CARBON SEQUESTRATION IN ARABLE SOILS IS LIKELY TO INCREASE NITROUS OXIDE EMISSIONS, OFFSETTING REDUCTIONS IN CLIMATE RADIATIVE FORCING

CHANGSHENG LI[1], STEVE FROLKING[1] and KLAUS BUTTERBACH-BAHL[2]

[1]*Institute for the Study of Earth, Oceans, and Space, University of New Hampshire, Durham, NH 03824, U.S.A.*
E-mail: steve.frolking@unh.edu
[2]*Institute for Meteorology and Climate Research, Atmospheric Environmental Research, Garmisch-Partenkirchen, D-82467 Germany*

Abstract. Strategies for mitigating the increasing concentration of carbon dioxide (CO_2) in the atmosphere include sequestering carbon (C) in soils and vegetation of terrestrial ecosystems. Carbon and nitrogen (N) move through terrestrial ecosystems in coupled biogeochemical cycles, and increasing C stocks in soils and vegetation will have an impact on the N cycle. We conducted simulations with a biogeochemical model to evaluate the impact of different cropland management strategies on the coupled cycles of C and N, with special emphasis on C-sequestration and emission of the greenhouse gases methane (CH_4) and nitrous oxide (N_2O). Reduced tillage, enhanced crop residue incorporation, and farmyard manure application each increased soil C-sequestration, increased N_2O emissions, and had little effect on CH_4 uptake. Over 20 years, increases in N_2O emissions, which were converted into CO_2-equivalent emissions with 100-year global warming potential multipliers, offset 75–310% of the carbon sequestered, depending on the scenario. Quantification of these types of biogeochemical interactions must be incorporated into assessment frameworks and trading mechanisms to accurately evaluate the value of agricultural systems in strategies for climate protection.

1. Introduction

Carbon sequestration has been highlighted recently as an important approach for mitigating the greenhouse effect by converting the atmospheric CO_2 into biotic or abiotic C sequestered in terrestrial ecosystems, underground reservoirs, the ocean, and as mineral carbonates (Lackner, 2003). Due to the tightly coupled cycles of carbon and nitrogen, anthropogenic changes in rates of the biotic C-sequestration in terrestrial ecosystems will directly affect N turnover processes in soils, and the biosphere–atmosphere exchange of gaseous N compounds. As highly managed systems, agricultural and forest ecosystems are being studied for their potential to sequester C in their soil and/or plant organic pools through management alternatives. For example, replacing conventional tillage with no-till generally results in net sequestration of soil organic carbon (SOC) (Lal et al., 1999; Robertson et al., 2000). National research programs seek the best strategies for C-sequestration in the terrestrial ecosystems (e.g., U.S.D.O.E., 1999). Lackner (2003) estimated the

Climatic Change (2005) 72: 321–338
DOI: 10.1007/s10584-005-6791-5

global storage capacity of soil carbon at roughly 100 Gt C, slightly greater than the potential for storage in woody biomass, and less than the potential for storage in the ocean. Agricultural soils generally have the capacity to store carbon, as their pre-cultivation SOC reserves were depleted in the first few decades after cultivation (e.g., Smith et al., 1997).

However, since the C- and N-cycles are closely coupled, and since the Kyoto Protocol (United Nations Framework Convention on Climate Change; see http://unfccc.int/resource/docs/convkp/kpeng.html) has set targets for a suite of greenhouse gases, including N_2O and CH_4, evaluating the potential of C-sequestration programs requires consideration of change in emissions of other greenhouse gases (Li, 1995; Robertson et al., 2000; Smith et al., 2001; Marland et al., 2003; Six et al., 2004). In addition, there are markets emerging for the trading of greenhouse gas emissions credits (Kyoto Protocol, Art. 17), and, to be effective, these markets will require full accounting of all gases (United Nations Conference on Trade and Development; see http://r0.unctad.org/ghg/sitecurrent/about_u/profile.html). Different greenhouse gases can be compared on a common basis by converting emissions values of non-CO_2 greenhouse gases into CO_2-equivalents via their global warming potentials (Ramaswamy et al., 2001). There is an urgent need to develop operational observational and modeling methodologies to allow the international community to improve the quality and quantity of greenhouse gas emission data.

Microbial production of N_2O in soils is tightly linked to SOC status (Sahrawat and Keeney, 1986), and soil C-sequestration will affect the production and/or consumption of N_2O. Numerous field measurements and laboratory experiments have established a positive correlation between N_2O production/flux and SOC. For example:

- Incubation experiments have identified SOC content as a key controller of N_2O production, along with nitrate content and oxidation–reduction potential (i.e., Eh) (Leffelaar and Wessel, 1988; Müller et al., 1980; Melillo et al., 1983; Federer and Klemedtsson, 1988).
- Soils with higher SOC generally have higher N_2O fluxes (Bremner and Shaw, 1958; Bowman and Focht, 1974; Burford and Bremner, 1975; Stanford et al., 1975; Pluth and Nommik, 1981; Terry et al., 1981; Duxbury et al., 1982; Goodroad and Keeney, 1984; Pang and Cho, 1984; Robertson and Tiedje, 1987; Klingensmith, 1987; Mosier et al., 1991; Vinther, 1992; Papen and Butterbach-Bahl, 1999; Bouwman et al., 2002).
- Soil N_2O emissions increase with additions of organic matter to the soil (Christensen, 1983; Goodroad and Keeney, 1984; Flessa et al., 1996; Clayton et al., 1997; Thomson et al., 1997; Dong et al., 2000; Wever et al., 2002; Hao et al., 2001; Khalil et al., 2002; Velthof et al., 2002), and the ratio of N_2O emitted to N applied also increases (Bouwman et al., 2002; Dong et al., 2000; Christensen, 1983), implying that the increase in N_2O emissions is due to both an increase in soil total N content and also in soil microbial activity. Organic

matter quality and other management practices also affect N_2O emission rates (Hao et al., 2001; Velthof et al., 2002).

- Spatial variation in N_2O fluxes has been related to SOC content (Federer and Klemedtsson, 1988; Ambus and Christenson, 1994; Christensen et al., 1990).

