

ORIGINAL ARTICLE

Quantifying greenhouse gas emissions from soils: Scientific basis and modeling approach

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Global climate change is one of the most important issues of contemporary environmental safety. A scientific consensus is forming that the emissions of greenhouse gases, including carbon dioxide, nitrous oxide and methane, from anthropogenic activities may play a key role in elevating the global temperatures. Quantifying soil greenhouse gas emissions is an essential task for understanding the atmospheric impacts of anthropogenic activities in terrestrial ecosystems. In most soils, production or consumption of the three major greenhouse gases is regulated by interactions among soil redox potential, carbon source and electron acceptors. Two classical formulas, the Nernst equation and the Michaelis–Menten equation, describe the microorganism-mediated redox reactions from aspects of thermodynamics and reaction kinetics, respectively. The two equations are functions of a series of environmental factors (e.g. temperature, moisture, pH, Eh) that are regulated by a few ecological drivers, such as climate, soil properties, vegetation and anthropogenic activity. Given the complexity of greenhouse gas production in soils, process-based models are required to interpret, integrate and predict the intricate relationships among the gas emissions, the environmental factors and the ecological drivers. This paper reviews the scientific basis underlying the modeling of greenhouse gas emissions from terrestrial soils. A case study is reported to demonstrate how a biogeochemical model can be used to predict the impacts of alternative management practices on greenhouse gas emissions from rice paddies.

Key words: biogeochemical model, greenhouse gas, inventory, mitigation, soil.

INTRODUCTION

Global climate change is becoming a hot issue in contemporary science as well as politics. There is a long-lasting debate about the cause of the climate change: anthropogenic activity versus the natural cycle (e.g. Crowley 2000; Cuffey 2004; Damon and Kunen 1976). However, driven by the rapidly accumulating observations worldwide, a scientific consensus is forming that the contemporary climate change (e.g. temperature increase) is mainly caused by anthropogenic emissions of the greenhouse gases, including carbon dioxide (CO₂), nitrous oxide (N₂O) and methane (CH₄) (Crutzen *et al.* 1979; Intergovernmental Panel on Climate Change 2001). Soil plays an important

role as a source or sink of greenhouse gases in almost all terrestrial ecosystems. Quantifying the impacts of mitigating strategies on soil greenhouse gas (GHG) emissions is a core task for identifying the best management practices in agriculture or forestry. This paper provides a review of the scientific basis and modeling approach for quantifying soil GHG emissions from terrestrial soils.

GREENHOUSE GAS PRODUCTION DRIVEN BY MICROBIAL ACTIVITY

Soil microorganisms play a key role in the production and consumption of CO₂, N₂O and CH₄ in most terrestrial ecosystems. The microbes survive and gain energy by breaking the carbon (C) bonds of dissolved organic compounds. To accomplish this process, electrons must be transferred from the dissolved organic carbon (DOC) to electron acceptors. Among the oxidants commonly existing in soils, oxygen (O₂) possesses the lowest Gibbs free energy and, hence, is the first candidate electron receiver. During the process of electron transfer, the ionized oxygen will be combined with the dissociated C to form

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CO₂ in the microbial cells. Under aerobic conditions, most soil microbes can use oxygen as an electron acceptor and release CO₂ into the atmosphere. This process leads to decomposition, a reaction that dominates the losses of soil organic carbon (SOC). Soil aeration status sensitively depends on soil physical conditions. For example, during a rainfall or irrigation event, the top layers of the soil can be saturated by water that will block the diffusion of atmospheric O₂ into the soil profile. While the soil microbes continuously consume the O₂ left in the soil pores, the soil O₂ partial pressure will drop rapidly. The depletion of O₂ will depress a wide range of decomposers, but will stimulate a group of special microbes, denitrifiers in the soil. The denitrifiers are capable of using nitrate (NO₃⁻), an oxidative form of nitrogen, as an electron acceptor. By receiving electrons, NO₃⁻ will become nitrite (NO₂⁻). Nitrite can be further reduced to nitric oxide (NO), nitrous oxide (N₂O) and finally dinitrogen (N₂). During the sequential reactions of denitrification, if the intermediate product N₂O can escape from the anaerobic micro-sites before it has been further reduced, a net emission of N₂O will occur. If a soil has been placed under anaerobic conditions for a long time (e.g. several days), the major oxidants, such as O₂, nitrate, manganese (Mn⁴⁺), iron (Fe³⁺) and sulfate, will be depleted by the decomposers, denitrifiers, manganese bacteria, iron bacteria and sulfur bacteria, respectively. In this case, methanogens will be activated to use hydrogen as an electron acceptor that will result in CH₄ production (Fig. 1). The above-described mechanisms have been observed by many researchers in their field and laboratory studies worldwide (e.g. Cappenberg 1974; De Groot *et al.* 1994; Gambrell and Patrick 1978; Jakobsen *et al.* 1981; Loveley and Phillips 1987; Takai and Kamura 1966).

