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Coupling soil water processes and nitrogen cycle across spatial scales: Potentials, bottlenecks and solutions

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Abstract

Interactions among soil water processes and the nitrogen (N) cycle govern biological productivity and environmental outcomes in the earth's critical zone. Soil water influences the N cycle in two distinct but interactive modes. First, the spatio-temporal variation of soil water content (SWC) controls redox coupling among oxidized and reduced compounds, and thus N mineralization, nitrification, and denitrification. Secondly, subsurface flow controls the movement of water and dissolved N. These two processes interact such that subsurface flow dynamics control the occurrence of relatively static, isolated soil solution environments that span a range of reduced to oxidized conditions. However, the soil water-N cycle is usually treated as a black box. Models focused on N cycling simplify soil water parameters, while models focused on soil water processes simplify N cycling parameters. In addition, effective ways to deal with upscaling are lacking. New techniques will allow comprehensive coupling of the soil water-N cycle across time and space: 1) using hydrogeophysical tools to detect soil water processes and then linked to electrochemical N sensors to reveal the soil N cycle, (2) upscaling small-scale observations and simulations by constructing functions between soil water-N cycle and ancillary soil, topography and vegetation variables in the hydropedological functional units, and (3) integrating soil hydrology models with N cycling models to minimize the over-simplification of N biogeochemistry and soil hydrology mechanisms in these models. These suggestions will enhance our understanding of soil water processes and the N cycle and improve modeling of N losses as important sources of greenhouse gas emission and water pollution.

Keywords
Biogeochemistry, Hydropedology, Nitrogen cycle, Soil Hydrology

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Coupling Soil Water Processes and Nitrogen Cycle across Spatial Scales: Potentials, Bottlenecks and Solutions

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Abstract: Interactions among soil water processes and the nitrogen (N) cycle govern biological productivity and environmental outcomes in the earth's critical zone. Soil water influences the N cycle in two distinct but interactive modes. First, the spatio-temporal variation of soil water content (SWC) controls redox coupling among oxidized and reduced compounds, and thus N mineralization, nitrification, and denitrification. Secondly, subsurface flow controls the movement of water and dissolved N. These two processes interact such that subsurface flow dynamics control the occurrence of relatively static, isolated soil solution environments that span a range of reduced to oxidized conditions. However, the soil water-N cycle is usually treated as a black box. Models focused on N cycling simplify soil water parameters, while models focused on soil water processes simplify N cycling parameters. In addition, effective ways to deal with upscaling are lacking. New techniques will allow comprehensive coupling of the soil water-N cycle across time and space: 1) using hydrogeophysical tools to detect soil water processes and then linked to electrochemical N sensors to reveal the soil N cycle, (2) upscaling small-scale observations and simulations by constructing functions between soil water-N cycle and ancillary soil, topography and vegetation variables in the hydropedological functional units, and (3) integrating soil hydrology models with N cycling models to minimize the over-simplification of N biogeochemistry and soil hydrology mechanisms in these models. These suggestions will enhance our understanding of soil water processes and the N cycle and improve modeling of N losses as important sources of greenhouse gas emission and water pollution.

1. Research progresses in soil water processes and N cycle

1.1 Soil Water Processes

Soil water processes can be described in terms of content (volumetric or gravitational; m^3 m^{-3} and kg kg^{-1}, respectively), potential (matric, osmotic and gravitational potentials; kPa) and movement (subsurface flows; m^3 in quantity or mm s^{-1} in speed). All of these descriptions are variable in time and space, yet the range may be restricted based on location and time. They are important controls on various biological, physical and chemical processes in the earth's critical zone (Vereecken et al., 2008; Lin et al., 2015; Ma et al., 2017). Therefore, intensive studies have been directed to observe, predict and explain variations in soil water processes across temporal and spatial scales. Direct measurements are typically used to measure water content (e.g., destructive: core sampling; non-destructive: neutron, gamma radiation, and time-domain reflectometry methods) and potential (e.g., tensiometer and pressure chamber). However, the spatial and temporal resolutions of these measurements are restricted due to cost. To measure and quantify soil water movement, measurements (e.g., dye tracing and runoff plot) are usually destructive and restricted to small spatial scales (e.g., pedon and hillslope).

Given these constraints, models (e.g., numerical models and distributed hydrological models) and hydrogeophysical tools (e.g., ground-penetrating radar or GPR, electromagnetic induction or EMI, and electrical resistivity tomography or ERT) are increasingly adopted as alternatives to direct measurements of soil water content, potential and movement (Vereecken et al., 2015, 2016). Due to the complexity of

parameters influencing soil water processes and the low spatial and temporal resolutions of the measurements, appropriate numerical models with pre-established scenarios have been used to understand soil water processes across spatial scales over the last decades (e.g., Fang et al., 2015; Lai et al., 2016). Hydrogeophysical tools have been widely needed to assist in the interpretation of subsurface soil water processes and calibrate the numerical models (Vereecken et al., 2015; Guo and Lin, 2018). The EMI measures soil apparent electrical conductivity, the GPR uses electromagnetic radiation in the microwave band and detects the reflected signals from the subsurface, and the ERT images subsurface conditions through measurements of subsurface electrical resistivity. Readings and outputs of these tools are correlated with various soil properties including direct measures of soil water including SWC and subsurface flow. Large amounts of data on subsurface soil water processes can be time and cost-efficiently collected and visualized using these tools. Therefore, these non-invasive geophysical tools have been successfully applied to investigate the SWC and subsurface flow paths from plot to catchment scales.

