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LETTER

Soil degradation determines release of nitrous oxide and dissolved organic carbon from peatlands

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Abstract

Carbon (C) and nitrogen (N) release from peatlands are closely related to water management and soil degradation. However, peat degradation has not been explicitly accounted for when estimating national greenhouse gas inventories. Here, we assembled a comprehensive dataset covering European, Russian and Canadian peatlands and introduced soil bulk density (BD) as a proxy for peat degradation to estimate nitrous oxide (N2O) and dissolved organic carbon (DOC) release. The results show that physical and biogeochemical properties of peat are sensitive to soil degradation. The BD is superior to other parameters (C/N, pH) to estimate annual N2O emissions and DOC pore water concentrations.

The more a peat soil is degraded, the higher the risk of air/water pollution in peaty landscapes. Even after rewetting, highly degraded soils may exhibit high N2O release rates. The estimated annual N2O–N emissions from European, Russian and Canadian degraded peatlands sum up to approximately 81.0 Gg. The derived BD-based functions can assist in computing global matter fluxes from peatlands.

1. Introduction

Peat soils are formed through the accumulation of decayed organic matter (OM) from Sphagnum and other non-moss species (Joosten and Clarke 2002). Peatlands only cover about 3% of the Earth’s terrestrial surface but play a major role in the global water and biogeochemical cycles (Limpens et al 2008, Rezanezhad et al 2016). Peatlands store approximately 10% of the global freshwater resources (Joosten and Clarke 2002), 644 Gt C or 21% of the global soil C pool (Yu et al 2010, Leifeld and Menichetti 2018), and 8–15 Gt N (Limpens et al 2006). At present, about 15% of the world’s peatlands are artificially drained for the purpose of establishing agriculture, forestry, peat extraction and bioenergy plantations (Joosten et al 2012). Peatland drainage creates oxic conditions and accelerates the mineralization of OM and N, which increases the emission of greenhouse gases (GHG) such as carbon dioxide (Tiemeyer et al 2016) and N2O. In addition, drained peatlands also act as a contributor to C losses through the aquatic fluxes of C (e.g. dissolved organic carbon, DOC) into downstream water bodies (Limpens et al 2008). The water-bound C loss equals around 15%–50% of the total GHG emissions (Evans et al 2016). It has been estimated that the world’s drained peatlands cumulatively release 80.8 Gt C and 2.3 Gt N (Leifeld and Menichetti 2018).

The releases of C and N from peatlands are closely linked to peat type such as bogs or fens (Martikainen et al 1993, Repo et al 2009), to anthropogenic activities such as land use and water management (IPCC 2014, Pärn et al 2018), and to peat degradation (Leifeld and Menichetti 2018). In previous studies, selected soil properties (e.g. C/N ratio of top soils (0–30 cm), carbon quality, soil pH) and vegetation types have been...
used to predict the magnitude of N$_2$O emissions and DOC concentrations or fluxes from peatlands (Aitkenhead and McDowell 2000, Klemetsdsson et al 2005, Couwenberg et al 2011, Sihi et al 2016, Kang et al 2018, Leifeld and Menichetti 2018, Leifeld 2018, Pärn et al 2018). To a certain extent, the mentioned parameters reflect peat degradation and the resulting biogeochemical responses. However, in addition to biogeochemical reactions, peatland degradation significantly alters the porous structure of the soil that controls soil moisture and water flow patterns, both of which are factors that impact N$_2$O emissions and DOC fluxes (Holden et al 2012, Pärn et al 2018). The stage of peat degradation has been neglected or insufficiently accounted for in the national and global estimates of N$_2$O and DOC fluxes (IPCC 2014, Leifeld and Menichetti 2018). Thus, there is a challenge to find an easily determinable parameter allowing fast and accurate calculations, taking peat degradation stages into consideration.

We compiled a comprehensive dataset covering entire European, Russian and Canadian peatlands. The dataset contains peat biogeochemical properties, including BD, OM, pore structure, carbon-to-nitrogen (C/N), oxygen-to-carbon (O/C) and hydrogen-to-carbon (H/C) ratios, annual N$_2$O emissions, DOC concentrations and fluxes. BD was used as a proxy for the degree of peat decomposition and degradation (Krüger et al 2013, Liu and Lennartz 2019). The main objectives of this study were to (1) systematically examine the physical (OM and pore structure) and biogeochemical properties (C/N, H/C, and O/C) of peat soils along BD gradients, (2) derive functions between physical (OM and pore structure), biogeochemical properties (C/N, H/C, and O/C) and BD, (3) elucidate the N$_2$O and DOC release as affected by peat degradation.