In general, CO₂, N₂O or CH₄ is a byproduct of the microbial survival, which is characterized by a transfer of electrons. This type of reaction is categorized as a reduction–oxidation (or redox) reaction. In soils, the major factors controlling the redox processes are redox potential (i.e. Eh in volts), DOC concentration, and the relevant electron acceptor (e.g. O₂, nitrate, Mn⁴⁺, Fe³⁺, sulfate or hydrogen) concentration. Soil Eh determines if a redox reaction can occur; and if it occurs, the contents of DOC and the electron acceptor will collectively determine the rate of the biogenic reaction. Any mitigation measure, if it can alter one or more of the three factors (i.e. Eh, DOC or electron acceptor), will change the emissions of CO₂, N₂O or CH₄. Thus, it is essential for quantifying GHG emissions to understand how the impacts of Eh, DOC and the electron acceptor on greenhouse gases can be quantitatively determined.

Two classical formulas, the Nernst equation and the Michaelis–Menten equation, can be used to describe the interactions between the three driving factors and the microbial activities. The Nernst equation is a basic thermodynamic formula defining soil Eh based on concentrations of the dominant oxidants and reductants existing in the soil liquid phase (Stumm and Morgan 1981):

$$Eh = Eo + RT/nF \times \ln([O]/[W]) \quad (1)$$

where Eh is redox potential (V), Eo is standard redox potential (V), R is gas constant, T is temperature in kelvin, n is number of electrons transferred in the redox reaction, F is Faraday constant, [O] is the concentration of the oxidant (mol L⁻¹) and [W] is the concentration of the reductant (mol L⁻¹).

The Michaelis–Menten equation is a widely applied formula describing the kinetics of microbial growth with dual nutrients (Paul and Clark 1989):

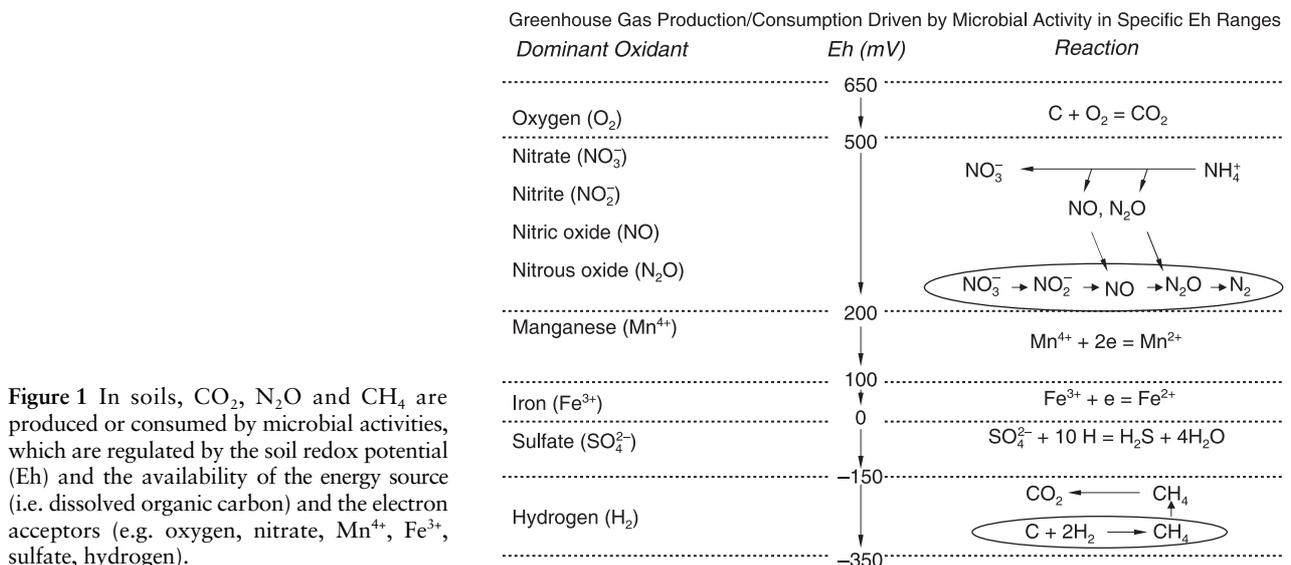


Figure 1 In soils, CO₂, N₂O and CH₄ are produced or consumed by microbial activities, which are regulated by the soil redox potential (Eh) and the availability of the energy source (i.e. dissolved organic carbon) and the electron acceptors (e.g. oxygen, nitrate, Mn⁴⁺, Fe³⁺, sulfate, hydrogen).

