NO_3^- + H_2O$$

If so, the cationic charge associated with NH_4^+ is transformed into an amount of anionic charge equivalent to $[NH_4]_{NO3}$, associated with NO_3^-, and an amount of cationic charge equivalent to $2[NH_4]_H$, associated with H^+. Hence:

$$[NH_4] = 2[NH_4]_H - [NH_4]_{NO3} \tag{16}$$

where the subscripts denote the chemical forms (H^+, NO_3^-) into which the initially present compound (NH_4^+) is transformed. From equations (15) and (16) it follows that, if all ammonium-nitrogen is nitrified:

$$\Sigma'[an] - \Sigma'[cat] + [NO_3] + [NH_4]_{NO3} = [H] + 2[NH_4]_H \tag{17}$$

or:

$$\Sigma'[an] - \Sigma'[cat] + [NO_3 + NH_4]_{NO3} = [H + 2NH_4]_H \tag{18}$$

where $[NO_3 + NH_4]_{NO3}$ denotes the amount of anionic charge associated with nitrate-nitrogen, both initially present and formed upon nitrification, and $[H + 2NH_4]_H$ denotes the amount of cationic charge associated with hydrogen ions. The left-hand side of equation (18) is equal to equation (2) for $\alpha = 1$.

Now assume that all nitrate-nitrogen disappears from the rooting zone of the soil, either by net uptake (f_u), denitrification (f_d) or leaching (f_l). The disappearance of all nitrate-nitrogen is associated with the disappearance of an equivalent amount of cationic charge, either from H^+ or other cations.

When nitrate ions are taken up, equivalent amounts of OH^- or HCO_3^- ions are excreted by the roots. However, because of the flow of nitrate ions into the plant roots, cations accumulate at the root surface. This may result in an excess uptake of cations, that is, an amount in excess of the amount of cations taken up when no additional nitrogen is applied. The coefficient α_u in equation (12) refers to the fraction of the nitrogen uptake that is associated with the excess uptake of cations, relative to a reference state in which no additional nitrogen is applied. If all additional nitrate-nitrogen taken up is compensated by an equivalent increase in the amount of cations taken up (other than H^+), it follows that $\alpha_u = 1$, and the fertilizer-applied nitrogen would have a maximum acidifying effect. If all additional nitrate-nitrogen taken up is compensated by an equivalent excretion of OH^- (or HCO_3^-) ions by the roots, it follows that $\alpha_u = 0$, and the fertilizer-applied nitrogen would have no acidifying effect at all.

The uptake of nitrate-nitrogen in fertilized plots may thus be represented by:

$$[NO_3^-] + (1 - \alpha_u)[H^+] + \alpha_u[cat^+] \rightarrow H_2O + \text{net plant uptake}$$

where α_u is the fraction of the nitrate-nitrogen taken up, associated with an equivalent uptake of cations, and cat^+ denotes a univalent cation. For cations with higher charge numbers, the cationic charge involved in plant uptake may be written as: $[n^{-1}cat^{n+}]$, where n is the charge number. Electroneutrality requires that:

$$f_u[NO_3 + NH_4]_{NO3} = f_u(1 - \alpha_u)[NO_3 + NH_4]_H + f_u\alpha_u[NO_3 + NH_4]_{cat} \tag{19}$$

where f_u is the fraction of the fertilizer-derived nitrate-nitrogen involved in the net uptake by crops.

Denitrification may be represented by:

$$NO_3^- + H^+ \rightarrow 0.5\,N_2 + 0.5\,H_2O + 2.5\,O$$

The coefficient α_d equals zero, because denitrification leads to an equivalent decrease in the amount of H^+ in the soil solution. Hence, from the condition of electroneutrality:

$$f_d[NO_3 + NH_4]_{NO3} = f_d[NO_3 + NH_4]_H \tag{20}$$

where f_d is the fraction of the fertilizer-derived nitrate-nitrogen involved in denitrification. Leaching of nitrate-nitrogen can be represented by:

$$[NO_3^-] + \alpha_l[cat^+] + (1 - \alpha_l)[H^+] \rightarrow \text{leaching}$$

The coefficient α_l is assumed to be close to 1, because when NO_3^- is leached an approximately equivalent amount of cations, other than H^+, is leached as co-ions. Electroneutrality requires:

$$f_1[NO_3 + NH_4]_{NO3} = f_1\alpha_1[NO_3 + NH_4]_{cat} + f_1(1-\alpha_1)[NO_3 + NH_4]_H \quad (21)$$

where f_1 is the fraction of the fertilizer-derived nitrate-nitrogen involved in leaching.

