

Ammonium Production in Submerged Soils and Sediments: The Role of Reducible Iron

K. L. Sahrawat *

International Crops Research Institute for the Semi-Arid Tropics
(ICRISAT), Patancheru, Andhra Pradesh, India

ABSTRACT

Submerged soils differ considerably from their arable counterparts. Lack of oxygen and the reduced state of submerged soils cause this. Submerging a soil under water triggers a series of physical, chemical and biological changes that greatly affect the dynamics of soil fertility and its use for growing crops such as wetland rice. Due to lack of oxygen, the nitrogen (N) mineralization process in submerged soils stops at ammonium production and nitrification is at low ebb. Most rice soils, especially those in the tropics are rich in iron (Fe). Redox cycling of Fe exerts a wide-ranging influence on the biogeochemistry of submerged rice soils and aquatic sediments where Fe is abundant. This article critically assesses the literature on the role of reducible Fe, which participates in redox reactions in submerged

*Correspondence: K. L. Sahrawat, International Crops Research Institute for the Semi-Arid Tropics (ICRISAT), Patancheru 502 324, Andhra Pradesh, India; Fax: +91 40 23241239/23296182; E-mail: klsahrawat@yahoo.com.

soils, on ammonium production or N mineralization in submerged soils and sediments. The reduction of Fe and organic matter oxidation in submerged soils and wetland sediments are interdependent. Results with freshwater sediments have indicated a direct correlation between Fe(III) oxide reduction rate constants and initial rates of organic C mineralization. The role of reducible Fe on N mineralization or ammonium production is not clearly understood. However, recent research with diverse West African rice soils showed that ammonium production in submerged soils is highly significantly correlated to reducible Fe. The results demonstrated that organic matter and reducible Fe control ammonium production in submerged soils. There is a need for future research to further elucidate the role and involvement of reducible Fe on N mineralization or ammonium production in submerged soils and sediments.

Key Words: Ammonium production; Iron redox couple; N nutrition of plants; Wetland soils and sediments; Role of electron acceptors; Organic matter; Redox potential.

INTRODUCTION

Globally, approximately 130 million ha land, constituting about 10% of the arable land, is temporarily flooded or submerged for wetland rice production.^[1] Flooding of soil under water greatly affects soil physical, chemical and biological properties through the well-known process of soil reduction caused by redox processes. The chemistry of submerged soils has been the subject of several reviews, covering various redox processes and nutrient transformations in these soils.^[2-6] The soil reduction-mediated physical, chemical and biological processes profoundly influence the quality of soil as a medium for the growth of crops such as wetland rice.^[2,3,5-7]

Iron (Fe) is present in large amounts especially in tropical soils.^[8] The Fe redox chemistry plays an important role in influencing the dynamics of soil fertility through its role on soil reduction and release of nutrient elements in solution of submerged rice soils.^[5,9]

Ammonium production is the key process for nitrogen (N) nutrition of wetland rice. The soil N supply plays a dominant role in the N nutrition of wetland rice. Over 50% of the N taken up by the rice crop even in N fertilized rice paddies come from the soil pool.^[10] The role of organic matter in controlling ammonium production or N mineralization in submerged soils is well known.^[10-14]



The role of Fe redox chemistry on N mineralization or ammonium production in wetland soils is not well understood. However, it is known that the most important geochemical change that takes place in many submerged soils and aquatic sediments is the reduction of Fe(III) to Fe(II) or Fe redox chemistry.^[2,3,9] The reduction of Fe(III) to Fe(II) greatly influences Fe and manganese (Mn) geochemistry and can have profound influence on a range of other important soil properties and fertility status of wetlands as a medium for plant growth and agricultural production.^[2,3,6,15,16]

Earlier reviews on the chemistry of submerged soils concentrated on iron redox reactions and its influence on chemical and electrochemical changes and exchange equilibria, soil properties and the dynamics of plant nutrients. This article focuses on the role of free or reducible iron, which participates in redox reactions in anaerobic or submerged environments, on organic matter mineralization and ammonium production in wetland soils and sediments. Nitrogen supply under submerged conditions, which depends on organic matter mineralization and ammonium production, is the key process for N nutrition of wetland rice and in regulating the primary productivity of wetlands.

This article is an attempt to critically review the role reducible Fe plays in ammonium production and N mineralization in wetland soils and sediments and suggests future areas of research. It is hoped that this review will stimulate further research in this important area because N supply (ammonium production) and cycling have a major influence on natural and agricultural wetlands' role in controlling productivity, regulating environmental quality, and determining the sustainability of wetland ecosystems.

