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The influence of increasing mineral fertilizer application on nitrogen leaching of arable land and grassland—results of a long-term lysimeter study

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Introduction: Despite various efforts to reduce nitrogen leaching from agricultural land, the permissible nitrate concentrations in groundwater have often been exceeded in the past. Intensive farming is often seen as the cause of the deterioration in water quality. Therefore, the present lysimeter study aimed to quantify nitrogen (N) leaching at different N fertilization levels for the agricultural land use systems of arable land and grassland to derive suitable management measures for improving groundwater quality.

Methods: The effects of three different of mineral fertilization treatments (50%, 100%, and 150%) in arable land and grassland use on four distinct soil types (loamy sand, sand, loam, loess) concerning seepage formation, nitrogen concentrations, nitrogen loads, dry matter yields and nitrogen balances were tested. The study was conducted at the lysimeter facility of the Helmholtz Centre of Environmental Research – UFZ at Falkenberg (northeast Germany). Twenty-four non-weighable lysimeters with a surface area of 1 m² and a depth of 1.25 m were managed as grassland and arable land with three different fertilization treatments since 1985.

Results and Discussion: For arable land use, N leaching differed between the studied soil types, with the highest N loads from the sand (36.6 kg ha⁻¹ yr⁻¹) and loamy sand (30.7 kg ha⁻¹ yr⁻¹) and the lowest N loads from loess (12.1 kg ha⁻¹ yr⁻¹) and loam soil (13.1 kg ha⁻¹ yr⁻¹). In contrast to grassland use, a reduction of N fertilization level by 50 % did not result in reduced N leaching for arable land, whereas a maximal 29% reduced dry matter yields was observed. An increase of N fertilization by 50 % did not cause significant enhanced N leaching at arable land use. Soil- and management-related factors (soil type, texture, soil tillage, crop rotation, and others) mask the effect of increased N fertilization rates in arable land using lysimeters. For arable land use, a reduction of N fertilizer levels as the only measure was insufficient to reduce NO₃⁻ leaching, and other strategies besides N fertilization levels are required to improve groundwater quality. Measures should be targeted to reduce N losses by mineralization processes.

KEYWORDS

Lysimeter, nitrogen leaching, seepage, mineral fertilization, land use

1 Introduction

Today, agricultural land use is often suspected of being responsible for the chemical contamination of groundwater that, in some cases, has been proven to exceed threshold values in many areas (1–3). Extensively applied pesticides and fertilizers that have not been implemented in a plant-physiological manner reach ground and surface waters via surface erosion or the seepage pathway in the case of intensive land cultivation and have a detrimental effect on water quality. In the case of mineral fertilizers, excess nitrogen and phosphorus, in particular, can cause significant water quality problems. The leaching of unused nutrients also represents a cost factor from a farm management point of view, which can be minimized by excellent site-adapted land management (4).

Numerous scientific publications are available regarding the transport and transformation processes of nitrogen (N) compounds in the unsaturated soil zone and leaching into groundwater (5–7). Nitrate (NO_3^-) has often been the focus of investigations since high NO_3^- concentrations in drinking water can adversely affect human health. On the one hand, NO_3^- can be converted to nitrite (NO_2^-) in the body under certain circumstances. This is especially critical for infants in the first months of life due to a possible cyanosis hazard. On the other hand, NO_2^- can form the so-called nitrosamines with a probabilistic cancer risk and the resulting disease burden (8). The risk of developing colorectal cancer significantly increased in the case of long-term exposure to more than 16.75 mg $\text{NO}_3^- \text{L}^{-1}$ (9). N loads into surface waters and seas have a eutrophic effect. As a result, algal blooms and oxygen deficiency occur more frequently. To achieve the European limit value for NO_3^- in groundwater of 50 mg $\text{NO}_3^- \text{L}^{-1}$ (10, 11), the existing NO_3^- surpluses must be reduced and, thus, the N inputs should be drastically reduced (12).

