

# A comparison of the isotope-dilution and the difference method for estimating fertilizer nitrogen recovery fractions in crops. I. Plant uptake and loss of nitrogen

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## Abstract

The apparent recovery fraction (ARF) of applied nitrogen (N) by a crop is calculated as the difference between the total N uptake by crops from fertilized and unfertilized treatments per unit N applied. The <sup>15</sup>N recovery fraction (<sup>15</sup>NRF) is calculated as the amount of <sup>15</sup>N-labeled N recovered in fertilized crops per unit <sup>15</sup>N-labeled N applied. The relationship between ARF and <sup>15</sup>NRF is discussed on the basis of a complete-mixing model for the distribution of <sup>15</sup>N-labeled N over different N pools in the soil-crop system. Mineralization-immobilization turnover in soil is not considered in the model. It is shown that in the lower range of ARF values, i.e., on soils high in available N, values of <sup>15</sup>NRF are likely to exceed those of ARF. This is because the fertilizer N mixes with the soil mineral N pool and thus the plant derives its N from applied as well as soil N, even if there is little or no crop response to applied N. In the higher range of ARF values, i.e., in N-deficient soils, values of <sup>15</sup>NRF may be lower than those of ARF due to an increased uptake efficiency of soil N in fertilized treatments. Loss of N, either from the fertilizer or from the mixed soil mineral N pool, reduces the range of values of <sup>15</sup>NRF and ARF. From an agronomic point of view, ARF is a meaningful quantity as it accurately reflects the overall effect of fertilizer application on crop N uptake, whereas <sup>15</sup>NRF is a meaningful quantity in <sup>15</sup>N tracer studies on N fertilizer use efficiency and N balances in soil-crop systems. In the absence of mineralization-immobilization turnover in soil, the fertilizer N recovery in the crop is accurately estimated by <sup>15</sup>NRF.

*Additional keywords:* apparent recovery fraction, <sup>15</sup>N recovery fraction, N fertilizer efficiency, mineral N, added N interaction, A-value

## Introduction

Stable nitrogen (N) isotope techniques are important research tools in N research in agriculture (Hauck, 1982; Jenkinson *et al.*, 1985). Their primary applications are in studies on N use efficiency by crops and in assessing biological N fixation by rhizobia in association with leguminous crops (Westerman & Kurtz, 1974; Hauck & Bremner, 1976; Hardarson *et al.*, 1991). However,  $^{15}\text{N}$  is also used in N balance studies in soil-crop systems, in assessing N mineralization potentials of soils and microbial biomass N, and, increasingly, in studying N transformations and N dynamics in soils (Shen *et al.*, 1984; Wickramasinghe *et al.*, 1985; Barraclough, 1991; Davidson *et al.*, 1991).

By comparing different fertilizer N sources and alternative types of application, the efficiency of fertilizer management practices can be assessed in terms of increasing the fertilizer N recovery in the crop and reducing losses of applied N from the soil-plant system. To assess the amount of fertilizer N recovered in crops, two methods may be used: (1) the difference or indirect method, and (2) the isotope-dilution or direct method. In the difference method, the amount of applied N taken up by a crop is estimated as the difference in total N uptake per unit N applied between fertilized and unfertilized plots. In the isotope-dilution method the amount of fertilizer N taken up by a crop is estimated from total N uptake and N isotope-ratio analysis of plant materials from fertilized treatments (Hauck & Bremner, 1976).

The difference method is generally assumed to give higher recoveries than the isotope-dilution method (Hauck, 1978; Hauck & Bremner, 1976; Jansson, 1971; Jansson & Persson, 1982), although this is not necessarily the case in the lower range of N recoveries. However, most experimental results appear to be in the medium to high range of N recoveries, e.g. typically 50–75% for crops and pasture (Hauck, 1982), where the difference method would tend to give higher results.

The question thus arose as to what method would give a more accurate estimate of plant uptake of applied N. Hauck & Bremner (1976) suggested that the determination of the recovery percentage of applied N could be made more accurately through the use of  $^{15}\text{N}$ , i.e., the isotope-dilution method. They indicated, however, that both methods make use of assumptions that may not be entirely valid under experimental or field conditions. Users of the difference method would have to assume that addition of N to the soil does not alter the amount of soil N taken up by the crop, whereas users of the isotope-dilution method would have to assume that their interpretation of  $^{15}\text{N}$  data is not confounded by the unknown extent of biological interchange of labeled N with unlabeled soil N. The discussion as to whether to use the difference or the isotope-dilution method continues (Rao *et al.*, 1992; Stout, 1995; Roberts & Janzen, 1990; Jokela & Randall, 1997; MacKown & Sutton, 1997). The quantities ARF and  $^{15}\text{NRF}$  are defined in different ways and therefore it seems that the question is not so much whether one recovery fraction is better or more accurate than the other, but rather when to apply which of the two methods and how to derive additional information from the difference between ARF and  $^{15}\text{NRF}$  in case both quantities are determined in an experiment.

The objective of this paper is to discuss a simplified model for the distribution of  $^{15}\text{N}$ -labeled N over different N pools in the soil-plant system, and to investigate how

these distributions would affect fertilizer-N recoveries by crops, calculated by the difference and the isotope-dilution method. Although it is known that biological interchange of  $^{15}\text{N}$ -labeled N with unlabeled soil organic N may confound the interpretation of fertilizer-N recovery fractions as calculated by the isotope-dilution method (Hauck & Bremner, 1976), mineralization-immobilization turnover in soil is not considered in the present paper. The treatment of mineralization-immobilization requires a different, time-dependent approach (Kirkham & Bartholomew, 1954, 1955; Jenkinson *et al.*, 1985; Hart *et al.*, 1986) and will be dealt with separately (Harmsen, 2003). The present paper's aim is to contribute to the understanding of how plant uptake and loss of N affect  $^{15}\text{NRF}$  and ARF, and what the relationship is between these two quantities. A better understanding of what each quantity really measures and how they complement each other may help to focus N research and obtain more information from experiments in which both quantities are determined. In a companion paper (Harmsen & Garabet, 2003), the models discussed here are compared with results of  $^{15}\text{N}$  research from field and greenhouse trials.

## Definitions and basic assumptions

In the difference method, the apparent recovery of the amount of fertilizer N taken up by a crop is defined as:

$$\text{ARF} = (\text{NP}_{\text{fn}} - \text{NP}_{\text{oh}}) / \text{NF}_i \quad (1)$$

where

ARF = the apparent recovery fraction (dimensionless),

$\text{NP}_{\text{fn}}$  = total N uptake by fertilized crops at harvest ( $\text{kg ha}^{-1}$ ),

$\text{NP}_{\text{oh}}$  = total N uptake by unfertilized crops at harvest ( $\text{kg ha}^{-1}$ ) and

$\text{NF}_i$  = the initial amount of N fertilizer applied ( $\text{kg ha}^{-1}$ ).

In this definition it is assumed that all fertilizer is applied at sowing and that all other factors affecting N uptake by crops are kept constant between fertilized and unfertilized treatments. Conventionally, all experimental treatments in N fertilizer experiments are supplied with required nutrients other than N to ensure that N is the only growth-limiting nutrient.

The term 'apparent recovery fraction' is not universally accepted. Some authors prefer the term 'recovery efficiency' (Bock, 1984; Simonis, 1987), whereas others prefer 'apparent recovery fraction' (Craswell & Godwin, 1984; Harmsen, 1984). The term 'recovery efficiency' refers to the related quantities of 'agronomic efficiency',  $\Delta Y_{\text{fn}} / \Delta \text{NF}_i$ , and 'physiological efficiency',  $\Delta Y_{\text{fn}} / \Delta \text{NP}_{\text{fn}}$ , where  $Y_{\text{fn}}$  and  $\text{NP}_{\text{fn}}$  denote crop yield ( $\text{kg dry matter ha}^{-1}$ ) and crop N uptake in fertilized plots at harvest ( $\text{kg ha}^{-1}$ ), respectively. Agronomic and physiological efficiencies are related to ARF by:

$$\Delta Y_{\text{fn}} / \Delta \text{NF}_i = (\Delta Y_{\text{fn}} / \Delta \text{NP}_{\text{fn}}) (\Delta \text{NP}_{\text{fn}} / \Delta \text{NF}_i)$$

where ARF has been written as  $\Delta \text{NP}_{\text{fn}} / \Delta \text{NF}_i$ .

Although there would be a case for referring to  $\Delta NP_{th}/\Delta NF_i$  as 'efficiency', the term 'apparent recovery fraction' is preferred here because of its analogy with the term ' $^{15}\text{N}$  recovery fraction', which is commonly accepted. The notion 'recovery fraction' refers to the fraction of the fertilizer N recovered by the crop, whereas the notion 'apparent' refers to the assumption that fertilized and control crops absorb the same amount of soil N (Craswell & Godwin, 1984).

In the isotope-dilution method, the amount of applied N taken up by a crop is estimated from:

$$^{15}\text{NRF} = c_{xp}NP_{th}/c_{xf}NF_i \quad (2)$$

where

$^{15}\text{NRF}$  = the  $^{15}\text{N}$  recovery fraction (dimensionless),

$c_{xp}$  = the atom%  $^{15}\text{N}$  excess concentration in fertilized crops (%) and

$c_{xf}$  = the atom%  $^{15}\text{N}$  excess concentration in the applied N fertilizer (%).

