



## Link between paddy soil mineral nitrogen release and iron and manganese reduction examined in a rice pot growth experiment

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### ABSTRACT

Paddy soil indigenous N supply is often poorly related to N status and our aim was to assess its linkage to reduction of Fe<sup>3+</sup> and Mn<sup>4+</sup>, primary terminal electron acceptors in submerged soils. Transplanted rice was grown in the greenhouse in four Bangladeshi paddy soils with distinct SOC to Fe<sub>ox</sub> ratio (1.6 to 4.9) for 72 days firstly under continuous and then intermittent flooding. Solution buildup rates of Fe<sup>2+</sup> and Mn<sup>2+</sup> in the first 2–3 weeks of flooding correlated negatively with soil organic carbon (SOC) to NH<sub>4</sub>-oxalate extractable Fe (Fe<sub>ox</sub>) and Mn (Mn<sub>ox</sub>) ratios ( $p < 0.01$ ). Mössbauer analysis detected ferrihydrite and goethite in all soils and with Mn<sub>3</sub>O<sub>4</sub> these are the likely source minerals. An electron (e<sup>-</sup>) balance calculated from soil C-emission rates suggested reductive Fe and Mn dissolution to relevant e<sup>-</sup>-accepting processes, probably responsible for no > 50% of e<sup>-</sup> capture, though. Reduction of abundantly present octahedral Fe<sup>3+</sup> in chlorites and vermiculite and their interstratified forms in these floodplain silty Inceptisols is hypothesized to also support microbial activity. Notwithstanding, a close temporal synergy existed between solution Fe and soil mineral N and their build-up rates were correlated ( $r: 0.77$  to  $0.90$ ;  $p < 0.01$ ) and with that of dissolved OC (DOC) ( $r: 0.84$  to  $0.96$ ;  $p < 0.01$ ), C emission rate ( $r: 0.99$ ;  $p < 0.01$ ) and SOC:Fe<sub>ox</sub> ( $r: -0.71$ ;  $p < 0.01$ ). These correlations suggest Fe<sup>3+</sup> reduction to be a relevant intermediary step in soil N mineralization, possibly through release of associated DOC, N or both. After switching to intermittent flooding dissolution of Fe, Mn and DOC were decoupled from mineral N release but since Eh remained in the Fe<sup>3+</sup>-reduction range in three out of four soils, possibly clay-Fe<sup>3+</sup> alternated with O<sub>2</sub> as e<sup>-</sup>-acceptor. Most importantly, in all soils N release slowed or halted after only 2 weeks of flooding but recommenced with intermittent flooding. As a next step, field experiments could verify if indigenous soil N supply also benefits from non-continuous irrigation management. Lastly, experimental proof is pending for release of clay-bound N and interlayer NH<sub>4</sub><sup>+</sup> following reduction of octahedral Fe<sup>3+</sup> with consequent increased negative charge or structural destabilization, possibly an important process in floodplain paddy soils in Bangladesh.

### 1. Introduction

Irrigated rice systems account for half of the total rice area and contribute 75% of the world's annual rice production (IRRI, 2011) since most rice varieties exhibit maximum yield potential under sufficient water supply. Nitrogen remains the most yield limiting and difficult nutrient to manage due to its complex biogeochemical cycle, poor fertilizer use efficiency (usually just 30%) and rapid losses via different

processes (Said-Pullicino et al., 2014). The availability of N for plant uptake depends on the balance between soil organic matter (SOM) mineralization, microbial immobilization, fertilizer-N application, NH<sub>4</sub><sup>+</sup> fixation or release, and N losses via nitrification-denitrification and NH<sub>3</sub> volatilization. A number of studies reveal that N uptake by rice plants, 15 to 22 kg N t<sup>-1</sup> rice (Dobermann and Cassman, 1996), originates mainly from soil organic N mineralization and not directly from fertilizer, even in sufficiently N-fertilized fields (Ando et al., 1992;