The relationships between mineral N fertilization and N leaching have been investigated in numerous research projects (13–23). It has been proven in several studies that there is a close link between NO_3^- leaching losses from fertilized crops and N application rates that exceed crop demand (excessive surpluses of fertilizer N) (5, 22, 24–26). Otherwise, several studies found no significant correlation between NO_3^- leaching and the fertilizer N surpluses (23, 27–29). As a reason for this, it was pointed out that a large part of the recently added residual N fertilizer is retained in soils in organic pools with intermediate resistance to mineralization (23, 29–31). Consequently, it was noted in different ^{15}N studies that most NO_3^- leaching from arable land originates from the mineralization of organic soil N (29, 32–34). However, very few long-term studies consider the fate of ^{15}N added in mineral fertilizers in soils, the long-term effects on the soil mineralization capacity, and associated leaching losses (31, 35).

Under the climatic conditions of Southern and Central Europe, climate change is predicted to result in reduced groundwater recharge rates and declining groundwater levels due to the expected development of drought effects. As a result of lysimeter investigations under the climatic conditions in northeastern Germany, a close interaction between the climatic conditions and

the nutrient leaching with seepage was proven. In dry years, no seepage formation was observed. The reappearance of seepage in subsequent wet periods was associated with exceptionally high NO_3^- concentrations, and leaching losses, which exceeded the applicable drinking water limits many times and could lead to a significant risk to water quality (36).

Excessive mineral fertilization, often associated with intensive land management, significantly increases water resource pollution (37–39). However, the leaching of NO_3^- from arable land is influenced by numerous factors. Weather, soil properties, land use, crop rotation, soil tillage intensity, amount, type, and timing of N fertilizer application are considered the main drivers (40, 41). It is known from several studies that NO_3^- leaching is reduced in grasslands compared to cropland due to high N efficiency and intense microbial biomass turnover under permanent vegetation (42–44). It has been reported that even with high N fertilizer applications, NO_3^- leaching remains low under mechanically managed grassland (45). Increased NO_3^- leaching from grassland was only monitored with extremely high N fertilizer surpluses (46, 47). Higher NO_3^- leaching was observed in several studies for no-till systems because of a higher proportion of macropores favoring preferential flow (48–51). Otherwise, higher N leaching in the treatments with intensive soil tillage compared to no-till was reported by others and explained by increased mineralization due to oxygen input by soil movement (52–54). Some studies, in turn, showed no significant effect of soil tillage on NO_3^- leaching (55–57). Hence, the impact of soil tillage on NO_3^- leaching was discussed controversially. Differences in NO_3^- leaching were observed between mineral and organic N fertilizers (29, 58). Animal manure is suspected to have a higher NO_3^- leaching risk compared to mineral fertilizers because of its low N use efficiency (29, 59).

Using a random forest model with a dataset of 48 farms, Dieser et al. (41) identified the factors of soil type, texture, precipitation in October, and crop rotation (main and subsequent crop) as the most relevant factors for the NO_3^- leaching potential of arable land. It was also confirmed by other authors that NO_3^- leaching is determined by a combination of environmental conditions (climate and soil) and cropping system management factors (crop rotation, tillage, and fertilization) (60, 61) whereas, among the environmental factors, NO_3^- leaching was positive correlated with precipitation during the hydrological year (April–March) and during early winter (October–December) (22). Main crops with high N requirements (e.g., winter wheat), large amounts of residues (e.g., rape, legumes), intensive soil movement during harvest (e.g., potatoes), and a high proportion of organic fertilizers (e.g., maize) led to high-soil mineral contents (0 cm–90 cm) in autumn (before the start of the annual leachate period) and can be regarded as a predictor for the NO_3^- leaching potential (41).

The main objectives of the present long-term lysimeter study were to

- i) quantify NO_3^- leaching and NO_3^- concentrations of seepage at the different agricultural land use systems, arable land, and grassland;

- ii) determine the effects of different soil types and different N fertilization levels on the NO_3^- leaching and NO_3^- concentrations of seepage and dry matter yields of both land use systems (arable land and grassland);
- iii) evaluate if a reduction of N fertilization is a suitable measure to improve the groundwater quality in the long term.

Long-term studies are scarce, considering the effect of reduced N fertilization on NO_3^- leaching at agricultural land use systems. However, these studies are necessary to derive efficient management strategies to improve groundwater quality while sustaining high crop productivity.

2 Materials and methods

2.1 Lysimeter facility

A combined multifactorial lysimeter experiment with the experimental components mineral fertilization, land use, and soil type was set up to clarify the relationships between the level of mineral N fertilization and the leachate-bound nutrient discharge (62). Lysimeter experiments are especially suited to investigate water and solute fluxes in soils (63–68). Therefore, a long-term lysimeter experiment was created at the Lysimeter facility of the UFZ in Falkenberg (coordinates 52.859780 N, 11.812595 E) in 1985. The site is located in the North-East German lowlands. It belongs to the temperate zone of Central Europe within the transition zone from maritime to continental climate with an average precipitation of 524.5 mm (1968–2007) and an average temperature of 9.2°C (1994–2007).