REDOX COUPLES AND ELECTRON ACCEPTORS IN WETLANDS

Principal redox couples in sequence in submerged soils are: O_2/H_2O , NO_3^-/N_2 , $Mn(IV, III)/Mn(II)$, $Fe(III)/Fe(II)$, SO_4^{2-}/H_2S , and CO_2/CH_4 .

The main electron-acceptors in submerged soils include dissolved O_2 , NO_3^- , $Fe(III)$, SO_4^{2-} , and CO_2 . The final products of reduction in submerged soils are Fe(II), H_2S , and CH_4 , although intermediate products such as dissolved H_2 and H_2S are also found in submerged soils and sediments.^[17]

As mentioned in the introduction, the redox reactions influence soil properties and fertility in submerged soils and sediments which affect



their primary productivity and environmental quality.^[2,16,18,19] It is known that when soil is submerged into water, oxygen in the system is rapidly consumed during aerobic microbial respiration. This is followed by successive use of NO_3^- , Mn(IV) , Fe(III) , and SO_4^{2-} as electron acceptors in anaerobic microbial respiration.^[3]

Also, in the absence of oxygen, facultative and obligate anaerobes use the dissimilation products of carbohydrates and proteins as electron acceptors in their respiration.^[3] In most rice soils, Fe is the main electron acceptor. Iron occurs as Fe(III) oxides and oxyhydroxides, and in the structure of clay minerals.^[20]

In a study of the long-term effects of intermittent flooding of a Vertisol, Favre et al.^[21] showed that cation exchange capacity (CEC) of the soil in the reduced state following flooding increased to twice that of its oxidized, unflooded state. The increase in CEC was caused by the increase in structural Fe(II) upon reduction and the removal of Fe oxyhydroxide coatings by reductive dissolution. Several authors have reported on the role of Fe on soil properties including CEC.^[22–25] It has been suggested that the changes in CEC and related redox reactions may be important in substantially modifying proton, anion, and cation balances in intermittently flooded soils.^[21]

Soil reduction in submerged soils and sediments is measured by redox potential (Eh) and expressed in mV. The measured Eh can be explained by the concentration of some redox systems. Dissolved O_2 , NO_3^- and SO_4^{2-} are relatively electrode-insensitive and others, especially Fe(III) play important role in the soil reduction, measured by Eh.^[17,26,27] In a field study, it was demonstrated that measured Eh showed a higher correlation to redox species Mn(II) ($r^2 = 0.76$), Fe(II) ($r^2 = 0.73$), and methane ($r^2 = 0.76$) than to dissolved oxygen, nitrate and sulfate-S.^[17]

The data on the range of redox potentials (Eh) encountered in soils and sediments ranging from well drained to submerged or flooded conditions are summarized in Table 1. The data can serve as a

Table 1. The range of redox potentials (Eh) encountered in well-drained and submerged soils and sediments.^[9]

Soil water condition	Redox potential (mV)
Well-drained (aerated)	+700 to +800
Moderately reduced	+400 to +200
Reduced	+100 to -100
Highly reduced	-100 to -300

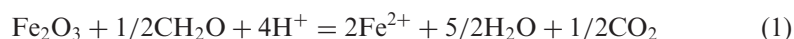


Table 2. Approximate redox potentials at which the main oxidized components in submerged soils and sediments become unstable.^[9,28]

Reaction	Redox potential (mV)
O ₂ -H ₂ O	+380 to +320
NO ₃ -N ₂ , Mn ⁴⁺ -Mn ²⁺	+280 to +220
Fe ³⁺ -Fe ²⁺	+180 to +150
SO ₄ ²⁻ -S ²⁻	-120 to -180
CO ₂ -CH ₄	-200 to -280

guideline for classifying soil reduction under diverse soil moisture regime.^[9] The approximate Eh values at which the main oxidized components in submerged soils become unstable are shown in Table 2. It is indicated that Fe(III) is reduced to Fe(II) at Eh value ranging from 150 to 180 mV.

