

Chapter 19

Soil Carbon and Nitrogen Interactions and Biosphere-Atmosphere Exchange of Nitrous Oxide and Methane

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Abstract Carbon (C) sequestration in terrestrial ecosystems and here specifically in soils is currently discussed as a potential strategy to contribute to reducing atmospheric carbon dioxide (CO₂) concentrations. However, increases in soil C stocks may also have adverse effects on the exchange of greenhouse gases (GHGs) between terrestrial ecosystems and the atmosphere. In view of the unprecedented perturbation of the global nitrogen (N) cycle, increases in soil C stocks and the ongoing saturation of terrestrial ecosystems with reactive forms of N (Nr) may result in a stimulation of soil nitrous oxide (N₂O) emissions. These largely unexplored ecosystem C-N interactions and their importance for biosphere-atmosphere GHG exchange need to be better understood to finally assess the climate benefits of C sequestration in soils.

Keywords CN interactions • N₂O • CH₄ • Nitrification • Denitrification

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Abbreviations

NH ₃	ammonia
NH ₄ ⁺	ammonium
C	Carbon
CO ₂	carbon dioxide
N ₂	dinitrogen
GHGs	greenhouse gases
CH ₄	methane
NO ₃ ⁻	nitrate
NO	nitric oxide
N	nitrogen
N ₂ O	nitrous oxide
Nr	reactive forms of N
SOC	soil organic carbon pools
SOM	soil organic matter

19.1 Introduction

Decomposition of soil carbon (C) stocks following conversion of natural soils to agricultural soils and intensification of agricultural management has significantly contributed to a depletion of soil organic carbon pools (SOC) worldwide. Lal (2004) pointed out that following cultivation of natural soils 60% or up to 75% of the SOC pools may be lost in temperate or tropical regions, respectively. It has been estimated that since 1850 changes in land use may have resulted in a loss of carbon (C) from terrestrial ecosystems of about 156 Pg C (Houghton 2007), with losses of SOC due to mineralization amounting to about one-third or 52 ± 8 Pg C and losses due to erosion to 26 ± 9 Pg C (Lal 2004). In the last years the reversion of historical SOC losses via the intentional implementation of improved land management practices – e.g., higher inputs of residue C, adoption of less intensive cropping systems or temporary vegetative cover between agricultural crops – is discussed as a potential strategy to contribute to reducing atmospheric carbon dioxide (CO₂) concentrations (Smith et al. 2008; Conant et al. 2011). However, increasing SOC stocks will not only affect ecosystem C cycling, but also ecosystem nitrogen (N) cycling and soil microbial processes involved in the production and oxidation of other non-CO₂ greenhouse gases (GHGs), namely nitrous oxide (N₂O) and methane (CH₄). In addition, changes in global N cycling as driven by human activities need also to be considered while implementing strategies to increasing soil C sequestration, since additional N inputs and increased availability of soil C may alter the net balance of the biosphere atmosphere exchange of GHGs at the ecosystem as well as at the landscape scale. This chapter will therefore provide examples of how C-N interactions and changes in N trace gas exchange between terrestrial ecosystems and the atmosphere may potentially affect the climate benefits of C sequestration in soils.

19.1.1 *Perturbation of the Global Nitrogen Cycling and Soil Carbon-to-Nitrogen Ratios*

Human activities have not only resulted in a perturbation of the global C cycle, but even more significantly in an unprecedented change of global N cycling. Following the introduction of industrial ammonia (NH_3) production via the Haber-Bosch process N fertilizers have increasingly been used to meet the demand of the growing world population for food and feed (Erisman et al. 2009). Furthermore, N fixing crops are increasingly cultivated also adding N to the biosphere. Finally, reactive N (Nr) compounds are also created incidentally during combustion processes. The human perturbation of the natural N cycle has led to an unprecedented accumulation of Nr in the biosphere (Erisman et al. 2009). For 2005 it is estimated that due to human activities, 187 Tg of N have been added to the global biosphere, which approximately relates to a doubling of global N cycling (Galloway et al. 2008). Not all of this N is ultimately denitrified, but a substantial, though hardly to quantify amount of Nr may remain in terrestrial ecosystems and here mainly in the soil. Galloway et al. (2004) estimated that from the 0.268 Pg N which were added annually during the 1990s to terrestrial ecosystems approximately 0.060 Pg N year⁻¹ may be stored in terrestrial ecosystems, i.e., 20–25% of the total input of Nr to terrestrial ecosystems. In contrast, it is assumed that in pre-industrial times Nr inputs and outputs were balanced. In conclusion, it is very much likely to assume that Nr is accumulating in the global biosphere and that Nr stocks of all terrestrial ecosystems are increasing.

