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Nitrous oxide emissions and biogeochemical responses to soil freezingthawing and drying-wetting



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ABSTRACT

Soil freeze-thaw (FT) and dry-wet (DW) cycles are brief transitory biophysical changes, but these events have important implications in determining the timing and magnitude of N₂O emissions and may represent a significant proportion of annual N2O emissions from agricultural systems. It is often assumed that FT and DW cycles influence the processes of N₂O production and emission in a similar manner, however, research has yet to systematically identify the similarities and differences in the mechanisms which lead to potentially higher N₂O fluxes during FT compared to DW cycles. Herein, we present the first review to do so; in addition, we identify strategic research areas required for improving the understanding of FT and DW processes leading to N₂O emissions. There are key differences between the mechanisms that contribute to N2O fluxes during FT and DW cycles, centered on the duration and spatial extent of anaerobiosis, temperature sensitivity of microbial activity, relative gas diffusivity, and soil water dynamics. These differences might increase the risk of N2O emissions during FT cycles relative to soil DW cycles. Current research gaps include (i) the identification of organic substrates made available due to FT and DW cycles, and their contribution to ensuing N₂O fluxes, (ii) an understanding of how cryosuction dynamics potentially influence N₂O production and emission, (iii) understanding and predicting the air-entry potential of soil as it relates to N_2O fluxes, (iv) identifying the relative significance of dissolved N_2O in soil water and its solubility changes during FT and DW phases, and (v) determining microbial community and functional changes across soil spatial and temporal scales. Advances in these areas are recommended for improving process descriptions in biogeochemical models in order to more accurately predict N₂O emissions from soils prone to FT and DW cycles.

1. Introduction

Understanding the mechanisms producing and leading to the emission of potent greenhouse gases, such as nitrous oxide (N₂O), is essential for accurate flux prediction and for developing effective adaptation and mitigation strategies in response to climate change. Nitrous oxide fluxes from soils are exceptionally sporadic at both spatial and temporal scales and thus remain challenging to predict. While the episodic nature of N₂O flux events has been closely related to transient changes in soil biophysical conditions in the field, such as freeze-thaw (FT) and dry-wet (DW) cycles (Burton and Beauchamp, 1994; Davidson, 1992a; Priemé and Christensen, 2001; Skiba and Smith, 2000), many uncertainties persist in our understanding of the mechanisms that lead to N₂O fluxes during these events. More frequent and severe soil FT and DW cycles are anticipated in the future due to climate change (Henry, 2013; IPCC, 2014; Sheffield and Wood, 2008), and without an improved understanding of the mechanisms generating N_2O emissions, our ability to derive predictions and develop recommendations for adapting to climate change and/or mitigating greenhouse gas emissions will be limited.

In agricultural systems, a significant portion (30–90%) of annual N_2O emissions are attributable to soil FT cycling (Abalos et al., 2016; Wagner-Riddle et al., 2007; Yanai et al., 2011). In contrast, the proportion of N_2O emissions resulting from soil DW cycling may be lower (Goldberg et al., 2010; Muhr et al., 2008) – possibly as low as 2% of the annual budget (Davidson, 1992b), yet the rewetting of dry soil remains the main driver for N_2O fluxes for seasonally dry or arid regions (Davidson, 1992a,b). The preciseness of these annual estimates are highly uncertain, because in addition to requiring careful measurement of spasmodic N_2O fluxes, numerous measurements should be recorded

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throughout entire annual cyles in order to better predict annual budget variability (Davidson and Kanter, 2014). Recent work, based on high-frequency micrometeorological measurements, suggests that neglecting N₂O emissions due to soil FT cycles in agricultural systems may lead to an underestimation of global N₂O emissions by 17–28% (Wagner-Riddle et al., 2017). A similar study for soil DW cycling is not available, but it is possible that the proportion of annual N₂O emissions due to soil DW cycling has been similarly underestimated. As such, further studying FT and DW cycles in detail will likely lead to a better understanding of N₂O production and emission.

It is common that N₂O fluxes from soil FT cycles are researched separately from DW cycles, as evidenced by a large body of research on soil FT cycles (Koponen et al., 2006; Matzner and Borken, 2008; Risk et al., 2013; Tatti et al., 2014; Teepe et al., 2001; VanBochove et al., 2000; Wertz et al., 2013) isolated from that of soil DW cycles (Beare et al., 2009; Borken and Matzner, 2009; Davidson, 1992a,b; Harrison-Kirk et al., 2013). Recently, one study characterized the influence of both FT and DW cycles on trace gas emissions and identified that N2O fluxes may be of greater amplitude and longer duration as a result of FT events when compared to DW events (Kim et al., 2012). Based on our compilation of various field-based studies from agricultural soils, the change in N₂O fluxes and peak N₂O fluxes tended to be greater from FT cycles than DW cycles (Fig. 1). Thus, to better understand N₂O production and emission dynamics, research should evaluate and compare the mechanisms contributing to emissions - an undertaking which was not performed by Kim et al. (2012) and that remains lacking.