This study is based on 24 non-weighing gravity-flow (free drainage) lysimeters (NWLYS). The simple NWLYS type is often used in Germany and other central European countries for applied research on land management and its impact on drainage water quantity and quality (69, 70). They were constructed in the form of a sheet steel vessel with a surface in the shape of a square with sides measuring 1 m × 1 m (surface area of 1 m²) and a total depth of 1.25 m. After the installation at the lysimeter facility, a 25-cm-thick filter layer (sand over gravel over stone gravel) was placed at the bottom of the vessels. A polyvinyl chloride drainage pipe (inner diameter of 63 mm) coated with well-filter gauze was laid within the filter layer to collect the seepage water and drain it into a storage tank in the lysimeter cellar (63, 71).

The lysimeter vessels were filled with soils representative of agricultural sites in the Elbe River basin in 1981 and 1982. The soil types, such as loamy sand, sand, loam, and loess, were used. The soil profiles were mined separately for topsoil (0 cm–30 cm) and subsoil (31 cm–100 cm) at the extraction sites and transported to the Falkenberg lysimeter station. There, the profiles were then installed in the lysimeter vessels in a largely profile-compliant manner. During the manual filling of the lysimeters, particular emphasis was placed on restoring the soil structure of the extraction site

within the lysimeter. The lysimeters were irrigated to accelerate the setting process after this filling procedure (approximately 100 mm of irrigation water per year).

The most essential soil-physical and soil-chemical properties of the soils in the lysimeter vessels are listed in Table 1.

The results presented here relate to the period 1985–2023. The progression of the experiment over time is shown in a timeline (see Supplementary Figure S1).

In addition, selected climate parameters were also measured at the UFZ lysimeter facility. Precipitation was measured daily at ground level and a height of 1 m above the ground using rain gauges with a collecting surface of 200 cm² (standard rain gauge of the German Meteorological Service).

2.2 Cultivation of the lysimeter soils

Table 2 provides an overview of the different agricultural management systems for arable land and grassland and the associated fertilizer applications. Mineral fertilizers were applied in liquid or solid form. The arable lysimeters were managed according to a grain-fodder crop rotation (clover-grass – winter wheat – winter barley – oats). After harvesting winter barley, a mixture of corn and sunflower was grown as a catch crop, which was harvested in late autumn and removed from the lysimeters. The cultivation was typical for agricultural production in Germany, while fertilization was more experimentally designed. Mineral fertilization (N, P, and K) was determined according to the recommendations for optimal supply valid in the mid-80s of the last century (this corresponds to the fertilization treatment 100%). A variation of mineral fertilization was carried out using increases and decreases of 50% each. Organic fertilization was not applied during the entire study period. The established crop rotation has been maintained ever since. The fertilization regime was not changed throughout the experimental period. In 1985, a stand consisting of 30% German ryegrass (*Lolium perenne*), 40% orchard grass (*Dactylis glomerata*), 20% timothy (*Phleum pratense*), and 10% bluegrass (*Poa pratensis*) was established on lysimeters managed as grassland. The grassland lysimeters were cut 4 times a year.

The lysimeters (both grassland and arable land) were irrigated from 1985 until 2003 according to the plant's physiological requirements for yield maximization. Depending on crop and specific climate conditions, irrigation water was additionally applied. This irrigation regime was changed in 2004. From this date onwards, the crops were irrigated exclusively to safeguard plant stocks, which significantly reduced the amount of irrigation water applied (up to 50 mm annually).