This positive correlation between SOC and N_2O flux arises from the coupled biogeochemical cycles of C and N. In nature, chemical elements typically act in coupled fashion determined by their geochemical abundance and atomic structure. C and N have been adopted by most life forms on Earth as the basic material for construction and metabolism. As green plants grow by assimilating CO_2, they require N to form amino acids and other essential compounds (Larcher, 1995). As plant tissues are incorporated into the soil after the plants die, decomposers decouple C and N as they derive energy from the breakdown of the organic compounds, ultimately re-mineralizing most C and N to CO_2 and inorganic N (e.g., ammonium or nitrate). The energy required by the soil microbes is usually generated by oxidation-reduction reactions, transferring electrons from the C atoms existing in the organic compounds to oxygen. If oxygen is unavailable, some microbes (e.g., denitrifiers) can use other oxidants as electron acceptors. After oxygen, the most ready-reduced oxidant is nitrate, and this denitrification process generates nitric oxide (NO), N_2O, and dinitrogen (N_2) (Conrad, 1996). N_2O is also produced during nitrification, the microbially-mediated oxidation of ammonium to nitrate. Consequently, increasing C-sequestration is directly linked with increasing N_2O-emissions. Jin and Gruber (2003) reached a similar conclusion for ocean carbon sequestration by modeling the impacts of iron fertilization on coupled C and N ocean biogeochemistry.

2. Methods

We used the DNDC agroecosystem biogeochemistry model (Li et al., 1992, 1996; Li, 2000) to evaluate the impacts of some common alternative cropland management practices on both SOC storage and N_2O emissions. DNDC is a plot-scale model that consists of two components: (1) three sub-models for soil climate, plant growth and decomposition which predict the dynamics of soil temperature, moisture, pH, Eh and substrate concentration profiles based on primary drivers (e.g., daily weather, soil properties, and crop management scenario); and (2) three sub-models for nitrification, denitrification, and fermentation which track production, consumption and emission of N_2O, NO, N_2, ammonia (NH_3) and methane, based on soil environmental factors.

DNDC has been tested against numerous field data sets of long-term SOC dynamics (Smith et al., 1997; also see Table I and Figure 1) and N_2O emissions (Figure 2 and Table II). For sites with SOC greater than 0.3 g C g^{-1} soil (organic soils in Florida and Germany), DNDC simulated high populations of soil microbes, including nitrifiers and denitrifiers, and high soil solution concentrations of dissolved

TABLE I
Site descriptions for long-term SOC simulations shown in Figure 1

Site	Duration	Cropping[a]	Management[b]	Soil[c]	References
Morrow Plots #3, IL, USA	1904–1989	• corn/corn/corn, • corn/oat + clover • corn/oat/clover	None	Silt loam; pH = 5.8; $\rho = 1.45$ g cm^{-3}	U.S.D.A. (1992); Odell et al. (1984)
Rothamsted, UK	1840–1984	Winter wheat	• None • 144 kg N ha^{-1} y^{-1} • 3 t FYM C ha^{-1} y^{-1}	Silt loam; pH = 7.0; $\rho = 1.5$ g cm^{-3}	Jenkinson (1991)
Bad Lauchstadt, Germany	1956–1991	Barley/potato/ wheat/sugarbeet	40/60/60/90 kg N ha^{-1} y^{-1} and 2.7/0/2.7/0 t FYM C ha^{-1} y^{-1}	Silt loam; pH = 6.6; $\rho = 1.35$ g cm^{-3}	Körchens and Müller (1996); Smith et al. (1997)
Waite, Australia	1925–1998	Wheat/fallow	None	Silt loam; pH = 6.0; $\rho = 1.3$ g cm^{-3}	Grace (1996); Smith et al. (1997)
Swift Current, SK, Canada	1967–1998	Spring wheat/fallow	6.8 kg N ha^{-1} y^{-1}	Silt loam; pH = 7.0; $\rho = 1.24$ g cm^{-3}	Grant et al. (2004)

[a]Annual crops in multi-year crop rotations are separated by a slash (/).
[b]All plots were tilled for each cropping; FYM: farmyard manure with C:N = 13; treatments separated by a slash (/) applied to different crops in multi-year rotation.
[c]ρ: bulk density of surface soil (0–10 cm).

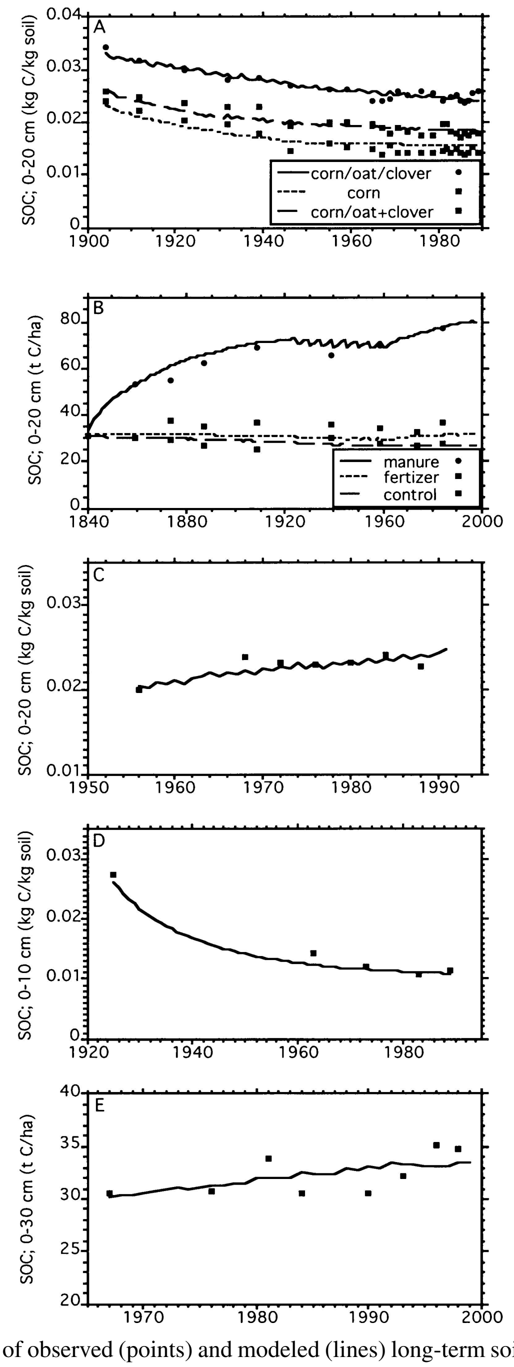

Figure 1. Comparison of observed (points) and modeled (lines) long-term soil organic carbon (SOC) dynamics at five agricultural sites in (A) the U.S.A., (B) the U.K, (C) Germany, (D) Australia, and (E) Canada (see Table I for site details and references). Note that units on the vertical scale are different between panels, based on units reported for the field data. The time-scale for changes in SOC is roughly 20–60 years.