Soil water processes can be categorized into static and dynamic aspects. The static aspect neglects water movement and describes instantaneous patterns and controlling factors on SWCs and potentials. The dynamic aspect addresses water movement and describes the mechanisms of water flow in the soil (matric and subsurface flows). These two aspects are intertwined. The subsurface flow, especially rapid flow, occurs when the SWC and potential reach certain thresholds (Lai et al., 2016; Zhu et al., 2014). On the other hand, the spatio-temporal distributions of SWC

can be altered by the timing of occurrence and paths of subsurface flow (Hopp et al., 2011; Zhu and Lin, 2009). These soil water processes transport and connect electron-donors and electron-acceptors that control redox potential and thus various biogeochemical processes.

(1) Static aspect: spatio-temporal variations in SWC and controlling factors.

Parameters extracted from statistical models, and particularly variograms (e.g., mean, standard deviation, and sill), have been used to characterize the state, distribution, variation and spatial structure of SWC (e.g., Brocca et al., 2010; Hu et al., 2011). The spatial variability of SWC is maximum under intermediate wetness condition and decreases at both dry and wet ends of SWC (Brocca et al., 2010; Gao et al., 2015). However, as the spatial scale of inference increases, the spatial variability of SWC also increases correspondingly (Famiglietti et al., 2008; Brocca et al., 2010). Common perceptions suggest that spatial variation in SWC is dominated by topography (e.g., slope, aspect, contribution area, and elevation) in wet periods, but by soil properties (e.g., soil texture, organic matter content, and rock fragment content) in dry periods (Grayson et al., 1997; Western et al., 1999).

Moreover, controlling factors and their contributions to SWC distribution are scale-dependent (Feng, 2016). Crow et al. (2012) summarized that soil property controls decreased from point scale to continental scale, topographic controls started at field scale, reached maximum at watershed scale, and then decreased at regional and continental scale, land use/cover controls were weak at watershed scale but increased at regional and continental scales, meteorological controls started at

watershed and regional scale and increased at continental scales. Temporal stability, which indicates the similarity of SWC spatial distribution pattern across time (Vachaud et al., 1985), has also been adopted to characterize the spatial variation of SWC. However, this spatial distribution pattern of SWC can change with wetness condition, season, precipitation and vegetative cover (Zhao et al., 2010; Penna et al., 2013; Lv et al., 2016). The temporal variation of SWC is mostly driven by meteorological properties like precipitation, temperature and evapotranspiration (e.g., Verma et al., 2011; Feng and Liu, 2015), occurrence of subsurface flow (Kim and Mohanty, 2016), and plant water uptake (e.g., Liancourt et al., 2012).

(2) Dynamic aspect: mechanisms and simulations of subsurface flow.

Subsurface flow (Fig. 1) has been recognized as an important pathway for the transport of water (e.g., Hopp et al., 2011; Liao et al., 2016; Guo and Lin, 2018) and solutes including nutrients and pollutants (e.g., van Verseveld et al, 2009; Daigh et al., 2015). Traditional methods used to investigate subsurface flow from pedon to catchment scales include *in situ* monitoring and *in silico* simulations (e.g., Vereecken et al., 2015, 2016). Hydrogeophysical survey (e.g., Robinson et al., 2012; Guo et al., 2014) and soil hydrology models (e.g., Fang et al., 2015; Lai et al., 2016) are two representative examples of *in situ* monitoring and *in silico* simulations, respectively. Monitoring has revealed that lateral subsurface flow always occurs at critical interfaces (e.g., soil-bedrock; between soil layers) and has strong spatial structure (Hopp et al., 2011; Guo and Lin, 2018). To trigger subsurface flow, thresholds of different controlling factors (e.g., precipitation and SWC) have been reported. For

example, regarding precipitation threshold, on a forest hillslope in Georgia, USA, little subsurface flow was observed when the rainfall amount was smaller than a threshold of 55 mm (Tromp-van Meerveld and McDonnell, 2006); regarding SWC threshold, on a tea garden hillslope in Taihu Lake Basin, China, subsurface flow occurred when the SWC researched a threshold of 0.18 m^3 m^{-3} (Liao et al., 2016). In addition, topography, soil properties, and vegetation have also been reported to influence the occurrence, network and quantity of subsurface flow. For example, as slope angle increased and soil depth decreased, subsurface flow quantity and connectivity both increased (Hopp et al., 2011); as the land use and evapotranspiration changed and thus the thresholds associated with the generation of subsurface flow changed (Lv et al., 2016).