Nitrogen was fertilized as calcium ammonium nitrate (CAN, 26% N, 10% Ca). The granulated fertilizer was annually spread (treatments according to the management schedule of Best Management Practice) in spring according to the development stages of the cultivated crops. The mineral fertilizer “Triple Super Phosphate” (Helm AG, Hamburg, Germany) containing 20% total P was applied for mineral P fertilization of the lysimeters. Grain

TABLE 1 Basic parameters of the lysimeter soils.

| Soil texture | Sand (S) | | Loamy sand (LS) | | Loam (L) | | Loess (Loe) | |
|-----------------------------------|----------|---------|-----------------|---------|----------|---------|-------------|---------|
| | Topsoil | Subsoil | Topsoil | Subsoil | Topsoil | Subsoil | Topsoil | Subsoil |
| Sand (%) | 88.2 | 91.2 | 73.6 | 75.2 | 50.4 | 57.6 | 4.7 | 2.8 |
| Silt (%) | 6.7 | 6.7 | 14.3 | 17.4 | 37.5 | 20.3 | 74.8 | 80.9 |
| Clay (%) | 5.1 | 2.1 | 12.1 | 7.4 | 12.1 | 22.1 | 20.5 | 16.4 |
| ζ_d (g cm ⁻³) | 1.34 | 1.66 | 1.48 | 1.84 | 1.62 | 1.76 | 1.25 | 1.22 |
| $K_s^{(1)}$ (cm d ⁻¹) | 106 | 200 | 21 | 43 | 21 | 12 | 32 | 45 |
| pH _{KCl} ⁽²⁾ | 5.6 | 6.0 | 4.8 | 5.6 | 6.3 | 6.5 | 6.9 | 7.0 |
| TOC (%) | 0.69 | 0.04 | 1.13 | 0.17 | 0.93 | 0.11 | 1.68 | 1.80 |

Soil texture class (WRB, 2006), soil texture with sand (2.0 mm–0.06 mm), silt (0.06 mm–0.002 mm), and clay (<0.002 mm). Bulk density (ζ_d), saturated conductivity (K_s), soil pH, total organic carbon (TOC) in topsoil (0 cm–30 cm), and subsoil (31 cm–100 cm) in the lysimeters measured at the agricultural sites from which the lysimeter soils were taken.

¹)Stationary procedure.

²)Potassium chloride method.

potash (K+S Minerals and Agriculture GmbH, Kassel, Germany) was used as potassium fertilizer (40% K₂O, 6% MgO, 3% Na, 5% S).

Plant protection products in the form of herbicides, fungicides, insecticides, and growth regulators were not used during the lysimeter experiment. Turning tillage (with a spade) was carried out to a tillage depth of 25 cm on the arable land lysimeters. No tillage was carried out on the grassland lysimeters. The crop residues were tilled into the soil after harvest, and weeds were treated mechanically with a cultivator.

The grass was cut 4 times a year in the second half of May, the second half of June, the end of July, and August. All treatments within the experiment were harvested manually on the same day. Dry matter yield was determined from all harvested products (grass, grain, and field forage). After measuring the fresh weight per lysimeter, approximately 500 g was taken and dried in a drying cabinet for 16h at 60°C to determine the dry-matter yield.

2.3 Leachate sampling, water analyzes, and assessments

Depending on the climatic conditions, leachate occurred in the lysimeters, mainly from November to April. The leachate was continuously collected in the storage tanks (two polyethylene canisters, each with a volume of 30 dm³) in the lysimeter cellar. The amount of leachate was determined monthly by weighing the leachate storage tanks once a month. Monthly samples were taken, filtered through 0.45- μ m Millipore syringe filters, and analyzed in the UFZ facility laboratory for concentrations of NO₂-N, NO₃-N, and ammonium (NH₄-N), according to German Industrial Standards (DIN 38405–38406) by ion-exchange chromatography (IC, Thermo Fisher Scientific, Idstein, Germany). Atmospheric deposition was determined using a bulk precipitation sampler BUS 100 from Eigenbrodt

TABLE 2 Overview on the experimental lysimeter management practices and mineral fertilization (W. = winter).