TABLE II

Site descriptions and annual N_2O emission (reported to two significant figures) for points shown in Figure 2

Site	Year	Land-use	Vegetation	Management	SOC (kg C/ kg soil)	Field N_2O (kg N/ ha/y)	Model N_2O (kg N/ ha/y)	References
Florida, USA	1979	Newly-drained org. soil	Fallow	Unfertilized	0.4	160	140	Terry (1986); Terry et al. (1981)
Florida, USA	1979	Newly-drained org. soil	Grass	Unfertilized	0.4	97	44	Terry (1986); Terry et al. (1981)
Florida, USA	1979	Newly-drained org. soil	Sugarcane	Unfertilized	0.4	48	41	Terry (1986); Terry et al. (1981)
Florida, USA	1980	Newly-drained org. soil	Fallow	Unfertilized	0.4	51	65	Terry (1986); Terry et al. (1981)
Florida, USA	1980	Newly-drained org. soil	Grass	Unfertilized	0.4	19	13	Terry (1986); Terry et al. (1981)
Florida, USA	1980	Newly-drained org. soil	Sugarcane	Unfertilized	0.4	7.6	1.9	Terry (1986); Terry et al. (1981)
Germany	1994	Decades-drained org. soil	Meadow	Unfertilized	0.37	20	18	Flessa et al. (1998)
Germany	1994	Decades-drained org. soil	Meadow	Fertilized	0.43	4.2	44	Flessa et al. (1998)
Germany	1994	Decades-drained org. soil	Rye-grass	Unfertilized	0.41	56	67	Flessa et al. (1998)
Germany	1994	Decades-drained org. soil	Maize-potato	Fertilized	0.34	16	130	Flessa et al. (1998)
Costa Rica	1995	Newly-cult, cropland	Maize	Fertilized	0.05	3.9	4.1	Crill et al. (2000)
Costa Rica	1995	Newly-cult. cropland	Maize	Unfertilized	0.05	1.0	0.90	Crill et al. (2000)
Rothamsted, UK	1992	Cropland	Winter wheat	Fertilized	0.033	3.8	2.9	Harrison et al. (1995)
Devon, UK	1995	Pasture	Pasture	Unfertilized	0.029	1.4	0.59	Yamulki et al. (2000)
Devon, UK	1995	Pasture	Pasture	Urine	0.029	3.9	0.79	Yamulki et al. (2000)
Devon, UK	1995	Pasture	Pasture	Manure	0.029	2.6	2.4	Yamulki et al. (2000)
Scotland, UK	1992	Ryegrass	Rye-grass	Fertilized	0.028	2.8	2.0	Clayton et al. (1997)
Hokkadio, Japan	1995	Cropland	Onion	Fertilized	0.032	8.0	7.9	Cai et al. (2003)

Hokkadio, Japan	1996	Cropland	Onion	Fertilized	0.032	3.5	4.2	Cai et al. (2003)
Hokkadio, Japan	1997	Cropland	Onion	Fertilized	0.032	5.6	7.0	Cai et al. (2003)
Hokkadio, Japan	1998	Cropland	Onion	Fertilized	0.032	4.8	4.9	Cai et al. (2003)
Hokkadio, Japan	1999	Cropland	Onion	Fertilized	0.037	16	11	Cai et al. (2003)
Hokkadio, Japan	2000	Cropland	Onion	Fertilized	0.037	16	12	Cai et al. (2003)
Tsukuba, Japan	1996	Cropland	Carrot	Fertilized	0.031	0.17	3.1	Cai et al. (2003)
New Zealand	2001	Pasture	Grass	Grazed	0.035	11	12	Saggar et al. (2004)
New Zealand	2001	Pasture	Grass	Ungrazed	0.035	1.8	2.0	Saggar et al. (2004)
Ontario, Canada	1993	Cropland	Wheat	Manure	0.026	1.6	1.3	Smith et al. (2002)
Beijing, China	1996	Long-term cropland	Maize-wheat	Fertilized	0.015	1.7	2.6	YH Zhuang[a]
Beijing, China	1996	Long-term cropland	Maize-wheat	Unfertilized	0.015	0.59	0.41	YH Zhuang[a]
China	1998	Semi-arid grassland	Grassland	Unfertilized	0.025	0.38	0.24	Xuri et al. (2003)
China	1998	Semi-arid grassland	Grassland	Unfertilized	0.02	0.30	0.19	Xuri et al. (2003)
China	1998	Semi-arid grassland	Grassland	Unfertilized	0.019	0.21	0.13	Xuri et al. (2003)
China	1995	Semi-arid grassland	Grassland	Unfertilized	0.025	0.61	0.28	Xuri et al. (2003)
China	1995	Semi-arid grassland	Grassland	Unfertilized	0.019	0.27	0.15	Xuri et al. (2003)
Colorado, USA	1979	Short-grass prairie	Grassland	Unfertilized	0.006	0.10	0.12	Mosier et al. (1981)
Colorado, USA	1991	Short-grass prairie	Grassland	Unfertilized	0.01	0.14	0.25	Mosier et al. (1991)
Colorado, USA	1991	Short-grass prairie	Grassland	Unfertilized	0.01	0.49	0.42	Mosier et al. (1991)
Germany	1992	Cropland	Barley	Fertilized	0.011	4.2	5.7	Flessa et al. (1995)

[a]Personal communication with YaHui Zhuang, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, 1998.

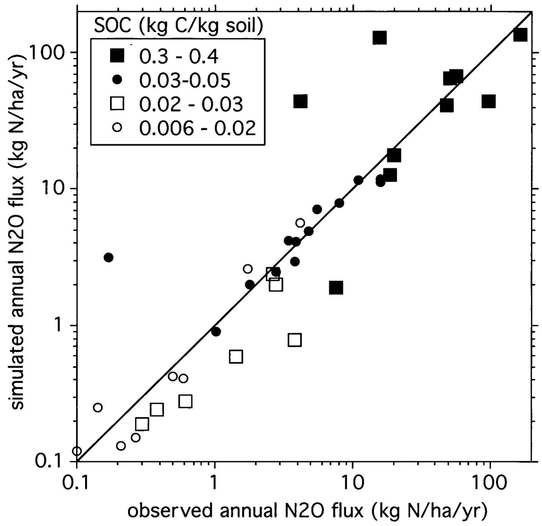

Figure 2. Comparison of observed and simulated estimates of annual N_2O emissions from agricultural sites around the world (see Table II for more information about field sites, and for field study references). Symbols indicate soil organic carbon (SOC) content in soil in kg SOC kg^{-1} soil (filled squares: SOC = 0.3–0.4; filled circles: SOC = 0.03–0.05; open squares: SOC = 0.02–0.03; open circles: SOC = 0.006–0.02); diagonal line is 1:1. A general trend to higher N_2O emissions for higher SOC is apparent in both field and simulated results, but a strong correlation between these two quantities is confounded by other factors (e.g., soil texture, weather, management). The correlation between the logarithms of modeled and measured annual N_2O emissions for these 38 sites was 0.82.