What is the consequence of this? Batjes (1996) estimated that global soil C stocks for 0–30 cm are in the range of 906–969 Pg C and that to 100 cm soil depth 2,150–2,300 Pg C may be stored. For soil N stocks the respective numbers are: =63–67 Pg N for the 0–30 cm soil layer and 133–140 Pg N for 0–100 cm soil layer, respectively. Following Galloway et al. (2004) that from the 0.268 Pg N year⁻¹ added to terrestrial ecosystems 0.060 Pg N year⁻¹ are remaining in ecosystems, and assuming that the major fate is its storage in soils this will lead to a narrowing of soil C:N ratios (Fig. 19.1). If soil C stocks are not increasing and 10% of the additional Nr is stored in the topsoil (0–30 cm) the global average soil C:N ratio may narrow for the first 30 cm of soil from 14.4 to 13.8 in the coming 100 years (Fig. 19.1). However, soil C:N ratios may further widen if soils get re-carbonized, whereas if soil C loss due to cultivation of soils continues the narrowing of soil C:N ratio may accelerate (Fig. 19.1).

Significant changes in forest floor and mineral topsoil soil C:N ratios following simulated increased atmospheric N deposition has been observed e.g., in a study by Andersson et al. (2002) for a series of *Picea abies* (L.) stands in Sweden or by Lovett and Godale (2011) for a mixed oak wood stand in Northeastern US. In a report for the German Environmental Agency, Kiese et al. (2009) provided data for a pine forest in the Northeastern German Lowlands where due to increased atmospheric N deposition forest floor C:N ratios decreased within 20 years from 31 to 27 and in the top 10 cm of the mineral soil from 26 to 23,

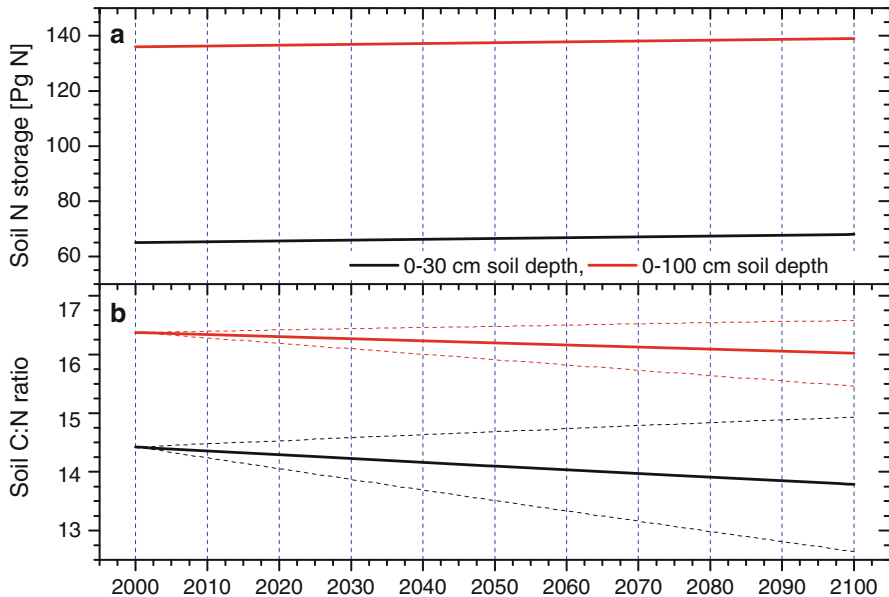


Fig. 19.1 Changes in global soil N stocks from 2000 to 2100 assuming that soil C stocks are remaining unchanged, anthropogenic N inputs are remaining at the same level as in the 1990s and that total N additions to terrestrial ecosystems are equaling $0.268 \text{ Pg N year}^{-1}$ (Galloway et al. 2004) and that about 10% or $0.030 \text{ Pg year}^{-1}$ of the N input to terrestrial ecosystems are either stored in the topsoil (0–30 cm) or in soils to 100 cm depth. (a) Shows changes in total soil N stocks, whereas (b) displays resulting changes in the soil C:N ratio either for 0–30 cm or 0–100 cm soil depth, respectively. Data for global soil N and soil C stocks were taken from Batjes (1996). Dashed lines in the (b) are indicating changes in the soil C:N ratio if C loss due to cultivation continues or if soils are re-carbonized. For both scenarios changes in soil C stocks (either 0–30 cm or 0–100 cm) at a rate of $78 \text{ Pg C } 100 \text{ year}^{-1}$ were assumed