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Manguiat et al., 1994; Khaokaew et al., 2007). Previous studies, however, have shown very inconsistent dependencies of N mineralization rate on general soil properties and management (Narteh and Sahrawat, 1997; Adhikari et al., 1999; Sahrawat, 2006; Kader, 2012; Kader et al., 2013). Improved estimates of indigenous soil N supply would allow for adapted N fertilizer application, thereby optimizing N use efficiency as well as minimizing adverse environmental impacts of excessive N (Mikha et al., 2006; Sharifi et al., 2007). In well-controlled lab incubations to assess N release, SOM decomposition evidently mainly depends on quality and quantity of SOM, but also very much on availability of oxidants (Cassman et al., 1996; White and Reddy, 2001; Li et al., 2010; Gao et al., 2014). Thus empirical predictions of  $\text{NH}_4^+$  production may be improved if co-based on contents of key oxidants, confirmed to drive anaerobic soil mineral N release. Usually  $\text{Fe}^{3+}$  reduction is seen to dominantly accept electrons released from organic matter, amounting up to 24% according to Jäkel and Schnell (2000), 66–84% according to Inubushi et al. (1984) and 58–79% according to Yao et al. (1999). Indeed, regression analysis indicated that inclusion of extractable Fe improved the prediction of mineralized N (Narteh and Sahrawat, 1997). For a set of 25 paddy soils in Bangladesh we previously found that anaerobic N mineralization correlated only with pH and pedogenic-Fe content and not with a myriad of other soil properties including soil C and N content (Kader et al., 2013). In our follow-up research (Akter et al., 2016) we instead found no correlations between anaerobic soil  $\text{NH}_4^+$  release and pedogenic Fe or Mn contents. However, soil exchangeable  $\text{NH}_4^+$  and dissolved Fe demonstrated a remarkable co-evolution over time and were positively correlated. But causality would need to be elucidated and confirmed in a more realistic soil environment than in small scale bare soil lab incubations as used by Akter et al. (2016) and Kader et al. (2013). Firstly, rice root-derived  $\text{O}_2$  input causes local rising of Eh, lowering of pH and re-oxidation and precipitation of  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$ . Secondly, without plant N-uptake paddy soil  $\text{NH}_4^+$ -N contents likely become inhibitive to further N-mineralization in bare soil setups. Thirdly, in a field setting, flooding is often not entirely continuous and frequent shorter periods of topsoil drying occur, deliberately or due to lack of irrigation water. Resulting variations of reduction or oxidation processes would strongly change the soil solution chemistry, especially reductive dissolution of Mn and Fe (hydr-)oxides, OM degradation and N mineralization. Experiments with non-continuously flooded growing rice plants allow further assessing linkages between soil N supply and other soil processes under more realistic conditions.

A first objective was to upscale our previous lab-incubations to rice pot-growth experiments and to complement measurements of soil mineral N release with ancillary information on the evolution of soil redox potential (Eh) and Fe and Mn reduction. Microbial activity, Fe- and Mn reduction are depth-dependent (Zschornack et al., 2011) in paddy soil and accordingly two depth increments were sampled to assess any vertical gradients.

We did not attempt to assess radial variation surrounding rice roots. A second objective was to investigate how a transition from continuous to alternate wetting and drying affects  $\text{Fe}^{3+}$  and  $\text{Mn}^{4+/3+}$  reduction in relation to soil net N supply. To address these objectives, we studied plant N uptake, the progressive dissolution of Fe and Mn as well as change in mineral N content in four young floodplain paddy soils of North-Bangladesh with wide variations in SOC to oxalate extractable Fe ratio ( $\text{Fe}_{\text{ox}}$ ). The  $\text{SOC}:\text{Fe}_{\text{ox}}$  ratio can determine net soil N supply in various ways. At lower  $\text{SOC}:\text{Fe}_{\text{ox}}$  ratio there is more OM adsorption onto the surface of Fe(hydr-)oxides, resulting in its stabilization against microbial decomposition (Hanke et al., 2014), and possibly also declined availability of the Fe(hydr-)oxides for reduction. Both mechanisms should result in a positive relation between  $\text{SOC}:\text{Fe}_{\text{ox}}$  and soil N supply. On the opposite, it could be argued that at lower  $\text{SOC}:\text{Fe}_{\text{ox}}$  ratio, more dissolved OM is co-released per equivalent  $\text{Fe}^{2+}$  produced into the soil solution, but this has not been confirmed yet. Likewise, a relatively lower content of reducible  $\text{Fe}^{3+}$  (approximated by  $\text{Fe}_{\text{ox}}$ ) might

limit microbially mediated OM decomposition and net soil N mineralization (Sahrawat, 2004). But in previous lab incubations (Akter et al., 2016) we could not reveal an effect of soil  $\text{Fe}^{3+}$  or  $\text{Mn}^{4+}$  amendment on net mineral N release. In sum, we thus expected a positive relation between  $\text{SOC}:\text{Fe}_{\text{ox}}$  and net soil N release.