$$R = R_{\max} \times \text{DOC} / (\text{K}_a + \text{DOC}) \times [\text{O}] / (\text{K}_b + [\text{O}]) \quad (2)$$

where R is reaction rate, R_{\max} is maximum reaction rate, DOC is concentrations of dissolved organic C, [O] is concentrations of oxidant, and K_a and K_b are half-saturation constants for substrates DOC and oxidant, respectively.

As the Nernst and Michaelis–Menten equations share a common item, oxidant concentration ([O]), the two equations can be integrated to track the feedback between Eh dynamics and the microbe-mediated redox reaction in a soil.

CONSTRUCTION OF VIRTUAL ECOSYSTEMS BASED ON BIOGEOCHEMICAL CONCEPTS

Redox potential, DOC and the electron acceptor are the major, but not the only, factors determining GHG emissions from soils. For example, temperature, moisture and pH can also affect the biochemical or geochemical reactions related to soil CO_2 , N_2O or CH_4 emissions. In general, there are seven environmental factors, namely gravity, radiation, temperature, moisture, pH, Eh and substrate concentration gradient, that control GHG production/consumption in soils. Most of the environmental factors are variable in space and time and are driven by a few ecological drivers, including climate, soil properties, vegetation and anthropogenic activity. The ecological drivers, the environmental factors and the reactions compose a complex system, which governs transport and transformation of chemical elements, including C and nitrogen (N), in ecosystems. Concepts of biogeochemistry have been adopted to cope with the complex systems.

Biogeochemistry is a scientific discipline studying the relationship between life and its environment through defining, understanding and tracking the movement of chemical elements (Vernadsky 1944). Biogeochemistry consists of four concepts, abundance, field, coupling and cycling, which regulate the interactions between life and its environment (Li 2001). Biogeochemical abundance explores the correlation between the elemental compositions of life and its inorganic environment (e.g. atmosphere, crust, soil, sea water). During the last several decades, various chemical and physical analytical methods have been used to identify the abundances of essential or trace elements in various life forms and the environment. These measurements have provided fundamental datasets for understanding the essential correlation between life and its environment through comparison of the biogeochemical abundances. Biogeochemical field is the assemblage of environmental forces or factors, including gravity, radiation, temperature, moisture, pH, Eh or redox potential,

and substrate concentration gradients. These factors construct a multi-dimensional, biogeochemical field in which the transport and transformation of chemical elements occur. The biogeochemical field plays a key role in integrating various factors and processes into an entirety, such as an ecosystem. A biogeochemical field is produced by primary drivers (e.g. climate, soil, topography, vegetation and anthropogenic activity) in a specific ecosystem, and it determines all of the relevant biochemical or geochemical processes and, hence, the ecosystem evolution. Mathematically expressing the biogeochemical field is a key step to predicting the transport and transformation of chemical elements in ecosystems. Biogeochemical modeling reconstructs the dynamics of the biogeochemical fields as they continually vary in space and time. Biogeochemical coupling describes how chemical elements must act in specific couples or groups during their movement in the environment. In most terrestrial ecosystems, the processes of coupling/decoupling of chemical elements are realized through a series of biochemical or geochemical reactions, including mechanical movement, dissolution/crystallization, decomposition/combination, oxidation/reduction, adsorption/desorption, complexation/decomplexation and assimilation/dissimilation. As do all chemical reactions, each of these processes has two directions to lead to elemental coupling or decoupling in various forms. The direction and rate of each reaction is usually controlled by more than one environmental factor. Elemental coupling and decoupling through the biochemical or geochemical reactions are driven by both internal factors (e.g. atomic structure, bond energies, electronegativities) and external factors (e.g. gravity, radiation, temperature, moisture, pH, Eh, substrate concentration gradients) forming the biogeochemical field. A myriad of coupling/decoupling phenomena shape the complexity of an element's biogeochemical cycle. Theoretical analyses of thermodynamics, chemical reaction kinetics, bond energy/enthalpy and quantum chemistry have been used to predict the coupling/decoupling phenomena occurring within the biotic bodies (e.g. antagonistic and synergistic effects of the elements) or the environment (e.g. Hedin *et al.* 1998; Li *et al.* 2000). Biogeochemical cycling represents the ultimate movement of chemical elements through transport in space and transformation in time at the interface between life and its environment, in response to the biogeochemical field and within the constraints of biogeochemical abundance and coupling. Through elemental cycling, life interacts with and shapes its environment by means of exchanges of energy, matter and information. These four concepts, biogeochemical abundance, field, coupling and cycling, are interwoven to define the scientific scope of biogeochemistry and, hence, determine its methodologies. Based on these concepts, the dynamics

