Comments on "Synthetic Nitrogen Fertilizers Deplete Soil Nitrogen: A Global Dilemma for Sustainable Cereal Production," by R.L. Mulvaney, S.A. Khan, and T.R. Ellsworth in the *Journal of Environmental Quality* 2009 38:2295–2314

Dear Editor,

Mulvaney et al. (2009) claim to present evidence that inorganic nitrogen fertilizer leads to a decline in the organic nitrogen (N) content of soil. This follows an earlier paper purporting to show a corresponding decline in soil organic carbon (C) resulting from N fertilizer applications (Khan et al., 2007). Both are based on an analysis of soil N and C data from the Morrow Plots, a long-term field experiment in Illinois, started in 1876. We submit that their conclusion is flawed because the authors did not take full account of past changes in the inputs of fertilizers and manures on the Morrow Plots. We further submit that their citation of data from Rothamsted and other long-term experiments worldwide, claimed to support their conclusion, is selective and misleading. The conclusions drawn by Mulvaney et al. (2009) also appear to be at variance with other accounts of changes in soil N and C in the Morrow Plots (Aref and Wander, 1998; David et al., 2009).

Mulvaney et al. (2009) compare soil N concentration in selected treatments of the Morrow Plots measured in 1955 and 2005, for three soil depths (0–15, 15–30, and 30–46 cm) and three crop rotations: continuous corn, corn–oats (oats replaced by soybean since 1967), and corn–oats–alfalfa hay. Within each crop rotation, three current fertilizer treatments are compared:

1. None: unamended since the start of the experiment in 1876.
2. NPK: unamended until 1955, then N–P–K applications started.
3. HNPK (high N–P–K): a higher rate of N than in the NPK treatment applied since 1967. From 1904 to 1966, the treatment was manure, together with limestone and rock phosphate; in 1967 manure was replaced by the higher rate of fertilizer N. We consider that the authors' misinterpretation of the data arises from ignoring this change.

The rates of N differ for each crop and have changed over time. For corn, the N rate in the NPK treatment was 168 kg ha^{-1} (1955–1966) and increased to 224 kg ha^{-1} from 1967. In the HNPK treatment, it was 336 kg ha^{-1} (1967–1997), reduced to 224 kg ha^{-1} from 1998. Thus, for the last 8 yr of the study period, the rate of N applied in HNPK and NPK were the same. For oats, the N rate is 28 kg ha^{-1} in both the NPK and the HNPK treatments. More detailed information on the previous treatments in the Morow Plots is given in Aref and Wander (1998).

In all except one of the crop rotation and fertilizer treatment combinations, there was some decrease in soil N concentration during the 51-yr period considered (1955–2005), ranging from a nonsignificant decrease of 0.007 g N kg^{-1} soil to a maximum of 0.502 g N kg^{-1} soil (Table 1 of Mulvaney et al., 2009). This is presumably because the entire site was still subject to a gradual decline in soil N following its conversion from natural grassland to arable cropping at the start of the experiment in 1876. Such long-term trends in soil N and C concentrations following a major change in management are commonly observed in long-term studies in temperate climates (e.g., Aref and Wander, 1998; Johnston et al., 2009). Mulvaney et al. (2009) argue that high rates of N fertilizer cause a decline in soil organic N and C based on the evidence that declines in N concentration were greatest in the HNPK treatment. However, this conclusion ignores the important difference in the longer-term history of the NPK and HNPK treatments summarized above. The treatment that became HNPK in 1967 had previously received manure over a period of 62 yr, and this increased soil N as shown in Table 1 of Mulvaney et al. (2009). For example, for continuous corn, N concentrations in 1955 in the NPK treatment (that had never received manure) were 1.376, 1.342, and 1.020 g kg^{-1} soil in the 0- to 15-, 15- to 30-, and 30- to 46-cm depths, respectively. In the same year, the corresponding concentrations in the HNPK treatment (that previously received manure) were 1.534, 1.568, and 1.476 g kg^{-1} soil (i.e., 11–45% greater than in the NPK treatment). It is inevitable that after stopping manure applications in 1966, organic N and C would decline as a result of decreased organic inputs. Thus, the observation that soil N and C declines in the HNPK treatment were greater than in the unamended and NPK treatments is entirely understandable on the basis of the cessation of manure applications. Manure applications stopped at the same time as the high fertilizer N rate started,

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so the effects are confounded. While it is impossible to conclude unequivocally which change of treatment is responsible for the greater decline in soil N concentration in the HNPK treatment, we suggest that the cessation of organic inputs in manure is a far more plausible explanation.