Hence, with equations (11) and (12), and $\alpha_d = 0$, it follows from equations (19)–(21):

$$[NO_3 + NH_4]_{NO3} = (1-\alpha)[NO_3 + NH_4]_H + \alpha[NO_3 + NH_4]_{cat} \quad (22)$$

That is, if all nitrate-nitrogen disappears from the rooting zone of the soil, the amount of anionic charge associated with nitrate-nitrogen, $[NO_3 + NH_4]_{NO3}$, reduces to zero, whereas $\Sigma'[cat]$ decreases to $\{\Sigma'[cat] - \alpha[NO_3 + NH_4]_{cat}\}$ and the amount of protonic charge decreases to $\{[H + NH_4]_H - (1-\alpha)[NO_3 + NH_4]_H\}$. Hence, equation (18) becomes:

$$\Sigma'[an] - \Sigma'[cat] + \alpha[NO_3 + NH_4]_{cat} = [H + 2NH_4]_H - (1-\alpha)[NO_3 + NH_4]_H \quad (23)$$

The left-hand side of equation (23) equals the Pierre-Sluijsmans equation (eq. 2). The right-hand side of equation (23) denotes that the amount of cationic charge associated with hydrogen ions, is equal to the sum of the amount of protonic charge initially present, [H], plus the amount of protonic charge formed upon nitrification of ammonium-nitrogen, $[2NH_4]_H$, minus the amount of protonic charge ‘consumed’ upon plant uptake, leaching or denitrification of nitrate-nitrogen, $(1-\alpha)[NO_3 + NH_4]_H$.

The coefficient α_u

To estimate the value of the coefficient α_u, ionic balances of the crops have to made up, both in fertilized and in unfertilized (reference) plots. Assuming that no ammonium-nitrogen is taken up, the amount of cations ($kmol_c\ ha^{-1}\ yr^{-1}$) taken up by the crop is:

$$\Sigma'_u[cat] = \Sigma[K^+ + Na^+ + Mg^{2+} + Ca^{2+}] \quad (24)$$

where H^+ is not taken into consideration. The amount of anions ($kmol_c\ ha^{-1}\ yr^{-1}$) taken up is:

$$\Sigma'_u[an] = \Sigma[H_2PO_4^- + SO_4^{2-} + Cl^-] \quad (25)$$

where $[NO_3^-]$ is not included in $\Sigma'_u[an]$. Equations (24) and (25) refer to the total amount of nutrients taken up by the crop and not to the nutrients present as ions in the plant tissue. An excess amount of bases, EB ($kmol_c\ ha^{-1}\ yr^{-1}$), is defined as:

$$EB = \Sigma'_u[cat] - \Sigma'_u[an] \quad (26)$$

If equivalent amounts of cations and anions are taken up by the crop and all nitrogen in the soil is present as nitrate-nitrogen, it follows from equations (24)–(26) that $EB = [NO_3^-]$. In general, however, part of the nitrate-nitrogen taken up results in a net excretion of OH^- ions by the roots, such that $EB < [NO_3^-]$.

The magnitude of the excess amount of bases depends on the additional amount of nitrate-nitrogen that is taken up by the crop, relative to the reference state, in which no additional nitrogen is applied to the soil. The excess amount of bases and the amount of nitrate-nitrogen taken up by the crop in the reference state are denoted by EB^0 and N_u^0, respectively. In the reference state a coefficient α_u^0 is defined according to:

$$\alpha_u^0 \equiv (EB^0/[N_u^0]) \quad (27)$$

If nitrate-nitrogen is added to the system, the amount of nitrate-nitrogen taken up by the crop increases and so does the excess amount of bases. The increase in EB, relative to the reference state, is given by:

$$\Delta EB = EB - EB^0 \quad (28)$$

The coefficient α_u is now defined as:

$$\alpha_u \equiv (\Delta EB/[\Delta N_u]) \quad (29)$$

where $[\Delta N_u]$ is given by equation (9a). From equations (27)–(29) it follows that:

$$EB = (\alpha_u^0 - \alpha_u)[N_u^0] + \alpha_u[N_u] \quad (30)$$

Hence, the coefficient α_u can be determined by regression of EB on $[N_u]$, at increasing levels of nitrogen applied.

Approximate estimation procedure

The notions of 'reference soil' and 'fertilized soil' may not be very practical from an experimental point of view, as unfertilized agricultural soils are quite rare under Dutch conditions. Hence, it may be difficult to obtain reliable estimates of the coefficients involved, in particular α_u.