organic carbon (DOC), ammonium and nitrate, due to the high decomposition rates. Under most environmental conditions simulated nitrification and denitrification rates were robust, producing higher N_2O fluxes with increasing SOC contents. In contrast, at the sites with SOC less than 0.03 g C g^{-1} soil (grassland soils in Colorado and Inner Mongolia, China), DNDC simulated low populations of nitrifiers and denitrifiers, and low concentrations of DOC, ammonium, and nitrate, due to the low decomposition rates. Even when soil temperature and/or moisture were favorable for nitrification or denitrification, simulated N_2O fluxes were low. For intermediate SOC contents (0.03–0.05 g C g^{-1} soil), the sites presented in Figure 2 do not show a strong correlation between SOC and N_2O flux in either the field data or the simulation results. The impacts on annual N_2O emissions of variability in other soil environmental factors (e.g., temperature, moisture, Eh, pH, inorganic N concentration) masked any SOC influence on annual N_2O emissions at the sites with intermediate SOC. However, field observations at contiguous plots with different SOC contents, or at the sites amended with manure or other organic matter, show that increased SOC correlates with higher N_2O fluxes, even within the moderate SOC range (Robertson and Tiedje, 1987; Mosier et al., 1991; Bouwman et al., 2002; Thomson et al., 1997; Flessa et al., 1996; Dong et al., 2000; Christensen, 1983). In sensitivity simulations with DNDC, an increase in SOC caused an increase in N_2O fluxes, if other environmental factors were held constant (Li, 1995).

For this study we simulated, for present climate conditions, different management alternatives for three cropland sites in North America, Asia, and Europe (Table III):

- We simulated two 20-year management scenarios for a maize/soybean rotation in Iowa, USA: conventional tillage and no-till. In the United States, the use of no-till agricultural has significantly grown over the past 10 years, with 35% of soybeans, 18% of maize, 14% of sorghum, and 12% of small grains fields nationally utilizing no-till practices (Conservation Technology Information Center, 2002).
- We simulated two 20-year management scenarios for a maize-winter wheat double-cropping rotation in Hebei, China: 15% and 90% crop residue incorporation. In China, it is common practice to burn crop residues (e.g., stems and other non-harvested above-ground biomass) as an easy means of disposal, with little crop residue incorporated back into the soil. This has caused significant SOC loss in some regions (Li et al., 2003). New policies in China prohibit burning crop residue (Zhang, 2000) and farmers are strongly encouraged to incorporate crop residue into the soil (Lin, 1998).
- We simulated two 20-year management scenarios for a continuous barley rotation in Bavaria, Germany: manure application at 0.0 and 2.0 t C ha^{-1} yr^{-1}. In many countries, disposal of livestock manure is an important environmental issue (Gollehon et al., 2001). One option, applying manure to cropland soils, can also increase C-sequestration.

In each case, we calculated mean annual net soil carbon sequestration and mean annual net N_2O emissions from and CH_4 uptake by the soil over the 20-year simulation. Net N_2O and CH_4 fluxes were converted to 'CO_2-equivalents' using the global warming potential (GWP) concept, with 100-year time horizon GWP values (296 for N_2O and 23 for CH_4, Ramaswamy et al., 2001). Changes in these CO_2-equivalent emissions resulting from alternative management were then compared directly to the change in net carbon sequestration resulting from alternative management.

3. Results and Discussion

In all the three cases, alternative management increased both SOC content (i.e., sequestered carbon), and increased N_2O flux relative to baseline management (Table IV, Figure 3), at ratios of 0.006–0.02 additional kg N_2O-N emitted per additional kg SOC sequestered. Net methane uptake was small and changed little with alternative management (Table IV). No-till management at the Iowa site led to SOC accumulation at the soil surface, where the soil microbial population was high,

TABLE III
Soil, crop rotation, and management alternatives for three sites

		Soil				Management[a]	
Crop rotation	Site	Initial SOC (g C/g soil)	pH	Bulk density (g/cm^3)	Texture	Standard (STD)	Alternative (ALT)
Maize/soybean[b]	Adair County, Iowa, USA	0.024	6.35	1.35	Silty clay loam	Conventional till	No-till
Maize-wheat[c]	Jixian County, Hebei, China	0.009	7.0	1.50	Sandy loam	15% residue incorporation	90% residue incorporation
Barley	Pfaffenhofen County, Bavaria, Germany	0.0105	6.2	1.45	Silty loam	No manure	2 t manure C ha^{-1} y^{-1}

[a]Additional management factors were: conventional tillage (20 cm depth at planting and harvest) in all scenarios except USA no-till; 80% crop residue incorporation for all U.S.A. and Germany simulations; N-fertilization (urea) at 79 kg N ha^{-1} y^{-1} for USA maize, 30 kg N ha^{-1} y^{-1} for USA soybeans, 240 kg N ha^{-1} y^{-1} for China maize-wheat; 80 kg N ha^{-1} y^{-1} for Germany barley. Default crop yields (t dry matter ha^{-1}) were: maize: 10.3; soybean: 3.6; wheat: 6.0; barley: 3.0. Manure C:N ratio = 13.5.
[b]A two-year rotation of maize and soybeans.
[c]A one-year double cropping of maize and winter wheat.

TABLE IV

Simulated annual average soil C-sequestration (ΔSOC), N_2O, and CH_4 emissions for standard (STD) and alternative (ALT) management (see Table III for details), and change in CO_2-equivalent emissions (ALT minus STD) for a 100-yr time horizon

	ΔSOC^a (kg C ha^{-1} y^{-1})		N_2O flux[b] (kg N ha^{-1} y^{-1})		CH_4 flux[b] (kg C ha^{-1} y^{-1})		ALT minus STD (kg CO_2-equiv. ha^{-1} y^{-1})			
Site	STD	ALT	STD	ALT	STD	ALT	ΔSOC^c	N_2O^d	CH_4^e	Total
Iowa	−420	−1200	+7.4	+12	−2.6	−2.7	−2800	+2100	−3.1	−670
Hebei	+150	−680	+13	+20	−1.7	−2.0	−3100	+3200	−9.2	+140
Bavaria	−97	−440	+8.8	+17	−0.96	−1.2	−1300	+4000	−7.4	+2700

All values reported to two significant figures.

[a]Negative values imply net annual sequestration of carbon in soil; positive values imply net annual loss of carbon from soil.

[b]Flux sign convention is positive for net emissions from soil, negative for net uptake by soil.

[c]kg CO_2-equivalents ha^{-1} y^{-1} = $(\Delta SOC_{ALT} - \Delta SOC_{STD}) \times$ (44 kg CO_2/12 kg C).

[d]kg CO_2-equivalents ha^{-1} y^{-1} = $(N_2O_{ALT} - N_2O_{STD}) \times$ (44 kg N_2O/28 kg N) $\times$ 296.