Analytical procedures for N isotope-ratio analysis seem to be well established (Bremner, 1965; Hauck & Bremner, 1976; Buresh *et al.*, 1982; Pruden *et al.*, 1985). The atom%  $^{15}\text{N}$  excess concentration in fertilized crops,  $c_{xp}$ , is calculated from the difference between the atom%  $^{15}\text{N}$  concentration in fertilized crops,  $c_{pf}$ , and the atom%  $^{15}\text{N}$  concentration in unfertilized crops,  $c_{po}$ . Hence:

$$c_{xp} = c_{pf} - c_{po}$$

Similarly, the atom%  $^{15}\text{N}$  excess concentration in the applied N fertilizer ( $c_{xf}$ ) is calculated from the difference between the atom%  $^{15}\text{N}$  concentration in the applied N fertilizer ( $c_f$ ) and natural abundance ( $c_o$ ), which is the  $^{15}\text{N}$  concentration in natural N (0.3663%) (Bremner, 1965). Hence:

$$c_{xf} = c_f - 0.3663$$

The atom%  $^{15}\text{N}$  concentration in unfertilized crops ( $c_{po}$ ) may not be exactly equal to natural abundance and may vary slightly between crops and growing conditions. The natural variations in N isotope abundance are measured as:

$$\delta_a^{15}\text{N} = 1000\{(^{15}\text{N}/^{14}\text{N})_s - (^{15}\text{N}/^{14}\text{N})_a\}/(^{15}\text{N}/^{14}\text{N})_a$$

where the subscript 's' refers to the sample and 'a' to the atmosphere, and where  $\delta_a^{15}\text{N}$  is expressed in ‰.

The absolute ratio of  $^{14}\text{N}/^{15}\text{N}$  in atmospheric N has been established as  $272 \pm 0.3$ , which is equivalent to an abundance of  $^{15}\text{N}$  of  $0.3663 \pm 0.0004$  atom% (Hauck & Bremner, 1976). Values of  $\delta_a^{15}\text{N}$  in natural soil and plant samples are generally restricted to a range of  $\pm 10$  ‰ (Rennie *et al.*, 1976; Doughton *et al.*, 1991). These variations are small relative to the enrichment of  $^{15}\text{N}$  used in N research, which would normally be in the range of 1–10 atom% excess  $^{15}\text{N}$ , where 1 atom% excess  $^{15}\text{N}$  would be equivalent to 1747.5  $\delta_a^{15}\text{N}$  units. Therefore, in general, in experiments using  $^{15}\text{N}$ -

enriched N sources, values of  $c_{po}$  determined from unfertilized crops may be considered approximately constant.

The effects of plant uptake and of loss of N on the relationship between  $^{15}\text{NRF}$  and ARF will be investigated for a number of simplified soil-plant systems. For this purpose it will be assumed that (1) application of fertilizer N is followed by instantaneous and complete mixing between initial soil mineral N and fertilizer-applied N, (2) the plant does not discriminate between labeled and unlabeled N, (3) only plant uptake of N and, if stated so, a particular N loss mechanism occur, and (4) mineralization-immobilization turnover does not occur (Figure 1).

In the case of loss of N from the soil-plant system, the assumption of complete mixing may be more realistic for denitrification and leaching of nitrate than for ammonia volatilization from surface-applied ammonium fertilizer or urea. For example, if an ammonium fertilizer or urea is applied to the soil surface, maximum losses due to ammonia volatilization are likely to occur shortly after application, before the applied N is leached down with the rain or the irrigation water and mixing with the initial soil mineral N could have occurred.

Therefore, three cases will be considered:

1. Plant uptake only (no losses);
2. Plant uptake and loss of N from the fertilizer only; and

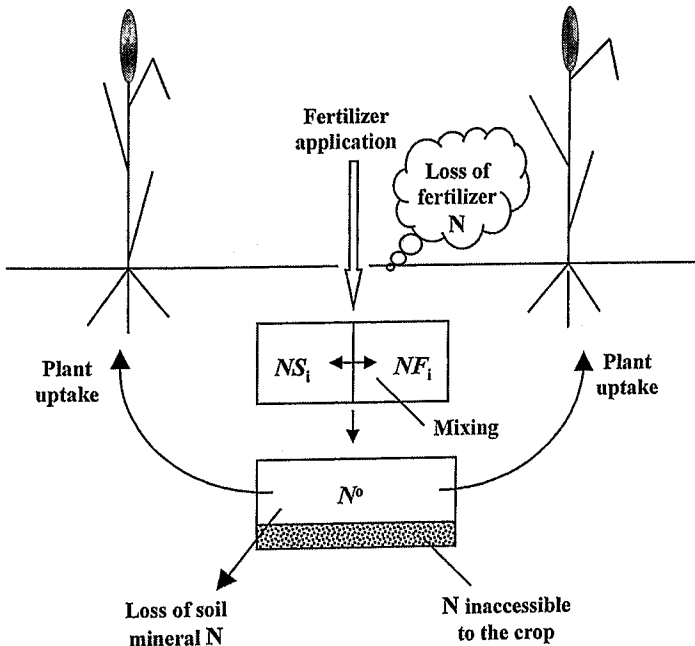


Figure 1. Schematic representation of the processes and transformations involving N considered in the present system: fertilizer N application ( $NF_i$ ), loss of fertilizer N before mixing has occurred, instantaneous mixing of initial soil mineral N ( $NS_i$ ) and fertilizer N ( $NF_i$ ), plant uptake and loss of N from the mixed soil mineral N pool, where the associated variables are given in parenthesis. It is assumed that a fraction of the soil mineral N pool ( $N^o$ ) is not accessible to the crop (shaded area).

3. Plant uptake and loss of N from the mixed soil mineral N pool.

Nitrogen losses under field conditions may be in between cases 2 and 3, depending on the loss mechanism involved and the conditions under which losses occur.

## Plant uptake only

The simplest possible system describing plant uptake of N consists of a soil, a plant and a soil mineral N pool. The plant is assumed to derive all of its N from the soil mineral N pool. Soil mineral N is constant with time and is only affected by plant uptake. All mineral N in the soil is assumed to be equally available to the crop. The amount of N taken up by an unfertilized crop at harvest is given by:

$$NP_{oh} = \epsilon_o NS_i \quad (3)$$

where  $\epsilon_o$  is an uptake coefficient for soil mineral N in unfertilized treatments at harvest (dimensionless), which is a measure for the N uptake efficiency of the unfertilized crop, and  $NS_i$  denotes initial soil mineral N ( $\text{kg ha}^{-1}$ ).

If only plant uptake occurs, the initial amount of soil mineral N remaining in the soil at harvest is:

$$NS_{oh} = (1 - \epsilon_o) NS_i \quad (4)$$

where  $NS_{oh}$  denotes soil mineral N in unfertilized plots at harvest ( $\text{kg ha}^{-1}$ ).

The uptake of N is assumed to be constant with time and thus  $\epsilon_o$  reflects the cumulative N uptake during the growing season. Within the present model  $\epsilon_o$  varies with the amount of soil mineral N available to the crop ( $NS_i$ ). The amount of N required by a crop to achieve its potential (maximum) yield under the soil and climatic conditions of the experiment is denoted by  $NP_{max}$ . It is assumed that a crop only takes up N until the cumulative N uptake equals  $NP_{max}$  and not thereafter. Hence, if  $NS_i \leq NP_{max}$ ,  $\epsilon_o$  is expected to be 1 unless part of the soil mineral N is inaccessible to the crop, or lost from the soil-crop system.

If indeed all N in the soil mineral N pool is available to the crop and there are no yield constraints such as drought (water deficiency) or some other nutrient deficiency (e.g. phosphorus), then  $\epsilon_o$  is expected to be 1, until the crop's demand for N is satisfied. Beyond that, the crop would take up little or no additional N and the 'effective'  $\epsilon_o$  would decrease with increasing N in the soil mineral N pool. In other words, if  $NS_i \leq NP_{max}$  it follows that  $\epsilon_o = 1$  and if  $NS_i > NP_{max}$  it follows that  $\epsilon_o = NP_{max}/NS_i < 1$ , where  $p = 1$  if all soil mineral N is accessible to the crop and  $0 < p < 1$  if not all soil mineral N is accessible to the crop.

The amount of N taken up by a fertilized crop at harvest can be expressed by:

$$NP_{fn} = \epsilon_r N^o \quad (5)$$

$$N^o = NS_i + NF_i$$

where  $N^0$  denotes soil mineral N after mixing of  $NS_i$  and  $NF_i$  ( $\text{kg ha}^{-1}$ ), and  $\epsilon_f$  is an uptake coefficient for soil mineral N in fertilized treatments at harvest (dimensionless), which is a measure for the cumulative N uptake efficiency of fertilized crops.

Hence, if  $NP_{\max} \geq N^0$  it follows that  $\epsilon_f = q$  and if  $NP_{\max} < N^0$  it follows that  $\epsilon_f = NP_{\max}/N^0 < q$ , where  $q = 1$  if all soil mineral N is accessible to the crop and  $0 < q < 1$  if not all soil mineral N is accessible to the crop.

Equation 5 thus assumes that the crop takes up N from both sources with equal efficiency, in proportion to their abundance in the soil mineral N pool. These assumptions are similar to those on which the A-concept is based (Fried & Dean, 1952) and the criticism of that concept (e.g. Hauck & Bremner, 1976) also applies to the present treatment.

Initial soil mineral N and fertilizer N not taken up by the crop at harvest will remain in the soil mineral N pool:

$$NS_m = (1 - \epsilon_f) N^0 \quad (6)$$

where  $NS_m$  denotes the amount of mineral N in the soil at harvest ( $\text{kg ha}^{-1}$ ).