Assuming that all fertilizer-applied ammonium-nitrogen is nitrified, and with $\alpha_d = 0$ and $\alpha_1 = 1$, it follows that the Pierre-Sluijsmans equation may be written as:

$$E = \Sigma'[\mathrm{an}] - \Sigma'[\mathrm{cat}] + \Delta EB + [\Delta N_1] \qquad (31)$$

Assuming that in the reference state the amounts of nitrate-nitrogen are very small relative to those in fertilized soils, that is, if:

$$[N_s^0] \ll [N_f] \qquad (32)$$

it follows that equation (31) reduces to:

$$E = \Sigma'[\mathrm{an}] - \Sigma'[\mathrm{cat}] + EB + [N_1] \qquad (33)$$

Hence, the acidifying effect of fertilizers can be estimated from the ionic balance of the fertilizers applied, the excess amount of bases taken up by the crop in fertilized plots and the amount of nitrate-nitrogen leached from fertilized soils. If the initial amount of nitrate-nitrogen in the reference state would not be negligible, equation (33) would tend to overestimate the acidifying effect of the fertilizers applied.

Discussion

In the derivation of the Pierre-Sluijsmans equation it is assumed that all added ammonium-nitrogen is nitrified in the soil, and that no net immobilization of fertilizer-nitrogen occurs, that is, the rate of immobilization of applied mineral-nitrogen is assumed to be equal to the rate of mineralization.

The assumption that all ammonium-nitrogen is nitrified in the soil, would be quite reasonable for agricultural soils, which are limed regularly (near-neutral pH), during the spring and summer periods, when temperature and moisture conditions are favourable for nitrification. During the winter period, or at high moisture contents, nitrification will be slowed down. During such periods, however, plant uptake would also be slowed down. In addition, most of the ammonium-nitrogen will be adsorbed in the soil, and thus be less available to the crop than nitrate-nitrogen. Therefore, unless crops would take up ammonium ions selectively, the presence of ammonium-nitrogen in the soil may be considered a temporary storage of mineral nitrogen, most of which is eventually transformed into nitrate-nitrogen.

The assumption that the rate of immobilization of nitrogen equals the rate of mineralization would apply to 'stable' soil and crop conditions: not during a single growing season, but averaged over a number of growing seasons or a series of crop rotations. Changes in the cropping system or in soil or crop management may result in temporary net immobilization or net mineralization, during one or more growing seasons. However, in the course of time, if soil and crop conditions stabilize, equilibrium would be reached between mineralization and immobilization.

Under equilibrium conditions, it may be assumed that N_s^0 is relatively small, as most mineral nitrogen in the soil is derived from fertilizer application or atmospheric deposition of nitrogen. Mineral nitrogen derived from mineralization of roots and crop residues during a particular season is largely due to fertilizer application in previous seasons, and thus considered fertilizer-derived nitrogen. Only in soils where net mineralization of soil organic-nitrogen would occur over a longer period of time, such as in certain organic (peat) soils upon the lowering of the groundwater table, would N_s^0 add significantly to the mineral nitrogen in the soil.

In agricultural soils the pH is maintained at near-neutral values through the application of lime. Therefore, low pH-values are unlikely to occur in the topsoil. Under such conditions, the coefficient α_1 may be assumed to be close to 1, because when NO_3^- is leached, an approximately equivalent amount of cations, other than H^+, is leached as co-ions. Only at very low pH-values, not normally encountered in the rooting zone of agricultural soils, would the leaching of H^+ become significant.

The coefficient α_d is taken equal to zero. This assumes that denitrification occurs in the topsoil, within the rooting zone of the crop. If denitrification occurs below the rooting zone, the nitrate-nitrogen is assumed to be leached, that is, the acidifying effect in the rooting zone of the soil is assumed to be at maximum ($\alpha_l = 1$). In general, denitrification is not considered to be a major loss mechanism of nitrate-nitrogen in arable soils, but it might be significant in pasture soils, in particular under conditions of water-logging or in the presence of a shallow water table.

Finally, it should be noted that the Pierre-Sluijsmans equation only estimates the acidifying effect of fertilizers and not the total acidification that may occur in soils, e.g. through the net mineralization and nitrification of organic-nitrogen compounds, oxidation reactions, etc. Also it should be emphasized that this equation does not add to the understanding of the nitrogen transformations or other processes that may occur in soils or soil-crop systems. On the contrary, an understanding and quantification of these processes is a necessary prerequisite for the application of this equation.

The Pierre-Sluijsmans equation does allow, however, to estimate the actual acidifying effect of fertilizers on agricultural soils, and may thus help to maintain proper pH-values in agricultural soils in The Netherlands.

Acknowledgement

Part of the research reported in this paper was supported by Grant no. 92 of the Dutch Priority Programme on Acidification.

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