[e]kg CO_2-equivalents ha^{-1} y^{-1} = $(CH_{4\,ALT} - CH_{4\,STD}) \times$ (16 kg CH_4/12 kg C) $\times$ 23.

and where drying/wetting shifts were more dramatic. This stimulated decomposition and N_2O emissions from both nitrification and denitrification, with an overall increase of 4.6 kg N_2O-N ha^{-1} y^{-1}. Higher N_2O fluxes have been measured in no-till versus conventionally-tilled cropland (Robertson et al., 2000; Aulakh et al., 1984). Denitrifier populations in the surface of no-till soils have been measured at 6–7 times that of populations in conventionally tilled soils (Aulakh et al., 1984; Doran, 1980). At the Hebei site, crop residue incorporation increased by ~800 kg C ha^{-1} y^{-1} and the SOC balance switched from a net loss to a net sink. Heavy fertilization of this double-cropped site (240 kg N ha^{-1} y^{-1}) provided favorable conditions for nitrification and denitrification when SOC increased, and N_2O emissions increased by 6.9 kg N ha^{-1} y^{-1}. Continuous amendment of readily decomposable manure at the Bavaria site increased the SOC content, and doubled N_2O emissions by increasing both nitrification and denitrification.

With alternative management the enhanced N_2O emissions, as CO_2-equivalents, offset 75–310% of the carbon sequestration in the soils, while the change in CO_2-equivalents due to changing CH_4 uptake was negligible (Table IV). The specific results for these simulations depended on site soil properties, details of management, and weather patterns; these would vary significantly if alternative management were applied across a region. For example, in regional DNDC simulations, we have typically found that simulated greenhouse gas fluxes vary by about 50% around the mean due to reported variability in soil properties, and that the impact of changing management on these fluxes has generally preserved this degree of variability while causing an overall shift in the mean fluxes (e.g., Li et al., 2002, 2003). Therefore, the numbers in Table IV should not be directly scaled to quantitative regional totals.

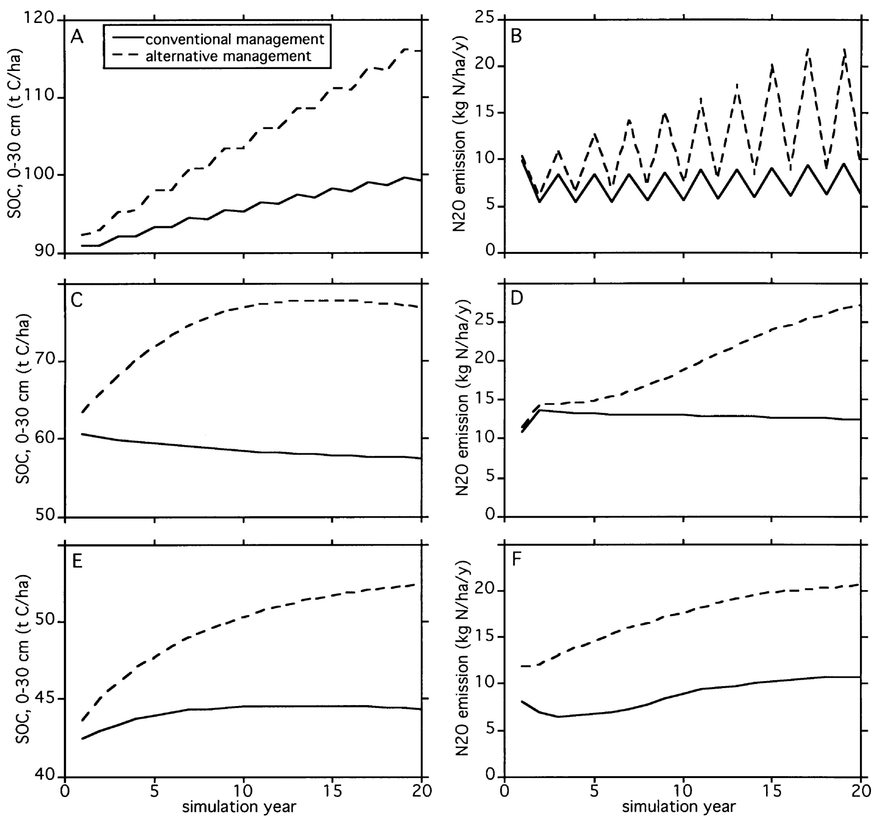

Figure 3. Simulated end-of-year soil organic carbon (SOC) content (0–30 cm) and total annual N_2O emission for conventional (solid lines) and alternative (dashed lines) management scenarios (see Table III for management details): (A) Iowa, USA, SOC; (B) Iowa, USA, N_2O; (C) Hebei, China, SOC; (D) Hebei, China, N_2O; (E) Bavaria, Germany, SOC; (F) Bavaria, Germany, N_2O. In each case the alternative management scenario led to increased SOC storage and to increased N_2O emissions. Note that vertical axis scales differ between panels. Changes in net methane uptake were small in all three cases. Overall average values and GWP calculations are presented in Table IV.

However, because there is a strong biogeochemical basis for N_2O flux varying with soil carbon sequestration, we believe that the general pattern of the results (i.e., increased N_2O emissions significantly offsetting the radiative forcing gains of any C-sequestration) is representative of the nature of the impact of widespread application of such mitigation scenarios.

Marland et al. (2003) noted that response of N_2O emissions to conversion from conventional to no-till can be highly variable, and in their analysis they adopted an increase in flux of 7 ± 15%, based on simulations with the NGAS model for 2639 cropland sites in the United States by Mummey et al. (1998). Six et al. (2004) reviewed published data on differences in fluxes of CO_2, CH_4, and N_2O for conventional tillage and no-till management, and they computed GWP values to compare the climate impacts of the three gases. Their analysis of the data indicated that, at least for the first decade or so, switching from conventional to no-till increased N_2O emissions by 2.5 ± 0.5 kg N ha^{-1} y^{-1} for humid environments (0.8 ± 1.0 kg N ha^{-1} y^{-1} for dry environments), which offset some of the C-sequestration gains in terms of radiative forcing of climate. Six et al. (2004) also found that, after 20 years, N_2O emissions returned to or dropped below conventional tillage fluxes. Methane GWP impacts also changed, but were small (Six et al., 2004). The DNDC simulations generated increases in N_2O emissions after five years of 1.7 kg N ha^{-1} y^{-1} for increased crop litter incorporation in China; 3.0 kg N ha^{-1} y^{-1} for conversion to no-till in the United States; and 7.9 kg N ha^{-1} y^{-1} for manure amendment (at a rate of 148 kg manure-N ha^{-1} y^{-1}) in Germany. The DNDC simulations also projected that these increases would continue to grow for at least 20 years (Figure 3), and probably until SOC pools stabilize at higher levels, which may take many decades (e.g., Li et al., 1994). The DNDC simulations were of well-fertilized plots, and did not account for interannual variability in weather or pest infestations that could influence field results.