The atom% excess  $^{15}\text{N}$  of the soil mineral N pool ( $c_{xs}$ ) immediately following fertilizer application ('zero-time control') may be calculated from the mass conservation equation in the form:

$$c_{xs} = c_{xf} NF_i / N^0 \quad (7)$$

Equation 7 could, in principle, be corrected for the differences in molecular weight between compounds of different isotopic composition, but these minor corrections are not considered in the present paper. Assuming that the crop derives all of its N from the soil mineral N pool without discrimination between  $^{14}\text{N}$  and  $^{15}\text{N}$ , it follows that:

$$c_{xp} = c_{xs} \quad (8)$$

From Equations 1, 3 and 5 it follows that:

$$ARF = \epsilon_f + (\epsilon_f - \epsilon_0) NS_i / NF_i \quad (9)$$

and from Equations 2, 5, 7 and 8 that:

$$^{15}\text{NRF} = \epsilon_f \quad (10)$$

Hence, in the present model,  $^{15}\text{NRF}$  would equal the N uptake efficiency by fertilized crops at harvest ( $\epsilon_f$ ). Furthermore, from Equations 9 and 10 it follows that:

$$^{15}\text{NRF} = (ARF + \epsilon_0 NS_i / NF_i) / (1 + NS_i / NF_i) \quad (11)$$

If mixing between  $NS_i$  and  $NF_i$  would be incomplete, Equation 5 could be modified as follows:

$$NP_{\text{th}} = \epsilon_{\text{ff}}NF_1 + \epsilon_{\text{fs}}NS_1$$

where the uptake coefficients  $\epsilon_{\text{ff}}$  and  $\epsilon_{\text{fs}}$  refer to the uptake of fertilizer- and soil-derived N, respectively. In that case, Equations 9 and 10 would become:

$$\text{ARF} = \epsilon_{\text{ff}} + (\epsilon_{\text{fs}} - \epsilon_0)NS_1/NF_1$$

$$^{15}\text{NRF} = NP_{\text{th}}/NF_1 = \epsilon_{\text{ff}}$$

such that:

$$^{15}\text{NRF} = \text{ARF} - (\epsilon_{\text{fs}} - \epsilon_0)NS_1/NF_1$$

where  $NP_{\text{th}}$  denotes the amount of fertilizer-derived N in the crop at harvest ( $\text{kg ha}^{-1}$ ). It can be seen that  $^{15}\text{NRF} = \text{ARF}$  if  $\epsilon_{\text{fs}} = \epsilon_0$ , as expected.

These equations may be of use in the interpretation of results of  $^{15}\text{N}$  tracer experiments in case the mixing model would not apply. In the present context, however,  $\epsilon_{\text{ff}}$  is assumed to be equal to  $\epsilon_{\text{fs}}$ , as this assumption is basic in the mixing model.

## Limiting cases of ARF and $^{15}\text{NRF}$

From Equations 9 and 10 it follows that:

$$^{15}\text{NRF} < \text{ARF} \quad \text{if } \epsilon_{\text{f}} > \epsilon_0$$

$$^{15}\text{NRF} = \text{ARF} \quad \text{if } \epsilon_{\text{f}} = \epsilon_0$$

$$^{15}\text{NRF} > \text{ARF} \quad \text{if } \epsilon_{\text{f}} < \epsilon_0$$

So ARF is equal to  $^{15}\text{NRF}$  only if  $\epsilon_{\text{f}} = \epsilon_0$ , i.e., if the amounts of initial soil mineral N taken up by the crop are the same for fertilized and unfertilized crops. In general, however,  $\epsilon_0$  is unlikely to be exactly equal to  $\epsilon_{\text{f}}$ . This will be illustrated by considering  $^{15}\text{NRF}$  in two limiting cases of ARF:  $\text{ARF} = 0$  and  $\text{ARF} = 1$ . The first case represents excess mineral N in the soil, i.e., conditions where N is not limiting crop growth and therefore little or no fertilizer N is taken up by the crop. The second case represents a N deficient situation where N is limiting crop growth and all applied and initial soil mineral N is taken up by the crop.

In the limiting case that  $\text{ARF} = 0$ , it follows that  $NP_{\text{th}} = NP_{\text{oh}}$  and thus:

$$\epsilon_0NS_1 = \epsilon_{\text{f}}N^0$$

from which it follows that  $\epsilon_0 > \epsilon_{\text{f}}$  if  $NF_1 > 0$ , as is expected in fertilizer experiments. Furthermore, from:

$$^{15}\text{NRF} = \epsilon_0(NS_1/NF_1)/(1+NS_1/NF_1)$$



it follows that  $^{15}\text{NRF} > 0$  if  $\epsilon_0 > 0$  and  $NS_i/NF_i > 0$ . The limiting case in which  $\text{ARF} = 0$  and  $^{15}\text{NRF} > 0$  could occur in practice if another nutrient or available moisture would become severely limiting at the same level of N in both fertilized and unfertilized treatments. In that case N uptake would be constant between treatments and presumably quite low. Another possibility is that there would be more than sufficient mineral N initially in the soil to satisfy the crop's N requirement. In this case, N uptake would also be approximately the same in both treatments (i.e.,  $NP_{\text{max}}$ ), as the crop would not take up more N than it needs.

The reason that  $^{15}\text{NRF}$  exceeds ARF in the lower range of ARF values is thus merely a matter of definitions: ARF is corrected for the N uptake in unfertilized plots, and if  $NP_{\text{oh}} = NP_{\text{fh}}$ , it follows that  $\text{ARF} = 0$ . In contrast,  $^{15}\text{NRF}$  is based on N uptake in fertilized plots only, and as fertilizer N and initial soil mineral N are fully mixed and taken up proportionally,  $^{15}\text{NRF}$  always assumes a positive value provided the crop takes up any N at all. This is illustrated in Figure 2 where  $^{15}\text{NRF}$  is plotted as a function of ARF according to Equation 11. In the lower range of ARF-values  $^{15}\text{NRF}$  exceeds ARF, whereas in the higher range the reverse is true, if  $\epsilon_0 < 1$ . From an agronomic point of view, ARF would be the more meaningful quantity in this particular case: one would

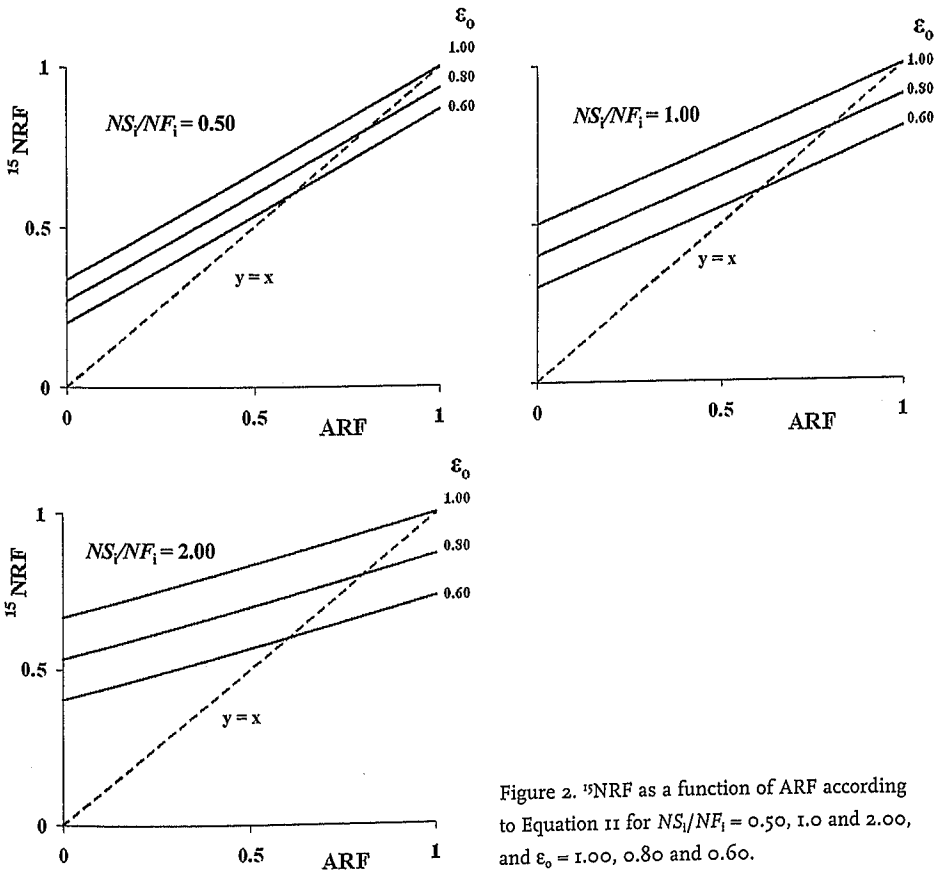


Figure 2.  $^{15}\text{NRF}$  as a function of ARF according to Equation 11 for  $NS_i/NF_i = 0.50, 1.0$  and  $2.00$ , and  $\epsilon_0 = 1.00, 0.80$  and  $0.60$ .

not normally recommend a farmer to apply N fertilizer if a crop response is not expected, irrespective of whether or not some fertilizer N would be taken up by the crop. On the other hand,  $^{15}\text{NRF}$  is a true reflection of the fertilizer N recovery, even though this does not show up in crop yield.

In case  $\text{ARF} = 1$ , i.e., if  $\text{NP}_{\text{th}} - \text{NP}_{\text{oh}} = \text{NF}_i$ , it follows that  $^{15}\text{NRF} = 1$  if  $\epsilon_o = \epsilon_f$  and  $\text{NS}_i, \text{NF}_i > 0$ , and that  $^{15}\text{NRF} < 1$  if  $\epsilon_o < \epsilon_f$  and  $\text{NS}_i, \text{NF}_i > 0$ .

One would expect both  $^{15}\text{NRF}$  and  $\text{ARF}$  to be close to 1 in soils deficient in N, although it is conceivable that the uptake efficiency would increase in fertilized plots because of better plant establishment and more prolific root development. The situation in which  $\epsilon_o < \epsilon_f$  would result in a 'real' added nitrogen interaction (Jenkinson *et al.*, 1985). In a highly N-deficient soil, crop growth and root development would be more prolific in the fertilized than in the unfertilized treatment, such that more initial soil mineral N would be taken up in the fertilized treatment, resulting in the inequality  $\epsilon_o < \epsilon_f$  to hold. In case  $\epsilon_o < \epsilon_f$ ,  $\text{ARF}$  would overestimate the fertilizer N recovery fraction, as more soil-derived N would be taken up in fertilized than in unfertilized treatments.