| Land management | Trial variant | Number of lysimeters | Soil texture | Crop rotation | Mineral fertilization (kg ha ⁻¹) | | |
|-----------------|--------------------------|----------------------|---------------|-------------------------|--|------|-----|
| | | | | | N | P | K |
| Grassland | Fertilization level 50% | 4 | LS, S, L, Loe | Grass | 140 | 20 | 85 |
| Grassland | Fertilization level 100% | 4 | LS, S, L, Loe | Grass | 280 | 40 | 170 |
| Grassland | Fertilization level 150% | 4 | LS, S, L, Loe | Grass | 420 | 60 | 255 |
| Arable land | Fertilization Level 50% | 4 | LS, S, L, Loe | W. Wheat | 65 | 12,5 | 55 |
| | | | | W. Barley & catch crops | 85 | 12,5 | 65 |
| | | | | Oats & underseed | 45 | 12,5 | 50 |
| | | | | Clover mixture | 0 | 15 | 75 |
| Arable land | Fertilization Level 100% | 4 | LS, S, L, Loe | W. Wheat | 130 | 25 | 110 |
| | | | | W. Barley & catch crops | 170 | 25 | 130 |
| | | | | Oats & underseed | 90 | 25 | 100 |
| | | | | Clover mixture | 0 | 30 | 150 |
| Arable land | Fertilization Level 150% | 4 | LS, S, L, Loe | W. Wheat | 195 | 37,5 | 165 |
| | | | | W. Barley & catch crops | 255 | 37,5 | 195 |
| | | | | Oats & underseed | 135 | 37,5 | 150 |
| | | | | Clover mixture | 0 | 45 | 225 |

(Hamburg, Germany). As the measuring device did not have an active cooling system, the rainwater collected by this device was extracted after significant rain events and stored in a container in the laboratory building at a temperature of 8°C. Here, too, monthly samples were prepared and analyzed according to the chemical methodology described.

The primary data collected were secured, and all work and observations were fully documented. Uniform measurement methods were used to determine the relevant quantitative and qualitative parameters for all lysimeters included in the lysimeter test. In 1993, the analytical method for determining concentrations of nitrate, nitrite, and ammonium was changed from photometric to ion exchange chromatography. Extensive parallel measurements were successfully carried out to ensure the comparability of the results. The analyzers used were serviced and checked annually by the manufacturer's service staff. In addition, the quality of the analyses performed was assured by participating in round-robin tests with other laboratories.

We calculated monthly loads based on monthly N concentrations and the amount of seepage water, which were finally used to calculate average annual N concentrations and annual loads (seepage-weighted averages), respectively.

2.4 Statistics

Data assessment was performed using ORIGIN software (OriginLab Corporation, Northampton, MA, USA). The available data were checked for normal distribution. As this was not the case, the available time series were statistically compared using the Kruskal–Wallis test (one-way analysis of variance on ranks), a non-parametric method used to test whether lysimeter data originated from the same distribution.

The available measurement series were compared using median values.

Due to the experimental design, only one lysimeter vessel could be used for each fertilization treatment, soil type, and cultivation method. There is no real repetition here. The natural variability is thus reflected to a limited extent.

The Change Point Analyses (CPA) app for OriginPro detected single and multiple changes within the seepage time series. This app can identify changes in mean, variance, or both. The CPA app is based on the R software package *changepoint* (72).

Time series analyses of the N leaching data (grassland and arable land) for the 38-year long-term series were performed with OriginPro 2024 (OriginLab Corporation, Northampton, MA, USA) with the provided apps “simple time series analysis (v. 1.20)” and “advanced time series analysis (v. 1.00)” to proof data series of stationarity, existing trends (linear regression), autocorrelation [Autocorrelation Function (ACF), Partial Autocorrelation Function (PACF), and Auto Regressive Integrated Moving Average (ARIMA)] and to identify seasonal variations of long-term data series. Thus, time series analyses were used for retrospective analyses of long-term data to detect seasonal patterns and structure of the time series and to compare different time series with each other.

3 Results

3.1 Seepage

Figure 1 presents the medians of seepage for the different soil types and fertilization treatments. Significant differences in seepage between the fertilization treatments could be proved neither for arable land nor for grassland (Figure 1). But, as expected, the amount of seepage was highly significantly different between arable land and grassland ($p < 0.0001$, cf. Supplementary Table S1). Under the specific conditions of our lysimeter experiment, grassland showed a higher seepage rate with a median value of 158.6 mm year⁻¹ compared to arable land (92.2 mm year⁻¹, Supplementary Table S1). Among soil types, lysimeters filled with sand had significantly higher seepage than those filled with loamy sand, loam, and loess (Supplementary Table S1). The 50% fertilization treatment showed, in most cases, higher seepage rates than the 100% and 150% fertilization treatments, although the differences were not significant. However, there was a trend of decreasing seepage amounts due to increased fertilizer application visible. This relates to increased water consumption by plants with higher nutrient supplies.