Fig. 1. Subsurface soil water movement at pedon, hillslope and watershed scales, including capillary flow, vertical flow and lateral flows in the soil matrix and along critical interfaces.

1.2 Nitrogen Cycle

The soil N cycle is recognized as one of the most important biogeochemical processes in the earth's critical zone. Inorganic nitrogen (N) typically limits biological production in terrestrial, freshwater, and marine ecosystems (Elser et al. 2007). Use of synthetic inorganic N fertilizers, which began in earnest in the mid-1900s, has significantly increased agricultural production (Erisman, et al., 2008). However, the use and overuse of N fertilizers and manure are associated with large losses of inorganic and organic N from cropping systems to the surrounding environment where they create numerous environmental problems including eutrophication,

acidification, radiative forcing, stratospheric ozone depletion, tropospheric ozone production and formation of particulate matter < 2.5 µm size (Denk et al., 2017). Soil water controls both N transport and transformation in the soil and the balance between retention and environmental loss of inorganic N.

Fertilization and atmospheric deposition are important external inputs of inorganic N to earth's critical zone (Fig 2). However, the largest source of inorganic N for crop uptake and environmental loss is often mineralized soil organic N (Stevens et al. 2005; Gardner and Drinkwater, 2009; Poffenbarger et al. 2018). Annual mineralization of soil organic N to NH_4^+ and NO_3^- far exceeds typical inputs of NH_4^+ and NO_3^- from atmospheric deposition, manure, fertilizers or biological N fixation (Booth et al., 2005; Osterholz et al., 2017). There are three key controls on soil organic N mineralization: the quantity and chemistry of SOM, soil temperature, and soil water (Probert et al. 1998)

Soil properties, topography, and management interact to affect these environmental controls on the pool sizes and fluxes of inorganic N compounds. Biological soil processes that transform N are associated with the microbial demand for energy and nutrients. Soil N mineralization, which is the depolymerization organic N (i.e., conversion of proteins and peptides to amino acids) that ultimately leads to the production of inorganic NH_3/NH_4^+, is a broad process performed by a range of soil biota under aerobic and anaerobic conditions (Schimel and Schaffer 2012). In contrast, the oxidation and reduction of inorganic N are relatively narrow processes that are largely restricted to aerobic and anaerobic environments, respectively. Nitrification is

performed by small groups of bacteria and archaea, largely under aerobic conditions, to produce NO_3^- that can be easily leached to ground and surface waters, taken up by plants or reduced by denitrifiers. Denitrification is an anaerobic process whereby NO_3^- is reduced to N_2 (Fig. 2). However, denitrification can leak N oxides to the atmosphere including N_2O, which is an ozone-depleting substance and a greenhouse gas. In contrast to NO_3^-, which is easily leached from soils due to its negative charge, the main path of $NH4^+$ loss is in surface runoff since the positively charged ions are attracted to the negatively charged surfaces of clays and humus (Fig. 2). In addition, release of NH_3 gas is can an important path of N loss in dry and hot soils with high pH and where NH_4^+ has accumulated in the surface (Tian, et al., 2018).

The retention of N within the soil-plant system and prevention of N loss to the surrounding environment occurs through plant and microbe N uptake and transformation. Plant roots can take up N in organic (peptides and amino acids) and inorganic (NO_3^- and NH_4^+) forms. This largely occurs through two mechanisms. First, N is taken up through the soil solution during the transpiration process (Engels & Marschner, 1995). Secondly, N is taken up by a diffusive flux driven by concentration gradients produced by the plant itself (Larcher, 1995). Nitrogen that is taken up by plants and microbes is transformed to a wide range of organic compounds. The N contained in plant and microbial residues can be stabilized on the surfaces of mineral particles or within soil aggregates where the N achieves long mean residence times that far exceed the mean residence time of the total soil N pool (e.g.,Mueller & Kogel-Knabner 2009).

Fig. 2. Diagram of soil N cycle and N transport at the interfaces of soil-air and soil-water. In this figure, N inputs, internal fluxes and outputs are illustrated as solid, dashed and dotted arrows, respectively.

By reviewing recent progress in research on soil water processes (static aspect: SWC spatio-temporal variation; dynamic aspect: subsurface flow) and soil N cycling, we can further elaborate the relationships between these two components, identify bottlenecks in our ability to link soil water and N processes, and develop possible solutions for coupling soil water processes with the N cycle.

2. Relationship between soil water processes and the N cycle

To describe how soil water processes affect the N cycle at microsite scale, we present a conceptual model of two discrete mcriosites (pools of solutes) representing extreme end-points of soil N cycling (Fig. 3). Soil water processes play an important role in regulating the abundance, duration, and timing of these microsites (Kleber et al., 2015). In turn, the abundance, duration and timing of these microsites play a critical role in the magnitude and vector of environmental N losses.

At one extreme, the soil solution is rich in dissolved organic matter (DOM), which is the most important energy source (i.e., electron donor) for soil microbes (Fig. 3, left panel). At the other extreme, the soil solution is rich in NO_3^-, which is an important electron acceptor (Fig. 3, right panel). Soil N dynamics will always proceed from the reduced, free energy-rich situation to the oxidized free energy-poor situation, but soil water processes are a key control.