Our proposal is supported by results from the long-term Hoosfield Barley Experiment at Rothamsted, UK, in which spring barley has been grown each year with different fertilizer and manure treatments since 1852. It includes a treatment in which manure was applied at 35 t ha^{-1} yr^{-1} for 20 yr (1852–1871) and none thereafter. Soil organic N and C increased during the 20 yr of manure application and declined steadily since. Figure 1 shows soil N concentrations in selected treatments over the period 1868 to 1998. More than 100 yr after manure applications ceased, soil N and C concentrations in this treatment continue to decline slowly but are still greater than in the treatments that have never received manure (Johnston et al., 2009; Jenkinson and Johnston, 1977). This evidence supports our conclusion that the greater decline in soil N concentration in the HNPK treatment in the Morrow Plots, compared with NPK and unamended, as reported by Mulvaney et al. (2009), are far more likely due to the cessation of manure application than to the application of N fertilizer. Indeed, the starting date for their comparisons, 1955, was when manure was still being applied. This point was made by Reid (2008) in relation to soil C concentration declines reported by Khan et al. (2008) but was ignored in the response by Khan et al. (2008).

If there was a genuinely deleterious effect of N fertilizer on soil organic N and C, one might expect to see declines in the NPK treatment of the Morrow Plots, compared with unamended during the 1955–2005 period, especially as the N application rate in the NPK treatment was fairly high (see above for rates applied to corn). But according to the data in Table 1 of Mulvaney et al. (2009), there was no such trend. Considering their data on "net change in 51 yr" in Table 1, nine comparisons of soil N concentration can be made between the NPK and unamended treatments (three crop rotations and three soil depths). In these nine comparisons, in only two cases was the decline in soil N concentration in NPK greater than in unamended; in one case it was larger in the unamended treatment, and in the remaining six cases they did not differ significantly from each other.

In Table 2 of Mulvaney et al. (2009), the authors present data from a large number of other long-term studies worldwide and claim that these support their contention of a decline in soil organic N caused by applications of inorganic fertilizer N. However, the data they present is highly selective. In all cases they, astonishingly, fail to include data on soil N changes in treatments given no fertilizers or given P and K but no N: it is therefore impossible to assess whether the decreases in soil N observed result from the effect of fertilizer N or from a general decline at the site that also occurs in the no N treatment.

Figure 2 shows data from the Broadbalk Wheat Experiment at Rothamsted, started in 1843 on a site that had been in arable cropping for at least 300 yr and probably much longer. Figure 2 extends earlier published data (Glendining et al., 1996; Glendining and Powlson, 1990) to 2005. The first sampling of soils in all treatments was in 1865. By 1881, 30 yr after the different fertilizer treatments had begun, soil N concentration in the plot receiving PK + 144 kgN ha^{-1} was measurably greater than in the plot receiving no fertilizer or PK alone (1.25 compared with 1.00 and 1.03 g N kg^{-1} soil, respectively; Fig. 2); these differences persisted until the most recent sampling in 2005. Figure 2 also shows data from a plot that received PK + 48 kgN ha^{-1} (in most years; occasionally 96 kg N ha^{-1}) from 1843 until 1967. Soil N in this plot was also greater than in the PK plot at most samplings between 1881 and 1966. In 1968, this plot was changed to a new higher rate of N fertilizer, PK + 192 kg N ha^{-1}. This increase in N fertilizer input led to an increase in soil N concentration; by 2005 it was larger than in the long-continued PK + 144 kg N ha^{-1} treatment (Fig. 2).