respectively (Fig. 19.2). These observed decreases could be simulated well by a biogeochemical model based on reported data of atmospheric N deposition at the forest site (Kiese et al. 2009).

19.1.2 Coupling of Carbon and Nitrogen Turnover Processes

As pointed out earlier the re-carbonization of the biosphere has been highlighted recently as an important approach to mitigate the increase and to consolidate atmospheric CO_2 concentrations by converting the atmospheric CO_2 into biotic or abiotic C sequestered in vegetation or soil pools. Due to the tight-coupling of C and N cycles in soils and ecosystems (Fig. 19.3) – e.g., mineralization of organic matter does not only lead to break down of C substrates and the release of CO_2 but also to the liberation of inorganic N – anthropogenic induced changes in rates of biotic C-sequestration in terrestrial ecosystems will directly affect N turnover processes

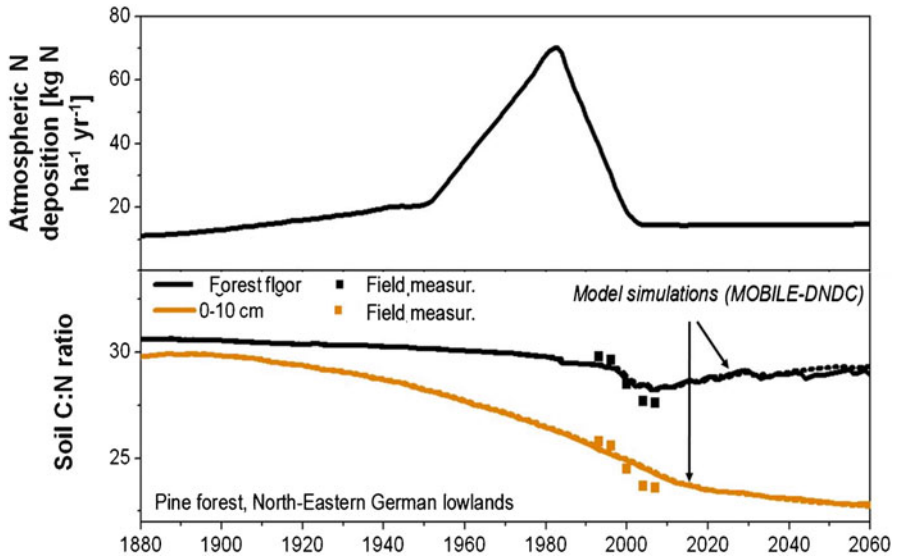


Fig. 19.2 Observed changes in forest floor and soil (0–10 cm) C:N ratios (*points lower graph*) of a pine stand in the Northeastern German Lowlands as a response to increased atmospheric N deposition (*upper graph*). The *lower graph* also displays simulated changes in soil C:N ratios using the biogeochemical model MOBILE-DNDC, and historic and predicted climate and N deposition data as drivers. For further details see Kiese et al. (2009)