Fig. 3. Soil N cycling in dissolved organic matter (DOM) rich microsites (left panel) and inorganic N-rich microsites (right panel). Over time, the N cycle proceed from the left to the right. Static and dynamic soil water properties affect the abundance, location, duration and timing of these microsites.

In the DOM-rich microsite, an individual pool of soil solution has a large pool of DOM and small pool of inorganic N. The majority of DOM in this scenario is derived from a plant source with a relatively high C/N ratio and free energy (Fig. 3, left panel). Therefore, microbial biomass grows as microbes rapidly mineralize the DOM and N is in high demand. There is little nitrification because DOM is abundant and autotrophic nitrifiers are poor competitors for NH_3/NH_4^+ with heterotrophic microbes that use DOM rather than NH_3/NH_4^+ as an electron donor (Booth et al., 2005). Environmental N losses are minimal due to conservative microbial N cycling (Mooshammer et al., 2014). Despite the small NO_3^- pool, denitrification can occur because rapid DOM mineralization can consume O_2 in the soil solution more rapidly than O_2 can diffuse into the soil solution in the microsite (Blagodatsky and Smith, 2012). Due to the relative abundance of electron donors (DOM) to acceptors (NO_3^-), denitrification will produce mostly N_2 but little NO_x (Firestone and Davidson 1989). In addition to microbial sinks for DOM, mineral soil surfaces are also a sink for DOM due to relatively high concentrations of DOM in solution vs. on mineral surfaces (Kothawala et al., 2009). Although the minerals serve as a net sink, there may be substitution of mineral-adsorbed organic matter with relatively low C/N ratios for plant- or microbe-derived DOM with relatively high C/N ratios (Kaiser et al., 2004).

These mineral-based equilibrium reactions partly drive the DOM-rich microsite to a N-rich microsite.

As DOM is consumed by all these processes, the microsites become enriched in N because the C in DOM is lost as CO_2, while N is cycled conservatively (at least initially). The abundance of DOM-rich microsites usually decreases with soil depth since plant C inputs are less and microbes have more opportunity process the DOM (Fig. 3). There are two ways DOM-rich microsites can be maintained, preventing procession towards the N-rich state: continued direct plant-C inputs or transport of DOM by dynamic soil water flow. The potential rate of DOM diffusion from mineral particles into the soil solution is slower than the rate of microbial consumption and mineral-associated DOM has a low C/N ratio (Kuzyakov and Blagodatskaya, 2015; Sollins et al., 2009). Thus, plant inputs and dynamic soil water flow are the key controls on the duration DOM-rich microsites. In addition, the static SWC can also control the duration of DOM-rich microsites. As soil dries, solute concentrations become higher (Gordon, et al., 2008). Thus, increasing DOM concentrations could increase heterotrophic respiration thereby maintaining relatively low O_2 concentrations despite greater soil air-filled pore space. This would extend the duration of a DOM-rich situation due to the relatively slow nature of anaerobic metabolism. With regard to time, DOM-rich microsites are mostly likely to occur when plants are actively growing or after plant senescence when litters are deposited on and in the soil. Indeed, environmental N losses tend to be small during these times (Lovett et al., 2000) and this pattern can be attributed to strong plant and microbial N

sinks.

In contrast, N-rich microsites are characterized by surpluses of N (Fig. 3 right panel). Although microbial metabolism is inherently limited by available energy (DOM), the scarcity of electron donors has major impacts on N cycle. Primarily, nitrification becomes an important process, which leads to N_2O emissions and an accumulation of NO_3^- pool. As long as the microsites remain aerobic, net nitrification is positive and thus the concentration of NO_3^- increases. Moreover, with relatively low O_2 consumption due to less DOM, there is great likelihood that the microsites will remain aerobic in the absence of a change in SWC. However, a switch to anaerobicity halts nitrification but initiates denitrification. In situations with high NO_3^-/DOM ratios, N_2O emissions tend to be greater because it is an obligate intermediate product in the reduction of NO_3^- to N_2 and electron transfer from NO_3^- to N_2O is greater than N_2O to N_2. Thus, when electron acceptors are plentiful and electron donors are few, NO_3^- is preferentially used as an electron acceptor. There is great opportunity for SWC to drive N losses from these N-rich sites to waterways and the atmosphere. A change in status of static SWC can turn a N-rich site anaerobic and dynamic soil water flow can transport NO_3^- to surface and ground water. N-rich microsites are mostly likely to occur at times and locations that are separated from plant C inputs.