There are additional greenhouse gas emissions associated with agricultural production that have not been included in our biogeochemical analysis. In particular, there are significant CO_2 emissions associated with production of fertilizer and pesticides, production and operation of farm machinery, operation of irrigation equipment, etc. Changes in these emissions associated with changing management practices can also significantly influence net greenhouse gas emissions, can persist after SOC pools have stabilized, and should be included in full greenhouse gas accounting (West and Marland, 2002; Marland et al., 2003). In addition, N_2O emission is only one of many fates for nitrogen in agro-ecosystems; others include volatilization as NO, N_2, or NH_3, leaching or erosional losses, removal in harvested material, changes in soil inorganic and organic N pools. In national scale simulations for croplands in China and the U.S. with DNDC, $<4\%$ of total N flux out of the soil was as N_2O (Li et al., 2003). A management scenario that mitigates climate change might have other negative or positive environmental impacts that can be analyzed with biogeochemical modeling.

Carbon sequestration in soil has benefits beyond removal of CO_2 from the atmosphere. No-till cropping reduces fossil fuel use, reduces soil erosion, and enhances soil fertility and water-holding capacity (Conservation Tillage Information Center, 1999). Organic matter amendments enhance soil fertility and water holding capacity (potentially reducing fertilization and irrigation requirements), reduce emissions of air pollutants from crop residue burning, and provide a depository for excess farmyard manure from animal feedlot operations.

Evaluating the greenhouse gas benefit of regional-scale changes in management practices aimed at C-sequestration requires analysis of interacting biogeochemical cycles, coupled with spatial datasets of weather data and soil properties. Unless these biogeochemical interactions are incorporated into a comprehensive assessment framework, the value of agricultural systems in strategies for climate protection cannot be accurately determined. Our analysis indicates that increased C-sequestration in soils, by any mechanism, will be generally accompanied by increased N_2O emissions, reducing or eliminating the usefulness of C-sequestration in soils as a greenhouse gas mitigation strategy.

Acknowledgments

We thank R.C. Harriss and W. Seiler for helpful comments. We also thank Gregg Marland and two anonymous reviewers for suggestions on improving the manuscript. This work was supported by NASA grants NAG5-10135, NAG5-12838, and NAG5-7631 to UNH, and by European Commission grant EVK2-CT2001-00106. The DNDC model (executable version, source code, and user's guide) is available for free over the Internet at www.dndc.sr.unh.edu.

References

Ambus, P. and Christenson, S.: 1994, 'Measurement of N_2O emission from a fertilized grassland: An analysis of spatial variability', *J. Geophys. Res.* **99**, 16549–16555.

Aulakh, M. S., Rennie, D. A., and Paul, E. A.: 1984, 'Gaseous nitrogen losses from soils under zero-till as compared with conventional-till management systems', *J. Environ. Qual.* **13**, 130–136.

Bouwman, A. F., Boumans, L. J. M., and Batjes, N. H.: 2002, 'Modeling global annual N_2O and NO emissions from fertilized fields', *Global Biogeochem. Cycles* **16**(4), doi:10.1029/2001GB001812.

Bowman, R. A. and Focht, D. D.: 1974, 'The influence of glucose and nitrate concentrations upon denitrification rates in a sandy soil', *Soil Biol. Biochem.* **6**, 297–301.

Bremner, J. M. and Shaw, K.: 1958, 'Denitrification in soil. II. Factors affecting denitrification', *J. Agric. Sci.* **51**, 40–52.

Burford, J. R. and Bremner, J. M.: 1975, 'Relationships between denitrification capacities of soils and total, water-soluble and readily decomposable soil organic matter', *Soil Biol. Biochem.* **7**, 389–394.

Cai, Z., Sawamoto, S., Li, C., Kang, G., Boonjawat, J., Mosier, A., and Wassmann, R.: 2003, 'Field validation of the DNDC model for greenhouse gas emissions in East Asian cropping systems', *Global Biogeochem. Cycles.* **17**(4), doi:10.1029/2003GB002046.

Christensen, S.: 1983, 'Nitrous oxide emission from a soil under permanent grass: Seasonal and dismal fluctuations as influenced by manuring and fertilization', *Soil Biol. Biochem.* **15**, 531–536.

Christensen, S., Simkins, S., and Tiedje, J. M.: 1990, 'Spatial variation in denitrification: Dependency of activity centers on the soil environment', *Soil Sci. Soc. Am. J.* **54**, 1608–1613.

Clayton, H., McTaggart, I. P., Parker, J., Swan, L., and Smith, K. A.: 1997, 'Nitrous oxide emissions from fertilized grassland: A 2-year study of the effects of N fertilizer form and environmental conditions', *Biol. Fert. Soils* **25**, 252–260.

Conrad, R.: 1996, 'Soil microorganisms as controllers of atmospheric trace gases (H_2, CO, CH_4, N_2O and NO)', *Microbiol. Rev.* **60**, 609–640.

Conservation Technology Information Center.: 1999, *National Survey of Conservation Tillage Practices*. (Conservation Tillage Information Center, West Lafayette, IN).

Conservation Technology Information Center.: 2002, *National Crop Residue Management Survey* (www.ctic.purdue.edu). (Conservation Tillage Information Center, West Lafayette, IN).

Crill, P., Keller, M., Weitz, A., Grauel, B., and Veldkamp, E.: 2000, ' Intensive field measurements of N_2O emissions from a tropical agricultural soil', *Global Biogeochem. Cycles* **14**, 85–96.

Dong, Y. S., Scharffe, D., Domroes, M., Qi, Y. C., and Zhang, S.: 2000, 'N_2O emissions from agricultural soils in the North China Plain: The effect of chemical fertilizer and organic manure', *J. Environ. Sci.* **12**, 463–468.

Doran, J. W.: 1980, 'Soil microbial and biochemical changes associated with reduced tillage', *Soil Sci. Soc. Am. J.* **44**, 765–771.

Duxbury, J. M., Bouldin, D. R., Terry, R. E., and Tat, R. L.: 1982, 'Emissions of nitrous oxide from soils', *Nature* **298**, 462–464.

Federer, C. A. and Klemedtsson, L.: 1988, 'Some factors limiting denitrification in slurries of acid forest soils', *Scand. J. For. Res.* **3**, 425–435.

Flessa, H., Dorsch, P., and Beese, F.: 1995, 'Seasonal variation of N_2O and CH_4 fluxes in differently managed arable soils in southern Germany', *J. Geophy. Res.* **100**, 23115–23124.

Flessa, H., Dorsch, P., Beese, F., Konig, H., and Bouwman, A. F.: 1996, 'Influece of cattle wastes on nitrous oxide and methane fluxes in pasture land', *J. Environ. Qual.* **25**, 1366–1370.