In theory, the processes of soil freezing during FT events can be considered analogous to soil drying during DW cycles with respect to the soil liquid water phase (Spaans and Baker, 1996) because in both cases the soil *liquid* water content decreases. Upon thawing or rewetting during FT and DW cycling, the soil liquid water content increases, often resulting in anaerobiosis and large N₂O fluxes. Thus, the mechanisms believed to cause high N₂O fluxes during FT cycles are similar to those suggested for DW cycles, such as increased substrate availability, anaerobiosis, and denitrifier activity (Davidson, 1992a,b; Priemé and Christensen, 2001; Stark and Firestone, 1995; Teepe et al., 2001; VanBochove et al., 2000). However, some mechanisms regulating N₂O are unique to FT or DW conditions. For example, in frozen soils physical blocking of soil pores by ice will impede the diffusivity of gases into the soil (e.g. O₂) or out of the soil (e.g. N₂O) until thaw (Risk et al., 2014), while during DW cycles only, soil hydrophobicity may affect mineralization and hence substrates for N2O fluxes especially in soils with

high organic matter (Kaiser et al., 2015). Furthermore, soil FT and DW events occur across different soil temperature ranges, consequently microbial function or community structure and the ensuing N₂O flux dynamics may be dissimilar (Lipson et al., 2002; Sharma et al., 2006; Wertz et al., 2013; Zogg et al., 1997). While N₂O fluxes resulting from FT and DW cycles may have similar drivers (e.g. substrate supply) there are likely important differences in the mechanisms and the balance of mechanisms that regulate N₂O emissions, and systematic evaluations are needed to address this research gap.

Many researchers have recently contributed to current knowledge of N₂O emissions by reviewing cutting-edge measurement techniques and the origins of spatial and temporal variability (Butterbach-Bahl et al., 2013; Henault et al., 2012), the uncertainty in projected emissions (Reay et al., 2012), the pathways for microbial N₂O production via heterotrophic denitrification, ammonia oxidation during nitrification, and nitrifier-denitrification (Hu et al., 2015), and by including N₂O as a part of a global meta-analysis on trace gas fluxes from FT and DW events (Kim et al., 2012). While the review by Kim et al. (2012) nicely quantified the influence of FT and DW events on N₂O emissions, their research did not evaluate the mechanisms or processes contributing to emissions. Herein, we focus entirely, and specifically, on the mechanisms and processes leading to N2O production and emission, and review the current understanding of N2O dynamics during FT as compared to DW cycles. Our goals are to (i) comprehensively synthesize the similarities and differences between soil FT and DW cycling on biogeochemical dynamics and N2O fluxes, to (ii) discuss potential underlying mechanisms which control N₂O fluxes, and to (iii) help shape future research needs and directions in order to better understand the sporadic nature of N₂O emissions during FT and DW cycles.

2. Soil physics behind soil freeze-thaw and dry-wet cycles

Before comparing the effects of soil FT and DW processes on N_2O fluxes and to provide context for readers working in a diversity of areas, we first describe the soil physical processes involved during soil freezing, thawing, drying, and wetting on the movement and redistribution of water and heat, and phase change. These processes are complex and controlled by a variety of factors such as soil temperature, antecedent soil water conditions, solute concentration, and soil texture.

In frozen soils, liquid water coexists with ice, even at very low temperatures, although the fraction of liquid water decreases with a decrease in soil temperature (Black and Tice, 1989; Spaans and Baker,



Fig. 1. The change in N₂O flux from before to after a DW or FT event (n = 11 and 18, respectively) and peak N₂O flux during the DW or FT event (n = 7 and 16, respectively) based on published literature focused on various agroecosystems (Barton et al., 2008; Congreves et al., 2017; Gelfand et al., 2015; Kessavalou et al., 1998; Kim et al., 2009, 2010; Liu et al., 2014; Maljanen et al., 2009; Röver et al., 1998; Vilain et al., 2010; Wagner-Riddle et al., 2007; Xu et al., 2015).

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