2. Data and methods

2.1. Data extraction

We searched the ISI Web of Science (http://apps.webofknowledge.com/) and Scopus database (https://scopus.com/) for articles including the terms ‘peat’ AND ‘OM content’ and ‘peat’ AND ‘OM quality’ OR ‘oxygen to carbon ratio’ OR ‘hydrogen to carbon ratio’ OR ‘carbon to nitrogen ratio’ and ‘peat’ AND ‘soil water characteristic curve’ OR ‘moisture retention’ and ‘peat’ AND ‘nitrous oxide emission’ and ‘peat’ AND ‘DOC fluxes’ in the title, abstract or keywords. The papers found by this search were refined by the following criteria: (1) studies from tropical peatlands were excluded because here we concentrate on boreal and temperate peatlands; (2) only articles giving details on physical properties such as BD, OM or ash content determined on undisturbed soil samples were selected; (3) the N$_2$O emissions from peatlands were at least monitored for one year; (4) DOC fluxes were reported from catchment studies.

In total, 68 published articles were selected for this study (supplementary table S1 is available online at stacks.iop.org/ERL/14/094009/mmedia). A dataset of peat soil properties was established including physical properties (peat type, e.g. bog or fen, soil sampling depth, soil pH, BD, OM, total porosity and macroporosity), biogeochemical properties (mole ratio of O/C and H/C and mass ratio of C/N), N$_2$O emissions, and DOC fluxes. All the data were basically extracted manually from the tables and texts of the publications.

We also collected pore water samples from 159 sampling locations (BD ranging from 0.04 to 0.44 g cm$^{-3}$) from 44 peatland field sites (11 bog sites and 33 fen sites) in European countries (Germany, Sweden, Poland and the UK). The determination of DOC concentrations in soil pore water (0–60 cm depth) was carried out employing the dialysis sampler technique (Zak and Gelbrecht 2007). In addition, BD, C/N ratio, and pH were analyzed for all peat water extraction locations.

2.2. Statistical analysis and model development

Nitrous oxide emissions are mainly related to top soil properties (Leifeld 2018). Thus, only top soil parameter values were used to compute C/N–N$_2$O and BD–N$_2$O relationships. In cases where several values of bulk density (BD) were reported for the top soil (e.g. 0–10, 10–20, 20–30 cm), one average value was calculated (0–30 cm). The dataset contains one negative N$_2$O emission value. Therefore, a positive value of 0.02 was added to all the values of annual N$_2$O emission before log-transformation. For the annual groundwater table, below ground surface is positive and above ground surface is negative. One negative value of ~1 was observed in the dataset, so a positive value of 2 was added to all values of groundwater table before log-transformation. Linear regression models were used to estimate annual N$_2$O emissions using BD and C/N of top soils (0–30 cm). In addition, a multiple regression model was employed to estimate N$_2$O emission from combined BD and groundwater table values. BD, C/N, and soil pH were tested for their power in predicting DOC pore water concentrations (linear regression model). All statistical analyses were performed using the default ‘stats’ package of R (R Core Team 2015).

2.3. Classification of peat degradation status using BD

No detailed map of BD is available for the considered regions/continents. In order to use the derived functions for upscaling N$_2$O emissions, we introduce a classification scheme of peat soil degradation. There is only very limited information available on a BD-based classification scheme of ‘peat degradation status’ (Lennartz and Liu 2019). From the obtained
relationships between BD and physical, as well as biogeochemical properties of peat soils, several critical points emerge: (1) BD of drained peat soils is never less than 0.05 g cm\(^{-3}\); from this, we classify here that pristine peat has a BD of <0.05 g cm\(^{-3}\); (2) BD of 0.2 g cm\(^{-3}\) is a critical point; above and below this value, macroporosity and C/N ratios follow different functions with BD (Liu and Lennartz 2019); (3) with increasing BD from 0.2 to 1.0 g cm\(^{-3}\), the C/N ratio remains almost constant but OM decreases proportionally. Based on above information, peat soil degradation status can be classified into four classes: pristine peat, BD ≤ 0.05 g cm\(^{-3}\); moderate degradation, 0.05 < BD ≤ 0.20 g cm\(^{-3}\); high degradation, 0.20 < BD ≤ 0.40 g cm\(^{-3}\); extreme degradation, BD > 0.4 g cm\(^{-3}\). Similar classes of peat degradation status were reported by Lennartz and Liu (2019).

