

Synthetic Nitrogen Fertilizers Deplete Soil Nitrogen: A Global Dilemma for Sustainable Cereal Production

R. L. Mulvaney,* S. A. Khan, and T. R. Ellsworth University of Illinois

Cereal production that now sustains a world population of more than 6.5 billion has tripled during the past 40 yr, concurrent with an increase from 12 to 104 Tg yr⁻¹ of synthetic N applied largely in ammoniacal fertilizers. These fertilizers have been managed as a cost-effective form of insurance against low yields, without regard to the inherent effect of mineral N in promoting microbial C utilization. Such an effect is consistent with a net loss of soil organic C recently observed for the Morrow Plots, America's oldest experiment field, after 40 to 50 yr of synthetic N fertilization that substantially exceeded grain N removal. A similar decline in total soil N is reported herein for the same site and would be expected from the predominantly organic occurrence of soil N. This decline is in agreement with numerous long-term baseline data sets from chemical-based cropping systems involving a wide variety of soils, geographic regions, and tillage practices. The loss of organic N decreases soil productivity and the agronomic efficiency (kg grain kg⁻¹ N) of fertilizer N and has been implicated in widespread reports of yield stagnation or even decline for grain production in Asia. A major global evaluation of current cereal production systems should be undertaken, with a view toward using scientific and technological advances to increase input efficiencies. As one aspect of this strategy, the input of ammoniacal N should be more accurately matched to crop N requirement. Long-term sustainability may require agricultural diversification involving a gradual transition from intensive synthetic N inputs to legume-based crop rotations.

SINCE the Green Revolution of the 1960s, substantial increases in cereal production have allowed an ongoing rise in world population, which now exceeds 6.5 billion (United Nations, 2006). The gain in agricultural productivity has been accomplished with the introduction of modern crop production practices that rely on high-yielding varieties and heavy inputs of fertilizers and pesticides. This approach is solely directed toward maximizing grain yield, without regard to long-term impacts on the soil resource that is crucial for sustainable cereal production.

Nitrogen is the most important mineral nutrient for cereal production, and an adequate supply is essential for high yields, especially with modern cultivars. Consequently, a dramatic escalation has occurred in global consumption of synthetic N, from 11.6 Tg in 1961 to 104 Tg in 2006 (FAO, 2009). This N is applied largely in the form of ammoniacal fertilizers produced via the Haber-Bosch process, an energy-intensive conversion of highly inert N₂ to highly reactive NH₃ that relies on natural gas for process energy and as a source of H₂ (Smil, 2001). Faced with rising energy costs and concomitant price increases for N fertilizers, grain producers are under growing pressure to maximize fertilizer N uptake efficiency (FNUE), defined herein as 100 × (fertilized yield – unfertilized yield) × grain N concentration/fertilizer N applied.

In many parts of the world, N fertilizer recommendations continue to follow a prescriptive approach using generic models of economic response, often without regard to site-specific variations in crop N requirement (Meisinger et al., 2008). These recommendations have been widely advocated by public institutions and the private sector as a cost-effective form of insurance against yield loss from N limitation (e.g., Aref et al., 1997; Fernández et al., 1998; Cui et al., 2008; Liang et al., 2008) and were deemed satisfactory as long as fertilizer N remained relatively inexpensive. The resulting N application rates, directly or indirectly subsidized in major markets of the world at considerable public expense (e.g., Narayan and Gupta, 1991; Bansil, 2004; Chanda, 2007; Hafiez, 2008; Liao, 2008), often exceed crop N requirement because (i) soil N availability is not adequately accounted for and (ii) fertilizer form, placement, or timing is unlikely to synchronize with soil

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*Corresponding author (lesssoiln@gmail.com).

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677 S. Segoe Rd., Madison, WI 53711 USA

R.L. Mulvaney, S.A. Khan, and T.R. Ellsworth, Dep. of Natural Resources and Environmental Sciences, Turner Hall, 1102 S. Goodwin Ave., Univ. of Illinois, Urbana, IL 61801. This study was performed with partial funding under Projects ILLU-875-375 and ILLU-875-397, Illinois Agric. Exp. Stn., and with additional support generated through the ¹⁵N Analysis Service.

Abbreviations: EONR, economically optimum nitrogen rate; FNUE, fertilizer nitrogen uptake efficiency; HNPk, high nitrogen–phosphorus–potassium fertilizer; ISNT, Illinois soil nitrogen test; SEM, standard error of mean; SOC, soil organic carbon.

and crop N dynamics (e.g., Lory and Scharf, 2003; Ladha et al., 2005; Cui et al., 2008). Not surprisingly, global FNUE has been estimated at only 33 to 36% for cereal grain production (Raun and Johnson, 1999; Ladha et al., 2005), in which case the unutilized N would represent an annual economic loss of more than \$90 billion, assuming the use of urea at \$650 per Mg (Fertilizer Market Bulletin, 2008). A substantial proportion of this N would be subject to loss, with environmental consequences from NO_3^- pollution of the hydrosphere and greenhouse gas emission into the atmosphere.

Agricultural leaching losses of NO_3^- have toxicological implications for animals and humans (e.g., Camargo and Alonso, 2006) and have generated public concern over escalating costs of compliance with municipal water quality standards. For the producer, NO_3^- leaching represents an economic loss, not only of a valuable nutrient, but also of the exchangeable bases that serve as counterions (e.g., Brye and Norman, 2004). The magnitude of this loss on a regional scale has been well documented for the Mississippi River Basin, with an annual average load of approximately 0.86 Tg of NO_3^- -N between 1996 and 2005 (Aulenbach et al., 2007), nearly threefold higher than the average flux between 1956 and 1965 (Goolsby and Battaglin, 2001) and attributed largely to N fertilization (Mississippi River/Gulf of Mexico Watershed Nutrient Task Force, 2008). Although the impact on the producer is primarily economic, there are broader ramifications, notably the growing global occurrence and extent in coastal waters of eutrophication, hypoxia, and denitrification (e.g., Turner and Rabalais, 1994; Naqvi et al., 2000; Diaz, 2001; Smith, 2003; Turner et al., 2008). Under such conditions, aquatic NO_3^- concentrations can be subject to rapid depletion and thus are not necessarily a reliable indicator of hypoxia (Naqvi et al., 2000) or riverine NO_3^- loading (Laursen and Seitzinger, 2004).

The modern era of synthetic N fertilization coincides with increasing rates of fossil fuel consumption and atmospheric enrichment by CO_2 and N_2O . Using a conversion factor of 1.2 kg C kg⁻¹ N (Schlesinger, 2000) for estimation of CO_2 produced during the manufacture and transport of N fertilizers, annual emissions currently exceed 458 Tg of CO_2 , representing approximately 5% of natural gas consumption (Smil, 2001). An additional 60 Tg of CO_2 are produced when liming neutralizes soil acidity generated from nitrification of NH_4^+ -N, assuming the 2005 global input of 70.9 Tg of NH_4^+ -N (FAO, 2009) and net production of 0.84 kg of CO_2 kg⁻¹ of N applied (Snyder et al., 2007). Upon conversion to NO_2^- or NO_3^- , excessive fertilizer N becomes subject to denitrification and thereby contributes to terrestrial emissions of N_2O , which have been found to increase with the rate of N fertilization (e.g., Eichner, 1990; McSwiney and Robertson, 2005; Parkin and Kaspar, 2006; Wagner-Riddle et al., 2007; Schlesinger, 2008). Further evidence of the latter trend has been provided by global budgets indicating a 3 to approximately 5% conversion of fertilizer N to N_2O -N (Crutzen et al., 2008), identified as an important factor in atmospheric concentrations that have risen at a rate of 0.8 nL L⁻¹ yr⁻¹ since 1980 (Prather et al., 2001). This enrichment has raised concern because N_2O is a potent greenhouse gas with nearly 300 times the global warming potential per unit weight of CO_2 (Ramas-

wamy et al., 2001) and has been implicated in stratospheric ozone depletion (Hahn and Crutzen, 1982).

The foregoing provides ample reason for concern about the impact on water and air quality of increasingly massive inputs of synthetic N, totaling 2727 Tg between 1961 and 2006 (FAO, 2009). The primary effect, however, is on the soil resource that is fundamental to food and fiber production, which in turn sustains economic prosperity, political stability, and ultimately civilization itself. For the sake of agricultural productivity and ecosystem stability, special attention must be given to soil organic matter because of its key role in maintaining soil aggregation and aeration, hydraulic conductivity and water availability, cation-exchange and buffer capacity, and the supply of mineralizable nutrients. The assertion has often been made that synthetic N fertilization maintains or increases soil organic C (SOC) by enhancing the production of crop residues (e.g., Melsted, 1954; Odell et al., 1984; Mitchell et al., 1991; Havlin et al., 2005). Yet the opposite effect was reported long before the modern era of chemical-based N management (White, 1927; Albrecht, 1938), which is fully consistent with evidence that mineral N enhances microbial decomposition of plant residues (e.g., Starkey, 1924; Waksman and Tenney, 1928; Tóth, 1977; Reinertsen et al., 1984; Schnürer et al., 1985; Green et al., 1995; Recous et al., 1995; Neff et al., 2002; Mack et al., 2004; Conde et al., 2005; Pikul et al., 2008; Poirier et al., 2009). Such evidence is likewise consistent with the decline of SOC we previously reported in a paper by Khan et al. (2007) that documented this trend for numerous baseline data sets involving nitrogen-phosphorus-potassium (NPK) fertilization and a wide variety of geographic regions, cropping systems, and tillage practices.