Flessa, H., Wild, U., Klemisch, M., and Pfadenhauer, J.: 1998, 'Nitrous oxide and methane fluxes from organic soils under agriculture', *Eur. J. Soil Sci.* **49**, 327–335.

Gollehon, N., Caswell, M., Ribaudo, M., Kellogg, R., Lander, C., and Letson, C.: 2001, *Confined Animal Production and Manure Nutrients*. Resource Economics Division, Economic Research Service, U. S. Department of Agriculture. Agriculture Information Bulletin No. 771. 39 pp.

Goodroad, L. L. and Keeney, D. R.: 1984, 'Nitrous oxide emission from forest, marsh, and prairie ecosystems', *J. Environ. Qual.* **13**, 448–452.

Grace, P. R.: 1996, 'The Waite Permanent Rotation Trial' in Powlson, D. S., Smith, P., and Smith, J. U. (eds.), *Evaluation of Soil Organic Matter Models Using Existing, Long-Term Datasets*, NATO ASI Series I, Vol. 38, Springer-Verlag, Heidelberg, pp. 335–340.

Grant, B., Smith, W. N., Desjardins, R., Lemkc, R., and Li, C.: 2004, 'Estimated N_2O and CO_2 emissions as influenced by agricultural practices in Canada', *Clim. Change* **65**, 315–332.

Hao, X., Chang, C., Carefoot, J. M., Janzen, H. H., and Ellert, B. H.: 2001, 'Nitrous oxide emissions from an irrigated soil as affected by fertilizer and straw management', *Nutr. Cycle Agroecosys.* **60,** 1–8.

Harrison, R. M., Yamulki, S., Goulding, K. W. T., and Webster, C. P.: 1995, 'Effect of fertilizer application on NO and N_2O fluxes from agricultural fields', *J. Geophys. Res.* **100**, 25923–25931.

Jenkinson, D.: 1991, 'The Rothamsted long-term experiments: Are they still of use?', *Agron. J.* **83**, 2–10.

Jin, X. and Gruber, N.: 2003, 'Offsetting the radiative benefit of ocean iron fertilization by enhancing N_2O emissions', *Geophys. Res. Lett.* **30**(24), 2249, doi:10.1029/2003/GL018458.

Khalil, M. I., Rosenani, A. B., Van Cleemput, O., Fauziah, C. I., and Shamshuddin, J.: 2002, 'Nitrous oxide emissions from an ultisol of the humid tropics under maize-groundnut rotation', *J. Environ. Qual.* **31**, 1071–1078.

Klingensmith, K. M.: 1987, 'Denitrification in floodplain successional soils of the Tanana River in interior Alaska', *Agroborealis* **19**, 39–42.

Körchens, M. and Müller, A.: 1996, 'The static experiment at Bad Lauchstädt, Germany', in Powlson, D. S., Smith, P., and Smith, J. U. (eds.), *Evaluation of Soil Organic Matter Models Using Existing, Long-Term Datasets*, NATO ASI Series I, Vol. 38, Springer-Verlag, Heidelberg, pp. 369–376.

Lackner, K. S.: 2003, 'A guide to CO_2 sequestration', *Science* **300**, 1677–1678.

Lal, R., Follett, R. F., Kimble, J. M., and Cole, C. V.: 1999, 'Management of U. S. cropland to sequester carbon in soil', *J. Soil Water Conserv.* **54**, 374–381.

Larcher, W.: 1995, *Physiological Plant Ecology*, 3rd Edn., Springer, Berlin, 506 pp.

Leffelaar, P. A. and Wessel, W. W.: 1988, 'Denitrification in a homogeneous, closed system: experiment and simulation', *Soil Sci.* **146**, 335–349.

Li, C.: 1995, 'Impact of agricultural practices on soil C storage and N_2O emissions in 6 states in the U.S.', in Lal, R., Kimble, J., Levine, E., and Stewart, B. A. (eds.), *Soil Management and Greenhouse Effect*, Lewis Publishers, Boca Raton FL, pp. 101–112.

Li, C.: 2000, 'Modeling trace gas emissions from agricultural ecosystems', *Nutr. Cycl. Agroecosys.* **58**, 259–276.

Li, C., Frolking, S., and Frolking, T. A.: 1992, 'A model of nitrous oxide evolution from soil driven by rainfall events: 1. Model structure and sensitivity', *J. Geophy. Res.* **97**, 9759–9776.

Li, C., Frolking, S., and Harriss, R. C.: 1994, 'Modeling carbon biogeochemistry in agricultural soils', *Global Biogeochem. Cycles* **8**, 237–254.

Li, C., Narayanan, V., and Harriss, R.: 1996, 'Model estimates of nitrous oxide emissions from agricultural lands in the United States', *Global Biogeochem. Cycles* **10**, 297–306.

Li, C., Qiu, J. J., Frolking, S., Xiao, X., Salas, W., Moore, B., Boles, S., Huang, Y., and Sass, R.: 2002, 'Reduced methane emissions from large-scale changes in water management of China's rice paddies during 1980–2000', *Geophys. Res. Lett.* **29**(20), doi:10.1029/2002GL015370.

Li, C., Zhuang, Y. H., Frolking, S., Galloway, J. N., Harriss, R. C., Moore, B., Schimel, D., and Wang, X. K.: 2003, 'Modeling soil organic carbon change in croplands of China', *Ecol. Appl.* **13**, 327–336.

Lin, X.: 1998, 'Status and management of soil organic matter in China', in Shen, S. (ed.), *Soil Fertility of China*, Chinese Agricultural Press, Beijing, pp. 111–159 (in Chinese).

Marland, G., West, T. O., Schlamadinger, B., and Canella, L.: 2003, 'Managing soil organic carbon in agriculture: The net effect on greenhouse gas emissions', *Tellus* **55B**, 613–621.

Melillo, J. M., Aber, J. D., Steudler, P. A., and Schimel, J. P.: 1983, 'Denitrification potentials in a successional sequence of northern hardwood forest stands', in Hallberg, R. (ed.), *Environmental Biogeochemistry*, *Ecol. Bull.* (Stockholm) **35**, 217–228.

Mosier, A., Schimel, D., Valentine, D., Bronson, K., and Parton, W.: 1991, 'Methane and nitrous oxide fluxes in native, fertilized and cultivated grasslands', *Nature* **350**, 330–332.

Mosier, A. R., Stillwell, M., Parton, W. J., and Woodmansee, R. G.: 1981, 'Nitrous oxide emissions from a native shortgrass prairie', *Soil Sci. Soc. Am. J.* **45**, 617–619.

Müller, M. M., Sundman, V., and Skujins, J.: 1980, 'Denitrification in low pH spodosols and peats determined with the acetylene inhibition method', *Appl. Environ. Microbiol.* **40**, 235–239.