2.4. Upscaling model results to regional scales

The \(\text{N}_2\O\) emissions at regional scale were estimated by the area of differently degraded peatlands and the according \(\text{N}_2\O\) emission factors. The total undegraded peatlands (pristine) in Europe, Russia and Canada cover an area of 148 160, 1304 090, and 1132 016 km\(^2\), respectively (Joosten 2010). The proportions of undegraded peatlands to total peatlands in Europe, Russia and Canada are 49%, 95%, and 99.8%, respectively (Joosten 2010). The \(\text{N}_2\O\) emissions from pristine peatland were set to zero as reported by IPCC (2014) and the equation in this study. Thus, \(\text{N}_2\O\) emissions from peatlands were computed for degraded peatlands only.

The area of differently degraded peatlands was estimated based on total degraded peatland area and the according proportions. The total degraded peatlands in Europe, Russia and Canada cover an area of 157 037, 71 600, and 1820 km\(^2\), respectively (Joosten 2010). From the top soil (0–30 cm) BD distribution map (figure S1 in the supporting information), the proportion of moderately degraded, highly degraded, and extremely degraded peatlands to total degraded peatlands in Europe, Russia, and Canada were estimated. The annual \(\text{N}_2\O\) emission factors for differently degraded peatlands were estimated using a respective average BD value of each degradation class (0.13, 0.30, and 0.55 g cm\(^{-3}\)) and the relationship between annual \(\text{N}_2\O\) emissions and BD. In addition, the maximum annual \(\text{N}_2\O\) emissions were computed, assuming that the already degraded peatlands will continue to degrade to an extreme degradation stage in the future.

3. Results and discussion

3.1. BD as a predictor of peat degradation

Peat degradation significantly reduced the soil OM content and porosity. A strong negative linear relationship was observed between the soil OM content and BD \(R^2 = 0.86, p < 0.001\); figure S2 in the supporting information) and between total porosity, macroporosity (equivalent pore diameter of 50 μm; Schindler et al 2003) and BD (figure S3 in the supporting information, \(R^2 = 0.70, p < 0.001\)). In natural peatlands, the soil pore structure is dominated by a large proportion of interconnected macropores (figure S3 in the supporting information), which are composed of peatforming plant residues. These macropores facilitate water movement in peat (Quinton et al 2008, Liu and Lennartz 2019). The peat parent plant material (e.g. sedge, woody or Spagnum peat) vanishes upon soil degradation and is partly replaced by fine mineral particles, resulting in a substantial decrease in OM, porosity and macroporosity and thereby also in hydraulic conductivity. With an increase in BD to a value greater than 0.2 g cm\(^{-3}\), macroporosity remained overall constant because of the formation and persistence of secondary macropores (e.g. root channels, wormholes, cracks, and piping).

Peatland degradation not only decreases the quantity of soil OM, it also has a strong effect on the quality of OM as denoted by changes in elemental ratios (C/N, O/C, and H/C). The C/N ratios (mass ratio) ranged between 10.8 and 130.6 (figure 1; table S2 in the supporting information). The observed wide range of C/N ratios (from 10.8 to 130.6) in peat soils is caused by differences in vegetation, the degrees of peat decomposition and degradation, peatland management and field conditions (Klemmedtsson et al 2005, Leifeld and Menichetti 2018, Leifeld 2018). The C/N ratios decreased with increasing BD and a strong linear-log relationship emerged \((R^2 > 0.50, p < 0.01)\), although a greater variance of C/N ratios was observed for peat soils with BD < 0.2 g cm\(^{-3}\). Peat degradation leads to the mineralization of organic carbon and relative enrichment of N (Krüger et al 2015). In contrast to bogs (figure 1(a)), fens have lower C/N ratios in peat soils with BD < 0.2 g cm\(^{-3}\) (figure 1(b)), indicating lower C and higher N accumulation in fens than bogs (Wang et al 2014, Zaccone et al 2017). The lower C/N ratios (less than 30) in fens occurred at almost all peat degradation stages.