Given the fundamental coupling of microbial C and N cycling, the dominant occurrence of both elements in soil organic forms, and the close correlation between soil C and N mineralization (Dou et al., 2008), the loss of SOC has serious implications for the storage of soil N. There is good reason for concern about sustaining world food production because crop N uptake originates largely from the soil rather than fertilizer, according to considerable evidence from ¹⁵N-tracer (e.g., Olson et al., 1979; Norman et al., 1992; Schindler and Knighton, 1999) and N-response studies (e.g., Blackmer et al., 1992; Lory and Scharf, 2003). Using long-term cropping experiments with baseline data and detailed records of crop and soil management, the present paper examines the effect of synthetic N fertilization on total and mineralizable soil N, soil N loss, and cereal grain production. Further insight regarding the efficiency of fertilizer N management was gained by computing FNUE for on-farm and long-term N response data sets.

Materials and Methods

Experimental Investigation

The long-term effect of synthetic N fertilization on changes in soil N content was evaluated by comparing total-N analyses of surface (0–15 cm) and subsurface (15–30 and 30–46 cm) samples collected in 1955 and 2005 from the Morrow Plots, America's oldest experimental field located on the University

of Illinois campus at Urbana-Champaign. These samples represent 9 of the 12 subplots that comprise the west half of the original experimental area, currently designated as NA (unfertilized since 1876), NB (fertilized since 1955 with urea, triple superphosphate, and KCl), and SA (heavily fertilized since 1967 with urea, triple superphosphate, and KCl following manure, limestone, and rock phosphate application from 1904 to 1966) within main plots cropped to continuous corn (*Zea mays* L.), a corn–soybean (*Glycine max* L. Merr.) rotation, or a corn–oats–alfalfa (*Medicago sativa* L.) hay rotation. Samples collected in 1955 had been stored as a single composite of at least five cores, whereas in 2005, triplicate samples were collected but not composited. Complete details regarding the Morrow Plots and their management, as well as processing and archival storage of soil samples, can be found in Khan et al. (2007).

Total-N analyses were performed in triplicate on approximately 0.2 g of soil by a permanganate-reduced Fe modification of a semimicro-Kjeldahl procedure (Bremner, 1996) using a Tecator Model 1016 digester and a Model 1012 autostep controller (Foss Tecator AB, Höganäs, Sweden) with a three-step program: Step 1, 90°C (ramp time, 15 min; plateau time, 45 min); Step 2, 250°C (ramp time, 15 min; plateau time, 2 h); Step 3, 350°C (ramp time, 15 min; plateau time, 5 h). The quantity of N in the digest was determined by acidimetric titration of NH_3 liberated on diffusion with 10 mol L^{-1} NaOH in a 473-mL (1-pint), wide-mouth glass Mason jar, following the procedure of Stevens et al. (2000) modified for analysis of the entire digest (^{15}N Analysis Service, 2000). To ensure the validity of soil N concentrations determined for 1955 and 2005, all determinations were performed jointly by the first and second authors, using samples that had been ground to <0.15 mm and weighed to 0.1 mg. Whenever the coefficient of variation exceeded 2%, the analyses were repeated.

To quantify the mass storage of soil N, total N concentrations were multiplied by the corresponding bulk density, obtained from direct measurements in 2005, or by using pedo-transfer functions to estimate 1955 values. A more detailed description is available in Khan et al. (2007).

Changes in potentially mineralizable soil N between 1955 and 2005 were estimated using the Illinois soil N test (ISNT) originated by Khan et al. (2001), which was performed in triplicate for all three sampling depths under investigation. Sample size was reduced from 1 to 0.5 g to minimize the amount of soil used, but otherwise the test procedure followed the protocol specified in a technical note (^{15}N Analysis Service, 2004).

Estimation of Crop Nitrogen Removal

Morrow Plot yield records in combination with literature-derived harvest index values provided the basis for estimating stover removal from the NA and SA subplots between 1955 and 1966, whereas stover was returned to the NB subplots during this period and to all subplots beginning in 1967 (see Khan et al., 2007). The corresponding N removals, and those for hay and grain harvested from 1955 to 2005, were calculated assuming textbook values for crude protein (Martin et al., 1976) and a protein N concentration of 160 g kg^{-1} .

Statistical Analyses

Data obtained for total N concentration and mass storage were analyzed statistically with PROC MIXED in SAS (SAS Institute, 2008), using the 2005 data to provide an appropriate variance for estimating standard error of mean (SEM) values obtained as five-core composites from 1955. The step-down Bonferroni adjustment of *P* values (Hochberg, 1988) was performed with PROC MULTTEST in SAS for multiple comparison tests of soil N concentration and mass differences and for evaluating ISNT changes between 1955 and 2005.

Evidence from Other Fertilizer Trials

To assess the fundamental interaction of soil and fertilizer N in a broader context, an extensive effort was made in compiling baseline sets of soil total N data from published cropping experiments with synthetic N fertilization. These data sets encompass an array of geographic regions, soil types, cropping systems, different N fertilizers, and a wide range of N application rates. In most cases, the same technique was used throughout the study period for total N analysis, which minimizes analytical artifacts that can arise from a difference in methodology (Mulvaney, 2008).

Results and Discussion

Changes in Soil Nitrogen Concentrations over Time

Historical yield records for the Morrow Plots reveal a dramatic and progressive increase in corn production since the shift to commercial fertilization in the 1950s, which has also been made possible by high populations of high-yielding hybrids and by ongoing improvements in other cultural practices. The use of synthetic N eliminated a major elemental constraint with respect to enriching the soil stock of organic C and N and was therefore assumed to contribute to the maintenance of soil fertility for sustained agricultural productivity, yet no evidence of soil C sequestration was found by Khan et al. (2007) after 40 to 50 yr of massive residue C inputs and synthetic N fertilization that exceeded corn grain N removal by 60 to 190%.

The excessive input of fertilizer N might logically be expected to build up soil organic N because typically 10 to 50% of the N applied is retained by microbial immobilization within the first year after fertilization, according to numerous ^{15}N -tracer studies (e.g., Kundler, 1970; Hauck, 1971; Olson, 1980; Shen et al., 1989; Balabane and Balesdent, 1992; Reddy and Reddy, 1993; Schindler and Knighton, 1999; Stevens et al., 2005a). Even at the lower limit of this range, synthetic N fertilization for 40 to 50 yr would be predicted to substantially increase total soil N within the Morrow Plots. These increases should be evident in Table 1, which compares total N concentrations for three depth intervals (0–15, 15–30, and 30–46 cm) sampled before and after a half century of continuous cropping with or without repeated NPK fertilization. Instead of accumulating, soil N declined significantly in every subplot sampled, according to the data in Table 1. These declines were more common for the subsurface soil (15–30 or 30–46 cm) than for the plow layer and were most extensive where manuring had been replaced by high NPK

Table 1. Effect of nitrogen–phosphorus–potassium fertilization on total soil nitrogen concentrations measured for the Morrow Plots between 1955 and 2005.

Rotation†	Fertilizer treatment‡	Sampling depth cm	Total soil N§			Net change in 51 yr¶	
			2005				
			1955	Mean	SD		
			g kg ⁻¹				
C-C	none	0–15	1.376	1.168	0.007	-0.208**	
		15–30	1.342#	1.068	0.008	-0.274***	
		30–46	1.020	0.996	0.006	-0.024	
	NPK	0–15	1.376	1.268	0.008	-0.108	
		15–30	1.342#	1.210	0.010	-0.132***	
		30–46	1.020	0.751	0.006	-0.269**	
	HNPK	0–15	1.534	1.421	0.014	-0.113	
		15–30	1.568	1.371	0.010	-0.197***	
		30–46	1.476	1.026	0.008	-0.450***	
C-O(S)	none	0–15	1.487	1.412	0.014	-0.075	
		15–30	1.638	1.312	0.008	-0.326***	
		30–46	1.460	1.284	0.009	-0.176*	
	NPK	0–15	1.487	1.500	0.013	+0.013	
		15–30	1.638	1.429	0.007	-0.209***	
		30–46	1.460	1.248	0.010	-0.212*	
	HNPK	0–15	1.890	1.449	0.005	-0.441***	
		15–30	1.707	1.426	0.008	-0.281***	
		30–46	1.408	1.061	0.010	-0.347***	
	C-O-H	none	0–15	1.695	1.688	0.008	-0.007
			15–30	1.661	1.526	0.014	-0.135***
			30–46	1.461	1.055	0.004	-0.406***
NPK		0–15	1.695	1.681	0.006	-0.014	
		15–30	1.661	1.630	0.010	-0.031	
		30–46	1.461	1.163	0.009	-0.298***	
HNPK		0–15	2.109	1.887	0.005	-0.222***	
		15–30	1.926	1.630	0.004	-0.296***	
		30–46	1.622	1.120	0.009	-0.502***	

*, **, *** Significant at $\alpha = 0.05, 0.01, \text{ or } 0.001$, respectively. Multiple comparison tests of total soil N differences between 1955 and 2005 were performed with PROC MULTTEST in SAS using the step-down Bonferroni adjustment of P values.