The relationship between  $^{15}\text{NRF}$  and  $\text{ARF}$  in the higher range of  $\text{ARF}$ -values is further illustrated in Figure 2. The intercept and slope of the curves in Figure 2 follow from Equation 11:

$$\text{intercept} = \epsilon_o(\text{NS}_i/\text{NF}_i)/(1+\text{NS}_i/\text{NF}_i)$$

$$\text{slope} = 1/(1+\text{NS}_i/\text{NF}_i)$$

Figure 2 shows, from left to right, that the intercept increases and the slope decreases with increasing  $\text{NS}_i/\text{NF}_i$  ratio, at constant  $\epsilon_o$ . Furthermore, the intercept decreases if  $\epsilon_o$  decreases, at a constant value of  $\text{NS}_i/\text{NF}_i$ .

If all N in the soil mineral N pool is fully mixed and equally available to the crop, the values of  $\epsilon_o$  and  $\epsilon_f$  would only decrease if the supply of N would be in excess of the crop's demand. The latter phenomenon is illustrated in Figure 3. Here it is assumed that the crop takes up all available soil N up to a level of 100 kg ha<sup>-1</sup>. Beyond that level, the crop does not take up any N from the soil mineral N pool. The uptake coefficients for unfertilized treatments may be calculated as follows:

$$\epsilon_o = 1 \quad \text{if } \text{NS}_i \leq 100 \text{ kg ha}^{-1}$$

$$\epsilon_o = 100/\text{NS}_i \quad \text{if } \text{NS}_i > 100 \text{ kg ha}^{-1}$$

and in a similar way for fertilized treatments, replacing  $\epsilon_o$  and  $\text{NS}_i$  by  $\epsilon_f$  and  $\text{N}^o$ , respectively. If  $\epsilon_o$  and  $\epsilon_f$  are smaller than 1, e.g. 0.80, their values may be calculated in an analogous manner:

$$\epsilon_o = 0.80 \quad \text{if } \text{NS}_i \leq 125 \text{ kg ha}^{-1}$$

$$\epsilon_o = 100/\text{NS}_i \quad \text{if } \text{NS}_i > 125 \text{ kg ha}^{-1}$$

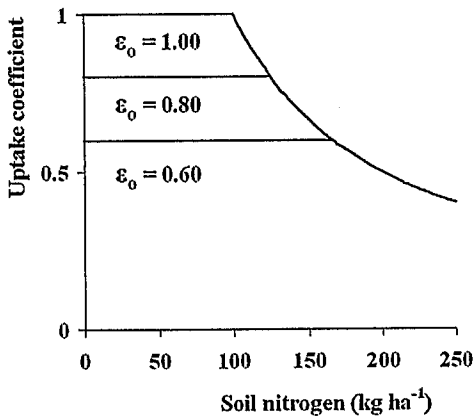


Figure 3. The effective N uptake coefficient ( $\epsilon_0$ ) as related to the initial and boundary conditions of the system for  $NP_{oh} \leq 100 \text{ kg ha}^{-1}$ , for three situations: all soil mineral N accessible to the crop ( $\epsilon_0 = 1.00$  if  $NS_i < 100 \text{ kg ha}^{-1}$ ), 20% inaccessible ( $\epsilon_0 = 0.80$  if  $NS_i < 125 \text{ kg ha}^{-1}$ ) and 40% inaccessible to the crop ( $\epsilon_0 = 0.60$  if  $NS_i < 167 \text{ kg ha}^{-1}$ ).

and in a similar way for fertilized treatments, substituting  $\epsilon_f$  and  $N^o$  for  $\epsilon_0$  and  $NS_i$ , respectively. Hence, at levels of soil mineral N in excess of the crop's demand, the values of the uptake coefficients decrease.

There are other reasons why an uptake coefficient might be smaller than 1. Part of the soil mineral N could not be accessible to the crop, e.g. because it would be beyond the reach of the crop's root system, either at depth in the soil or inside soil aggregates where the roots do not penetrate. Also drought could make N in the topsoil temporarily inaccessible to the crop. However, in such cases the question comes up whether the assumption of complete mixing would be realistic, i.e., whether it is realistic to assume that fertilizer N would fully mix with soil N beyond the reach of the crop, such as in soil aggregates in heavy-textured soils.

Finally, an uptake coefficient could be 'effectively' smaller than 1 because of losses of N occurring from the soil mineral N pool, through ammonia volatilization, denitrification or deep leaching. Other processes, such as ammonium fixation by clay minerals or immobilization of N by the heterotrophic biomass, would have the same effect, as N is removed from the available N pool and becomes effectively inaccessible to the crop. In fact, even uptake of N by weeds would have the same effect, but weeds are not considered in the present treatment.

## Relationships between ARF and $^{15}\text{NRF}$

The relationships between  $^{15}\text{NRF}$  and ARF as shown in Figure 2 should be interpreted with some caution. Not all possible combinations of  $\epsilon_0$ ,  $\epsilon_f$  and  $NS_i/NF_i$  are physically realistic nor are they independent, i.e., one cannot treat  $\epsilon_0$ ,  $\epsilon_f$ ,  $NS_i$  and  $NF_i$  as if they were independent parameters ( $\epsilon_0$ ,  $\epsilon_f$ ) or variables ( $NS_i$ ,  $NF_i$ ). For example,  $\epsilon_0$  and  $\epsilon_f$  decrease at soil mineral N levels in excess of the crop's demand (Figure 3), whereas the ratio  $NS_i/NF_i$  increases with increasing fertilizer rates. Therefore, to better understand the relationship between  $^{15}\text{NRF}$  and ARF, their pair-wise values were calculated for a very simple but physically not unrealistic system. The system consists of independent variables ( $NS_i$ ,  $NF_i$ ), dependent variables and functions ( $NP_{oh}$ ,  $NP_{th}$ , ARF and

<sup>15</sup>NRF) and parameters ( $\epsilon_o$ ,  $\epsilon_f$ ). Calculations were done for the following conditions:

$$\begin{aligned}
 &NS_i, NF_i = 0, 10, 20, \dots, 100 \text{ kg ha}^{-1} \\
 &NP_{oh}, NP_{fh} \leq 100 \text{ kg ha}^{-1} \\
 &\epsilon_o = \epsilon_f = 1.00 \quad \text{if } NS_i, N^o \leq 100 \text{ kg ha}^{-1} \\
 &\epsilon_o = 100/NS_i \quad \text{if } NS_i > 100 \text{ kg ha}^{-1} \\
 &\epsilon_f = 100/N^o \quad \text{if } N^o > 100 \text{ kg ha}^{-1}
 \end{aligned}$$

This calculation thus generates 100 numbers for each of the variables and parameters considered. Only <sup>15</sup>NRF-ARF data-pairs for which  $NP_{fh} = 100 \text{ kg ha}^{-1}$ , <sup>15</sup>NRF < 1 and ARF > 0 are plotted in Figure 4. This is because many combinations of  $NS_i$  and  $NF_i$  result in ARF = <sup>15</sup>NRF = 1.00, whereas at ARF = 0 there is a large number of different values of <sup>15</sup>NRF that fit the model. Data-pairs for which  $NS_i/NF_i$  is constant conform to linear curves, as follows from Equation 11. However, the data in Figure 4 do not conform to a single straight line, because  $NS_i/NF_i$  ranges from 0.10 to 10 and the value of  $\epsilon_f$  decreases when the crop's N requirement has been satisfied. The linear regression equation calculated from the data in Figure 4 is:

$${}^{15}\text{NRF} = 0.55 + 0.40 \text{ ARF}$$

It should be noted, though, that the numerical values of the regression coefficients depend on the values of  $NS_i$  and  $NF_i$  in the example calculations and on the selection of the values included in Figure 4. For example, if values for <sup>15</sup>NRF = ARF = 1 at  $NP_{fh} = 100$  would have been included in Figure 4, the linear regression equation would have been very close to Equation 11 for  $\epsilon_o = 1$  and  $NS_i/NF_i = 1.0$ . Nevertheless, Figure 4 illustrates how variations in the ratio  $NS_i/NF_i$  as well as decreasing values of  $\epsilon_f$  affect the relationship between <sup>15</sup>NRF and ARF. Also, by following the same logic in all example calculations, there is a basis for comparison between them. Figure 4 thus shows that <sup>15</sup>NRF tends to be higher than ARF, over the entire range of ARF values, in accordance with Equation 11 for  $\epsilon_o = \epsilon_f = 1$ .

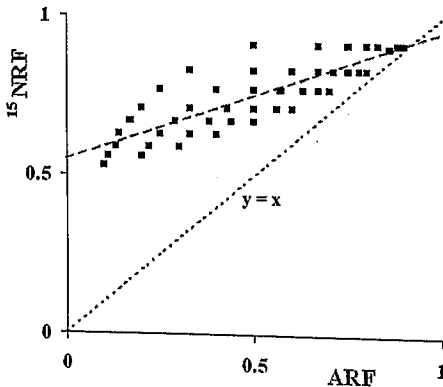


Figure 4. Calculated <sup>15</sup>NRF-values as a function of ARF for  $\epsilon_o = 1.00$ ,  $\epsilon_f = 1.00$  and  $NP_{oh}, NP_{fh} \leq 100 \text{ kg ha}^{-1}$ . The broken line represents the linear regression equation of <sup>15</sup>NRF on ARF.