At larger spatial scales (e.g., aggregate, field and watershed), soil water is the solvent for N. Without water, there is no medium for N uptake by plants or microbes. Thus, soil water processes influence the soil N cycle by controlling the biological transformation and physical transport of N compounds. In many ways, these controls

are interactive. From a biological perspective, static aspects of SWC regulate redox potential over longer time periods while dynamic aspects of SWC temporarily or chronically alter redox potential by transporting and connecting electron donors and acceptors that limit N transformations (Hedin et al., 1998; Schade et al., 2001; McClain et al., 2003) (Fig. 4). From a physical perspective, water flow can lead to the coupling or decoupling of electron acceptors and donors or transport dissolved N from the soil system to surface and ground waters (Fig. 3). Therefore, the static and dynamic properties of SWC could interact create hot spots and hot moments of N losses through leaching and gas emissions by affecting plant N uptake, redox potential, and the transport of dissolved N (Dharmakeerthi et al., 2005; Schmidt et al., 2008; Zhu et al., 2012; Keiluweit et al., 2017).

Fig. 4. The diagram of how static and dynamic soil water processes control soil N cycle through biological and physical perspectives.

Static aspects of SWC can create barriers to the connection of electron donors (DOM-rich sites) and acceptors (O_2 and NO_3^--rich sites). First, the low spatial variation of soil water at wet ends of SWC may indicate that electron donors and acceptors have few barriers to connecting with one another. Second, the low spatial variation of SWC and greater solute concentrations under dry conditions may indicate that there are many sites that have accumulated either surpluses of electron acceptors or donors due to stoichiometric mismatches of solutes in isolated microsites. Static aspects of spatial variation in SWC have been clearly shown to create spatial and

temporal segregation between different N transformation processes. Across time, Klefoth et al. (2014) confirmed that switching between N_2O consumption and production was a consequence of the water filled pore space and soil bulk density effects on gas diffusivities, which was an indicator of anaerobic and aerobic conditions. Across space, Hefting et al. (2004) found that where the water table level is <10 cm, 10-30 cm, and >30 cm deep, ammonification, denitrification, or nitrification is the dominant N transformation process, respectively. Alternatively, dynamic aspects of SWC directly affect N losses by transporting N from the plant-soil system and indirectly affect gaseous N losses by connecting pools of electron acceptors and donors.

Subsurface flow can be an important driving force of N transport (Fig. 3). Dissolved N, especially NO_3^-, is easily transported to ground and surface waters through vertical and lateral subsurface flow. This is due to the fact that soil NO_3^- has low exchange affinity with positively charged sites on mineral soil particles and humus. For example, based on long-term monitoring data from 13 catchments in Norway, Bechmann (2014) observed that N concentrations in subsurface flow were 2-4 times higher than in surface flow. Zhu et al. (2012) demonstrated that lateral subsurface flow at the critical soil-bedrock and fragipan interfaces are also important paths of N loss.

There are two primary ways that rapid subsurface flow paths can transport NO_3^- from surface to subsurface. Firstly, NO_3^- can be transported at rates that overwhelm stoichiometric sinks for NO_3^- such that the rate of NO_3^- transport through DOM-rich

locations exceeds the potential rate of immobilization despite high capacity for immobilization (Castellano et al., 2013). Secondly, fast subsurface flows (e.g., preferential flow) may disconnect bulk soil water flow through DOM-rich surface soils, thereby reducing soil NO_3^- losses. Moreover, as soil water moves downward, dissolved organic C concentrations decrease but inorganic N concentrations increase (Fig. 5). As a result, as NO_3^--rich soil solution is transported downward, the probability of denitrification or immobilization in plant or microbial sinks decreases. This results in the soils change from DOM-rich to N-rich in as the depth increasing. Dynamic soil water flow controls these outcomes such that soil solution experiences less change per unit transport in rapid vs. slow flow paths. In addition, antecedent SWC controls infiltration rate that regulates the surface runoff (Defersha and Melesse, 2012; Ouyang et al., 2017). According to previous studies, surface runoff has been recognized as an important pathway of dissolved and particulate N (e.g., NH_4^+ and organic N) losses (Volk et al., 2006; Zhu et al., 2012).

Fig. 5. Patterns of free energy and water chemistry change as the soil water moves downward through the soil profile by slow, moderate and rapid flows.

Given the importance of soil water processes to N transport and transformation, the factors affecting soil water processes can explain significant variation in N dynamics. Topography and soil properties affect the SWC spatial distribution in wet and dry seasons at field scales, respectively (Grayson et al., 1997; Western et al., 1999). These factors have also been reported to be critical in soil N transportation and

transformation. For example, Stewart et al. (2014) revealed that topography is a key landscape-level driver of N mineralization, nitrification and denitrification processes through its control on SWC, soil temperature and nutrient availability. Sogbedji et al. (2000, 2001) found that by controlling the saturation and O_2 content of the soil, drainage class and soil texture determined the main paths of soil NO_3^- loss (poorly drained and fine-textured: denitrification and gaseous emission, well drained and coarse-textured: nitrification and leaching). Poblador et al. (2017) observed that N_2O emissions showed a remarkable spatial gradient with elevation as a result of SWC variation in a Mediterranean riparian forest soil.