Mummey, D. L., Smith, J. L., and Bluhm, G.: 1998, 'Assessment of alternative soil management practices on N_2O emissions from US agriculture', *Agric., Ecosys. Environ.* **70**, 79–87.

Odell, R. T., Melsted, S. W., and Walker, W. M.: 1984, 'Changes in organic carbon and nitrogen of morrow plot soils under different treatments, 1904–1973', *Soil Sci.* **137**, 160–171.

Pang, P. C. K. and Cho, C. M.: 1984, 'Oxygen consumption and denitrification activity of a conifer forest soil profile', *Soil Sci. Soc. Am. J.* **48**, 393–399.

Papen, H. and Butterbach-Bahl, K.: 1999, 'Three years continuous record of N-trace gas fluxes from untreated and limed soil of a N-saturated spruce and beech forest ecosystem in Germany: I. N_2O-emissions', *J. Geophys. Res.* **104**, 18487–18503

Pluth, D. J. and Nommik, H.: 1981, 'Potential denitrification affected by nitrogen source of a previous fertilization of an acid forest soil from Central Sweden', *Acta Agric. Scand.* **31**, 235–241.

Ramaswamy, V., Boucher, O., Haigh, J., Hauglustaine, D., Haywood, J., Myhre, G., Nakajima, T., Shi, G. Y., and Solomon, S.: 2001, 'Radiative forcing of climate change', in Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., Dai, X., Maskel, K., and Johnson, C. A. (eds.), *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, Cambridge, UK, pp. 350–416.

Robertson, G. P. and Tiedje, J. M.: 1987, 'Nitrous oxide sources in aerobic soils: nitrification, denitrification and other biological processes', *Soil Biol. Biochem.* **19**, 187–193.

Robertson, G. P., Paul, E., and Harwood, R. R.: 2000, 'Greenhouse gases in intensive agriculture: Contributions of individual gases to the radiative forcing of the atmosphere', *Science* **289**, 1922–1925.

Saggar, S., Andrew, R. M., Tate, K. R., Hedley, C. B., Rodda, N. J., and Townsend, J. A.: 2004, 'Modelling nitrous oxide emissions from dairy-grazed pastures', *Nutr. Cycling Agroecosys.* **68**, 243–255.

Sahrawat, K. L. and Keeney, D. R.: 1986, 'Nitrous oxide emission from soils', *Adv. Soil. Sci.* **4**, 103–147.

Six, J., Ogle, S. M., Breidt, F. J., Conant, R. T., Mosier, A. R., and Paustian, K.: 2004, 'The potential to mitigate global warming with no-tillage management is only realized when practiced in the long term', *Global Change Biol.* **10**, 155–160.

Smith, P., Smith, J. U., Powlson, D. S., Arah, J. R. M., Chertov, O. G., Coleman, K., Franko, U., Frolking, S., Gunnewick, H. K., Jenkinson, D. S., Jensen, L. S., Kelly, R. H., Li, C., Molina, J. A. E., Mueller, T., Parton, W. J., Thornley, J. H. M., and Whitmore, A. P.: 1997, 'A comparison of the performance of nine soil organic matter models using datasets from seven long-term experiments', *Geoderma* **81**, 153–225.

Smith, P., Goulding, K. W., Smith, K. A., Powlson, D. S., Smith, J. U., Falloon, P., and Coleman, K.: 2001, 'Enhancing the carbon sink in European agricultural soils: Including trace gas fluxes in estimates of carbon mitigation potential', *Nutr. Cycling Agroecosys.* **60**, 237–252.

Smith, W. N., Desjardins, R. L., Grant, B., Li, C., Lemke, R., Rochette, P., Corre, M. D., and Pennock, D.: 2002, 'Testing the DNDC model using N_2O emissions at two experimental sites in Canada', *Can. J. Soil Sci.* **82**, 365–374.

Stanford, G., Vander Pol, R. A., and Dzienia, S.: 1975, 'Denitrification rates in relation to total and extractable soil carbon', *Soil Sci. Soc. Am. Proc.* **39**, 284–289.

Terry, R. E., Tate, R. L., and Ducbury, J. M.: 1981, 'Nitrous oxide emissions from drained, cultivated organic soils of south Florida', *J. Air Pollut. Control Assoc.* **31**, 1173–1176.

Terry, R. E.: 1986, 'Nitrogen transformations in histosols', in Chen, Y., and Avnimelech, Y. (eds.), *The Role of Organic Matter in Modern Agriculture*, Martinus Nijhoff Publishers, Dordrecht, Netherlands, pp. 55–70.

Thomson, P. E., Parker, J. P., Arah, J. R. M., Clayton, H., and Smith, K. A.: 1997, 'Automated soil monolith-flux chamber system for the study of trace gas fluxes', *Soil Sci. Soc. Am. J.* **61**, 1323–1330.

U.S.D.A. (United States Department of Agriculture): 1992, Su*pp. to the Morrow plots: A century of Learning*, Bull. No. 775, Agriculture Experiment Station, University of Illinois, Champaign-Urbana, IL.

U.S.D.O.E. (United States Department of Energy): 1999, *Carbon Sequestration: Research and Development*. Office of Science, Office of Fossil Energy, U.S. Department of Energy, Washington, DC.

Velthof, G. L., Kuikman, P. J., and Oenema, O.: 2002. 'Nitrous oxide emission from soils amended with crop residues', *Nutr. Cycl. Agroecosys.* **62,** 249–261.

Vinther, F. P.: 1992, 'Measured and simulated denitrification activity in a cropped sandy and loamy soil', *Biol. Fert. Soils* **14**, 43–48.

West, T. O. and Marland, G.: 2002, 'A synthesis of carbon seqeuestration, carbon emissions, and net carbon flux in agriculture: Comparing tillage practices in the United States', *Agric. Ecosys. Environ.* **91**, 217–232.

Wever, H., Mussen, S., and Merckx, R.: 2002, 'Dynamics of trace gas production following compost and NO_3^- amendments to soil at different initial TOC/NO_3^- ratios', *Soil Biol. Biochem.* **34**, 1583–1591.

Xuri, X., Wang, M., and Wang, Y.: 2003, 'Using a modified DNDC model to estimate N_2O fluxes from semi-arid grassland in China', *Soil Biol. Biochem.* **35**, 615–620.

Yamulki, S., Wolf, I., Bol, R., Veldcamp, E., Grant, B., Brumme, R., and Jarvis, S.: 2000, 'Effects of dung and urine amenedments on the isotopic content of N_2O released from grasslands', *Rapid Commun. Mass Spectr.* **14**, 1356–1360.
Zhang, B.: 2000, 'Severe status of agricultural eco-environment in China', *China News*, 6 June 2000 (in Chinese).

(Received 29 December 2003; in revised form 25 October 2004)