As a second example, values of  $^{15}\text{NRF}$  and ARF were calculated for two cases, similar to the one considered in Figure 4, except for the following:

- (1)  $NS_i = 0, 10, 20, \dots, 130 \text{ kg ha}^{-1}$   
 $\epsilon_0 = 0.80$   
 $\epsilon_f = 0.85$  ( $NF_i = 10$ ),  $0.90$  ( $NF_i = 20$ ),  $0.95$  ( $NF_i = 30$ ) and  $1.00$  ( $NF_i \geq 40 \text{ kg ha}^{-1}$ )

and:

- (2)  $NS_i = 0, 10, 20, \dots, 170 \text{ kg ha}^{-1}$   
 $\epsilon_0 = 0.60$   
 $\epsilon_f = 0.65$  ( $NF_i = 10$ ),  $0.70$  ( $NF_i = 20$ ),  $\dots, 1.00$  ( $NF_i \geq 80 \text{ kg ha}^{-1}$ )

The situation in which  $\epsilon_0 < \epsilon_f$  could be interpreted as a case of a 'real' added nitrogen interaction: a fraction of the initial soil mineral N, which is not accessible to the unfertilized crop, can be taken up by crops in fertilized treatments, e.g. because of a more prolific root development. As a consequence, the example calculation generates some ARF values larger than 1. In the example it is assumed that  $\epsilon_f$  increases linearly with increasing fertilizer rate from  $\epsilon_0 = \epsilon_f = 0.6$  or  $0.8$  at  $NF_i = 0$  until it reaches 1. This is done because the assumption that  $\epsilon_0 = 0.6$  or  $0.8$  implies that 20–40% of the mineral N pool would not be accessible to the crop in unfertilized plots. In such a situation, where  $NS_i$  ranges from 10–170  $\text{kg ha}^{-1}$ , it is unlikely that the addition of  $NF_i = 10 \text{ kg ha}^{-1}$  would suddenly make the entire additional 20–40% of the soil mineral N pool accessible to the crop. So a gradual increase in  $\epsilon_f$  seems physically more realistic.

It follows from Figure 5 that if  $\epsilon_0 < 1$ , values of  $^{15}\text{NRF}$  tend to be lower than their corresponding values in case  $\epsilon_0 = 1$ . The slopes of the linear regression equations for  $\epsilon_0 = 0.8$  and  $0.6$  are virtually the same as in case  $\epsilon_0 = 1$ , thus confirming that changes in  $\epsilon_0$  do not affect the slope of the relationship between  $^{15}\text{NRF}$  and ARF (Equation 11). The lower intercept in case  $\epsilon_0 < 1$  implies that  $\text{ARF} > ^{15}\text{NRF}$  in the higher range of ARF values, i.e.,  $\text{ARF} > 0.6$  ( $\epsilon_0 = 0.6$ ) or  $\text{ARF} > 0.8$  ( $\epsilon_0 = 0.8$ ). It further follows from Figure 5 that the intercept decreases with decreasing value of case  $\epsilon_0$ , in accordance with Equation 11.

## Added nitrogen interaction

The concept of 'added N interaction' (ANI) is defined by Jenkinson *et al.* (1985) as "any increase (or decrease) in the quantity of soil-derived N in a compartment caused by added N", where 'compartment' is defined as "a particular assemblage of N atoms". The term 'compartment' in the definition of ANI could be replaced by 'pool', where 'pool' is defined as "a compartment containing material that is chemically indistinguishable and equally accessible to plants (or to the soil population)". From the definition of ANI it follows that for plant uptake it can be written as:

$$\text{ANI} = NP_{\text{fsh}} - NP_{\text{oh}}$$

where  $NP_{fsh}$  denotes soil-derived N in fertilized crops at harvest ( $kg\ ha^{-1}$ ). From Equation 1 and from

$$NP_{fh} = NP_{fsh} + NP_{fn}$$

$${}^{15}NRF = NP_{fn} / NF_i$$

it follows that:

$$ANI/NF_i = ARF - {}^{15}NRF$$

This expression for ANI ('relative ANI') would generally apply to ANI's whether they be negative or positive and apparent or real, and has been applied to quantify ANI's in field experiments (Stout, 1995). In the case of plant uptake only, the relative ANI reduces to:

$$ANI/NF_i = (\epsilon_r - \epsilon_o) NS_i / NF_i$$

From the example in Figure 5, two cases will be taken to illustrate the notion of  $ANI/NF_i$ :

- (1)  $\epsilon_o = 0.8$   
 $NS_i = 50$  and  $NF_i = 0, 10, \dots, 250\ kg\ ha^{-1}$
- (2)  $\epsilon_o = 0.6$   
 $NS_i = 50$  and  $NF_i = 0, 10, \dots, 250\ kg\ ha^{-1}$

The results for  $ANI/NF_i$  are plotted in Figure 6 from which it can be seen that at

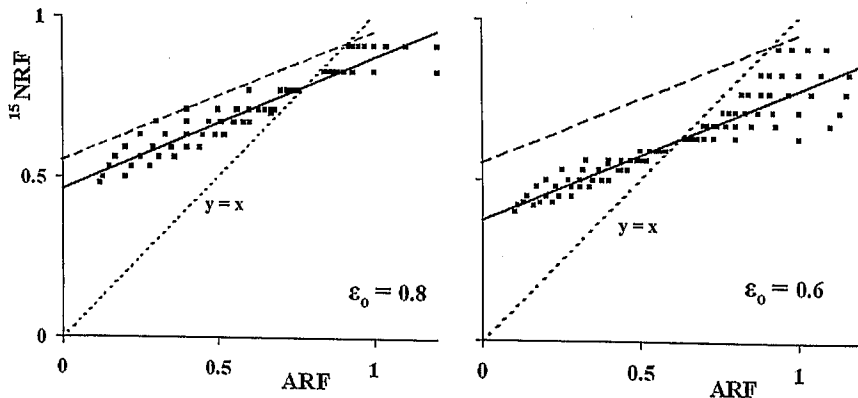


Figure 5. Calculated  ${}^{15}NRF$ -values as a function of ARF for  $\epsilon_o = 0.80$  (left) and  $0.60$  (right),  $\epsilon_r = 1.00$  and  $NP_{oh}$ ,  $NP_{fn} \leq 100\ kg\ ha^{-1}$ . The solid lines represent the linear regression equations of  ${}^{15}NRF$  on ARF, the broken lines represent the linear regression equations of  ${}^{15}NRF$  on ARF for  $\epsilon_o = 1.00$  and  $\epsilon_r = 1.00$  (taken from Figure 4).

low rates of  $NF_i$ , where  $\epsilon_o < \epsilon_f$ , the real ANI is positive. This is because more soil mineral N becomes available when  $\epsilon_f$  increases in fertilized treatments. At higher rates of  $NF_i$ , when the crop's N requirement has been met,  $\epsilon_f$  decreases while the proportion of  $^{15}N$  in the soil mineral N pool and in the crop continues to increase, causing  $NP_{ff}$  to increase and  $NP_{fs}$  to decrease, which results in a negative ANI. The latter phenomenon is common to all example calculations in the present paper, where  $\epsilon_f < \epsilon_o$  when the crop's demand for N has been satisfied and  $NF_i$  continues to increase. The positive ANI is unique to the present example, where  $\epsilon_o < \epsilon_f$  at low rates of fertilizer application.

### Plant uptake and loss of fertilizer nitrogen before mixing

If urea or ammonium fertilizers are applied to the surface of calcareous or weakly buffered soils, losses of N due to ammonia volatilization can occur shortly after fertilizer application, i.e., before mixing with the initial soil mineral N has occurred. If the N is applied in the form of large granules or in the form of slow-release fertilizers it can take even longer before mixing with soil N is complete. Similarly, if surface application of urea is followed by a dry period it may take considerable time before the urea can enter into the soil and mix with soil mineral N.

If only applied N is subject to losses, the situation in unfertilized treatments would remain the same as in the case of plant uptake only (Equations 3 and 4). In the case of fertilized treatments, however, the situation would change as follows:

$$\begin{aligned}
 NP_{fh} &= \epsilon_f \{ NS_i + (1 - \lambda_{ff}) NF_i \} \\
 NL_{fm} &= \lambda_{ff} NF_i \\
 NS_{fh} &= (1 - \epsilon_f) \{ NS_i + (1 - \lambda_{ff}) NF_i \}
 \end{aligned}
 \tag{12}$$

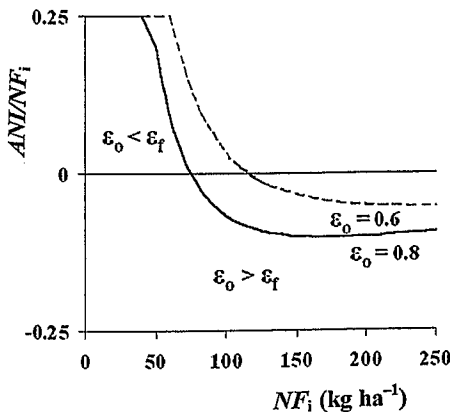


Figure 6. The relative added N interactions ( $ANI/NF_i$ ) calculated for  $NS_i = 50$ ,  $NF_i = 0-250$  kg N  $ha^{-1}$ ,  $NP_{oh}$ ,  $NP_{fs} \leq 100$  kg N  $ha^{-1}$  and two cases: (left) one in which 20% ( $\epsilon_o \leq 0.80$ ) and (right) one in which 40% of the soil mineral N in the unfertilized treatments is inaccessible to the crop ( $\epsilon_o \leq 0.60$ ). In the fertilized treatments, all soil mineral N is assumed to be accessible to the crop.

where  $NL_{fh}$  is the amount of fertilizer-derived N lost from fertilized treatments at harvest ( $\text{kg ha}^{-1}$ ) and  $\lambda_{ff}$  is a loss coefficient for fertilizer-derived N in fertilized treatments at harvest (dimensionless).

From Equations 1, 3 and 12 it follows that:

$$ARF = \epsilon_f(1-\lambda_{ff}) + (\epsilon_f - \epsilon_o)NS_i/NF_i \quad (13)$$

To calculate  $^{15}\text{NRF}$ , Equation 7 has to be corrected for the loss of fertilizer N:

$$c_{xs} = c_{xf}(1-\lambda_{ff})NF_i / \{NS_i + (1-\lambda_{ff})NF_i\} \quad (14)$$

Hence, from Equations 2, 8, 12 and 14:

$$^{15}\text{NRF} = \epsilon_f(1-\lambda_{ff}) \quad (15)$$

which shows that in this case  $^{15}\text{NRF}$  is no longer equal to the uptake coefficient for N in fertilized plots ( $\epsilon_f$ ), but smaller by a factor  $1-\lambda_{ff}$ . The relationship between  $^{15}\text{NRF}$  and ARF becomes:

$$^{15}\text{NRF} = (ARF + \epsilon_o NS_i / NF_i) / \{1 + NS_i / (1-\lambda_{ff}) NF_i\} \quad (16)$$

As a consequence, if losses of fertilizer N occur,  $^{15}\text{NRF}$  would be lower by a factor  $1-\lambda_{ff}$  relative to the case of plant uptake only, but ARF would also be lower by a factor  $1-\lambda_{ff}$ , if  $\epsilon_o = \epsilon_f$ . Hence, if  $\epsilon_o = \epsilon_f$ , the entire dataset would be 'condensed', i.e., the surface area reduced by a factor  $(1-\lambda_{ff})^2$ .