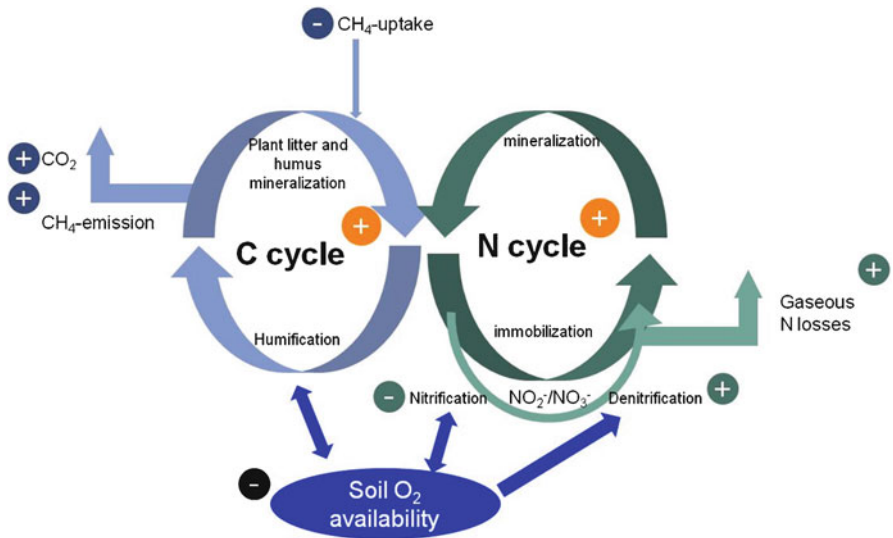


Fig. 19.3 Coupling of ecosystem C and N cycling and possible consequences of an acceleration of both cycles with regard to soil-atmosphere C and N exchange. +: enhancement -: attenuation (Figure modified following Blagodatsky et al. 2011)

in soils, and thus, also the biosphere–atmosphere exchange of gaseous N and C compounds (Li et al. 2005).

Consequently, increased rates of C sequestration in soils are likely to accelerate not only C but also N turnover, in particular when agricultural N fertilizer use and atmospheric N deposition in many regions globally remain at the present high levels. Increased C and N cycling may result in increased soil anaerobiosis, due to increased microbial oxygen consumption during mineralization of organic matter. This may lead to increases in soil anaerobiosis, which may result in elevated soil N₂O emissions and a weakening of the sink strengths of upland soils for atmospheric CH₄ (Butterbach-Bahl et al. 2011), due to a stimulation of denitrification and methanogenesis versus plant and microbial N immobilization or CH₄ oxidation in soils.

19.2 Soil Organic Carbon and Magnitude of Nitrous Oxide Emissions

In their literature review on C:N interactions, Li et al. (2005) revealed a very significant positive relationship between SOC content of soils and the magnitude of N₂O emissions (Fig. 19.4). There are several reasons why SOC contents are a major control of soil N₂O emissions. First, organic C is the basis for any biotic mineralization process and with increasing SOC contents – except for conditions of strict

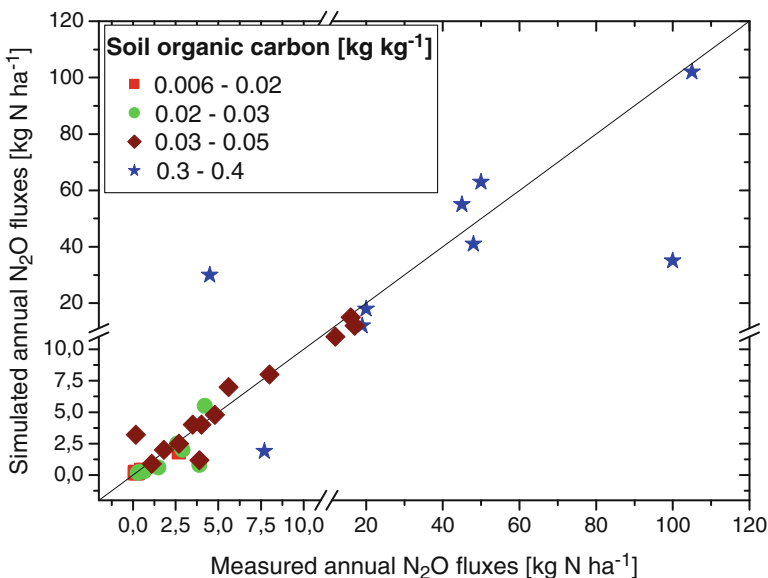


Fig. 19.4 Comparison of observed and simulated annual N₂O emissions from agricultural soils. Symbols indicate soil organic carbon (SOC) content in soil in kg SOC kg⁻¹ soil. A general trend to higher N₂O emissions for higher SOC is apparent in both field and simulated results (Figure adapted from Li et al. 2005)