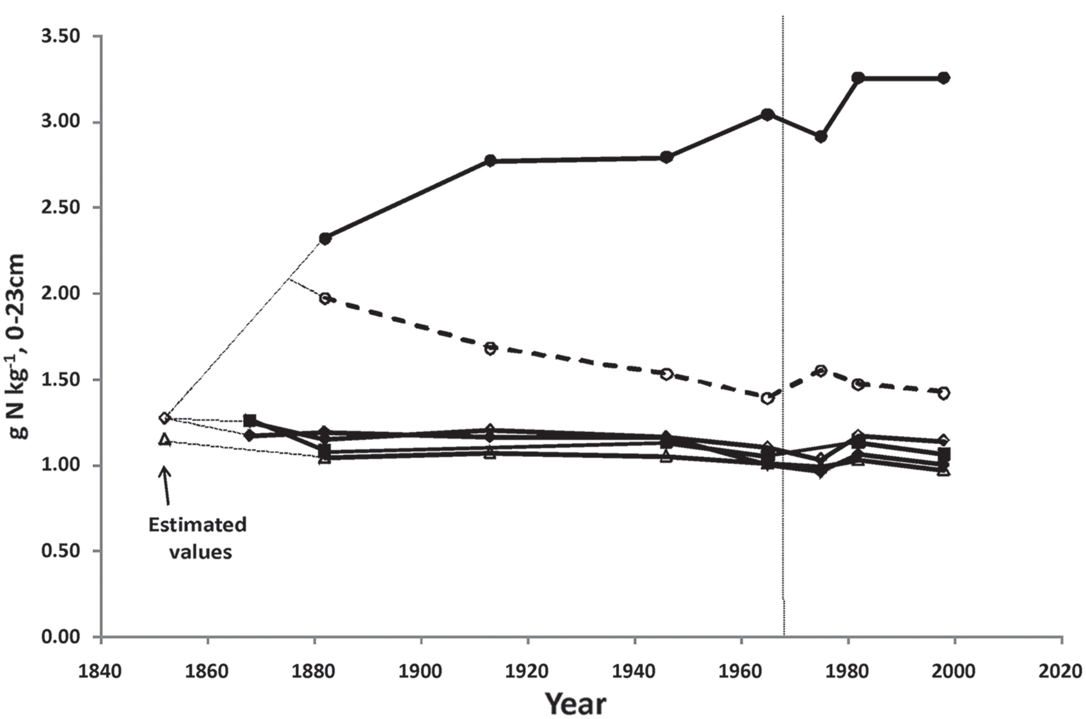

Fig. 1. Hoosfield Barley Experiment, Rothamsted, UK. Total N, g kg^{-1}, in topsoil, 0–23 cm. The treatments (ha^{-1} yr^{-1}) are (△) nil; (◇) PKMg since 1852; (◆) PKMg + 48 kg N as ammonium sulfate since 1852; (■)PKMg + 48 kg N as sodium nitrate since 1852; (●) farmyard manure, 35 t, since 1852; and (O) farmyard manure residues, 35 t, 1852–1871 only, none since. Since 1968 (indicated by vertical line) all treatments have received, on average, 72 kg N ha^{-1} yr^{-1}.

Presumably, this reflects increased organic inputs to soil from crop residues (stubble, roots, root exudates: straw is removed in these treatments) in the new higher N rate treatment: grain yields with N at 192 kg ha^{-1} averaged more than 2 t ha^{-1} greater than with N at 48 kg ha^{-1} in the relevant sections of the plots in the period 1968 to 1987 and ~0.5 t ha^{-1} greater than with N at 144 kg ha^{-1}. Thus, data from the Broadbalk experiment lead to the opposite conclusion to that drawn by Mulvaney et al. (2009)—soils receiving N fertilizer consistently contain *more* organic N than those not receiving N.

The results from the Broadbalk and Hoosfield experiments illustrate the degree of variability that occurs in soil N data in sequential samplings of long-term experiments. Within-plot soil variability and changes in sampling technique can lead to differences in results at different sampling times. For this reason, when using data from long-running, unreplicated experiments and attempting to quantify small differences between treatments, it is best to compare paired data (i.e., with and without the treatment) on a number of occasions over a long run of years.