In this case,  $^{15}\text{NRF}$  would underestimate the uptake efficiency of fertilizer-derived N from the soil mineral N pool ( $\epsilon_f$ ), but it would correctly estimate the over-all uptake efficiency of fertilizer N if the loss of N is considered part of that uptake efficiency. So it is a matter of definition whether one considers  $^{15}\text{NRF}$  a true reflection of fertilizer uptake efficiency or not. If one compares fertilizer management practices aimed at reducing fertilizer N losses, it seems appropriate to use  $^{15}\text{NRF}$  for an overall-evaluation of such practices. However, to quantify the losses in each situation, one would have to supplement such research on  $^{15}\text{N}$  recoveries with  $^{15}\text{N}$  balance studies. Otherwise it would be difficult to understand why one fertilizer management practice recovers more  $^{15}\text{N}$  than the other, and to be able to extrapolate the results across different crop, soil and management conditions.

Equation 16 is plotted in Figure 7 for different values of  $\lambda_{ff}$ . In this case both the intercept  $[\epsilon_o(NS_i/NF_i) / \{1 + NS_i / (1-\lambda_{ff}) NF_i\}]$  and the slope  $[1 / \{1 + NS_i / (1-\lambda_{ff}) NF_i\}]$  decrease with increasing values of  $\lambda_{ff}$ , at constant  $\epsilon_o$  and  $NS_i/NF_i$ . It follows from Equations 13 and 15 that ARF and  $^{15}\text{NRF} \leq 0.8$  ( $\lambda_{ff} = 0.2$ ) and  $\leq 0.6$  ( $\lambda_{ff} = 0.4$ ).

Values of  $^{15}\text{NRF}$  and ARF were calculated for the system considered earlier (see Figure 4), except for:

$$(1) \epsilon_f = 1.00, \lambda_{ff} = 0.20$$

$$(2) \epsilon_f = 1.00, \lambda_{ff} = 0.40$$



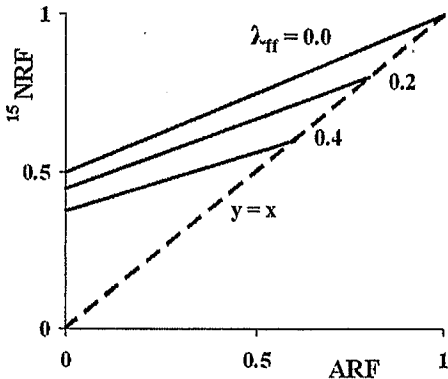


Figure 7. <sup>15</sup>NRF as a function of ARF according to Equation 16, for  $NS_i/NF_i = 1.0$ ,  $\epsilon_o = 1.00$  and 3 values of  $\lambda_{ff}$ : 0.0, 0.20 and 0.40.

Figure 8 includes the data-pairs for which  $NP_{fn} = 100$ ,  $^{15}NRF < (1 - \lambda_{ff})$  and  $ARF > 0$ . It can be seen that  $^{15}NRF, ARF < 0.8$  if  $\lambda_{ff} = 0.2$  and  $^{15}NRF, ARF < 0.6$  if  $\lambda_{ff} = 0.4$ . The intercept decreases with increasing values of  $\lambda_{ff}$ , in accordance with Equation 16. One would also expect the slope to decrease with increasing values of  $\lambda_{ff}$ . However, in Figure 8 this effect appears to be somewhat masked by the variation in the values of  $NS_i/NF_i$ . Nevertheless, the slope of the linear regression equations decreases slightly from 0.404 ( $\lambda_{ff} = 0$ ) to 0.387 ( $\lambda_{ff} = 0.2$ ) and 0.350 ( $\lambda_{ff} = 0.4$ ).

### Plant uptake and loss of soil nitrogen after mixing

The case of loss of soil N, after mixing of initial soil mineral N and fertilizer N is complete, may occur in practice when denitrification or leaching takes place during

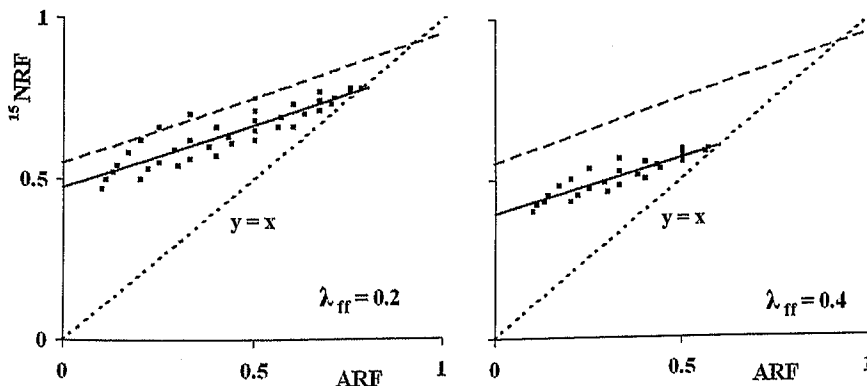


Figure 8. Calculated <sup>15</sup>NRF-values as a function of ARF for  $\epsilon_o = \epsilon_f = 1.00$ ,  $NP_{ob}, NP_{fn} \leq 100 \text{ kg ha}^{-1}$  and 2 values of  $\lambda_{ff}$ : 0.20 (left) and 0.40 (right). The solid lines represent the linear regression equations of <sup>15</sup>NRF on ARF; the broken lines represent the linear regression equation of <sup>15</sup>NRF on ARF for  $\epsilon_o = 1.00$  and  $\epsilon_f = 1.00$  (taken from Figure 4).

the growing season. If losses of N occur from the mixed soil mineral N pool, the N in unfertilized treatments at harvest will be divided over three pools that can be represented by:

$$\begin{aligned} NP_{oh} &= \varepsilon_o(1-\lambda_o)NS_i \\ NL_{oh} &= \lambda_oNS_i \\ NS_{oh} &= (1-\varepsilon_o)(1-\lambda_o)NS_i \end{aligned} \quad (17)$$

where  $NL_{oh}$  denotes the amount of N lost from the unfertilized treatment at harvest ( $\text{kg ha}^{-1}$ ) and  $\lambda_o$  is a loss coefficient for N in unfertilized treatments (dimensionless).

Equation 17 assumes that the crop takes up N at a lower rate than in the case of plant uptake only, as the N concentration in the soil mineral N pool (i.e., soil solution) presumably is lower by a factor  $1-\lambda_o$  because of the losses occurring from this pool. Hence, in N-deficient soils there would be 'competition' between crop uptake and loss of N.

The situation for fertilized treatments may be represented by:

$$\begin{aligned} NP_{fh} &= \varepsilon_f(1-\lambda_f)N^o \\ NL_{fh} &= \lambda_fN^o \\ NS_{fh} &= (1-\varepsilon_f)(1-\lambda_f)N^o \end{aligned} \quad (18)$$

where  $NL_{fh}$  denotes the amount of N lost from fertilized treatments at harvest ( $\text{kg ha}^{-1}$ ) and  $\lambda_f$  is a loss coefficient for N in fertilized treatments (dimensionless).

The way the loss of N from the soil mineral N pool is represented in Equations 17 and 18 is somewhat arbitrary. In deriving these equations it is essentially assumed that (1) there is an interaction between loss of N and plant uptake, i.e., if losses occur plant uptake will be lower, (2) there is no interaction between plant uptake and loss of N, i.e., loss of N is not affected by plant uptake, and (3) losses occur from the entire soil mineral N pool, including any part beyond the reach of the root system. The latter assumption could apply (1) to loss of N through denitrification if it occurs inside soil aggregates where the roots cannot penetrate, or (2) to leaching of nitrate at depth in the soil beyond the reach of the crop's roots.

An alternative way of expressing losses from the mixed soil mineral N pool would be:

$$\begin{aligned} NP_{fh} &= \varepsilon_f(1-\lambda_f)N^o \\ NL_{fh} &= \lambda_f\varepsilon_fN^o \\ NS_{fh} &= (1-\varepsilon_f)N^o \end{aligned} \quad (18a)$$

in which case it is assumed that loss of N is limited to that part of the soil mineral N pool that is accessible to the root system. This would apply to leaching of soluble N in the larger soil pores within the rooting zone or to denitrification occurring in the rhizosphere of the crop. A disadvantage of Equation 18a would be that  $NL_{fh}$  decreases if  $\varepsilon_f$  decreases, if the quantity of N in the soil mineral N pool exceeds the crop's demand, whereas in that case one would expect  $NL_{fh}$  to remain constant or even increase. This

drawback could, in principle, be remedied by using an expression of the type:

$$NL_{\text{th}} = [\lambda_f \{1 - \varepsilon_f (1 - \lambda_f)\}]^{1/2} N^0$$

in which case  $NL_{\text{th}}$  would increase from  $\lambda_f N^0$  at  $\varepsilon_f = 1$  to  $(\lambda_f)^{1/2} N^0$  at  $\varepsilon_f = 0$ .