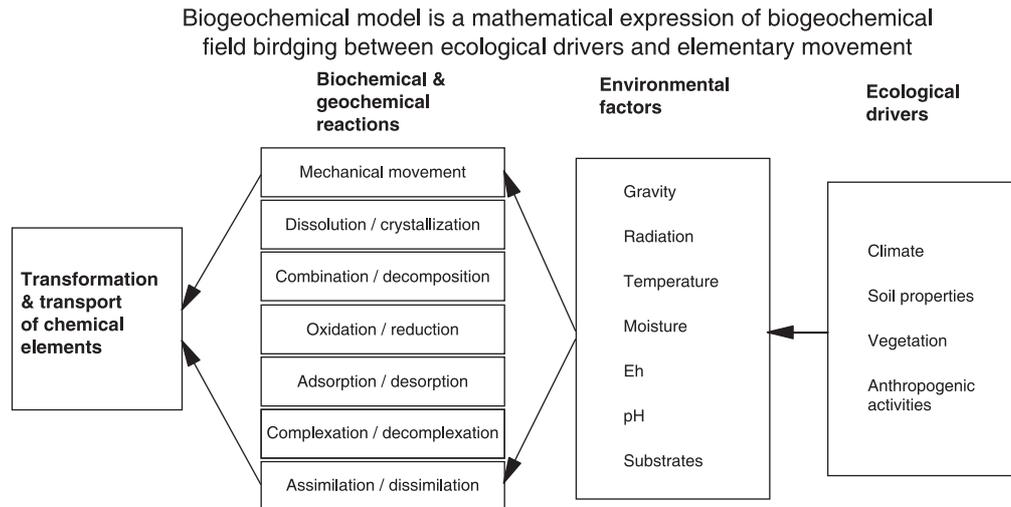


Figure 2 Biogeochemical field consists of seven environmental factors and bridges between the ecological drivers (e.g. climate, soil, vegetation and anthropogenic activity) and the biochemical or geochemical reactions that determine elemental coupling, decoupling and cycling in ecosystems.

of an ecosystem can be disaggregated into four components: primary drivers, biogeochemical field, biogeochemical coupling/decoupling and biogeochemical cycles (Fig. 2). Quantifying the biogeochemical field for each specific ecosystem is an essential task for predicting biogeochemical processes or ecosystem evolution. Any single change in the primary drivers, such as climate, soil, vegetation or human activity, can simultaneously cause changes in several of the environmental factors (e.g. radiation, temperature, moisture, redox potential and substrate concentration gradients); and any single change in the environmental factors can simultaneously affect several biochemical or geochemical reactions, which collectively determine the patterns and rates of elemental cycles in the ecosystem. For example, a change in precipitation could simultaneously alter radiation, soil temperature, moisture, Eh and substrate concentration gradients; these changes will simultaneously and collectively affect decomposition, nitrification and denitrification, which interact to determine how much N_2O will ultimately be emitted from the soil. It is almost impossible to determine a quantitative relationship between the cause (a change in precipitation) and the consequence (N_2O fluxes) through simple correlation or regression analysis. In addition, the extreme spatial and temporal heterogeneity of many of the primary drivers has obscured the relationship between the causes and effects for many of the biogeochemical processes. That is why most of the correlations between a deviation in the primary drivers and the changes in biogeochemical cycles caused by the deviation are inherently non-linear, and even sometimes random or chaotic. The challenge is to describe this kind of complexity in a mathematical framework for

prediction. The four concepts of biogeochemistry were developed to provide the intellectual framework to help undertake this challenge.