In contrast, this relationship was not observed for the soil type loamy sand and sand used as arable land at a fertilization treatment of 50%. The seepage measured here decreased compared to the 100% and 150% fertilization treatment at the same use. The 2004 change in the irrigation regime (irrigating only for stand protection) did not change the seepage time series. As a result of a change point analysis, changes in the amount of seepage water occurred in the lysimeters of both grassland and arable land as a consequence of the years 2018, 2019, and 2020, which were characterized by low precipitation and high air temperatures during the growing season (cf. Supplementary Figures S2 and S3).

The quantities of seepage water measured varied considerably from year to year. Seepage fluxes were very closely linked to the climatic conditions, especially the precipitation regime. In wet years, leachate quantities of up to 470 mm were measured (1994, grassland use on sandy soil, fertilization level 100%). In contrast, seepage water fluxes came to a standstill following the dry year of 2018, except for the sand-filled lysimeters in grassland use. In the study period from May 1984 to April 2023, seepage formation was very closely correlated with the amount of precipitation. For grassland use, this relationship was direct, that is, in the result of a simple time series analysis using cross-correlations, the highest correlation coefficients r of 0.26–0.35 were calculated for a time lag of 0 or 6 months. In the case of arable land use, the highest correlation coefficients of 0.22–0.27 were calculated exclusively for the investigated soil types at a time lag of 6 months. The amount of leachate was, thus, significantly influenced by the precipitation 6 months earlier. High-annual seepage volumes corresponded to years with high rainfall and vice versa.

3.2 Nitrogen concentrations

The N concentrations measured during the study period varied considerably and were subject to significant annual fluctuations.

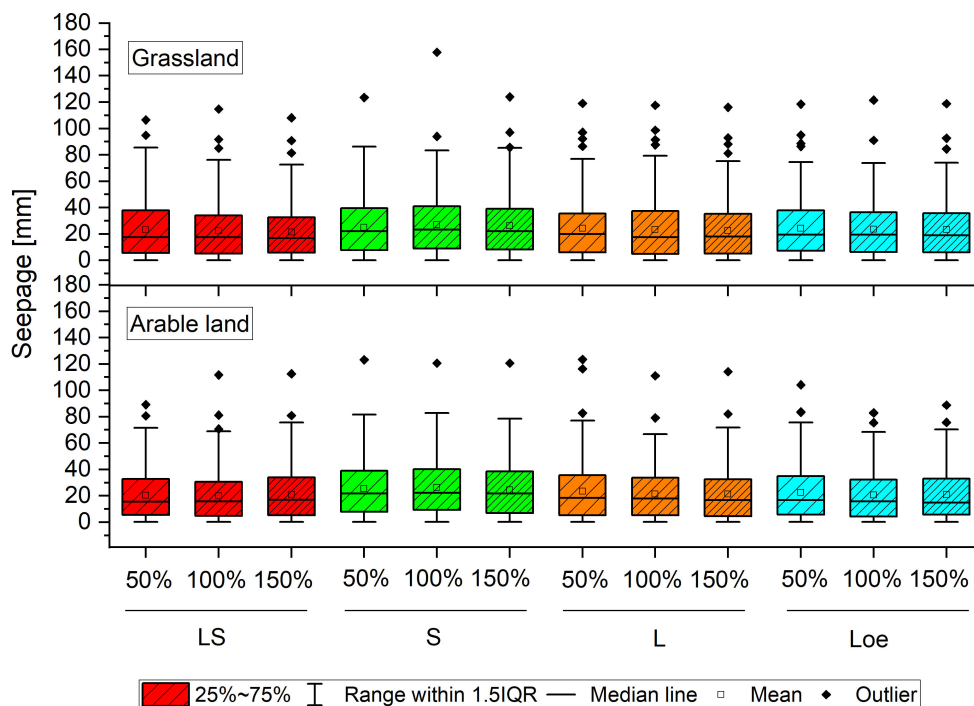


FIGURE 1

Boxplot of seepage rates measured in lysimeters filled with the soil types loamy sand, sand, loam, and loess under three fertilization treatments (50%, 100%, and 150%) for arable and grassland management (asterisk with bracket as significance mark) for the 38 years observation period 1985–2023.