Finally, losses from the mixed soil mineral N pool could be expressed by:

$$\begin{aligned} NP_{\text{th}} &= \varepsilon_f N^0 \\ NL_{\text{th}} &= \lambda_f N^0 \\ NS_{\text{th}} &= (1 - \varepsilon_f - \lambda_f) N^0 \end{aligned} \tag{18b}$$

in which case  $\varepsilon_f$  and  $\lambda_f$  would have to satisfy the inequality  $0 \leq \varepsilon_f + \lambda_f \leq 1$ . In this case there would be no interaction between plant uptake and loss of N, and one could not distinguish between  $\varepsilon_f$  being smaller than 1 because of inaccessibility of part of the soil mineral N pool to the crop's root system, or because of N losses from the soil mineral N pool. Therefore, the representation of losses as expressed by Equation 18 or 18a is preferred. Equations 18 and 18a result in the same equations for ARF and  $^{15}\text{NRF}$  and the same relationship between  $^{15}\text{NRF}$  and ARF. Therefore the difference between the expressions for  $NL_{\text{th}}$  in Equations 18 and 18a is somewhat academic at this point and the use of either one of them would depend on the presumed loss mechanism and on whether losses occur from the plant-accessible part of the soil mineral N pool or from the entire pool.

If the loss mechanisms involved do not discriminate between N isotopes, Equations 7 and 8 would hold, and ARF and  $^{15}\text{NRF}$  follow from Equations 17 and 18:

$$\text{ARF} = \varepsilon_f (1 - \lambda_f) + \{\varepsilon_f (1 - \lambda_f) - \varepsilon_o (1 - \lambda_o)\} NS_i / NF_i \tag{19}$$

$$^{15}\text{NRF} = \varepsilon_f (1 - \lambda_f) \tag{20}$$

and the relationship between  $^{15}\text{NRF}$  and ARF becomes:

$$^{15}\text{NRF} = \{\text{ARF} + \varepsilon_o (1 - \lambda_o) NS_i / NF_i\} / (1 + NS_i / NF_i) \tag{21}$$

Hence, the case of plant uptake and loss of N from the mixed soil mineral N pool yields essentially the same results as the case of plant uptake only (Equations 9–11), if  $\varepsilon_o (1 - \lambda_o)$  is substituted for  $\varepsilon_o$  and  $\varepsilon_f (1 - \lambda_f)$  for  $\varepsilon_f$  in the relevant equations. Equation 21 is plotted in Figure 9 for different values of  $\lambda_o$  and  $\lambda_f$ . The intercept and slope are given by:

$$\text{intercept} = \{\varepsilon_o (1 - \lambda_o) NS_i / NF_i\} / (1 + NS_i / NF_i)$$

$$\text{slope} = 1 / (1 + NS_i / NF_i)$$

Hence, the intercept decreases with increasing values of  $\lambda_o$ . The intercept and slope are not directly affected by  $\lambda_f$ , but this coefficient does affect  $^{15}\text{NRF}$  and ARF in that their numerical values are reduced by a factor  $1 - \lambda_f$  (see Equations 19 and 20).

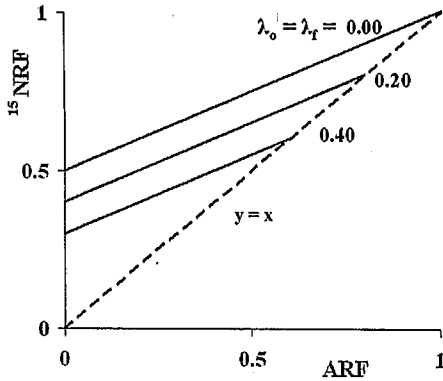


Figure 9.  $^{15}\text{NRF}$  as a function of ARF according to Equation 21, for  $NS_i/NF_i = 1.0$ ,  $\epsilon_o = 1.00$  and 3 values of  $\lambda_o = \lambda_f$  0, 0.20 and 0.40.

Values of  $^{15}\text{NRF}$  and ARF were calculated for the system considered earlier (Figure 4), except for:

- (1)  $NS_i = 10, 20, \dots, 130 \text{ kg ha}^{-1}$   
 $\epsilon_o = 1.00$ ,  $\lambda_o = 0.20$   
 $\epsilon_f = 1.00$ ,  $\lambda_f = 0.20$

and:

- (2)  $NS_i = 10, 20, \dots, 170 \text{ kg ha}^{-1}$   
 $\epsilon_o = 1.00$ ,  $\lambda_o = 0.40$   
 $\epsilon_f = 1.00$ ,  $\lambda_f = 0.40$

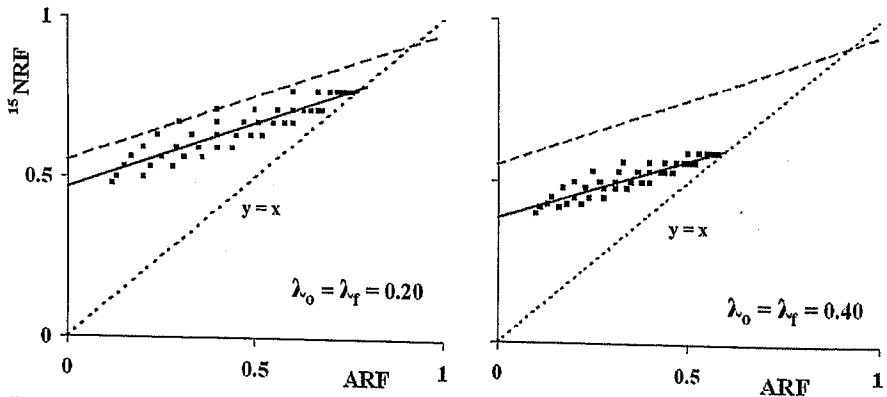


Figure 10. Calculated  $^{15}\text{NRF}$ -values as a function of ARF for  $\epsilon_o = 1.00$ ,  $\epsilon_f = 1.00$ ,  $NP_{oh}$ ,  $NP_m \leq 100 \text{ kg ha}^{-1}$  and 2 values of  $\lambda_o = \lambda_f$  0.2 (left) and 0.4 (right). The solid lines represent the linear regression equations of  $^{15}\text{NRF}$  on ARF, the broken lines represent the linear regression equation of  $^{15}\text{NRF}$  on ARF for  $\lambda_o = \lambda_f = 0$  (taken from Figure 4).

This generates 55 data-pairs for which  $NP_f = 100$ ,  ${}^{15}\text{NRF} < 0.80$  ( $\lambda_o = \lambda_f = 0.20$ ) or  $0.60$  ( $\lambda_o = \lambda_f = 0.40$ ) and  $\text{ARF} > 0$  (Figure 10). In principle, the present dataset is very similar to the dataset that was obtained for Figure 4, except that all values of  $\text{ARF}$  and  ${}^{15}\text{NRF}$  are 'compressed' by a factor 0.8 if  $\lambda_o = \lambda_f = 0.20$  or by a factor 0.60 if  $\lambda_o = \lambda_f = 0.40$ . Figure 10 confirms that the intercept decreases with increasing values of  $\lambda_o$  and that the slope remains approximately constant.

## Discussion

The mixing model considered in the present paper is a strongly simplified representation of a natural soil-plant system. All processes in soil depend on moisture, temperature, nutrient status and other factors, and are functions of space and time, and thus a simple static model does not do justice to the dynamic nature of N interactions in soil. In addition, one would have to distinguish between different forms of mineral N. The question is, however, whether the assumptions are such that conclusions can be drawn that can help us to better understand natural systems.

The model under consideration requires that initial soil mineral N and fertilizer N are completely mixed in a dimensionless 'box' or such that the distribution of N is uniform with depth. In practice such a situation will rarely be encountered, although it may be approached for the top layer of the soil in situations where plant uptake of N starts later in the growing season, several months after fertilizer N has been applied. For example, in the Mediterranean environment rainfed crops are sown in late November–early December and remain in a stage of dormancy during winter. Fertilizer N enters the soil with the winter rains and mixes with initial soil mineral N in the upper part of the soil profile. By early spring, crops enter into a stage of rapid development and start taking up significant amounts of N from the topsoil. Later in spring they start using stored soil moisture (and N) from deeper layers (Garabet, 1995; Harmsen & Garabet, 2003). On the other hand, under low-rainfall, dry farming conditions, fertilizer-derived soil N is likely to concentrate in the upper part of the soil profile such that the lower part is relatively high in soil-derived mineral N. Early in the season, the crop takes up most of its N from the upper part of the soil profile, where  $NS_i/NF_i$  is relatively low. Later in the season, however, when rainfall ceases and the crop starts using stored soil moisture, it takes up N from deeper layers, where  $NS_i/NF_i$  would be relatively high. As a result,  $\text{ARF}$  increases quite strongly during that period, but almost solely because of an increase in  $NP_f$ . In contrast,  ${}^{15}\text{NRF}$  hardly increases during that period, as  $NP_f$  remains practically constant (Garabet, 1995; Harmsen & Garabet, 2003). The conclusion of this might well be that complete mixing of  $NS_i$  and  $NF_i$  in the soil is unlikely to occur under any field conditions, although there may be situations that approach complete mixing in part of the soil profile. Under low-rainfall, dry-farming conditions this is more likely to happen in the topsoil, whereas under high-rainfall or irrigated conditions, mixing could occur over the entire depth of the soil profile. Even though the conditions for applying the mixing model might not be fully met under field conditions, the model could help to understand the dynamics of  $\text{ARF}$  and  ${}^{15}\text{NRF}$  during the season and the differences between treatments.

The assumption that there is only one soil mineral N pool and that all mineral N is equally available to the crop, assumes that  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  behave similarly, that the crop does not distinguish between them and that loss mechanisms are indifferent with regard to the chemical form of mineral N. Most agricultural soils contain both  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  as the major forms of mineral N and although crops may use both forms of mineral N in the course of the growing season, loss mechanisms are definitely specific to either  $\text{NH}_4\text{-N}$  or  $\text{NO}_3\text{-N}$ . For example, ammonia volatilization and ammonium fixation by certain types of clay minerals (and, in fact, immobilization of ammonium by the heterotrophic biomass) are specific to  $\text{NH}_4\text{-N}$ , whereas denitrification and leaching of nitrate are specific to  $\text{NO}_3\text{-N}$ . Therefore, if  $^{15}\text{N}$  is not evenly distributed over the ammonium and nitrate pools, the occurrence of these processes may affect soil- and fertilizer-derived N differently and affect the distribution of  $^{15}\text{N}$  over the ammonium and nitrate pools in soil, thus obscuring the interpretation of the results of  $^{15}\text{N}$  tracer studies. Under such circumstances Equations 7 and 8 would not hold and it would be required to consider the ammonium and nitrate pools separately. If the relative abundance of  $^{15}\text{N}$  in the ammonium and nitrate pools were not constant in time, it might be useful to calculate 'mean pool abundances' of  $^{15}\text{N}$  in these pools (Barraclough *et al.*, 1985; Barraclough, 1991).