3. Bottlenecks in coupling soil water processes and the N cycle

3.1 Knowledge gaps exist between soil water and N cycling studies

Although soil water processes have large effects on soil N dynamics, they have not been comprehensively and fundamentally integrated into N cycling investigations. Traditional work on soil N dynamics has focused on the effects of land management (e.g., Dharmakeerthi et al. 2005; Gutlein et al., 2018), soil properties (e.g., Sogbedji et al., 2000; Sogbedji et al., 2001; Poblador et al., 2017), topography (e.g., Stewart et al., 2014), microorganisms (e.g., Wang et al., 2015), vegetation or crop (e.g., Saha et al., 2016), and weather (e.g., Iqbal et al., 2018). Detailed soil water processes and their effects on soil N cycle have not been fully addressed in previous studies. Parameters like SWC, drainage, and water table were usually used to construct empirical relationships between soil water and N cycle (e.g., Schmidt et al., 2007; Sogbedji et al., 2001; Zhu et al., 2015), but mechanisms remain unclear.

There is significant potential to understand how static and dynamic aspects of

SWC affect soil N dynamics. Anaerobic microsites in well-drained soils have a disproportionally large effect on carbon mineralization rates and can produce significant denitrification (Keiluweit et al., 2017). Moreover, the response of soil N dynamics to changes in SWC is often nonlinear and abrupt. In studies of nitrification, denitrification and N_2O emissions, very small changes in soil water potential or aerobicity can alter N fluxes by more than 100% (Castellano et al., 2010; Zhu et al., 2013). Rewetting of dry soil produces extremely large bursts of C and N mineralization accounting for large fluxes during short periods of time (Birch, 1958; Fierer and Schimel, 2002). Recent work in modeling SWC at small scales (µm) has accurately predicted the occurrence and magnitude of these effects (Evans et al., 2016). Dynamic soil water flow can transport large amounts of dissolved N to subsoils. The amount and chemical composition of the dissolved N are partly an outcome of static soil water processes. Moreover, dynamic flow of dissolved inorganic N can lead to water pollution while transport of dissolved organic N can lead to the accumulation of subsoil organic matter (Kalbitz et al., 2000). These effects can also impact productivity and resource use efficiency of agricultural systems. Zhu et al. (2015) reported that at the farm scale, areas with subsurface flow paths and high SWC temporal variations were the hot spots of NO_3^- leaching and denitrification and thus had lower crop yield and higher optimum N fertilizer requirements.

3.2 Upscaling soil water-N cycle interactions

Previous studies have reported that SWC (static) and subsurface flow (dynamic) have critical impacts on soil N transport and transformation (e.g., Zhu et al., 2012;

Keiluweit et al., 2017; Ouyang et al., 2017). Yet, in process models and management decisions, impacts of dynamic and static soil water processes are usually simplified or substituted by some indirect parameters. For example, drainage class, irrigation, SWC, and water table have been used to interpret the soil N cycle and plant N availability (e.g., Sogbedji et al., 2000; Hefting et al., 2004; Schmidt et al., 2007; Poblador et al., 2017). When studying the N cycle from a hydrological perspective, controlling effects of soil water dynamics on the N cycle were usually investigated without digging into the underlying biological mechanism (e.g., Zhu et al., 2012, 2015; Ouyang et al., 2017; Zhang et al., 2018).

Instead of statistically linking soil N cycling parameters (e.g., N_2O emission, nitrification, denitrification, and N leaching) to parameters of soil water processes, studies proposed that the mechanisms inside the black box should be mathematically formulated and visually displayed (Lin et al., 2015). However, in large-scale soil N cycling investigations, researchers still prefer to rely on the mature models and approaches. For example, area means of soil hydraulic parameters were used in these models, and their spatial heterogeneities were ignored. In addition, more attention to upscaling observations and critical mechanisms at small scales has also been proposed (Lin et al., 2015; Crow et al., 2012).

The hysteresis phenomenon, as it influences soil water dynamics and effects on the N cycle, should be also addressed. For example, in previous studies, N losses through subsurface flow have been reported to increase after dry periods (van Verseveld et al., 2008; Loecke et al., 2017). Explanations for this are varied. First, at

the rainfall event scale, the dry antecedent SWC condition may increase the soil N mineralization and nitrification, which may in turn create a more abundant source of N for leaching (Goldberg and Gebauer, 2009; Castellano et al., 2013; Ouyang et al., 2017). Second, at the yearly scale, dry conditions may limit plant N uptake and result in more legacy N from previous N input (manure, fertilizer etc.) (Loecke et al. 2017; Iqbal et al., 2018). Third, under dry or wet initial conditions, the major component of water source (e.g., lateral flow, shallow ground water flow and matrix flow) in the hydrograph can be different, which could result in different N concentrations in leachate and in the stream (van Verseveld et al., 2008). However, the governing mechanisms for this phenomenon can be vary with respect to different temporal and spatial scales, climate conditions, and geological and pedological backgrounds.