anaerobiosis like in peatlands or if soil moisture/temperature is hampering microbial activity like in arid and semi-arid regions – mineralization activities are increasing too. The oxygen consumption by microbial mineralization may result in an increased frequency and increased spatial extend of anaerobic microsites, where mineralized and oxidized inorganic Nr may be transformed by nitrification and denitrification processes into N_2O . Second, the availability of labile C substrates is a prerequisite for denitrification besides the availability of oxidized Nr substrates. Simultaneous availability of sufficient concentrations of readily available C and Nr substrates for denitrification is more likely to occur in soils rich in organic matter, while in soils with low organic C content denitrification activity and, thus, also N_2O formation, may even be hampered due to missing C substrates. Such a situation is e.g. occurring in many arable soils in the North China Plain, one of the key regions of food production in China. For this region, several reports indicate, that soil N_2O emissions are rather low at mineral fertilizer application of up to $600 \text{ kg N ha}^{-1} \text{ year}^{-1}$ with annual cumulative emission rates of $<4 \text{ kg N}_2\text{O-N ha}^{-1} \text{ year}^{-1}$ (Ju et al. 2011; Liu et al. 2010). Also the N fertilizer N_2O emission factor of 0.5–0.7% of added fertilizer (Ding et al. 2007) for wheat-maize (*Triticum spp.-Zea mays* L.) rotations in the North China Plain is significantly below the global average of 1% as suggested by IPCC (2006). The main reason for low N_2O emissions seems to be the low C content of the soils and not the availability of nitrate (NO_3^-), which accumulates in the soil. Most likely C substrate supply limits denitrification and, thus, also N_2O production by denitrification. If these soils are re-carbonized, the large stocks of inorganic Nr in surface and subsurface soils may largely get denitrified, thereby potentially releasing significant amounts of N_2O as by-products of denitrification. Comparable situations, i.e., that the availability of readily available C is limiting denitrification and N_2O formation has been described for agro-ecosystems as well as for natural ecosystems (Morley and Baggs 2010; Kammann et al. 2008; Butterbach-Bahl and Dannenmann 2011).

19.3 Soil C:N Ratio and Microbial N Turnover Processes

The major long-term sink of atmospheric Nr input into ecosystems is soil organic matter (SOM), as was shown in many ^{15}N -tracing experiments (Morier Jaquet et al. 2008; Nadelhoffer et al. 1999; Tietema et al. 1998). Since Nr deposition via atmospheric pathways is often exceeding plant N demand in natural and semi-natural ecosystems, the atmospheric N input has led to N saturation of natural and semi-natural ecosystems, resulting in a decrease in the soil C:N ratio both in the organic layer and in the mineral soil of natural and semi-natural ecosystems within large regions of Europe, Asia and North America (Galloway et al. 2004; Corré et al. 2007; Velthoff et al. 2011; Butterbach-Bahl et al. 2011). Even if atmospheric Nr input in terrestrial ecosystems may not further increase in future, the C:N ratio in mineral topsoils will further decrease (Fig. 19.1). The N saturation and associated decline in soil C:N ratios involves detrimental effects such as soil eutrophication, -acidification,

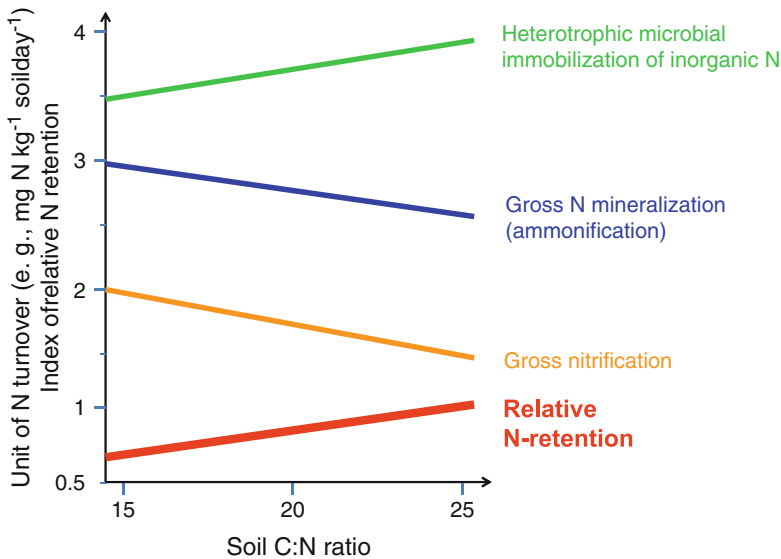


Fig. 19.5 Schematic representation of the relationships between soil C:N ratio and gross rates of inorganic N production (ammonification, nitrification) and consumption (microbial immobilization). When C:N ratios narrow below a value of approximately 25, e.g. as a consequence of atmospheric Nr deposition, gross ammonification as well as nitrification will increase, while heterotrophic microbial immobilization of inorganic Nr will decrease. Thus, the index of microbial Nr retention in the soil (i.e., the ratio between gross microbial immobilization and total gross inorganic N production) is decreasing, indicating an opening of the N cycle and increased N₂O emissions. Mechanisms of impacts of the C:N ratio on single N cycle processes are provided in the text