The assumption that  $\text{NS}_i$  and  $\text{NF}_i$  mix instantaneously again might not be easily realized in practice. Most commonly  $^{15}\text{N}$  is applied in the form of  $^{15}\text{N}$ -labeled urea [ $(^{15}\text{NH}_4)_2\text{CO}$ ] or an ammonium salt, such as ammonium sulphate [ $(^{15}\text{NH}_4)_2\text{SO}_4$ ] or ammonium nitrate ( $^{15}\text{NH}_4\text{NO}_3$ ). If  $^{15}\text{N}$  is applied in the form of  $^{15}\text{NH}_4\text{-N}$ , the occurrence of ammonia volatilization or fixation of ammonium by certain types of clay minerals can result in the loss of  $^{15}\text{NH}_4\text{-N}$  from the soil mineral N pool, which in turn can affect the interpretation of the results of  $^{15}\text{N}$  tracer studies. In the case of urea, the fertilizer first hydrolyses to form ammonium and then, in most well-aerated soils, it nitrifies upon which nitrate is formed. The rate of urea hydrolysis depends on a number of factors, such as temperature, moisture content and the presence of the enzyme urease. Similarly, the rate of nitrification depends, amongst other things, on temperature, moisture, and the presence of nitrifying organisms and a source of decomposable organic carbon. Therefore, it may take some time before labeled urea- or ammonium-N is nitrified in the soil. So the assumption of complete and instantaneous mixing, resulting in an even distribution of  $^{15}\text{N}$  over the ammonium and nitrate N pools throughout the soil profile, will at best be approximate under most field conditions.

If one limits the discussion to crops that do not derive part of their N from biological N fixation (e.g. rhizobia in association with legume crops) and to systems where the role of free-living N-fixing micro-organisms is limited, then the assumption that the crop derives all of its N from the mineral N pool in the soil seems reasonable. Also, there is no reason to assume that the crop would discriminate between  $^{14}\text{N}$  and  $^{15}\text{N}$  to a significant extent. The slightly higher atom%  $^{15}\text{N}$  concentrations in crops as compared with natural abundance are corrected for by using the measured value of the atom%  $^{15}\text{N}$  in unfertilized crops. Also, loss mechanisms such as nitrate leaching, denitrification and ammonia volatilization would not discriminate to a significant extent between N isotopes

Under field conditions, fertilizer N will infiltrate into the soil with rain or irriga-

tion water. Therefore, some fertilizer N may be lost before mixing has occurred or is fully realized. This means that for losses of N the situation may well be in between the two cases considered: (1) losses from fertilizer N only, and (2) losses from the soil mineral N pool after mixing has occurred. For ammonia volatilization the situation may be closer to case (1) and for leaching of nitrate and denitrification, the situation may be better described by case (2). <sup>15</sup>NRF may underestimate the fertilizer N recovery in the crop of the fertilizer N that actually enters the soil and mixes with the soil pool, if loss of fertilizer N is not accounted for. To some extent this is also a matter of definitions: if one would be interested in the uptake efficiency of fertilizer-derived N from the soil mineral N pool then one would have to correct for losses of N. Alternatively, if one is interested in the fertilizer N under actual field conditions, which includes losses of N, then one would not correct the <sup>15</sup>NRF and accept that losses result in lower recoveries.

It follows from Table 1 that several quantities can lower the intercept of the relationship between <sup>15</sup>NRF and ARF. However, in the case of losses of N, the values of <sup>15</sup>NRF are reduced by a factor  $1-\lambda_{ff}$  or  $1-\lambda_f$ , and in the case of ARF they are reduced by a factor  $1-\lambda_{ff}$  if  $\epsilon_o = \epsilon_f$  or  $1-\lambda_f$  if  $\epsilon_o(1-\lambda_o) = \epsilon_f(1-\lambda_f)$ , such that the situation that  $ARF > ^{15}NRF$  does not occur in the case of losses of N. In the higher range of ARF values, the situation that  $ARF > ^{15}NRF$  can only be caused by  $\epsilon_o < \epsilon_f$  or  $\epsilon_o(1-\lambda_o) < \epsilon_f(1-\lambda_f)$ , in combination with  $NS_i/NF_i > 0$ . Therefore, on the basis of the present mixing model, one would generally expect  $^{15}NRF > ARF$ . The slope of the relationship between <sup>15</sup>NRF and ARF is only affected by  $\lambda_{ff}$  and by the ratio  $NS_i/NF_i$ .

Table 1. The effect of N uptake and loss coefficients and the ratio  $NS_i/NF_i$  on the intercept and slope of the relationship between <sup>15</sup>NRF and ARF, according to Equation 11 in the case of plant uptake only, Equation 16 in the case of loss of fertilizer N prior to mixing with soil mineral N, and Equation 21 in the case of loss of N from the mixed soil mineral N pool. An increase in a quantity is denoted by ++, a decrease by -- whereas +/- denotes either a decrease or an increase.

Quantity	Change	Effect on intercept	Effect on slope	Remarks
$\epsilon_o$	--	--	none	
$\epsilon_f$	+/-	none	none	
$NS_i/NF_i$	++	++	--	
	--	--	++	
$\lambda_{ff}$	++	--	--	reduces ARF and <sup>15</sup> NRF
$\lambda_o$	++	--	none	
$\lambda_f$	+/-	none	none	reduces ARF and <sup>15</sup> NRF

## Conclusions

It is concluded that in the lower range of ARF values,  $^{15}\text{NRF}$  values are likely to be higher than those of ARF, because the fertilizer N mixes with the soil pool and the plant will take up both soil- and fertilizer-derived N from the soil pool in proportion to their relative abundance, even if there is little or no yield response to applied N. In this case the  $^{15}\text{NRF}$  would accurately estimate the N fertilizer recovery fraction.

In the higher range of ARF values,  $^{15}\text{NRF}$  values would only be lower than those of ARF in case of an increased uptake efficiency of soil-derived mineral N in fertilized treatments, i.e.,  $\varepsilon_o < \varepsilon_f$  or  $\varepsilon_o(1-\lambda_o) < \varepsilon_f(1-\lambda_f)$ . In that case, ARF would tend to overestimate the N fertilizer recovery in the crop, as the estimates would include some of the additional soil-derived N, not accessible or available in unfertilized treatments. Except for the effect of isotopic exchange – not dealt with here –  $^{15}\text{NRF}$  would correctly estimate N fertilizer recovery by the crop. In the case of loss of N fertilizer shortly after application,  $^{15}\text{NRF}$  correctly reflects the overall N recovery fraction by the crop, but the loss of N would appear as a lower recovery fraction, i.e., as seemingly lower uptake efficiency rather than as a loss.

From an agronomic point of view, ARF would be preferred, as it is a measure for the overall effect of N fertilizer application on N uptake or yield, even if some of the increased uptake in fertilized treatments may be due to increased soil mineral N uptake. If one is interested in the fate of  $^{15}\text{N}$  itself in soil-plant systems, then obviously the isotope-dilution method is the appropriate methodology.

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## Appendix

### List of symbols

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All quantities of N are in  $\text{kg ha}^{-1}$ , atom%  $^{15}\text{N}$  (excess) concentrations are in % and recovery fractions and N coefficients are dimensionless. Symbols that occur only once are defined in the text and are not repeated here.

ARF	= apparent N recovery fraction at harvest
$c_f$	= atom% $^{15}\text{N}$ concentration in N fertilizer
$c_o$	= atom% $^{15}\text{N}$ concentration in natural N (natural abundance)
$c_{pf}$	= atom% $^{15}\text{N}$ concentration in fertilized crop at harvest
$c_{po}$	= atom% $^{15}\text{N}$ concentration in unfertilized crop at harvest
$c_{xf}$	= atom% $^{15}\text{N}$ excess concentration in N fertilizer
$c_{xp}$	= atom% $^{15}\text{N}$ excess concentration in fertilized crop at harvest
$c_{xs}$	= atom% $^{15}\text{N}$ excess concentration in the soil mineral-N pool
$N^o$	= $NS_i + NF_i$
$NF_i$	= initial N fertilizer applied to the soil
$NL_{fh}$	= N lost from fertilized treatments at harvest
$NL_{oh}$	= N lost from unfertilized treatments at harvest
$NL_{ffh}$	= fertilizer-derived N lost from fertilized treatments at harvest
$NP_{fh}$	= N uptake by fertilized crop at harvest
$NP_{oh}$	= N uptake by unfertilized crop at harvest
$NP_{ffh}$	= uptake of fertilizer-derived N by fertilized crop at harvest
$NP_{fsh}$	= uptake of soil-derived N by fertilized crop at harvest
$NS_i$	= initial soil mineral N
$NS_{fh}$	= soil mineral N in fertilized treatments at harvest
$NS_{oh}$	= soil mineral N in unfertilized treatments at harvest
$^{15}\text{NRF}$	= fertilizer N recovery fraction in the crop at harvest
$\epsilon_f$	= uptake coefficient for N in fertilized treatments at harvest
$\epsilon_o$	= uptake coefficient for N in unfertilized treatments at harvest
$\epsilon_{ff}$	= uptake coefficient for fertilizer-derived N in fertilized treatments at harvest
$\epsilon_{fs}$	= uptake coefficient for soil-derived N in fertilized treatments at harvest
$\lambda_f$	= loss coefficient for N from fertilized treatments
$\lambda_o$	= loss coefficient for N from unfertilized treatments
$\lambda_{ff}$	= loss coefficient for fertilizer-derived N from fertilized treatments