The reduction of Fe(III) oxides to Fe(II) can be represented by the following equation, in which Fe(III) oxides serve as the source of reducible Fe and organic matter (CH₂O) serve as the electron donor:



The reduction rate of Fe is affected by surface chemical reactions and is directly related to the concentration of organic reductant.^[29] For example, Gao et al.^[17] observed that in submerged paddy field the reduction rates of Fe(III) to Fe(II) were faster in straw-incorporated plots as higher soluble, reduced Fe(II) was detected. Precipitation reaction tends to increase Fe(III)/Fe(II) and derives the reduction (Fe³⁺ = Fe²⁺ + H⁺) reaction forward. Moreover, application of organic matter has an inhibitory effect on the crystallization of amorphous ferric hydroxides and keeps Fe in the amorphous and relatively reducible form.^[30]

Narteh and Sahrawat^[6] showed that at 4 weeks after flooding of West African soils, the soil solution Eh can be predicted from the concentration of Fe(II) in soil solution and soil solution pH:

$$\text{Eh} = 409 - 4.09 \log \text{Fe(II)} - 59\text{pH}; \quad R^2 = 0.99 \quad (2)$$

Furthermore, the changes in soil solution pH corresponded to changes in soil solution Eh. The stability in Eh-pH relationship was



recorded at the 4 week after flooding and the relationship was described by the following equation:^[6]

$$\Delta E_h = -16 - 48 \Delta \text{pH}; \quad R^2 = 0.84 \quad (3)$$

IRON REDOX AND CARBON METABOLISM

Geological evidence suggests that Fe(III) reduction has been an important process for organic matter oxidation since early in the Earth's biotic history. The strong correlation in the occurrence of isotopically light carbonates and magnetite in the pre-Cambrian banded Fe formations indicates that organic matter oxidation was coupled to Fe(III) reduction in this environment (for review, see Lovley^[16]). It has been suggested that Fe may have been central to the overall carbon (C) flow in these pre-Cambrian environments because Fe(II) may have also served as the electron donor for photosynthesis.^[31] According to this model, the organic matter and Fe(III) that were produced as the result of photosynthesis in the water column would then settle out where dissimilatory Fe(III)-reducing microorganisms could oxidize the organic matter with the reduction of Fe(III). This results in the production of magnetite as well as in the regeneration of dissolved Fe(II) for further photosynthesis.

A number of studies in freshwater and marine sediments indicate that dissimilatory microbial reduction of Fe(III) oxides contributes substantially to C metabolism.^[32,33] It has been suggested that microbial Fe(III) oxide reduction could play a major role in suppressing methanogenesis in freshwater environments. This applies to both in surface sediment^[34] and in the rhizosphere of aquatic plants^[35] where oxygen input from plant roots derives a dynamic Fe redox cycle. Oxygen from the rice tops is conducted to rice roots via aerenchyma or air-conducting channels in the plant tissue of aquatic plant.^[2] In addition to Fe redox, C metabolism in wetland sediments is also affected by other terminal electron-accepting reactions involving oxygen, nitrate, manganese and sulfate reduction.^[36-38]

Roden and Wetzel^[39] studied freshwater wetland sediments and demonstrated a direct correlation between first-order Fe(III) oxide reduction rate constants and initial rates of organic C mineralization (amount of carbon dioxide and methane accumulated). The results provide empirical support for existing approaches to modeling organic matter decay-dependent Fe(III) oxide reduction kinetics in sediments.



Simulation of Fe(III) oxide reduction in freshwater wetland sediment where amorphous Fe(III) oxide is the dominant form of Fe(III) available for microbial reduction can be conducted with the simple organic matter decay-dependent first-order rate model described by the authors.^[39]

In a recent study of 15 West African rice soils, Sahrawat and Narteh^[40] demonstrated that the concentration of macro- and micro-nutrients released under submerged conditions in these soils, measured by EC of soil solution, was significantly correlated to organic C and reducible Fe extracted by EDTA. These results indicate the importance of reducible Fe on the release of plant nutrients in soil solution of submerged rice soils that are rich in Fe.

REDUCIBLE IRON AND AMMONIUM PRODUCTION

Ammonium fixation and release in submerged tropical rice soils is affected by reducible Fe and it has been suggested that ammonium dynamics in submerged soils is greatly influenced by Fe redox reactions.^[41] During submergence, ammonium fixed under nonsubmerged conditions by amorphous Fe, is released following the reduction of Fe(III) to Fe(II). Under reduced conditions in soils and sediments, the sorbed ammonium is released by both Fe(III) reduction to Fe(II) and exchange with Fe(II).^[15,41,42]

A study of N mineralization in diverse West African rice soils, showed that although extractable Fe was not directly correlated to ammonium produced under waterlogged condition, its inclusion in the multiple regression analysis improved the prediction of ammonium production.^[13]

In a recent study with 15 diverse West African soils, Sahrawat and Narteh^[43] showed that mineralizable N or ammonium production under anaerobic condition was significantly correlated to organic C ($r=0.79$, $n=15$) and Fe extracted by EDTA ($r=0.86$) or ammonium oxalate ($r=0.75$). Ammonium released in soil solution of the submerged soils was also significantly correlated to EDTA extractable Fe ($r=0.60$) or ammonium oxalate-extractable Fe ($r=0.60$) although the correlation coefficients were lower than those with the mineralizable N. Multiple regression analysis of the data showed that ammonium production in soils or ammonium released in soil solution can be predicted from organic C and EDTA or ammonium oxalate extractable Fe.