As shown in Fig. 2, a virtual ecosystem consists of two components as two bridges linking the ecological drivers and the environmental factors and the environmental factors and the biochemical or geochemical reactions, respectively. Scientific laws in physics, chemistry and biology as well as equations empirically generated from experiments can be incorporated in the framework to express the quantitative relationships among the linked factors or processes. Within the virtual ecosystem, any change in a single factor could cause variation in multiple factors or processes in the system. For example, a change in irrigation could simultaneously alter the soil temperature, moisture, Eh and substrate concentrations; the altered environmental factors could collectively affect dissolution/crystallization, oxidation/reduction, adsorption/desorption and other reactions that will finally change the production and consumption of CO_2 , N_2O or CH_4 in the soil. Converting the virtual ecosystem into a computable program will build a biogeochemical model to serve GHG or other ecological or environmental studies.

A BIOGEOCHEMICAL MODEL FOR SIMULATING GREENHOUSE GAS EMISSIONS

During the past 15 years with support from US federal agencies (e.g. Environmental Protection Agency (EPA), National Science Foundation (NSF), National Aeronautics and Space Administration (NASA), National Oceanic

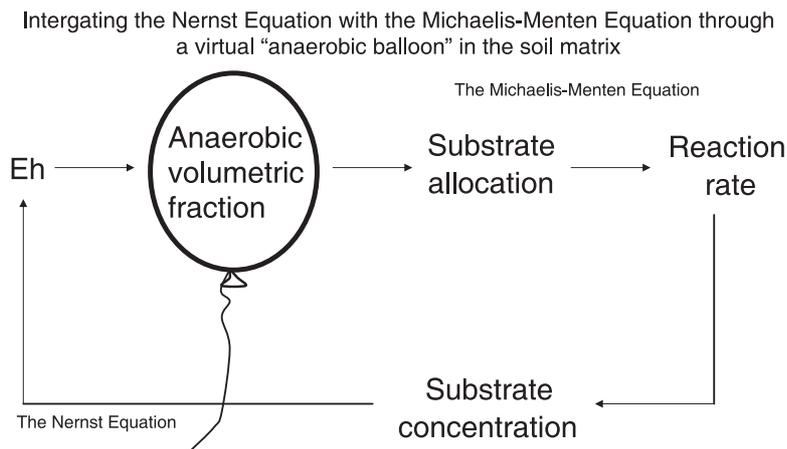


Figure 3 A computable kinetic scheme “anaerobic balloon” was invented to link the Nernst equation to the Michaelis–Menten equation for tracking the interaction between the soil redox conditions and the microbial activities.

and Atmospheric Administration (NOAA) and United States Department of Agriculture (USDA)) in collaboration with a wide range of international researchers, a process-based model, Denitrification–Decomposition or DNDC, was developed based on the biogeochemical concepts. The model was originally developed for estimating carbon sequestration and trace gas emissions from US agriculture (Li *et al.* 1992, 1994, 1996), but was extended to more ecosystems and regions. The core of DNDC is a soil biogeochemical model with integrated Nernst and Michaelis–Menten equations to track microbial activities. A computing scheme, “anaerobic balloon”, was developed in DNDC to realize the integration of the two equations in the modeling framework. The anaerobic balloon is defined as the volumetric fraction of anaerobic microsites in a soil. The size of the balloon varies between 0 and 1, representing fully aerobic and anaerobic conditions, respectively. With the Nernst equation, DNDC calculates soil Eh based on concentrations of oxygen, nitrate or other dominant electron acceptors in the soil. And then the size of the anaerobic balloon is defined based on the modeled Eh. Microsites within the balloon are anaerobic and those outside the balloon are aerobic. The substrates (e.g. DOC, ammonium, nitrate) will then be proportionally allocated into the aerobic and anaerobic fractions. Only the substrates allocated in the aerobic fraction will participate in the oxidative reactions (e.g. nitrification, CH_4 oxidation); and the substrates allocated in the anaerobic fraction will participate in the reductive reactions (e.g. denitrification, CH_4 production). Given the substrate contents partitioned into the aerobic and anaerobic microsites, rates of the relevant oxidative and reductive reactions will be calculated based on the Michaelis–Menten equation. Based on the modeled consumption rates of the substrates involved in the redox reactions, DNDC will reestablish the substrate concentrations and the new Eh value. Through the computing loop of “Eh definition–substrate