† C, corn; H, alfalfa hay; O, oats; S, soybean. Since 1967, the two-crop rotation has involved soybean instead of oats.

‡ NPK, nitrogen–phosphorus–potassium fertilization (168 [1955–1966] or 224 [since 1967] kg N ha⁻¹ for corn, 28 kg N ha⁻¹ for oats) with no amendment before 1955; HNPK, high nitrogen–phosphorus–potassium fertilization (336 [1967–1997] or 224 [since 1998] kg N ha⁻¹ for corn, 28 kg N ha⁻¹ for oats) after manure, limestone, and rock phosphate application from 1904 to 1966.

§ Determined by the Kjeldahl method. SD, standard deviation, which averaged 0.021 g kg⁻¹ for triplicate determinations of 1955 samples.

¶ Obtained by difference. Values expressed as cumulative change rather than as a time-averaged rate, which may not have been constant throughout the study period.

Analyses were performed on a sample collected in 1953, representing the same management practiced in 1955.

(HNPK) fertilization. Both findings are consistent with the corresponding and highly correlated ($R^2 = 0.80^{***}$) declines in SOC reported in Table 1 of Khan et al. (2007) and can thus be attributed to the loss of organic matter.

Table 1 reveals a shift from surface to subsurface N depletion in comparing unfertilized with fertilized soils under continuous corn, and a similar shift is evident for the unfertilized subplots

where corn was grown in a 2- or 3-yr rotation rather than continuously. Higher plant populations, documented by Aref and Wander (1998), can be identified as a key factor in both cases that would have increased rooting density, producing a more extensive rhizosphere that stimulated mineralization through microbial activities further enhanced by favorable temperature and moisture conditions in the subsoil (Rovira and Vallejo, 1997; Herman et al., 2006). There is an obvious implication that soil fertility and organic matter evaluation should not be confined to the plow layer, an expedient practice that has traditionally been followed in production agriculture and is often a major limitation in assessing the long-term impact of production practices (e.g., VandenBygaert and Angers, 2006; Baker et al., 2007; Khan et al., 2007).

To ascertain whether soil N depletion is unique to the six subplots within the Morrow Plots, baseline changes in total soil N were compiled from published field experiments using synthetic N, which encompass a wide range of soil and climatic conditions, cropping systems, and management practices. The resulting database reveals that synthetic fertilization has often been ineffective for preventing soil N depletion, even in cases involving an ample input of N and the incorporation of crop residues (Table 2). This trend is consistent with the declining N content of the Morrow Plots (Table 1), as is the finding of Rasmussen and Parton (1994) that N losses were more extensive for the subsoil than the surface soil.

Even without subsoil data, the detrimental impact of synthetic N fertilization is readily apparent when residues were removed from highly fertile soils, which led to the serious N losses documented in Table 2 for studies by White (1932, 1955), Mattingly et al. (1975), Kirchmann et al. (1994), and Persson and Kirchmann (1994). Substantial soil N losses likewise occurred when synthetic N inputs were used for biomass production of sugarcane (Ribón Carrillo et al., 2003), silage corn (Vitosh et al., 1997; Sainju et al., 2002), or tea (Gokhale, 1959), in which case SOC oxidation was promoted by residue removal. Soil N depletion was no less common with the return of aboveground residues, although a shift toward enrichment was more apt to occur by this practice than by removing residues, which was evident from direct comparisons in Sweden (Kirchmann et al., 1994; Persson and Kirchmann, 1994) and Germany (Stumpe et al., 2000). No such benefit was detected by Campbell and Zentner (1993, 1997) and Rasmussen and Parton (1994) for Mollisols with a high N content or by Vanotti et al. (1997) and Sainju et al. (2002) in studies following several years of cropping to alfalfa. On the contrary, synthetic N fertilization was accompanied by a serious loss of soil N.

In a few cases, soil N content was increased by synthetic N fertilization. These increases, documented in Table 2, are open to question because sampling was usually confined to the surface soil. This is a critical limitation according to Table 1, which shows that subsoil N depletion far exceeded any enrichment of the plow layer observed with NPK or HNPK fertilization of the Morrow Plots. Regarding the surface soil, N accumulation has been observed in the absence of liming to control acidity generated during oxidation of fertilizer-derived $\text{NH}_4^+\text{-N}$ (Mercik et al., 1993; Mihaila and Hera, 1994; Vanotti et al., 1997), with the result that mineralization of organic N is impeded. Waterlogging has a similar effect

Table 2. Continued.

Location	Soil order‡	Cropping system§	Study period	Fertilizer N applied		Sampling depth#	Total soil N††			Reference(s)	
				Form(s)¶	Mean rate		Initial	Final	Net change		
England	Alfisol(s) (sicl)	W _w -Rc-B-Gcl (17)‡‡	1988–2004	8	100	0–20	1.17	1.12	–0.05	Jenkinson and Johnston (1977) Glendining and Powlson (1990) Mattingly et al. (1975) Last et al. (1985) Morel et al. (1984) Schmalfuß (1957) Körschens (1978) Stumpe et al. (2000) van Fassen and Lebbink (1994), Lebbink et al. (1994) Uhlen (1991) Mercik et al. (1993) Mihaila and Hera (1994) Kirchmann et al. (1994), Persson and Kirchmann (1994) Petterson and Wistinghausen (1979) Mattsson (1987) Djokoto and Stephens (1961) Jones (1971) Yan et al. (2007) Zhang et al. (2008) Gokhale (1959) Digar (1958) Mandal et al. (1984) Manna et al. (2005) Kundu et al. (2007), Prakash et al. (2007) Singh et al. (2000) Kundu and Samui (2000) Bhandari et al. (2002) Manna et al. (2005)	
		W _w -Rc-B-Gcl (10)‡‡	1988–2004	8	150	0–20	1.14	1.10	–0.04		
		W _w -Rc-B-Gcl (4)‡‡	1988–2004	8	200	0–20	1.11	1.03	–0.08		
		B-B‡‡	1868–1975	1, 8	48	0–23	1.02	0.98	–0.04		
		W _w -W _w ‡‡	1881–1987	1, 4, 8	48	0–23	1.15	1.13	–0.02		
	Alfisol(s) (sl)	B-B‡‡	1888–1972	1	96	144	0–23	1.26	1.24		–0.02
					46	144	0–23	1.31	1.26		–0.05
					5	46	0–23	1.43	0.63		–0.80
					5	46	0–23	1.43	0.73		–0.70
					5	46	0–23	1.31	0.70		–0.61
Inceptisol(s) (sl)	Sb-W _w -B‡‡	1964–1983	4, 8	75	0–25	1.02	0.99	–0.03			
France	Alfisol(s) (scl)	W _s -Sb§§	1959–1980	1, 4, 5	87	0–25	1.02	1.04	+0.02		
						0–20	1.53	1.29	–0.24		
Germany	Mollisol(s) (sl)	Ry-Ry‡‡	1929–1953	1, 5, 8	40	0–20	0.80	0.94	+0.14		
						0–20	1.49	1.55	+0.06		
	Mollisol(s) (sil)	Sb-B-P-W _w ‡‡	1930–1976	1, 5, 6	65	0–20	1.2(6)	1.0(8)	–0.1(8)		
						0–20	1.1(7)	1.1(5)	–0.0(2)		
Netherlands	Entisol(s) (sil)	W _w -Sb-B-PS§§	1985–1991	5, 6	149	0–20	1.2(6)	1.0(8)	–0.1(8)		
						0–20	1.1(7)	1.0(5)	–0.1(2)		
						0–25	1.2(7)	1.3(0)	+0.0(3)		
						0–25	1.02	1.04	+0.02		
Norway	Inceptisol(s) (cl)	B-O-W _s §§	1966–1984	6	80	0–20	2.9(0)	3.0(2)	+0.1(2)		
						0–20	2.8(2)	2.9(5)	+0.1(3)		
Poland	Spodosol(s) (ls)	Ry-Ry§§	1957–1992	1	74	0–25	0.50	0.46	–0.04		
						0–25	0.52	0.53	+0.01		
						0–25	0.42	0.45	+0.03		
Romania	Mollisol(s) (cl)	C-W _w	1967–1992	4	135	NR	1.55	1.67	+0.12		
Sweden	Inceptisol(s) (cl)	Ce§§	1956–1991	6	107	0–20	4.90¶¶	5.03¶¶	+0.13¶¶		
						0–20	4.90¶¶	4.23¶¶	–0.67¶¶		
						0–20	4.90¶¶	4.21¶¶	–0.69¶¶		
	Alfisol(s) (sicl)	W _s -Gcl-P-Bt	1958–1976	4, 6	28	0–10	2.67	2.45	–0.22		
						25–35	0.90	0.99	+0.09		
						0–10	2.60	2.61	+0.01		
						25–35	0.90	1.24	+0.34		
Africa	Inceptisol(s) (sil)	B-L-Os-W _w -Sb§§	1972–1981	4	86	0–10	2.49	2.61	+0.12		
						25–35	0.90	0.88	–0.02		
						0–20	2.0	1.9	–0.1		
						0–22	0.72	0.67	–0.05		
						0–22	0.77	0.69	–0.08		
						0–30	0.96	0.87	–0.09		
						0–30	0.50	0.50	0		
Asia	Alfisol(s) (cl)	Ce-Y-Gn (21)‡‡	1948–1953	1	27	0–15	0.32	0.26	–0.06		
						0–15	0.32	0.26	–0.06		
India	Ultisol(s) (cl)	Ri-W _w ‡‡	1980–2005	7	300	0–15	1.43	1.68	+0.25		
						0–20	1.07	0.95	–0.12		
	Inceptisol(s) (sl)	W _w -C‡‡	1990–2005	7	300	0–23	1.00	0.84	–0.16		
						0–15	0.83	0.82	–0.01		
	Oxisol(s) (l)	T‡‡	1937–1956	1	90	0–22	0.78	0.66	–0.12		
						0–22	0.72	0.67	–0.05		
	Entisol(s) (c)	Ri-Ri‡‡	1945–1954	1	84	0–22	0.77	0.69	–0.08		
						0–22	0.72	0.67	–0.05		
	Inceptisol(s) (sl)	Ri-W _w -J‡‡	1972–1979	1	50	0–30	0.96	0.87	–0.09		
						0–30	0.96	0.87	–0.09		
Alfisol(s) (scl)	S-W _w §§	1971–2002	7	105	0–15	0.45	0.64	+0.19			
					0–15	0.45	0.64	+0.19			
Inceptisol(s) (sl)	S-W _w ‡‡	1973–2005	7	20	0–15	0.45	0.64	+0.19			
					0–15	0.45	0.64	+0.19			
Mollisol(s) (cl)	Ri-W _w ‡‡	1977–1995	7	80	0–20	1.04	0.71	–0.33			
					0–20	1.04	0.68	–0.36			
Entisol(s) (sl)	Ri-W _w ‡‡	1986–1996	7	100	0–15	0.50	0.52	+0.02			
					0–15	0.56	0.58	+0.02			
Inceptisol(s) (ls)	Ri-W _w ‡‡	1988–1997	7	240	0–15	0.57	0.50	–0.07			
					0–15	0.57	0.50	–0.07			
Vertisol(s) (c)	Sr-W _w ‡‡	1988–2002	7	110	0–15	0.8(3)	0.6(7)	–0.1(6)			
					0–30	0.44	0.49	+0.05			