reduced plant biodiversity and reduction in tree population, while it has a large potential to increase C sequestration in plant biomass and eventually also in soil (Snyder et al. 2009; Velthoff et al. 2011).

Besides these effects, the C:N ratio is – as well as total C and N content and -availability – a major driver of soil microbial N turnover processes. This is not only the case for processes such as ammonification, nitrification, denitrification and microbial immobilization of inorganic N, but for the relative importance or even dominance of single processes such as nitrification over competing processes such as microbial ammonium (NH₄⁺) immobilization. In particular, the relative importance of nitrification and denitrification over the processes of microbial immobilization of inorganic Nr (and mycorrhizal/plant uptake) determine whether there are large or small losses of N₂O from soil (Tietema and Wessel 1992; Stockdale et al. 2002; Butterbach-Bahl et al. 2011). By regulating this delicate balance, the C:N ratio plays a crucial role in the characterization of the N cycle, i.e., if it is open (high N loss along gaseous and hydrological pathways) or closed (internal N cycling facilitates ecosystem N retention). In the following section, the effect of the C:N ratio on ecosystem Nr retention and loss is explained at the level of the single N cycle processes (Fig. 19.5).

The lower the soil C:N ratio, the larger the N yield during depolymerization and mineralization of a given amount of SOM. Thus, ammonification will increase with decreasing C:N ratio (Frankenberger and Abdelmagid 1985; Hart et al. 1994). Subsequently, the balance of partitioning of NH_4^+ to the potentially competing microbial processes of heterotrophic immobilization into biomass of free living microorganisms and autotrophic nitrification (i.e. conversion to NO_3^-), as well as plant or mycorrhizal uptake of NH_4^+ , determines whether N is retained in the ecosystem or subject to increased risk of N loss via gaseous or hydrological pathways.

Due to the long residence time of Nr in plant biomass, plant Nr uptake results in persistent ecosystem N retention until disturbances such as harvest or fire. However, plants are in general poor competitors for Nr against microbes at the process level (Rennenberg et al. 2009). Even in fast growing spruce (*Picea* spp) forests, plant N uptake may at least be one order of magnitude lower than gross N mineralization, thus consuming only for a minor part of inorganic N produced in soil (Kreutzer et al. 2009).

Heterotrophic NH_4^+ assimilating microorganisms generally tend to outcompete autotrophic nitrifiers (Verhagen and Laanbroek 1991; Hart et al. 1994; Verhagen et al. 1995; Booth et al. 2005). However, this may not necessarily be the case in agricultural soils, when there is no NH_4^+ limitation (Burger and Jackson 2003). The availability of labile C compounds is the major controller of heterotrophic microbial immobilization of NH_4^+ -N (Woodmansee and Duncan 1980; Compton and Boone 2002; Booth et al. 2005; Accoe et al. 2004, 2005). At decreasing C:N ratios, microbial N uptake may be C-limited, leaving more NH_4^+ available to the process of autotrophic nitrification, thus opening pathways to potential N loss such as N_2O emission to the atmosphere.

In contrast to nitrification, the incorporation of NH_4^+ -N into microbial cell walls, characterized by short residence times, leads to retention of N in the ecosystem. This may be of short-term, when there is dieback and remineralization of microbial residues leading to a rapid and repeated re-allocation of mineral N to the soil NH_4^+ pool. However, despite the rapid turnover of microbial biomass in soil, microbial immobilization can also lead to long-term stabilization of N. Among the processes are microbially mediated long-term N stabilization by the accumulation of bacterial-derived residues such as proteins, amino acids and amino sugars on the surface of clay minerals (Sollins et al. 2006). Decreasing soil C:N ratios could also reduce the long-term N stabilization in soil via reducing microbial immobilization, which could contribute to increased risk of N loss. Hence, the C:N ratio is of crucial importance for the balance of NH_4^+ partitioning to processes promoting either N retention or N loss. Beside its effect on nitrification at the substrate level, the C:N ratio is also affecting the balance of consumption of the end product of nitrification. Specifically, heterotrophic microbial NO_3^- immobilization will also be dependent on C availability, i.e., decreasing with decreasing C:N ratio, promoting accumulation of soil NO_3^- , which would be available for denitrification and associated N_2O production and -loss.