The O/C and H/C ratios of organic soils are widely used as indicators of the relative abundance of different groups of compounds such as lipids, lignins and carbohydrates (Preston and Schmidt 2006, Bader et al 2018). For bogs, both O/C and H/C ratios decreased initially with peat degradation and the subsequent BD increase from 0.01 to ~0.20 g cm\(^{-3}\). Beyond this threshold, no clear tendency was observed (BD values of 0.20 to 0.66 g cm\(^{-3}\)). For fens, higher O/C and H/C ratios were more likely to appear at highly degraded peat sites and the relationships between BD and O/C, H/C ratios are unclear. Pristine bogs contain high amounts of carbohydrate-like materials (figure 2(a)) because of the accumulation of easily degradable compounds during the peat formation process. An increase in BD from 0.05 to 0.2 g cm\(^{-3}\) marks a preferred decomposition of the easily
degradable compounds, leading to the accumulation of recalcitrant compounds (lignin-like materials, figure 2(a)). The lower carbon quality constrains C mineralization rates, slowing down the reduction of C/N ratios with peat degradation (figure 1). Nonetheless, higher H/C and O/C ratios were sometimes observed in bogs and fens with greater BD (e.g. BD > 0.2 g cm$^{-3}$), carbohydrate-like materials, figures 2(a) and (b)), possibly because of inputs of fresh plant litter to the topsoil; thus, highly degraded peat soils are often rich in carbohydrates (Kögel-Knabner 2002, Bader et al 2018). Although the OM quality in peat is supposedly a depth-dependent property (Leifeld et al 2012, Zaccone et al 2007, 2017), the generated dataset revealed no correlation between the elemental ratios (O/C and H/C) and soil sampling depths (Pearson’s correlation coefficient, $r < 0.1; p > 0.05$). In a natural state, BD is expected to increase with depth, while with progressing degradation, the top soil becomes more compacted (Zak and Gelbrecht 2007, Liu and Lennartz 2019).

3.2. Annual N$_2$O emission from peatlands

The overall annual N$_2$O emissions from peatlands (from assembled dataset) ranged from −0.01 to 61.1 kg N ha$^{-1}$ yr$^{-1}$. In natural peatlands, the observed annual N$_2$O emissions were low, ranging from 0.01 to 1.46 kg N ha$^{-1}$ yr$^{-1}$. In contrast, the mean annual N$_2$O emission from drained peatlands was significantly higher than that from natural peatlands ($p < 0.001$), with values of 8.94 ± 11.25 kg N ha$^{-1}$ yr$^{-1}$ (mean ± standard deviation). The gas release from drained peatlands differed significantly according to land use. Croplands emitted higher N$_2$O fluxes (16.8 ± 14.8 kg N ha$^{-1}$ yr$^{-1}$) than grassland (10.7 ± 10.7 kg N ha$^{-1}$) and forest (8.5 ± 9.4 kg N ha$^{-1}$ yr$^{-1}$). It is likely that most
As peat degradation becomes more severe, peat soils under agricultural use (cropland) have low C/N ratios (<30) and N as well as phosphorus fertilizer applications may additionally enhance N₂O emissions (Liimatainen et al. 2018, Saurich et al. 2019). In peatland forest, most of the high N₂O emission values (>10 kg N ha⁻¹) originate from afforested agricultural peatlands, indicating that land use history is another important factor affecting N₂O emission. The dataset did not reveal a significant difference in N₂O emission between boreal and temperate forest peatlands. The N₂O emissions from top soils (0–30 cm) with BD < 0.15 g cm⁻³ were comparable with those from natural peatlands. For these peat soils, N₂O emission were constrained by high C/N ratios and low carbon quality such as lignin-like materials (Hume et al. 2002, Millar and Baggs 2004). As peat degradation becomes more severe (e.g. BD > 0.20 g cm⁻³), N₂O emission increases rapidly because the lower C/N ratios (<30) promote N mineralization and nitrification (Klemetsson et al. 2005).