(cont'd)

Table 2. Continued.

Location	Soil order†	Cropping system‡	Study period	Fertilizer N applied		Sampling depth#	Total soil N††			Reference(s)
				Form(s)¶	Mean rate		Initial	Final	Net change	
Japan	Entisols (c)	Ri-Ri§§	1933–1985	1	105	0–15	2.4(5)	2.6(9)	+0.2(4)	Suzuki et al. (1990)
Philippines	Inceptisols (c)	Ri-Ri (3)§§	1968–1986	7	280	0–20	0.8	0.(9)	+0.(1)	De Datta et al. (1988)
	Mollisols (c)	Ri-Ri (4)§§	1968–1986	7	280	0–20	1.4	1.(8)	+0.(4)	
	Vertisols (c)	Ri-Ri (6)§§	1968–1986	7	280	0–20	1.2	0.(9)	–0.(3)	
Australasia										
Australia	Vertisols (c)	W ₃ -W ₃ (NT)§§	1989–1994	7	50	0–10	0.71	0.74	+0.03	Dalal et al. (1995)
		W ₃ -W ₃ (CT)§§	1989–1994	7	50	0–10	0.74	0.69	–0.05	
Fiji	Oxisols	Sc-Sc (3)††	1978–1983	1	108	0–12	3.1	1.6	–1.5	Masilaca et al. (1986)
						30–40	1.1	1.1	0	

† Most data as reported from fertilization with NPK.

‡ Surface texture designated parenthetically as c (clay), cl (clay loam), l (loam), ls (loamy sand), s (sand), scl (sandy clay loam), silcl (silty clay loam), sil (silt loam), and sl (sandy loam).

§ A, alfalfa (*Medicago sativa* L.); B, barley (*Hordeum vulgare* L.); Bt, beet; C, corn (*Zea mays* L.); Ce, cereal; Cl, clover; C_s, silage corn; CT, conventional tillage; Ct, cotton (*Gossypium hirsutum* L.); Cv, cover crop; Ep, eggplant (*Solanum melongena* L.); F, fallow; Gcl, grass/clover mixture; Gg, gamba grass (*Andropogon gayanus* Kunth); Gn, groundnut (*Vigna subterranea* L. Verdc.); H, hay; J, jute (*Corchorus olitorius* L.); L, ley; MT, mulch tillage; NT, no-tillage; O, oats (*Avena sativa* L.); Os, oilseed; P, potato (*Solanum tuberosum* L.); Rc, root crop; Rcl, red clover (*Trifolium pratense* L.); Ri, rice (*Oryza sativa* L.); Ry, rye (*Secale cereale* L.); S, soybean (*Glycine max* L. Merr.); Sb, sugarbeet (*Beta vulgaris* L.); Sc, sugarcane (*Saccharum officinarum* L.); Sg, small grain; Sr, sorghum (*Sorghum bicolor* L. Moench); T, tea (*Camellia sinensis* L.); Ti, timothy (*Phleum pratense* L.); To, tomato (*Lycopersicon esculentum* L.); W_d, durum wheat (*Triticum turgidum* L.); W_s, spring wheat (*Triticum aestivum* L.); W_w, winter wheat (*Triticum aestivum* L.); Y, yam (*Dioscorea* L.). Subscripted values indicate the number of consecutive years a given crop or crop sequence was grown. Values in parentheses indicate the number of separate trials summarized.

¶ 1, (NH₄)₂SO₄; 2, NH₄H₂PO₄; 3, (NH₄)₂HPO₄; 4, NH₄NO₃; 5, NaNO₃; 6, Ca(NO₃)₂; 7, urea; 8, CaNH₄(NO₃)₃; 9, CaCN₂.

NR, not reported.

†† Parentheses indicate uncertainty in estimating data reported in figures.

‡‡ Aboveground residues removed.

§§ Aboveground residues returned.

¶¶ Values expressed as Mg ha⁻¹.

Initial and final values for total soil N obtained by different methods.

on the latter process and may promote a buildup of N in fertilized paddy soils (De Datta et al., 1988; Suzuki et al., 1990; Kundu and Samui, 2000; Manna et al., 2005; Yan et al., 2007).

Serious consideration must be given to the widespread depletion of soil N documented by Table 2 for studies with high as well as low N rates, which is consistent with the decrease in total soil N reported by Table 1 for Morrow Plots with or without fertilization beyond grain N removal. A loss of soil N is to be expected in the absence of fertilizer N inputs or if fertilizer inputs are exceeded by crop N requirement. On the contrary, this type of interpretation cannot explain why soil N would decline when fertilization supplies more N than the crop removes.

Fertilizer Effects on Soil Nitrogen Mineralization

Such a decline is readily explained on the basis of C and N cycling by heterotrophic soil microorganisms. During residue decomposition, mineral N is immobilized for the synthesis of biomass, producing a labile pool of organic N that exists in equilibrium with a larger and more stable pool associated with humus (e.g., Jansson, 1958; McGill et al., 1981; Jansson and Persson, 1982; Sarawad et al., 2001). This equilibrium is shifted toward immobilization (assimilation) by increasing the input of C relative to N and toward mineralization (decomposition) by increasing the input of N relative to C. The ultimate effect in the latter case is a net loss of organic N (Drinkwater et al., 1998) through profile transport of dissolved organic N (Murphy et al., 2000; van Kessel et al., 2009) or through crop uptake, leaching, or denitrification of NO₃⁻.

The inherent potential of synthetic N for enhancing microbial decomposition is apparent from Table 3, in which a global data set has been compiled to compare net mineralization with and without N fertilization or among different N rates, representing five soil orders, 16 cropping systems, and nine N fertilizers. This data set is remarkably consistent in documenting more rapid mineralization for fertilized than unfertilized soils, and in many cases there was a positive effect from increasing the N rate applied. Table 3 shows that the effect was particularly marked with annual N fertilization for monoculture cropping to corn (Stanford and Smith, 1972; Vanotti et al., 1997; Jordan et al., 2004), wheat (El-Haris et al., 1983; Janzen, 1987; Shen et al., 1989), rye (Garz et al., 1982), or sugarcane (Graham et al., 2002) and for soils cropped to a rice–wheat rotation (Yan et al., 2007). Such findings reveal a dominance of mineralization over immobilization despite a heavy input of highly carbonaceous residues and can only be interpreted within the context that the newly immobilized N does not contribute toward a buildup of nonlabile N in fertilized soils. Rather, Tables 1 and 2 show that synthetic N fertilization often has a negative effect on soil N content, reflecting a shift of native organic N toward the labile pool.

This potentially mineralizable pool plays a critical role in supplying N for crop uptake and can be estimated from soil N liberated by alkaline diffusion or distillation, which is closely correlated with net N mineralization (Cornfield, 1960; Mulvaney et al., 2001; Sharifi et al., 2007; Bushong et al., 2008). The ISNT (Khan et al., 2001) was used for this purpose in our work, so as to compare potentially mineralizable N before and after imposing 51 yr of NPK fertilization on the Morrow

Table 3. Effect of synthetic nitrogen fertilization on soil nitrogen mineralization in long-term cropping experiments.