Via its effects on ammonification, nitrification as well as on microbial immobilization, the soil C:N ratio is a crucial determinant of the microbial Nr retention

capacity (Booth et al. 2005). The effect of the soil C:N ratio on microbial N retention was shown in some field studies, e.g., for a Rendzic Leptosol in a mountainous beech (*Fagus sylvatica* L.) forest in Southern Germany. In this study, variations in soil C:N ratio triggered by different microclimate and management were found to be correlated with microbial N retention, i.e., decreased C:N ratios lead to decreased relative importance of microbial N retention (Dannenmann et al. 2006, 2007).

By promoting nitrification, the decreasing C:N ratio may also promote the formation of N_2O as a by-product. Besides its effect on nitrification, also the formation of N_2O as a facultative end-product of denitrification may be increased, when a narrow C:N ratio is leading to NO_3^- accumulation in soil, promoting denitrification in general, while impairing the last step of denitrification, i.e. the conversion of N_2O to the end product dinitrogen (N_2) catalyzed by the enzyme N_2O reductase (e.g., Groffman et al. 2006; Wang et al. 2011). Thus, narrowing C:N ratios favour both N_2O production by nitrification and denitrification. However, denitrification requires also labile C substrates. In this context, increased root exudation of monomeric C compounds, promoted by generally improved plant growth as a consequence of atmospheric Nr deposition and, in particular, due to increased atmospheric CO_2 levels, may further promote denitrification (Kammann et al. 2008; Butterbach-Bahl and Dannenmann 2011). Denitrification may be further promoted by increased soil respirations and plant water use efficiency, both increasing the soil anaerobic volume in a changing climate (Butterbach-Bahl and Dannenmann 2011). Hence, in a changing climate, NO_3^- accumulation in soil as a consequence of Nr deposition and narrowed C:N ratios may increasingly meet environmental conditions favourable for denitrification, bearing the potential for high N_2O emissions from soil.

19.4 Soil Carbon-to-Nitrogen Ratios and Soil Nitrogen Trace Gas Emissions

The most important soil microbial processes involved in the formation of N_2O and nitric oxide (NO) are nitrification and denitrification. The magnitude of both processes will strongly depend on the availability of Nr substrates. For autotrophic nitrification NH_3 is the relevant Nr substrate, while for denitrification inorganic Nr oxides as well as labile C substrates are needed. Heterotrophic nitrifiers may use also organic N compounds instead of NH_3 to produce NO_3^- and produce as side products as well NO and N_2O . Therefore, a close link between Nr and C availability and N_2O emissions can be expected. Specifically, close relationship between soil C:N ratios and magnitude of N trace gas emissions have been shown for forest soils in various climate zones (Fig. 19.6). Based on soil emission measurements at various tropical rain forest sites in Queensland, Australia, and a detailed analysis of soil properties, Breuer et al. (2000) showed that topsoil C:N ratios are a significant predictor for understanding the spatial variability of soil N_2O emissions. Comparable results were also reported by Pilegaard et al. (2006) while analyzing environmental controlling variables of soil N_2O and NO emissions for 15 different forest ecosystem sites across Europe (Fig. 19.6). At least for N_2O a significant relationship was found