3.3 Models lack robust hydrological or biogeochemical mechanisms

When simulating the soil N cycle in most models at field, watershed and regional scales (e.g., using DNDC, DayCent, SWAT and CLM), soil water processes are usually simplified as parameters easily to be measured or predicted by empirical functions. This includes three main steps. First, soil texture, bulk density and organic matter are used to derive soil hydraulic properties (saturated hydraulic conductivity, SWCs at saturation, field capacity and wilting point) from pedotransfer functions (PTFs). Second, these soil hydraulic properties, as well as other meteorology, vegetation and soil parameters, are used in models to simulate the SWC, water filled pore space, oxygen content and thus oxidization-reduction potential. Third, the oxidization-reduction potential is then used to calculate the nitrification and

denitrification rates based on formulas like the Michaelis-Menten equation.

Uncertainty surrounding soil hydraulic parameters has been recognized as one of the major obstacles for achieving optimal simulations (Chirico et al., 2010; Baroni et al., 2017). Direct measurements of soil hydraulic parameters are costly and time-consuming in the laboratory and field, thus restricting the availability of high spatial resolution data for model simulations (Chirico et al., 2010). Indirect methods, especially the PTFs, have been developed to estimate the SHPs from easily measurable soil physical and chemical properties (Saxton et al., 1986). However, previous studies have demonstrated that applying existing PTFs always introduced substantial uncertainty as they were used outside the datasets used to develop them (Chirico et al., 2010; Liao et al., 2014). To improve simulation, accuracies and spatial heterogeneities of soil hydraulic parameters should be considered in modelling (Sciuto and Diekkrüger, 2010). In addition, factors like subsurface flow, soil matric potential, and soil drying-wetting processes should also be properly mathematically formulated and incorporated in model simulations (Lin et al., 2015).

4. Possible solutions

4.1 Hydrogeophysical tools to study the coupled soil water-N cycle

Non-invasive hydrogeophysical tools have been applied to investigate soil water processes from plot to catchment scales (Fig. 4). Although the success of using these tools to detect soil water processes are restricted by various factors like wetness, salinity, soil texture, etc (Doolittle, et al., 2007; Doolittle and Brevik, 2014). For example, Zhou et al. (2001) used ERT to detect the three-dimensional spatio-temporal variations of SWC at the field scale, while Greer et al. (2017) used the same tool to

visualize wetting fronts of subsurface flow in coal mine valley fill. Robinson et al.
(2012) and Zhu et al. (2010) relied on repeated EMI surveyes to derive the subsurface
soil water redistribution and flow paths at the farm scale. Guo et al. (2014) used
time-lapse GPR survey to reveal the subsurface lateral preferential flow network at
the hillslope scale.

Fig. 6. Scheme of coupling the soil water-N cycle across different spatial scales. Geophysical
tools are used to detect the soil water processes at small scale; soil hydrology and N cycling
models are used to derive critical parameters and functions through small scale study;
hydropedological functional units are the basis the small scale monitoring and simulation, as
well as the key to upscaling.

Because hydrogeophysical tools can be used to determine the spatio-temporal
variation of soil water processes, they have great potential to reveal subsurface N loss
paths and hot spots/moments of N biogeochemistry associated with soil water
processes (Fig. 4). Some attempts have been made to use hydrogeophysical tools to
study the soil N cycle. For example, Kennedy et al. (2018) used GPR to map the
thickness of peat and then, coupled with conventional measurements of peat N
concentrations, calculated the N stock. McDaniel et al. (2017) built a relationship
between measured apparent electrical conductivity and N_2O flux and then used this
relationship to map the spatial pattern of N_2O flux. However, most of these studies
focused on indirect measurement and mapping of soil N stock and flux based on
relationships with soil properties that affected the readings of these hydrogeophysical
tools.

In the future, hydrogeophysical tools will be linked to innovations in soil N-sensing techniques (Fig. 4). New electrochemical tools promise to allow *in situ* measurement of N dynamics without disturbance to soil structure as current methods demand (e.g., Ali et al., 2017). The U.S. National Academy of Science identified the development of 'highly sensitive, field-deployable sensors' as one of five breakthroughs that are required to meet challenges facing food and agricultural sciences (National Academies of Sciences, Engineering, and Medicine, 2018). However, several difficulties remain in this aspect. First, knowledge gaps between soil water processes and the N cycle have not been addressed and fully resolved. Further analyses are still needed by soil hydrologists, biogeochemists and microbiologists to interpret the outputs of these tools. Second, errors and uncertainties have also been reported when using these tools to detect SWC, subsurface flow and soil N parameters (Doolittle et al. 2007; Zhu and Lin, 2009). New technologies and methods of analysis will be required to meet these challenges. Coupled use of automatic high-resolution and high-accuracy hydrogeophysical tools and electrochemical N sensors will create an enormous amount of data. With further development of machine learning and artificial intelligent techniques, we would expect a bright future for using these tools to visualize the spatial and temporal variations of the soil N cycle.