The assembled dataset shows that the N₂O emissions from rewetted peatlands (by blocking drainage ditches and/or raising the water table; BD > 0.20 g cm⁻³) ranged from 2.3 to 27.4 kg N ha⁻¹ year⁻¹. In rewetted and degraded peatlands, the average annual groundwater table (10–30 cm below ground surface) and the soil water content (0.5 to 0.7 cm³ cm⁻³) together with the high BD create both aerobic and anaerobic conditions, allowing N₂O emissions from both nitrification and denitrification processes (Repo et al. 2009). The relationships between BD and annual N₂O emissions are comparable under dry (drainage) and wet conditions (natural and rewetted) for highly degraded peat soils, indicating that restoration measures such as blocking ditches or raising of the water table are ineffective at reducing N₂O emissions (figure 3 and Höper et al. 2008). The reduction of N₂O emissions from peatlands requires a water table at ground surface or above, which constrains nitrification and denitrification. Furthermore, the produced N₂O is efficiently reduced to N₂ under totally water saturation conditions. Thus, the stages of degradation should be considered when initially planning peatland restoration measures.

The C/N ratio of top soils (0–30 cm) is often used to predict the magnitude of N₂O emissions (Klemetsson et al. 2005, Leifeld 2018). The C/N ratio may be determined more precisely than the BD (possible compaction during sampling); however, the experimental analysis to determine the C/N ratio takes much more effort than BD measurements. Considering a regional or continental scale, BD values are more readily available than C/N ratios. In this study, we demonstrated that prediction of N₂O emissions based on the BD of top soils (0–30 cm depth) is superior (r² = 0.56, p < 0.001; figure 4(a)) to computations using C/N ratios (r² = 0.40, p < 0.001; figure S4 in the supporting information) and soil pH (r² = 0.19, p < 0.001). Addition of the groundwater table to the model improves the estimation accuracy (r² = 0.60, p < 0.001; figure 4(b)). Over an increase in BD from 0.2 to 1.0 g cm⁻³, the C/N ratio remains almost constant, while the soil OM content decreases from 80 to 20 wt% due to mineralization. Thus, we assume that BD better reflects OM mineralization than the C/N ratio. The abundance of micro-pores or dead-end pores in highly degraded peat soils favors a more frequent occurrence of local anaerobic conditions and...
consequently an increase in denitrification-derived N\textsubscript{2}O emissions. This phenomenon has been reported earlier for mineral soils (Sey et al 2008, Laudone et al 2011, Wang et al 2018).

Based on the relationship between BD and annual N\textsubscript{2}O emission (figure 4(a)), we estimated that the average annual N\textsubscript{2}O emission factor for moderately degraded peat, highly degraded peat, and extremely degraded peat are 0.68, 3.94 and 14.1 kg N ha\textsuperscript{-1} yr\textsuperscript{-1}, respectively. According to the annual N\textsubscript{2}O emission factors and according areas of differently degraded peatland (table 1), we estimated that the annual N\textsubscript{2}O–N emissions from European, Russian and Canadian peatlands are 68.6, 12.1 and 0.24 Gg, respectively. In this study, the estimated annual N\textsubscript{2}O–N emissions from European peatlands is comparable to that estimated using IPCC emission factors, but lower than that estimated using a fuzzy logic model (Leppelt et al 2014). The N\textsubscript{2}O emissions from Canadian peatlands are low because pristine and moderately degraded peat soils cover almost the entire area (Moore 1994). Peat degradation continues under climate change scenarios because the high frequency and intensity of droughts will promote carbon cycling (Fenner and Freeman 2011). The reasonable assumption of a further increase in the state of peat degradation of already degraded peatlands to extreme degradation level would result in an emission of approximately 324 Gg N\textsubscript{2}O–N yr\textsuperscript{-1}. The N\textsubscript{2}O emission from peatlands are closely related to peat types (bogs and fens) but this information is not available from most publications. The accuracy of estimation could be improved with detailed BD maps, peat type and land use history information.