Glendining and Powlson (1995) reviewed results from numerous long-term studies worldwide, specifically to assess the impact of long-continued applications of inorganic N fertilizer on soil organic N content. In this review, topsoil N concentrations in treatments receiving NPK were compared with treatments receiving PK, but not N; in the vast majority of experiments, subsoil data were not published. In 29 out of 34 sites lasting over 40 yr, soil N in NPK treatments was greater than in the treatment with PK but no N; in only 5 sites was it less. In these 29 sites, the mean increase was 8% of the value in the PK treatment (range 2–21%) with an increase of >10% in about one-third of the cases. Taking all 34 comparisons, the average increase was 6%. In another set of 36 comparisons, based on experiments of 7- to 40-yr duration, soil N concentration in NPK treatments was greater than in PK at 30 sites, with the increase being at least 10% of the value in PK-treated soil in almost half these cases. At six sites, there was no difference or a decrease in soil N. There was considerable evidence to indicate that these increases in soil N where N fertilizer was applied were associated with increased crop growth. For example, at two sites where irrigation was included, increases in soil N with N fertilizer were greater in the irrigated treatments than in unirrigated, reflecting increased crop growth with irrigation and greater crop response to added N fertilizer. And where several rates of N were tested, increases were usually larger with the larger N rates. Overall, this large body of data indicates that even if there is any increased soil organic matter decomposition caused by added inorganic N, it is outweighed by the tendency for soil N and C to increase due to the extra organic inputs from larger crops.

Unlike the Broadbalk Experiment, the Hoosfield Barley Experiment at Rothamsted does not contain a series of treatments with different rates of N fertilizer applied over a long period. However, the four treatments not given manure shown in Fig. 1 do provide a comparison of nil (no fertilizer), PKMg, and PKMg + 48 kgNha^{-1}, where the N was applied as either ammonium sulfate or sodium nitrate. Although, overall, there is a very small downward trend in soil N among these treatments, there is no clear indication that fertilizer N either increased or decreased this slight decline (Fig. 1). This is in contrast to the Broadbalk Experiment, where fertilizer N increased soil N. There are several possible reasons for this difference: fewer cultivations are required to produce a seedbed for an autumn sown crop compared to a spring sown crop, winter wheat is in the ground for longer than spring barley and yields of wheat on Broadbalk, and thus returns of crop residues, are larger than on Hoosfield. Also, in long-running, unreplicated experiments such as these there is always the possibility of inherent differences in soil properties within the site so it is unwise to place too much emphasis on comparisons where measured differences are small. The observations of very small decreases in soil N concentration in N-fertilized soils, that contrasts with the