allocation–microbial activity–substrate consumption–Eh change”, DNDC tracks dynamics of soil Eh as well microbial activity to quantify the production and consumption of greenhouse gases at hourly or daily time steps (Fig. 3). The DNDC simulates a series of anaerobic balloons driven by different electron acceptors. If a soil is fully aerobic (Eh +350 mV), O_2 will be used as the dominant electron acceptor by the soil microbes. In this case, CO_2 is the major gas produced in the soil. During a rainfall or irrigation event, the soil O_2 can be gradually depleted to drive the oxygen-driven anaerobic balloon to swell. When the soil O_2 is totally gone, the oxygen-driven balloon will reach its maximum and burst, and then a new balloon will appear driven by the next electron acceptor, nitrate. Within the nitrate-driven anaerobic balloon, denitrification will occur to produce nitrite, NO, N_2O and N_2 sequentially. When the soil nitrate is depleted, the nitrate-driven balloon will burst and another balloon will arise driven by Mn^{4+} . If the soil anaerobic conditions last long enough, the major electron acceptors (e.g. O_2 , nitrate, Mn^{4+} , Fe^{3+} and sulfate) will be sequentially depleted, which will drive the soil Eh to -150 mV or lower. In this case, methanogens will be stimulated and CH_4 will be produced using hydrogen as the electron acceptor. Thus, by tracking swelling and shrinking of a series of anaerobic balloons, DNDC is able to simulate the soil Eh dynamics as well the production and consumption of CO_2 , N_2O and CH_4 at different stages of the soil Eh evolution.

By linking the balloon scheme with other algorithms defining soil climate, crop growth and farming management practices (e.g. tillage, fertilization, manure amendment, irrigation, grazing), DNDC estimates GHG emissions across climatic zones, soil types and management regimes. During the past decade, DNDC has been independently tested by a number of researchers worldwide with promising results (Brown *et al.* 2002; Butterbach-Bahl *et al.* 2001; Cai *et al.* 2003; Grant *et al.* 2004; Jagadeesh

Babu *et al.* 2006; Kesik *et al.* 2005; Kiese *et al.* 2005; Pathak *et al.* 2005; Saggarr *et al.* 2003; Smith *et al.* 1997, 2004; Xu-Ri *et al.* 2003; Zhang *et al.* 2006). The results indicate that computer models equipped with biogeochemical processes could form a reliable tool for quantifying GHG emissions.

BRINGING UNCERTAINTY UNDER CONTROL FOR UPSCALING

One of the major purposes for developing process-based models is to extend our understanding gained at specific sites to regional, national or global scales. In fact, policy decisions can only be made based on their effectiveness at regional or national scales. When a process-based model is applied to a regional scale, an issue of uncertainty will arise even when the model may have been well calibrated and validated at a site scale. The uncertainty mainly results from spatial heterogeneity of the input parameters used for the upscaling. For applying this type of model to a region, the region is routinely divided into many grid cells or polygons, with an assumption that each grid cell is uniform in all its properties. This assumption is against the fact that some of the input parameters, especially soil properties, usually vary even within small scales, such as a county or a farm. Averaging the variations of the input data may not resolve the problem because the correlation between the modeled output (e.g. GHG flux or other concerned items) and the drivers is non-linear. There are a number of measures to cope with this challenge. A relatively simple approach, the Most Sensitive Factor (MSF) method, was recently developed with DNDC applications (Li *et al.* 1996, 2004). Through sensitivity tests, the most sensitive factors for a concerned output (e.g. CO₂, N₂O or CH₄) can be identified. For example, initial SOC content has been identified as the most sensitive factor for soil N₂O emissions in many case studies (Li *et al.* 2002). During the input database construction, the most sensitive factor (e.g. SOC content) was assigned two values for each grid cell, the maximum and minimum values observed within the cell. During the model simulation, DNDC runs twice for each grid cell with the two extreme values (e.g. the maximum and minimum SOC contents) in the cell. The two runs will produce two N₂O fluxes to form a range, which is assumed to be wide enough to include the “real” N₂O flux with a high probability. This methodology has been verified against the Monte Carlo method, a classical tool for uncertainty tests. The results indicated that 60–90% of the Monte Carlo method induced CO₂, N₂O or CH₄ fluxes were located within the ranges produced by the MSF method (Li *et al.* 2004) (Fig. 4). By constructing the regional database with the maximum and minimum values for