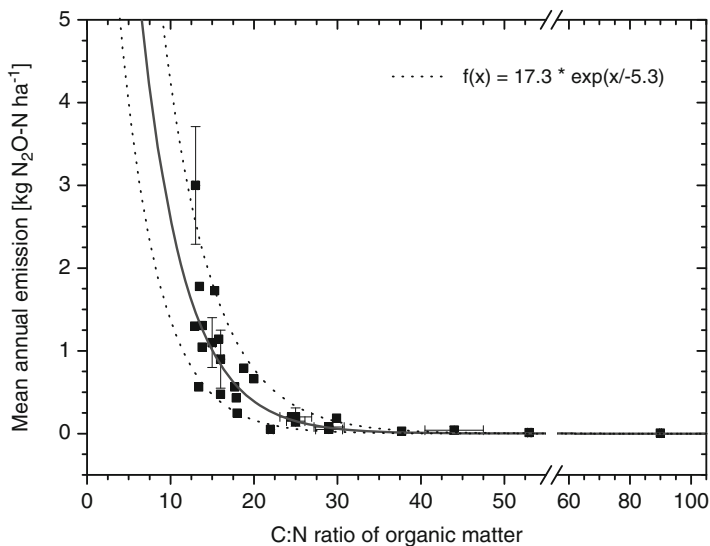


Fig. 19.6 Correlation between topsoil C:N ratio and mean annual nitrous oxide emissions for different forest sites across Europe (Data were taken from Table 1 of Klemetsson et al. (2005) and Tables 2 and 5 from Pilegaard et al. (2006))

with exponential increasing soil N_2O emission rates with decreasing soil C:N ratios in the topsoil. The study of Klemetsson et al. (2005) on N_2O emissions from drained forested histosols in Sweden shows that a strong negative relationship between N_2O emissions and soil C:N ratios exist. This relationship has been successfully used to estimate emissions at other sites in Finland and Germany.

The exponential increase in forest soil N_2O emissions with decreasing C:N ratios is in good agreement with the observation that the risk for elevated NO_3^- leaching from forest soils is increasing exponentially if soil C:N ratios are smaller than 25 (Gundersen et al. 2006; Butterbach-Bahl et al. 2011). In both cases the ecosystem N retention capacities are likely to be exceeded and losses of N to the hydrosphere and atmosphere are increasing. With regard to the re-carbonization of soils this clearly indicates, that the N status of the ecosystem needs to be explored, e.g., by recording soil C:N ratios as indicators – and it will be necessary to consider total ecosystem N inputs either by atmospheric N-input and/or by atmospheric-N-input plus organic or inorganic fertilizers or even weathering of bedrock N (Morford et al. 2011).

19.5 Nitrogen Availability and Ecosystem Carbon Sequestration

Input of Nr to terrestrial ecosystems is also a measure to increase ecosystem C sequestration. For agricultural systems Nr has been reported to play an important role in soil C storage either by promoting crop dry matter production and/ or by

chemically stabilizing C in the soil (Snyder et al. 2009). However, though several studies have shown that nitrogen fertilization results in higher levels of soil C over time (Paustian et al. 1992; Wilts et al. 2004) the stimulating effect may be rather small (about 2%) as shown in the meta-data analysis by Liu and Greaver (2009). Besides agricultural ecosystems, several recent studies have shown that Nr deposition to European forest ecosystems has resulted in increased forest growth and C sequestration (De Vries et al. 2006, 2011; Schulze et al. 2010). With regard to the net climate balance of atmospheric Nr deposition, C sequestration was thereby largely outweighing reductions in soil CH₄ uptake or stimulations in soil N₂O emissions (De Vries et al. 2011). However, long-term assessments are still missing and the stimulation of forest growth may cease following a few decades while stimulative effects on soil N₂O emissions may persist over longer periods of time.

19.6 Conclusions

Re-carbonization of terrestrial ecosystems is likely to be a sustainable strategy to stabilize or even lowering of atmospheric CO₂ concentrations. Nevertheless, it still needs to be assessed if specifically an increase in soil C stocks will indeed lead to a net reduction of GHG emissions from terrestrial ecosystems, i.e., if benefits due to C sequestration are not specifically outweighed by the stimulation of N₂O emissions from soils. The reason for this is the unprecedented perturbation of the global N cycle. If current rates of direct (fertilization) and indirect reactive N inputs (mainly atmospheric N deposition) continues, C:N ratios in soils may further narrow even if additional C is sequestered in soils. Both, C accumulation as well as the narrowing of soil C:N ratios may result in a stimulation of microbial N turnover and associated emissions of N₂O, which has as a global warming potential of about 300 over a 100-year period as compared to CO₂. Therefore, a thorough quantification of the biogeochemical interaction of C and N needs to be incorporated into assessment frameworks to accurately evaluate the climate benefits of a re-carbonization of the terrestrial environment.

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