We also monitored soil temperature, moisture, pH, Eh, DOC,  $\text{CO}_2$  and  $\text{CH}_4$  emission to obtain detailed insight into the evolution of soil reductive processes. During the first weeks after onset of flooding, often with a major part of soil mineral N release, we estimated the potential share of donated electrons accepted by various soil oxidants. We used Mössbauer spectroscopy to identify potential source minerals of reducible Fe.

## 2. Materials and methods

### 2.1. Soils

Four typical young floodplain paddy soils were collected from farmers' fields in May 2014 in Northern Bangladesh (24°37' N to 24°49' N, 90°02' to 90°25' E). Soil Types (Soil Taxonomy) of the studied soils were Aeric Haplaquepts (Sonatala-1), Aeric Fluvaquepts (Melandoho), Mollic Haplaquepts (Balina) and Ultic Ustochrepts (Noaddah-2) (Table 1). The subtropical monsoon climate is characterized by an annual mean temperature and rainfall of 25.8 °C and 2427 mm, respectively (BMD, 2015). Agricultural production is dominated by single or double rice cropping with rain-fed and supplemental irrigated production in the Aman season (Jul–Nov) and irrigated production in the dryer Boro season (Jan–Apr). Puddle layer (0–15 cm) soil was collected from 15 spots per field by spade. The field-moist soils were broken, air dried, ground and sieved through a 2 mm mesh sieve prior to shipment to the University of Ghent, Belgium. Previously determined general soil properties are shown in Table 1. In addition, plant-available nutrients in air dried soils were determined by  $\text{NH}_4$ -lactate (Ca, K, Na, Mg and P)

**Table 1**

Physical and chemical properties of four Bangladeshi floodplain paddy soils used in the greenhouse rice growth pot experiment.

Properties	Sonatala-1	Melandoho	Balina	Noaddah-2
Latitude (N) <sup>a</sup>	24°43'14.5"	24°48'42"	24°49'28"	24°37'19.2"
Longitude (E) <sup>a</sup>	90°25'52.3"	90°24'49"	90°21'09"	90°02'17"
Cropping pattern <sup>a</sup>	Rice-fallow-rice	Rice-fallow-rice	Rice-fallow-rice	Rice-fallow-rice
Soil type <sup>a</sup>	Aeric Haplaquepts	Aeric Fluvaquepts	Mollic Haplaquepts	Ultic Ustochrepts
Soil texture <sup>a</sup>	Silt loam	Silt loam	Silty clay loam	Silt loam
SOC (g kg <sup>-1</sup> )	23.6	5.5	16.5	11.5
Total N (g kg <sup>-1</sup> )	2.1	0.8	1.9	1.3
pH-KCl	5.5	4.2	4.0	4.2
$\text{Fe}_{\text{ox}}$ (g kg <sup>-1</sup> )	4.8	3.5	8.4	4.5
$\text{Mn}_{\text{ox}}$ (g kg <sup>-1</sup> )	0.20	0.05	0.22	0.25
$\text{SO}_4^{2-}$ (mg kg <sup>-1</sup> ) <sup>a</sup>	19.3	17.5	58.2	12.6
CEC (cmol <sub>c</sub> kg <sup>-1</sup> ) <sup>a,b</sup>	44	25	43	35
$\text{NH}_4^+$ -N (mg kg <sup>-1</sup> )	7	16	13	6
$\text{NO}_3^-$ -N (mg kg <sup>-1</sup> )	4	8	12	2
Ca-lac (mg kg <sup>-1</sup> )	1814	460	1652	401
K-lac (mg kg <sup>-1</sup> )	24	32	61	22
Mg-lac (mg kg <sup>-1</sup> )	554	95	339	42
Na-lac (mg kg <sup>-1</sup> )	143	26	52	18
P-lac (mg kg <sup>-1</sup> )	24	56	5	127
Cu-DTPA (mg kg <sup>-1</sup> )	1.8	1.0	2.4	0.4
Fe-DTPA (mg kg <sup>-1</sup> )	141	174	165	240
Mn-DTPA (mg kg <sup>-1</sup> )	97	21	128	92
Zn-DTPA (mg kg <sup>-1</sup> )	0.1	0.1	0.2	0.5
Si-DTPA (mg kg <sup>-1</sup> )	43	21	40	35
Mo-DTPA (mg kg <sup>-1</sup> )	0.1	0.1	0.1	0.1
$\text{SOC}:\text{Fe}_{\text{ox}}$	4.9	1.6	2.0	2.6

<sup>a</sup> Data taken from Kader et al. (2013).

<sup>b</sup> CEC: cation exchange capacity.

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