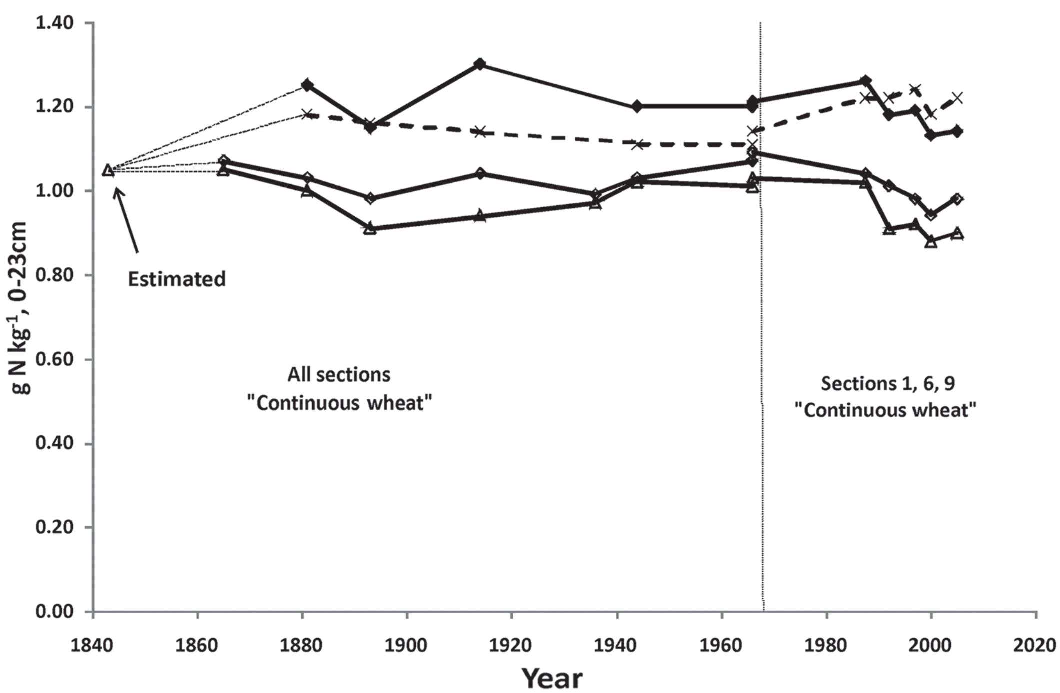

Fig. 2. Broadbalk Wheat Experiment, Rothamsted, UK. Total N, g kg^{-1}, in topsoil, 0–23 cm. The treatments (ha^{-1} yr^{-1}) are (△) nil; (◇) PKMg since 1852; (◆) PKMg + 144 kg N since 1852; and (×) PKMg + 192 kg N since 1968 only, previously PKMg + 48 or 96 kg N. In 1968, the experiment was divided into 10 sections, some of which remained in continuous wheat, others going into wheat grown in rotation. The discontinuity in each line reflects this change, the latter data points being the mean of three of the sections in continuous wheat.

vast body of other data, in no way substantiates the conclusions of Mulvaney et al. (2009).

A further criticism of the paper is the treatment of N fertilizer impacts on rates of N mineralization. In their Table 3, Mulvaney et al. (2009) summarize N mineralization rates, assessed from laboratory incubations, from a range of long-term experiments. In almost all cases, soils from plots regularly receiving N fertilizer had higher mineralization rates than soil from zero N plots. And at sites with multiple N rates, there was a tendency for rates to be higher in the soils receiving higher rates of N. The authors cite this as evidence of enhanced soil organic matter decomposition where inorganic N fertilizer is regularly applied. But they fail to note that these increased N mineralization rates in N-fertilized soils are associated with an *increased* concentration of total soil N. If they had included soil total N concentrations of the zero N treatments in their Table 1, this would have been apparent. There are presumably two reasons for the increase in mineralization on N-fertilized plots: first, that inputs of organic N and C are greater, due to increased plant growth, and second, that these inputs have a narrower C/N ratio, so that *immobilization* of mineral N is less. The overall balance of increased organic inputs and increased mineralization is a small increase in N concentration in soil. This trend was clear at many sites in the review of Glendining and Powlson (1995). Jenkinson (1991, their Fig. 9) illustrated the point using data from Broadbalk (Shen et al., 1989). As N fertilizer application increased, total soil N and mineralizable N both increased: this finding is in complete contrast to the contention of Mulvaney et al. (2009).

We propose that the conclusion drawn by Mulvaney et al. (2009), that inorganic N fertilizer causes a decline in soil organic N concentration, is false and not supported by the data from the Morrow Plots or from numerous studies worldwide. Their contention that the depletion of soil N by inorganic fertilizers causes a "global dilemma for sustainable cereal production" is equally false based on the evidence presented here. Of course, their conclusion, that more accurately matching inputs of N fertilizer to crop requirements to avoid overfertilization and unnecessary environmental pollution, is entirely correct, as is the need to better recycle N from organic residues and better incorporate legumes into cropping systems. But these conclusions in no way result from their flawed analysis.

On a more positive note, the observation of significant soil C and N declines in subsoil layers is interesting and deserves further consideration. Richter et al. (1999) observed an unexpected and counterintuitive decrease in subsoil organic C during conversion of arable land to forest in the Calhoun Experimental Forest, South Carolina. However, in the Broadbalk Wheat Experiment, there was no evidence of changes in subsoil N or C concentrations in arable plots resulting from N fertilizer applications between 1881 and 1999 (Jenkinson et al., 2008).

Using data from long-term experiments to explore fundamental issues relevant to sustainable food production and environmental protection is of great value (Johnston et al., 2009; Rasmussen et al., 1998; Richter et al., 2007). It is to be regretted that Mulvaney et al. (2009) and Khan et al. (2007) have so badly misinterpreted data from valuable long-term sites, thus causing scientific confusion that would have serious consequences for global food security—if taken seriously.

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