Location	Soil order†	Cropping system‡	Fertilizer N applied		Net N mineralization¶	Reference					
			Form(s)§	Mean rate kg ha ⁻¹ yr ⁻¹							
North America											
Canada	Mollisols	W _s -W _s #	4	0	1.12	Janzen (1987)					
				45	1.48						
		F-W _s -W _s #	4	0	0.88						
				30	1.09						
USA											
Colorado	Mollisols	W _w -F#	7	0	0.27§§	Kolberg et al. (1999)					
				16	0.28§§						
				32	0.27§§						
				48	0.33§§						
				W _w -C-F#	7		0	0.11§§			
							22	0.14§§			
							45	0.16§§			
									67	0.19§§	
				Georgia	Ultisols		C-C	NR	0	0.2(9)	Stanford and Smith (1972)
									180	0.4(0)	
Missouri	Alfisols	C-C#	4	0	0.8	Jordan et al. (2004)					
Oregon	Mollisols	W _w -F#	4	156	4.1	Rasmussen et al. (1998)					
				0	0.36						
				22	0.45						
				45	0.43						
				0	0.2(4)						
Washington	Mollisols	W _w -W _w #	4	34	0.3(3)	El-Haris et al. (1983)					
				67	0.4(2)						
				135	0.5(7)						
				270	0.7(6)						
Wisconsin	Alfisols	C-C#	4	0	0.34	Vanotti et al. (1997)					
				68	0.31						
				135	0.36						
				271	0.45						
				0	0.30						
				84	0.47						
				168	0.50						
Europe											
Bulgaria	Ultisols	C-W _w	4	0	0.3(8)	Ikonomova et al. (1999)					
				200	0.5(3)						
Czech Republic	Alfisols	W _s -Sb	1	0	0.13	Körschens et al. (1998)					
England	Alfisols	W _w -W _w ††	4	104	0.25	Shen et al. (1989)					
				0	0.26						
				47	0.28						
				94	0.36						
				141	0.42						
				182	0.46						
				1	0.55						
				10	0.48						
France	Alfisols	W _s -Sb††	4	0	0.16	Houot et al. (1987)					
				87	0.19						
Germany	Mollisols	Ry-Ry#	1, 8	0	0.43	Garz et al. (1982)					
				40	0.80						
				0	0.15	Garz and Hagedorn (1990)					
				40	0.20						
	Alfisols	Rc-Ce-Ce	5	0	0.44	Bosch and Amberger (1983)					
			1	72	0.42						
			6	72	0.50						
			9	72	0.40						
	Mollisols	Sb-B-P-W _w ††	8	0	1.18	Peschke et al. (1987)					
			8	128	1.13						
	Mollisols	Sb-B-P-W _w ††	8	0	0.26	Körschens et al. (1998)					
			8	111	0.27						

(cont'd)

Table 3. Continued.

Location	Soil order†	Cropping system‡	Fertilizer N applied		Net N mineralization¶	Reference	
			Form(s)§	Mean rate			
Sweden	Inceptisols	Ce††		0	0.10	Schnürer et al. (1985)	
			6	80	0.12		
	Mollisols (4)	B-Os-W _w -Sb#		6	80	0.08	Bjarnason (1989)
			8	150	0.47		
				0	0.54		
Inceptisols (2)	B-Os-W _w -Sb#		8	150	0.48		
			0	0.64			
Africa	Vertisols	Sc-Sc#		0	1.21§§	Graham et al. (2002)	
			1	140	1.80§§		
Asia							
China	Ultisols	Ri-W _w ††		0	0.1(0)	Yan et al. (2006)	
			7	315	0.1(1)		
				405	0.1(5)		
				495	0.1(5)		
				585	0.1(7)		
	Ultisols	Ri-W _w ††		0	0.69	Yan et al. (2007)	
			7	300	0.91		
India	Inceptisols	C-W _w -Cp††		7	300	0.86	Kanchikerimath and Singh (2001)
				0	0.91		
			7	130	1.10		
	Inceptisols	Ri-W _w ††			260	1.48	Tirol-Padre et al. (2007)
					390	1.74	
			3, 7	0	0.56		
				240	0.53		
		Ri-W _w ‡‡		240	0.83		

† Values in parentheses indicate the number of separate trials summarized.

‡ B, barley; C, corn; Ce, cereal; Cp, cowpea (*Vigna unguiculata* L.); F, fallow; Os, oilseed; P, potato; Rc, root crop; Ri, rice; Ry, rye; Sb, sugarbeet; Sc, sugarcane; W_s, spring wheat; W_w, winter wheat.

§ 1, (NH₄)₂SO₄; 3, (NH₄)₂HPO₄; 4, NH₄NO₃; 5, NaNO₃; 6, Ca(NO₃)₂; 7, urea; 8, CaNH₄(NO₃); 9, CaCN₂; 10, KNO₃; NR, not reported.

¶ Estimated by incubation. Parentheses indicate uncertainty in estimating data reported in figures.

Aboveground residues returned.

†† Aboveground residues removed.

‡‡ Wheat straw returned.

§§ Expressed as kg ha⁻¹ d⁻¹.

Plots. The results (Fig. 1) show a substantial decline that became progressively more serious with increasing applications of synthetic N that averaged, on an annual basis, 224 kg ha⁻¹ for continuous corn, 112 kg ha⁻¹ for a corn-soybean rotation, and 76 kg ha⁻¹ for a corn-oats-hay rotation. In each case, the decline in potentially mineralizable N was more extensive for the subsoil than the plow layer and was also more extensive than the corresponding decline in total N (Table 1). The latter finding is consistent with reports that the impact of synthetic N fertilization is more pronounced for labile soil N than for the passive N pool (Campbell and Souster, 1982; Shevtsova et al., 2003). There are serious implications for agricultural productivity and sustainability because crop N uptake is often greater from the soil than fertilizer, according to numerous ¹⁵N recovery studies with corn (e.g., Bigeriego et al., 1979; Blackmer and Sanchez, 1988; Balabane and Balesdent, 1992; Schindler and Knighton, 1999; Stevens et al., 2005b), wheat (e.g., Olson et al., 1979; Carranca et al., 1999; Tran and Tremblay, 2000; López-Bellido et al., 2006), barley (e.g., Vos et al., 1993; Glendining et al., 1997), and rice (e.g., Norman et al., 1992; Luong et al., 2002).

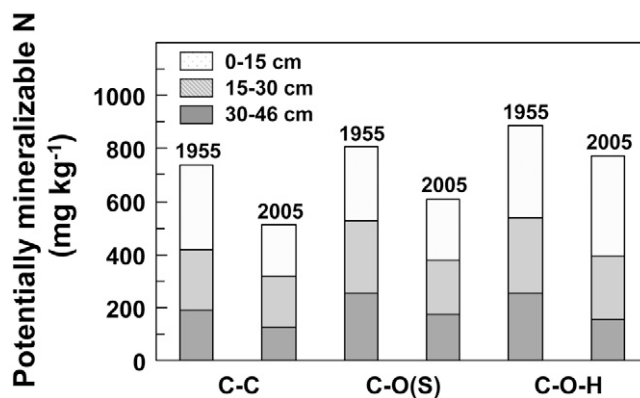


Fig. 1. Potentially mineralizable soil N before and after 51 yr of nitrogen-phosphorus-potassium fertilization of previously unfertilized Morrow Plots cropped to continuous corn [C-C], a corn-oats (1876–1966) or corn-soybean (since 1967) rotation [C-O(S)], or a corn-oats-alfalfa hay rotation [C-O-H]. Fertilizer N was applied as urea to corn (168 [1955–1966] or 224 [since 1967] kg N ha⁻¹) and oats (28 kg N ha⁻¹). Potentially mineralizable N was estimated by the Illinois soil N test. Every change was significant at $\alpha = 0.001$ by the step-down Bonferroni procedure, and all were negative, except for the 0- to 15-cm depth in the C-O-H rotation.

Implications for Fertilizer Nitrogen Uptake Efficiency

The inherent value in substituting biologically fixed N for synthetic fertilization is documented by Table 4, which compares N balance sheets constructed for Morrow Plots where corn was grown continuously or following soybean or alfalfa. Remarkably, the N balance was substantially negative when the input of fertilizer N far exceeded grain N removal by continuous corn, whereas a positive N balance usually occurred for the two rotations, in which case cropping removed more N than was applied by fertilization. More importantly for the producer, Table 4 shows that corn yields have been reduced considerably with monoculture cropping, as compared to either rotation. This reduction has occurred despite a considerably greater input of fertilizer N (Table 4) and residue C (Khan et al., 2007) and persists when the N rate was increased from 224 (NPK treatment) to 336 (HNPK treatment) kg N ha⁻¹. Regardless, corn yields decreased in the order corn–oats–hay > corn–soybean > continuous corn, which is consistent with rotational differences in total soil N (Tables 1 and 4) and potentially mineralizable N (Fig. 1) as well as SOC (Khan et al., 2007). Such conformity is what would be expected if crop N uptake originates largely from the soil rather than fertilizer but also reflects the numerous physical, chemical, and biological benefits associated with SOC.