![Figure 4. Annual N\textsubscript{2}O flux regression models of different sites, calculated using the top soil bulk density BD; (a), (b) using BD and groundwater table (WT). The dashed plane (b) represents the multiple-regression model: \( \ln(\text{N}_2\text{O–N}) = 1.82 + 1.84 \ln(\text{BD}) + 0.49 \ln(\text{WT}) \), \( R^2 = 0.60, p < 0.001 \).](image-url)
3.3. DOC concentrations and fluxes

The peat pore water DOC concentrations ranged widely, from 9.3 to 288 mg L\(^{-1}\). The large variance of DOC concentrations was related to land management and degradation stages (figure 5(a)). In natural peatlands, the average pore water DOC concentration was 36.7 ± 40.4 mg L\(^{-1}\) (mean ± standard deviation), which was significantly lower than those in drained peatlands (69.4 ± 29.0 mg L\(^{-1}\)) and rewetted peatlands (101.2 ± 71.4 mg L\(^{-1}\)). No significant differences were detected between DOC concentrations in drained and rewetted peatlands (\(p > 0.05\)), which is also evident from the data of Fiedler et al. (2008) and Holl et al. (2009) in figure 5(a). The DOC concentration was found to depend more strongly on BD (Pearson’s correlation coefficient, \(r = 0.76, p < 0.01\)) than on soil pH (\(r = 0.42, p < 0.01\)) and the C/N ratio (\(r = 0.33, p < 0.01\)). A positive linear relationship between BD and DOC concentrations was obtained (figure 5(a); \(R^2 = 0.62, p < 0.001,\) own data). Higher DOC concentrations in highly decomposed and degraded peat (especially under restoration) are probably related to the dissolution of redox-sensitive Fe(III)-OM compounds and to some extent also to the lowered hydraulic conductivities (Zak and Gelbricht 2007). During drainage and peat mineralization refractory, OM compounds are accumulated due to chemical binding of amorphous and more crystalline ferric compounds, also formed under oxic drained conditions (Zak et al. 2018). The expected duration of the suspected DOC mobilization caused by iron reduction or other biogeochemical processes after rewetting of degradation of peatlands is uncertain. Therefore, stages of degradation should be considered when DOC export was estimated using the differences between DOC concentrations before and after drainage or rewetting (IPCC 2014, Evans et al. 2016).

The DOC fluxes (\(n = 45;\) most values are from bogs) from peatland catchments varied from 35 to 804 kg C ha\(^{-1}\) year\(^{-1}\). The average DOC fluxes from drained peatlands (470 kg C ha\(^{-1}\) yr\(^{-1}\)) are significantly higher than those from natural undrained sites (139 kg C ha\(^{-1}\) yr\(^{-1}\); \(p < 0.001;\) figure 5(b)) because discharge increases and DOC concentration increased after drainage. Peatland rewetting reduces the hydraulic gradient and thereby the discharge volume. The limited dataset suggests that DOC fluxes after peatland restoration decreased by 0.45%–82.6%, depending on the fluxes prior to rewetting (Pearson’s correlation coefficient, \(r = 0.76, p < 0.01\)). However, positive effects of restoration measures might be impaired by preferential pathways (Turner et al. 2013, Liu et al. 2017) or reduced acid deposition (Monteith et al. 2007, Evans et al. 2012) maintaining high DOC transport rates. Currently, DOC fluxes cannot be upscaled based on peat degradation stage because BD information is not available in the analyzed studies. The observed relationship between BD (peat degradation) and DOC pore water concentrations provides a first step to calculate DOC fluxes at larger scales. However, volume fluxes from the considered peatlands would be required.

4. Conclusions

This is the first time that soil physical and biogeochemical properties and the related N\(_2\)O and DOC release are systematically explored as a function of peat degradation using BD as a proxy. The derived simple, though robust model is useful to estimate annual N\(_2\)O emissions and DOC concentrations across a wide range of land management regimes. This study highlights the importance of peat degradation as the main driver of N\(_2\)O and DOC release. BD as a proxy for peat degradation is superior to other independent variables because it mirrors both biogeochemical and physical transformation processes. The BD explains >50% of
the variation of biogeochemical properties, annual N\textsubscript{2}O fluxes and DOC concentrations, which enables upscaling of the N\textsubscript{2}O and DOC release from individual sites to regional and continental scales. Until today, the stages of degradation remains unconsidered in the development of emission factors for peatlands (IPCC 2014), which may result in an over- or under-estimation of N\textsubscript{2}O and DOC release at a national scale. Highly degraded peatlands are at risk of becoming major sources of N\textsubscript{2}O release. Our work suggests that restoration measures may be unsuccessful because the soil moisture of top soil is still favorable for N\textsubscript{2}O production.

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Data availability statement

The data that support the findings of this study are available from the corresponding author upon reasonable request. The data are not publicly available for legal and/or ethical reasons.

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