4.2 Upscaling based on the concept of hydropedological function units

To couple the soil water process with soil N cycle, observations and simulations at small spatial scale should be properly upscaled (Fig. 6). The concept of hydropedological function units can be useful in the upscaling of soil water-N cycling

dynamics (Lin, 2011). Soil water-N cycle is jointly and interactively affected by topographic, pedological and hydrological properties (Sogbedji et al., 2000; Stewart et al., 2014; Zhu et al., 2015). The concept of hydropedological function units is used to delineate a large study area into small units and assumes that topographic, pedological and hydrological properties controlling soil water-N cycle are comparatively homogeneous within each unit. Critical soil water and N cycling parameters and mechanisms of these units can be upscaled by constructing the empirical functions between these parameters and ancillary soil, topography and vegetation variables, and then incorporating them into the large scale models and investigations. This upscaling can be based on time stability concepts and block kriging (Crow et al., 2012), and hydrological connections and flows among different units should be addressed at large scales. For example, to upscale the N leaching simulation, Schmidt et al. (2008) delineated so-called "nitrogen response units" and regressed N leaching with percentages of cereals in crop rotations and livestock. In addition, critical functions determined at small scales can also be used to revise or add new modules to the large scale models. For example, to incorporate the small scale observations of subsurface flow at soil-bedrock interface, Fu et al. (2014) revised the SWAT model by adding the corresponding module.

4.3 Coupling soil hydrology and N biogeochemistry models

Coupling soil hydrology models with N biogeochemistry models can be another strategy for integrating soil water-N cycles to improve simulations (Fig. 4). Models are usually developed for certain purposes and to capture specific processes. For this

reason, nearly no model can claim that it could describe all different processes well and satisfy all the requirements of the clients. For example, although DNDC is good at simulating N biogeochemical processes, it sometimes performs poorly in capturing soil water processes since it is based on the simple cascade approach instead of the Richards' equation (Kröbel et al., 2010). Conversely, although Hydrus simulates the percolation and SWC well, it yielded worse simulations of N uptake and N concentration than models addressing the crop growth and N dynamics (Doltra and Muñoz, 2010). Therefore, by coupling models with different purposes, weaknesses of these models can be minimized, and thus the soil water-N cycle can be better simulated. For example, to solve the deficits of DNDC, Kraus et al (2015) suggested coupling it with more complex soil hydrology models based on the Richards' equation.

Recently, Vereecken et al. (2016) proposed that a new generation of models based on a systemic approach should be developed to comprise relevant physical, chemical, and biological processes to address knowledge gaps in understanding soil processes and their interactions. Model improvements have also been attempted by revising the soil water module in N biogeochemistry models, revising the N biogeochemistry module in soil hydrology models, or constructing new models that integrate sophisticate hydrological and biogeochemical processes. For example, Li et al. (2007) built the water and N management model based on the processes of water dynamics, soil temperature, C and N cycles in soils and crops, crop growth, and agricultural management practices. Zhang et al. (2016) the extended the distributed

time variant gain model by integrating detailed processes of soil biogeochemistry ecology. However, we prefer coupling existing models to improve the simulation of soil water-N cycle since it can be easier and more time-efficient than building new models and revising existing models (Jones et al. 2001). One model in this category is the Agricultural Productions Systems sIMulator (APSIM; Holzworth et al. 2014). It operates on a daily time step and has a modular platform, allowing users to add or remove submodels of various complexity. For example, SWC can be simulated with a relatively simple 'tipping bucket' model or with the SWIM model that is based on numerical solution of the Richards' equation and advection-dispersion equations (Stewart et al. 2006; Dietzel et al. 2016).

5. Summary

Soil water processes are critical controls of the soil N cycle. We reviewed the research progresses of SWC spatio-temporal variation and subsurface flow, the soil N cycle, and relationships between these two components. Bottlenecks to coupling the soil water-N cycles, include not considering soil water processes in the soil N cycle, treating soil water-N cycle as a black box, a lack of upscaling, and simplifying soil water parameters in N modeling. Based on these, we proposed possible solutions for coupling the soil water -N cycle. Possible solutions include the use hydrogeophysical tools to better detect soil water-N processes, upscaling the small scale monitoring and simulation based on hydropedological functional units, and coupling soil hydrology models with N cycle models.

This paper provides an alternative approach to studying the soil water-N cycles

that will benefit and enhance our understanding of critical zone sciences. This approach will benefit efforts to better understand how soil and water interact to produce sources of greenhouse gas emission and water eutrophication. Specifically, by opening the black box of the soil water-N cycle across spatial scales, the temporal and spatial heterogeneities of N losses can be better predicted and explained. This information is critical to better manage the N cycle for food production and environmental quality.

An understanding of the coupled water-N cycling mechanisms of NO_3^- leaching and N_2O emissions can aid the development of strategies to reduce these N losses. The concepts of static and dynamic water properties could be used to guide precision N fertilizer (or manure) and irrigation managements that maintain the crop production while minimize the negative N environmental impacts. For example, farmers can apply N fertilizer in narrow concentrated 'bands' or homogenous 'broadcast' applications, and this decision could be informed by the importance of dynamic flow processes; irrigation could be planned around SWC thresholds that produce large N losses. In addition, by opening this black box, strategies (e.g., from different expertise like agricultural, engineering, ecological and sociological) to intercept non-point N contamination and to remove and immobilize excessive inorganic soil N can be targeted on the hot spots and hot moments of N losses. This will greatly improve the effectiveness and pertinence of these strategies.

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