If the deleterious impact of synthetic N fertilization on soil N storage is to be minimized, the logical emphasis would be on

strategies to improve FNUE. The need for such improvement is evident for corn grain production at the Morrow Plots, according to FNUE values of 20 to 36% that were higher for continuous corn than for the two rotations (Table 4). This difference, also reflected in the apparent percentage of N derived from fertilizer, can be attributed to a higher soil content of total (Table 1) and labile (Fig. 1) N for the legume-based cropping systems, with the result that fertilizer N was subject to more extensive dilution by mineralized soil N. Such dilution would have been promoted by fertilizer-induced mineralization (Table 3) that was intensified with increasing N application rate or soil N reserves. Both factors affect FNUE in Table 4, which also documents the usual trend toward lower FNUE with heavy N fertilization.

The foregoing findings from the Morrow Plots emphasize that excessive N fertilization has a detrimental impact on FNUE and does not promote soil N storage. Given the world's extensive inputs of synthetic N for cereal grain production, further evidence of the same consequences should be available for other areas where long-term cropping experiments have been conducted. This is verified by Table 5, which summarizes several such experiments documenting grain yield increases from N fertilization, usually accompanied by a substantial increase in total net N loss, which was directly related to the quantity of N applied in studies by Dyke et al. (1983), Rasmussen and Parton (1994), Vanotti et al. (1997), Samui et al. (1998), and Kundu and Samui (2000).

Table 4. Efficiency of nitrogen–phosphorus–potassium fertilization for increasing corn grain production on the Morrow Plots, as related to soil nitrogen storage and loss and crop nitrogen removal.

Rotation†	Treatment‡	Fertilizer N input (1955–2005)‡ Mg ha ⁻¹	Average corn grain yield§ Mg ha ⁻¹ yr ⁻¹	Apparent		Total soil N††				Estimate for 1955–2005		
				NDFFF¶	FNUE#	0–15 cm		0–46 cm		Soil N loss‡‡	Crop N removal§§	Apparent N balance¶¶
				%		Mg ha ⁻¹						
C-C	none	0	2.9			3.1	2.6	8.5	7.6	0.9**	2.1	1.2
	NPK	10.8	8.6	66	36	3.1	2.9	8.5	7.3	1.2***	5.8	-6.2
	HNPK	12.5	9.2	69	28	3.3	3.2	9.9	8.7	1.2***	6.2	-7.5
C-O(S)	none	0	5.3			3.2	3.2	10.0	9.2	0.8**	6.1	5.3
	NPK	5.7	10.4	49	32	3.2	3.2	10.0	9.3	0.7*	10.0	3.6
	HNPK	6.5	10.5	50	23	3.9	3.2	10.6	8.8	1.8***	9.6	1.3
C-O-H	none	0	6.5			3.6	3.6	10.4	9.3	1.1**	2.4	1.3
	NPK	4.1	11.7	44	33	3.6	3.6	10.4	9.5	0.9**	5.4	0.4
	HNPK	4.8	11.0	41	20	4.1	3.9	11.5	9.7	1.8***	6.4	-0.2

*, **, *** Significant at $\alpha = 0.05, 0.01, \text{ and } 0.001$, respectively.

† C, corn; H, alfalfa hay; O, oats; S, soybean. Since 1967, the two-crop rotation has involved soybean instead of oats. NPK, nitrogen–phosphorus–potassium fertilization (168 [1955–1966] or 224 [since 1967] kg N ha⁻¹ for corn, 28 kg N ha⁻¹ for oats) with no amendment before 1955; HNPK, high nitrogen–phosphorus–potassium fertilization (336 [1967–1997] or 224 [since 1998] kg N ha⁻¹ for corn, 28 kg N ha⁻¹ for oats) after manure, limestone, and rock phosphate application from 1904 to 1966.

‡ Cumulative totals for the 51-yr study period. Values reported for the HNPK treatment include estimated inputs of manure N between 1955 and 1966 on the basis of the assumption that the manure contained 4.5 g N kg⁻¹ (Mullins et al., 2005).

§ Yield data were averaged for 7 yr between 1967 and 2005 when corn was grown in all three rotations and are reported at market standard moisture content (155 g kg⁻¹).

¶ NDFFF, nitrogen derived from fertilizer. Estimated from corn yield data as $100 \times (\text{fertilized yield} - \text{unfertilized yield}) / \text{fertilized yield}$, assuming that grain N concentration was unaffected by fertilization.

FNUE, fertilizer nitrogen uptake efficiency. Estimated from corn yield data as $100 \times (\text{fertilized yield} - \text{unfertilized yield}) \times \text{grain N concentration} / \text{fertilizer N rate}$, where grain N concentration was calculated assuming 90 g protein kg⁻¹ grain (the value reported by Martin et al. [1976] was adjusted to a moisture content of 155 g H₂O kg⁻¹) and 160 g N kg⁻¹ protein.

†† Total N data reported in Table 1 were converted to soil N mass per hectare using bulk density data collected after the 2005 growing season.

‡‡ Obtained by difference for 0–46 cm.

§§ Estimated as the total amount of N removed in the harvested portion of each crop grown, using textbook values for crude protein (Martin et al., 1976) and assuming 160 g N kg⁻¹ protein.

¶¶ Estimated as crop N removal – (fertilizer N input + soil N loss).

Further examination of Table 5 reveals the same pattern noted previously for the Morrow Plots (Table 1) and elsewhere (Table 2) in that a net loss of soil N often occurred with synthetic N fertilization. There are, however, indications that the detrimental effect becomes less serious when NO₃⁻ accounts for an increasing proportion of the N applied. Of particular interest is the study by Dyke et al. (1983) using CaNH₄(NO₃)₃,

which shows a progressive shift from a net loss to a net gain of soil N as fertilization was increased. In contrast, the opposite trend occurred with urea (Samui et al., 1998; Kundu and Samui, 2000), presumably reflecting the pronounced preference that exists for heterotrophic immobilization of NH₄⁺ over NO₃⁻ (e.g., Jansson et al., 1955; Jansson, 1958; Rice and Tiedje, 1989; Recous et al., 1990).

Table 5. Efficiency of nitrogen fertilization for increasing cereal grain production and estimated nitrogen losses in long-term cropping experiments with a static plot design.

Location/ soil order†	Cropping system‡	Study period	Fertilizer N			Grain yield#	Apparent		Net N loss			Reference(s)
			Form(s)§	Mean rate	Application¶		NDF‡††	FNUE‡‡	From fertilizer§§	From soil¶¶	Total	
			kg ha ⁻¹ yr ⁻¹			Mg ha ⁻¹ yr ⁻¹	%		kg ha ⁻¹ yr ⁻¹			
England												
Alfisols (sicl)	B-B†††	1882–1961		0		0.7			1 (23)	1	Warren and Johnston (1967)	
			1	48	spring	2.0	67	61	19	4	23	
			5	48	spring	2.1	69	65	17	-1	15	
Alfisols (sicl)	W _w -W _w †††	1882–1925		0		0.6			5 (23)	5	Garner and Dyke (1969)	
			1	48	spring	1.1	41	26	35	2	37	
			5	48	spring	1.4	55	42	27	3	30	
Alfisols (sicl)	W _w -W _w †††	1970–1978		0		1.4			7 (23)	7	Dyke et al. (1983)	
			8	48	spring	3.0	51	62	18	10	28	
			8	96	spring	4.1	69	66	33	0	33	
			8	144	spring	4.5	76	63	53	-2	51	
			8	192	spring	4.7	78	53	91	-10	81	
India												
Entisols (sl)	Ri-W _w †††	1986–1996		0		2.0			-22 (15)	-22	Samui et al. (1998), Kundu and Samui (2000)	
			7	100	split (4)	3.2	36	26	74	-5	69	
			7	200	split (4)	4.8	62	39	122	17	139	
USA												
Missouri	W _s -W _s †††	1914–1938		0		0.6			15 (18)	15	Smith (1942)	
Alfisols (sil)			5	86	spring	1.2	52	27	63	8	71	
Nebraska	C-C†††	1953–1972		0		2.0			-7 (30)	-7	Anderson and Peterson (1972)	
Mollisols (sl)			4	180	spring	5.4	62	27	132	-18	114	
Oklahoma	W _w -W _w †††	1930–1938		0		0.9			9 (15)	9	Harper (1959)	
Mollisols (l)			5	16	spring	1.3	32	80	3	9	12	
Oregon	W _w -F†††	1932–1966		0		2.2			24 (60)	24	Rasmussen and Parton (1994)	
Mollisols (sil)			1, 5, 6	34	fall§§§	2.6	22	30	24	7	31	
		1967–1986		0		2.8			16 (60)	16		
			4	45	fall	3.9	41	52	22	14	36	
			4	90	fall	4.4	55	46	49	12	61	
South Dakota	C-Sg-	1915–1939		0		1.1			39 (36)	39	Hutton (1938), Puhr (1945)	
Mollisols (l)	Rcl†††		5	64	spring	1.3	16	10	57	57	114	
Wisconsin	C-C†††	1967–1989		0		2.8			46 (20)	46	Vanotti et al. (1997)	
Alfisols			4	68	spring	5.2	62	70	20	56	76	
(sil)##			4	136	spring	5.8	68	45	74	36	110	
			4	273	spring	6.3	71	26	202	32	234	

† Surface texture designated parenthetically as l (loam), sicl (silty clay loam), sil (silt loam), sl (sandy loam).