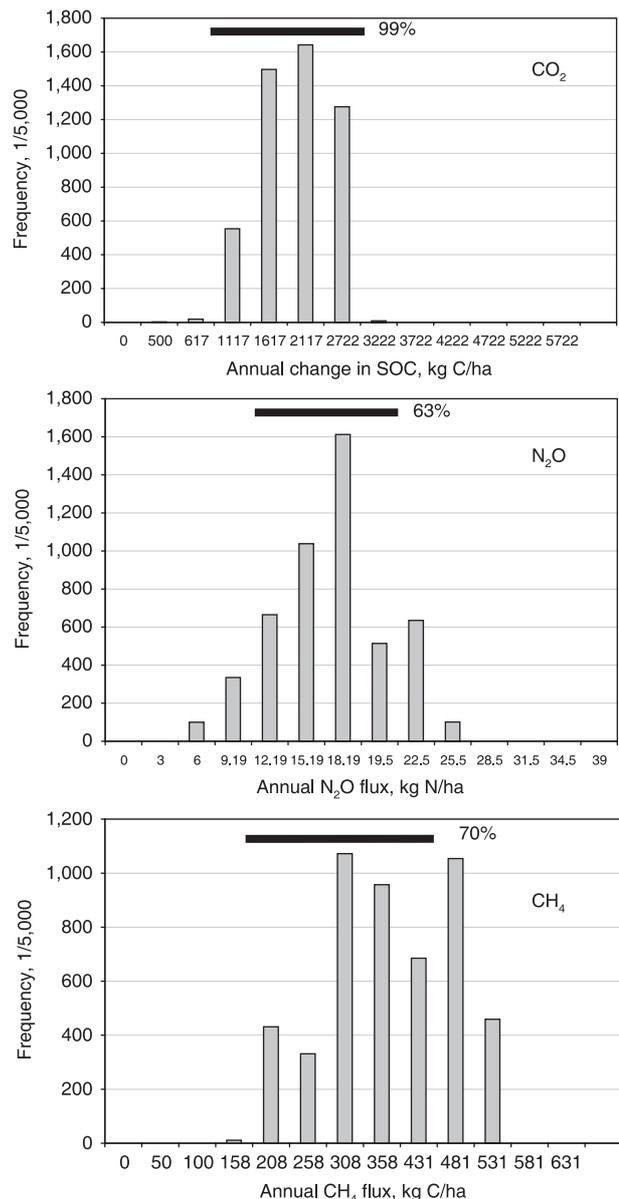


Figure 4 The Denitrification–Decomposition (DNDC)-modeled annual net CO₂, CH₄ and N₂O fluxes from rice paddies using the Monte Carlo approach (the vertical gray bars) and the Most Sensitive Factor (MSF) method (the horizontal black lines) for Colusa County, California, USA.

soil properties or other sensitive factors, the uncertainties induced by the spatial heterogeneity of input parameters can be brought under control.

A CASE STUDY ON MITIGATING GHG EMISSIONS FROM PADDY SOILS IN CHINA

It has long been known that paddy soil is an important emitter of methane. In fact, GHG emissions from paddy

soils have been intensively studied to date. Water management has been investigated as one of the options for mitigation of GHG emissions (Kanno *et al.* 1997; Nugroho *et al.* 1994; Wassmann *et al.* 1993; Yagi *et al.* 1994, 1996, 1997). A study was recently carried out to upscale the impacts of change in water management on crop yield, soil C sequestration and GHG emissions for the entire rice fields of China (Li *et al.* 2006). An evolution of water management for rice agriculture is under way in China. Continuous flooding was the dominant practice before the 1980s, which constantly kept the rice fields flooded with a water layer of approximately 5–10 cm during the rice growing season. From 1980 to 2000, the traditional practice was gradually replaced by midseason drainage, in which the rice fields were drained 2–5 times during a rice growing season. Each of the draining periods lasted for 5–10 days. The midseason drainage increased the rice yield by enhancing plant root development, depressing over-tillering and increasing mineralization of soil organic nitrogen. After 2000, a new practice, shallow flooding, emerged in rice cultivation in China. In this practice, the rice fields are marginally flooded with the soil surface barely covered by a thin water layer, even sometimes having the water table

drop below the soil surface for days. This management practice saves water and improves soil aeration (Chen 2004). This decades-long evolution in water management raised an interesting question: how would the changes in water management affect GHG emissions from rice production in China?

The DNDC was used to answer this question. Thirty million hectares of rice paddies in China were simulated with several scenarios of alternatives, varying in management practices of water, fertilizer and rice straw. The DNDC was run with 21-year alternative management schemes for each of the ~2,500 counties in the country. The modeled results indicate that: (1) replacing continuous flooding with midseason drainage in 1980–2000 has substantially reduced CH₄ emissions from rice production in China by approximately 4 Tg CH₄-C per year, (2) despite large scale adoption of midseason drainage currently, there is still a large potential for additional CH₄ reductions from Chinese rice paddies of 20–60% from 2000 to 2020 by applying shallow flooding (Fig. 5), (3) changes in management for reducing CH₄ emissions simultaneously affect soil carbon dynamics as well as N₂O emissions and can thereby reorder the ranking of technical mitigation effectiveness, (4) most