Several authors have suggested that EDTA and ammonium oxalate solutions extract fractions of amorphous Fe that are easily reducible and



thus can be utilized as the measure of the activity of poorly crystalline Fe(III) oxides in soils.^[44,45] Moreover, research has shown that the amorphous Fe fractions contribute to Fe(II) production in reduced (Eh 150–180 mV) soils and sediments. The intensity of Fe(II) production increases with a decrease in the crystallinity of the pedogenic Fe(III) oxides and hydroxides and with an increase in the easily mineralizable organic substances.^[30,46–49]

Based on research conducted with West African rice soils, Sahrawat and Narteh^[50] concluded that a chemical index based on organic matter and reducible Fe contents can be used for predicting ammonium production in submerged soils. Multiple regression equations relating organic C and reducible Fe (extracted by EDTA or ammonium oxalate) to ammonium N released can be used for determining the N mineralization potential of wetland soils.

The relationships of organic C and reducible Fe with ammonium produced in 15 West African soils were represented by the following regression equations:

$$\begin{aligned} &\text{Mineralizable N (mg kg}^{-1}\text{ soil)} \\ &= 16.4 + 1.320 \text{ Organic C (g kg}^{-1}\text{)} \\ &\quad + 0.0369\text{EDTA-Fe (mg kg}^{-1}\text{)}; \quad R^2 = 0.85 \end{aligned} \quad (4)$$

$$\begin{aligned} &\text{Mineralizable N (mg kg}^{-1}\text{ soil)} \\ &= 11.14 + 1.805 \text{ Organic C (g kg}^{-1}\text{)} \\ &\quad + 0.00469 \text{ Ammonium oxalate-Fe (mg kg}^{-1}\text{)}; \quad R^2 = 0.81 \end{aligned} \quad (5)$$

It was found that soils high in both organic C and reducible Fe were high in mineralizable N. On the other hand, soils low in organic C or reducible Fe had relatively lower contents of mineralizable N (Table 3).

Table 3. Distribution of 15 West African rice soils according to mineralizable N (Min-N) released under anaerobic incubation and associated organic C and EDTA (EDTA-Fe) or ammonium oxalate extractable Fe (Amox-Fe) contents.^[50]

Min-N mg kg ⁻¹ soil	No. of soils	Organic C g kg ⁻¹ soil	EDTA-Fe	Amox-Fe
			mg kg ⁻¹ soil	
86–166	4	23.0–46.0	150–2200	1875–11412
55–77	5	9.2–23.2	325–800	1100–6750
21–50	6	7.4–15.6	125–600	925–3562



These results are supported by highly significant correlations between mineralizable N and organic C and reducible Fe extracted by EDTA or ammonium oxalate.^[43]

PERSPECTIVES

The research discussed in this article indicates those Fe redox reactions, controlled by reducible Fe, influence ammonium production or N mineralization in submerged soils and sediments. Reducible Fe along with organic matter controls the N mineralization process in submerged rice soils where Fe is abundant. It appears that the availability of electron acceptors such as Fe(III) oxides might greatly influence ammonium production in reduced soils. However, since Fe is present in high amounts especially in tropical rice soils, it exerts a dominant role not only in soil reduction but also in the release of ammonium in submerged soils and sediments. In the absence of oxygen, ferric Fe serves as an electron acceptor and affects organic matter oxidation and ammonium production,^[16,51] although the mechanisms involved, especially in ammonium production are not fully understood.

A better understanding of the involvement of reducible Fe in N mineralization in submerged soils and sediments will lead to improved methods for assessing their N supplying capacity. There is an obvious need to conduct further research in understanding the role of Fe redox reactions in influencing N supply and cycling in submerged soils and sediments. Because N supply and cycling has a major influence in controlling the wetlands' role in controlling productivity, regulating environmental quality and determining the sustainability of the wetland ecosystems. Obviously, the research in this area is of scientific, environmental and agricultural interest. It is hoped this article will stimulate further research in this area.

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