‡ B, barley; C, corn; F, fallow; Rcl, red clover; Ri, rice; Sg, small grain; W_s, spring wheat; W_w, winter wheat.

§ 1, (NH₄)₂SO₄; 4, NH₄NO₃; 5, NaNO₃; 6, Ca(NO₃)₂; 7, urea; 8, CaNH₄(NO₃)₃.

¶ Value in parentheses indicates the number of applications.

Averaged over the study period and expressed on a dry-weight basis.

†† NDF, nitrogen derived from fertilizer. Estimated as 100 × [(fertilized grain yield – unfertilized grain yield) × grain N concentration + ∞ × (fertilized stover yield – unfertilized stover yield) × stover N concentration] / (fertilized grain yield × grain N concentration + ∞ × fertilized stover yield × stover N concentration), where ∞ = 1 if residue was removed and 0 otherwise.

‡‡ FNUE, fertilizer nitrogen uptake efficiency. Estimated as 100 × [(fertilized grain yield – unfertilized grain yield) × grain N concentration + ∞ × (fertilized stover yield – unfertilized stover yield) × stover N concentration] / fertilizer N rate, where ∞ = 1 if residue was removed and 0 otherwise. If not reported within the reference cited, grain and stover N concentrations were calculated from textbook values for crude protein (Martin et al., 1976).

§§ Estimated as fertilizer N rate × (1 – FNUE/100).

¶¶ Estimated from soil total N change during the study period, using bulk density data reported. Values in parentheses indicate the depth of sampling (in cm).

Analysis based on time-weighted data for fertilizer N rate and yield.

††† Aboveground residues removed.

‡‡‡ Aboveground residues returned.

§§§ Spring application before 1944.

On account of this fundamental distinction, a higher FNUE would be expected for fertilizer N applied as NO_3^- , relative to fertilizer-derived NH_4^+ -N, provided that immobilization is not inhibited by soil acidity or other factors and that plant uptake of NO_3^- is not limited by leaching or denitrification. This difference is indeed apparent from ^{15}N recovery data collected in several greenhouse and field studies using labeled fertilizer (e.g., Broadbent and Nakashima, 1968; Rennie and Rennie, 1973; Dev and Rennie, 1979; Powlson et al., 1986; Recous et al., 1988, 1992; Shen et al., 1989; Crozier et al., 1998) and from two long-term studies cited in Table 5 that directly compare yield responses by barley or winter wheat to NaNO_3 and $(\text{NH}_4)_2\text{SO}_4$ (Warren and Johnston, 1967; Garner and Dyke, 1969). The use of NO_3^- was particularly beneficial when fertilization supplied N during a period of active growth by an established wheat crop (Garner and Dyke, 1969), whereas the benefit was reduced considerably when N was applied before sowing barley (Warren and Johnston, 1967), presumably reflecting N losses during a period when there was very little, if any, crop N uptake (e.g., Recous and Machet, 1999). Long-term studies in Norway (Uhlen, 1991) and Sweden (Kirchmann et al., 1994; Persson and Kirchmann, 1994) have documented a gain in total C and N for surface soils under cereal cropping by annual fertilization with $\text{Ca}(\text{NO}_3)_2$ and incorporation of aboveground residues. Such gains are consistent with findings by Bosch and Amberger (1983) and Schnürer et al. (1985) that long-term use of this fertilizer decreased net N mineralization, relative to an unfertilized control (Table 3).

Modern cereal production relies heavily on ammoniacal fertilizers, largely for economic reasons arising from universal dependence on the Haber-Bosch process. Besides the economic motivation, ammoniacal fertilization reduces the short-term potential for N loss by leaching or denitrification, providing rationale for the convenience of fertilizer application well in advance of crop N demand. Greater care is called for when fertilizing with NO_3^- , such that application rate and timing must be more closely matched to crop uptake. If this is done, a potential advantage arises because NO_3^- is transported by the plant more readily than NH_4^+ and rapidly accumulates in the vacuole (e.g., see Salsac et al., 1987), which often increases dry matter production despite a higher energy requirement for assimilation.

If the primary purpose of N fertilization is to ensure that plant N availability does not limit yield while FNUE is maximized, then there is an inherent contradiction in today's massive consumption of ammoniacal fertilizers for cereal production. The management of these fertilizers is necessarily complicated by microbial competition for NH_4^+ , such that crop N requirement is subject to numerous interactions involving such factors as the quantity and quality of residue inputs, the timing of fertilization relative to these inputs, tillage, and fertilizer placement. The consequences are documented more realistically by on-farm response trials (Table 6) than by long-term static plot studies (Table 5) because fertilized and unfertilized plots share the same fertilization history, such that yield differences are not inflated as nutrient depletion intensifies over time (e.g., Varvel and Wilhelm, 2003). The FNUE values in Table 6 are lower than most of those documented by Table 5 and likely have been inflated over time by the loss of soil organic N. Closer

examination of Table 6 reveals a clear tendency for FNUE to decrease with an increase in unfertilized yields, which are inherently linked to soil N availability and management practices.

The FNUE values reported in Table 6 reflect N rates typically applied by grain producers, without knowing the soil's productive capacity in the absence of N fertilization. These values leave no doubt that such knowledge is indispensable for optimizing fertilizer N management because yields without fertilizer N were usually greater than the yield increase with optimal N fertilization (delta yield), except when corn was grown on coarse-textured soils (Williams et al., 2007) or when wheat was grown after the incorporation of oat straw (Bélanger et al., 1998). In each case, limited soil N availability substantially increased the need for N fertilization to optimize yield. In contrast, the economically optimum N rate (EONR) was lower for more productive Mollisols or Alfisols under corn production in Illinois, Iowa, or Wisconsin, and a further decrease in EONR occurred when soil N supplies had been enhanced by previous or recent manuring. Unfortunately, such soils are likely to receive more N than the crop needs according to yield-based recommendations commonly used for corn N fertilization since the 1970s (e.g., Hoefl and Peck, 2002) or the grouped economic approach advocated more recently by Sawyer et al. (2006) and Fernández et al. (2009).

Both strategies, and also the blanket N recommendations widely used in developing countries, are fundamentally flawed because soil N cycling is not taken into account, the usual result being excessive N applications (e.g., Lory and Scharf, 2003; Mulvaney et al., 2006) that seriously reduce FNUE (Raun and Johnson, 1999). The extent of overfertilization is evident from the disparity between fertilizer N requirement (EONR) and typical N rates listed in Table 6 and is quantified in terms of the extent of N fertilization beyond crop requirement. Considering that most of the on-farm FNUE values in Table 6 are well below the 33 to 36% global estimates of Raun and Johnson (1999) and Ladha et al. (2005), the collective economic cost of unutilized N could be substantially higher than the \$90 billion cited previously and would be accompanied by escalating societal and ecological costs attending depletion of SOC and soil N reserves.

The importance of these reserves is apparent from the indigenous fertility of Mollisols, which account for a disproportionate share of world cereal production with high grain yields per unit land area (Troeh and Thompson, 2005). Such soils are inherently high in their yield potential without fertilization compared with Alfisols or Ultisols, where fertilization is more beneficial for increasing delta yield, EONR, and FNUE. This distinction was elucidated by Cassman et al. (2003) and is evident from Table 6 because all three parameters were greater when corn was grown on North Carolina Ultisols as opposed to Mollisols in Illinois or Iowa. A further distinction should also be noted, in that heavy N fertilization of these Ultisols did not fully compensate for an inherent limitation in nutrient-supplying power.

Consequences for Food and Environmental Sustainability

The foregoing analysis provides ample reason for concern over the maintenance of soil productivity, considering the soil N declines documented for the Morrow Plots (Table 1) and numerous

Table 6. Efficiency of nitrogen fertilization for increasing grain yield in on-farm nitrogen-response experiments.