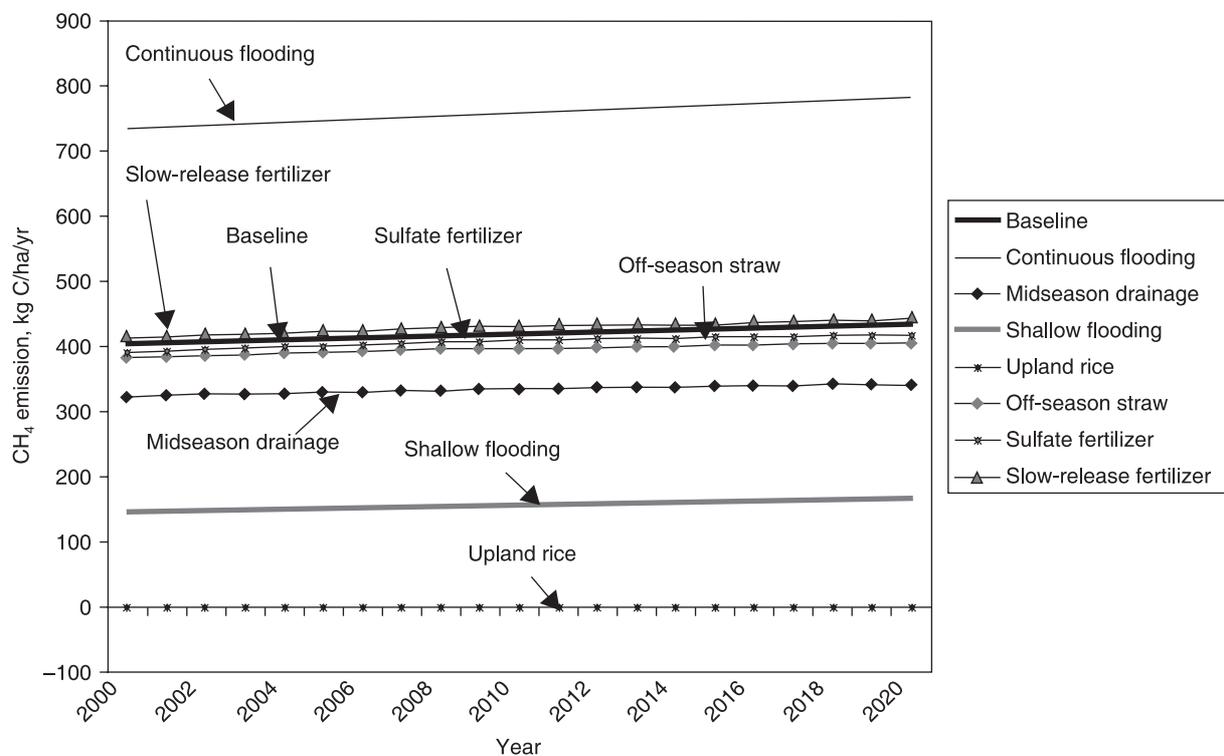


Figure 5 Impacts of management alternatives on nationally averaged CH₄ emissions from rice paddy fields in China from 2000 to 2020. The continuous flooding scenario produced the highest CH₄ emissions. Applying midseason drainage reduced more than half of the emissions resulting from continuous flooding. Shallow flooding further reduced CH₄ emissions by 200–300 kg CH₄-C per ha. From 2000 to 2020, CH₄ emissions slightly increased because of the soil C accumulation.

of the management alternatives produced rice yields comparable to the baseline, however, continuous flooding and upland rice significantly reduced yields, (5) water management strategies appear to be the most technically promising GHG mitigation alternatives, with shallow flooding providing additional benefits of both water conservation and increased yields (see details in Li *et al.* 2006). This national-scale study indicates that integrating process-based models with spatial databases can form a powerful tool to assess the effectiveness of GHG mitigation strategies at large scales.

In summary, quantifying soil GHG emissions is a scientific challenge that requires capacity to extend the understanding at a microbial or soil chemical scale to a regional or global scale. Fortunately, the latest developments in biogeochemistry, spatial data acquisition and computing techniques have provided potentials to integrate the research effort across the scales in space and time. The business of biogeochemical modeling is still in the early stages, although few researchers now doubt its irreplaceable role in contemporary ecosystem studies. I hope this brief review has touched on the basic issues of biogeochemical modeling and will draw more scientists to the new development.

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