Location	Cropping system†	No. of trials	Fertilizer N		Yield without fertilizer N¶	Delta yield#	EONR††	Typical N rate‡‡	Apparent			Reference(s)
			Form(s)§	Applications§					NDFF§§	FNUE¶¶	NBCR##	
					Mg ha ⁻¹ yr ⁻¹		kg ha ⁻¹		%		kg ha ⁻¹	
Corn												
Illinois	C-C	11	4, 7	sidedress	7.2	2.9	111	190	27	25	79	Mulvaney et al. (2006)
	C-C (M ₁)	8	4, 7	sidedress	12.0	0.1	10	69	1	2	78	
	C-C (M ₂₋₅)	12	4, 7	sidedress	10.3	0.8	45	209	7	6	164	
	C-S	33	4, 7	sidedress	8.2	4.2	136	165	33	39	26	
	C-S (M ₁)	13	4, 7	sidedress	11.0	0.8	30	93	6	12	57	
	C-S (M ₂₋₅)	8	4, 7	sidedress	8.8	2.9	131	184	25	23	47	
Iowa	C-S	30	4	spring	8.9	2.6	108	161	21	24	53	Barker et al. (2006a,b)
	C-S (M ₁₋₁₂)	10	4	spring	10.7	1.2	28	150	10	11	122	
North Carolina	C-C	13	4, 7	sidedress	4.4	5.8	196	174	56	47§§§	-22	Williams et al. (2007)
Pennsylvania	C-C	10	4	spring	5.3 (21)	3.7	138	185	39	28	47	Fox and Piekielek (1983)
Wisconsin	C-C (M _{>3})	18	4	spring	7.9	1.4	76	190	15	10	114	Bundy and Andraski (1995)
Wheat												
Canada	O-W ₅	6	4	spring	1.6	2.6	135	55	62	38§§§	-80	Bélangier et al. (1998)
China	C-W _w (i)	121†††	7	split (2)	4.8	1.2	128	325	20	7	197	Cui et al. (2008)
	Ri-W _w	3	7	split (3)	2.2	2.2	218	150	50	29§§§	-68	Liang et al. (2008)
Greece	NR	253†††	NR	split (3)	2.0	1.4	172	110	41	16§§§	-62	Velemis et al. (1998)
Pakistan	Ri-W _w	43†††	7	preplant	1.8	1.2	87	85	40	28§§§	-2	Aslam et al. (1993)
Pennsylvania	O-W _w	5	4	split (2)	3.5	1.5	81	67	29	17§§§	-14	Roth et al. (1989)
Rice												
California	NR	25†††	1	preflood	3.5	3.8	149	168	52	26	19	Roberts et al. (1993)
India	Ri-Ri	27†††	7	split (3)	5.1	1.9	88‡‡‡	120	27	18	32	Yadvinder-Singh et al. (2007)
Philippines	Ri-Ri	38†††	NR	split (2)	2.3	0.9	57	<20	28	18§§§	<-37	Mandac and Flinn (1983)

† C, corn; NR, not reported; O, oats; Ri, rice; S, soybean; W_s, spring wheat; W_w, winter wheat. Manuring denoted parenthetically by M, with a subscript to indicate the number of years relative to the growing season studied, within which the most recent application was made. Irrigation denoted by (i).

‡ 1, (NH₄)₂SO₄; 4, NH₄NO₃; 7, urea; NR, not reported.

§ Values in parentheses indicate the number of applications.

¶ Reported at market standard moisture content (155 g kg⁻¹ for corn, 135 g kg⁻¹ for wheat, 120 g kg⁻¹ for rice). Value in parentheses indicates pre-season application of fertilizer N (in kg ha⁻¹).

Economically optimum yield – yield without fertilizer N. The value reported for C-C in Pennsylvania was uncorrected for pre-season N fertilization.

†† EONR, economically optimum nitrogen rate. Estimated by regression analysis unless otherwise specified.

‡‡ If not reported by the reference(s) cited, this value was obtained from fertilizer recommendations relevant to the crop, location, and study period.

§§ NDFF, nitrogen derived from fertilizer. Estimated as 100 × delta yield / (delta yield + yield without fertilizer N), assuming that grain N concentration was unaffected by fertilization.

¶¶ FNUE, fertilizer nitrogen uptake efficiency. Estimated as 100 × delta yield × grain N concentration / typical N rate, where delta yield determined for the EONR was assumed to hold for the typical N rate, and grain N concentration was calculated from textbook values for crude protein (Martin et al., 1976), assuming 160 (corn or rice) or 175 (wheat) g N kg⁻¹ protein.

NBCR, nitrogen fertilization beyond crop requirement. Estimated as typical N rate – EONR.

††† NDFF and FNUE estimated from mean data representing all trials reported.

‡‡‡ Estimated by averaging cumulative N rates from four to five split applications according to leaf color.

§§§ Calculated for the EONR.

other sites throughout the world (Table 2). Any such decline would contribute toward a growing dilemma confronting chemical-based cereal production: World population and grain demand are growing more rapidly than grain yield. This disparity has occurred despite increasing inputs of fertilizer N and is documented by widespread reports of yield stagnation or even decline (e.g., Byerlee and Siddiq, 1994; Cassman et al., 1995, 2003; Cassman and Pingali, 1995; Pingali et al., 1997; Calderini and Slafer, 1998; Cassman, 1999; Dawe and Dobermann, 1999; Aggarwal et al., 2000; Dawe et al., 2000; Duxbury et al., 2000; Ladha et al., 2000a, 2000b, 2003a, 2003b; Regmi et al., 2000, 2002; Yadav et al., 2000a, 2000b; Kumar and Yadav, 2001; Pathak et al., 2003; Barah, 2004; Manna et al., 2005; Bationo, 2007; International Rice Research Institute, 2008; Zhang et al., 2008). Declining yields likely have other causes, but there is evidence from long-term NPK trials that a loss of indigenous soil N supply is a contributing factor (e.g.,

Cassman et al., 1995, 1998; Cassman and Pingali, 1995; Singh et al., 1998, 2000; Ladha et al., 2000b, 2003a, 2003b; Ram, 2000; Yadav et al., 2000b; Bhandari et al., 2002; Manna et al., 2005), which may reflect a change in soil organic matter quantity (e.g., Singh et al., 1998, 2000; Dawe et al., 2000; Ram, 2000; Yadav et al., 2000a; Byerlee et al., 2003; Ladha et al., 2003a, 2003b; Bationo, 2007) or quality (Kretschmann et al., 1991; Olk et al., 1996; Manna et al., 2005; Šimon, 2005).

The degradation of soil C and N resources necessarily increases reliance on synthetic N fertilization (e.g., Singh et al., 1998), but given the value of organic matter for improving the chemical, physical, and microbial properties of soil, this strategy cannot be expected to maintain current levels of productivity (Cassman et al., 2003; Tong et al., 2003). Despite ongoing genetic and cultural improvements, a 66% global decrease has occurred over the past 40 yr in the agronomic efficiency of fer-

tilizer N (Tilman et al., 2002; Raun and Schepers, 2008). Excessive N fertilization no doubt contributes to the latter trend, but soil degradation is a more important concern for sustaining world food supplies (Tong et al., 2003).

The issue of sustainability is of paramount importance for a soil resource that has been subjected to escalating chemical inputs for the past four decades (Tilman et al., 2002). World population has doubled during this period, while cereal production has tripled as an eightfold increase has occurred in global use of synthetic N (FAO, 2009). Besides having an adverse effect on the soil resource, the latter input has contributed to NO_3^- pollution of ground and surface waters (e.g., Goolsby and Battaglin, 2001; Zhu et al., 2003; Rupert, 2008) and has been linked to a growing occurrence of O_2 depletion in coastal waters receiving riverine discharge, notably affecting up to 22,000 km^2 of the northern Gulf of Mexico (Turner et al., 2008) and many other areas, including 180,000 km^2 of the western Indian continental shelf (Naqvi et al., 2000), 20,000 km^2 of the Changjiang Estuary (Wei et al., 2007), and up to 9000 km^2 of the southern North Sea (Conley et al., 2007). There have also been atmospheric consequences from increasing reliance on synthetic N fertilization, which has been implicated in escalating atmospheric N_2O enrichment observed since 1980 (Prather et al., 2001) and the loss of SOC under grain production (Khan et al., 2007; Pikul et al., 2008; Poirier et al., 2009).

The degradation of soil, air, and water resources by chemical-based cereal production will likely intensify if crop residues are used as a bioenergy feedstock, a practice increasingly advocated for short-term economic gain by the public and private sectors. The long-term consequences of the latter practice are readily apparent from the developing world, where soils have been depleted by many centuries of continuous cropping without the return of aboveground residues and are now subject to more rapid depletion with intensive cereal production systems.

Conclusions

There is a prevailing view that global food and fiber production will continue to expand because of modern agricultural management systems with improved cultivars and intensive chemical inputs dominated by synthetic ammoniacal fertilizers. The use of these fertilizers has led to concerns regarding water and air pollution but is generally perceived to play an essential role for sustaining agricultural productivity, not only by supplying the most important nutrient for cereal production but also by increasing the input of crop residues for building soil organic matter. The scientific soundness of the buildup concept has yet to be substantiated empirically using baseline data sets from long-term cropping experiments.

The present paper and a companion study by Khan et al. (2007) provide many such data sets that encompass a variety of cereal cropping and management systems in different parts of the world. Overwhelmingly, the evidence is diametrically opposed to the buildup concept and instead corroborates a view elaborated long ago by White (1927) and Albrecht (1938) that fertilizer N depletes soil organic matter by promoting microbial C utilization

and N mineralization. An inexorable conclusion can be drawn: The scientific basis for input-intensive cereal production is seriously flawed. The long-term consequences of continued reliance on current production practices will be a decline in soil productivity that increases the need for synthetic N fertilization, threatens food security, and exacerbates environmental degradation.

This dilemma calls for an international effort by agricultural scientists to thoroughly review, evaluate, and revise current cereal production and management systems and policies. The immediate need is to use scientific and technological advances that can increase input efficiencies. One aspect of this strategy would be to more accurately match the input of ammoniacal N to crop N requirement by accounting for site-specific variations in soil N-supplying capacity and by synchronizing application with plant N uptake. In the long term, a transition may be required toward agricultural diversification using legume-based crop rotations, which provide a valuable means to reduce the intensity of ammoniacal fertilization with the input of less reactive organic N. For further insight about these and other possible strategies, researchers should consider and exploit the work described and cited herein.

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