The N concentrations showed a minimum of 0.002 mg L^{-1} (grassland use, soil type loam, and fertilization treatment 50%) and a maximum of 488.0 mg L^{-1} (grassland use, soil type loam, and fertilization treatment 150%). Further concentration peaks of a comparable magnitude occurred, especially with the 150% fertilization treatments, when leachate resumed after dry periods. Cross-correlation did not establish a relationship between the precipitation regime and the measured N-concentrations. The lowest median N concentration (sum of $\text{NO}_3^- \text{ N}$, $\text{NO}_2^- \text{ N}$, and $\text{NH}_4^+ \text{ N}$ concentration) of 1.04 mg L^{-1} was found in grassland use on the loam soil at the 50% fertilization treatment. In contrast, the highest median N concentration, 40.8 mg L^{-1} , occurred in the loess soil managed as grassland at the 150% fertilization treatment. The median N concentrations averaged over all fertilizer treatments showed significant differences between grassland (5.08 mgN L^{-1}) and arable land (21.6 mgN L^{-1}) (Supplementary Table S2).

In the case of grassland use, there were significant differences between the fertilization treatments for all soil types considered. The lowest median N concentrations of 1.4, 2.9, 1.0, and 5.4 mg L^{-1} were measured for the 50% fertilization treatment on soil types such as loamy sand, sand, loam, and loess, respectively. On the other hand, the fertilization treatment of 150% showed the highest median concentrations of 6.3, 16.2, 7.9, and 40.8 mg L^{-1} on soil types such as loamy sand, sand, loam, and loess, respectively. For all soil types investigated, the significantly higher N concentrations of the 150% fertilization treatment compared to the 50% and 100% fertilization treatments were particularly pronounced (Figure 2).

In the case of arable land, the median N concentrations of the investigated soil types showed a significant differentiation. The lowest median concentrations were found in the loam-filled lysimeter with 16.4 mgN L^{-1} at the 50% fertilization treatment. The highest median concentration of 36.5 mg L^{-1} occurred in the soil type loess at the 150% fertilization treatment. The lowest median N concentrations for the 50% fertilization treatments were observed in each case. As expected, the fertilization treatment of 150% showed the highest concentration on soil types such as loamy sand, loam, and loess, with 31.6, 22.5, and 36.5 mg L^{-1} , respectively. For the soil type sand, the highest N concentration was found in fertilization treatment 100% with 20.7 mg L^{-1} . No significant differences between fertilization treatments were evidenced in the soil types except for the 50% and 150% fertilization treatments in the loess soil (Figure 2).

The monthly median values of N concentration calculated as a result of the 38-year study period are presented in Table 3. After a soil passage of 1.25 m, the N concentrations of lysimeters used as grassland with a fertilization level of 50% for all soil types tested did not exceed the limit value currently valid in the European Union (11) for the nitrate concentration in groundwater of $50 \text{ mgNO}_3 \text{ L}^{-1}$ (approximately 11.3 mgN L^{-1}). This could also be proven for grassland use for the soil types such as loamy sand, sand, and loam in the fertilization treatment 100%. In contrast to the treatments with 50% and 100% fertilization, applying mineral fertilizer at 150% resulted in significantly increased N concentrations for grassland use.

In arable land use, monthly N concentrations were high for all soil types, and fertilization treatments studied and exceeded the limit values. Peak median values of up to 76.7 mg L⁻¹, mainly caused by NO₃⁻, occurred above all in arable land use in the fertilization of 150% in October to March within the leaching period. In contrast, lower N concentrations tended to occur from April to September during the vegetation period (Table 3).

3.3 Nitrogen loads

Annual N loads showed a high variation between the investigated study years, particularly for grassland use (Figure 3). Here, maximal values of up to 476 kg ha⁻¹ year⁻¹ were observed at the 150% fertilization treatment on the soil type sand in 2011, combined with high precipitation and seepage amounts. Minimal annual N loads of ≤ 0.01 kg N ha⁻¹ were observed for the soil type loamy sand for all fertilization treatments in the dry year 2019 and the loess soil in 2022 at the 100% fertilization treatment. For arable land, annual N loads differed between ≤ 0.01 kg N ha⁻¹ (sand soil and loamy soil in 2019 and 2012) and maximal 395 kg N ha⁻¹ (loamy sand at 150% fertilization treatment in 2011) (Figure 3). High-annual N loads corresponded to years with high precipitation and seepage amounts and vice versa.

Analyses of time series showed a markedly pronounced seasonal variation of N leaching for grassland and arable land, with the highest N leaching occurring from January to March and November to December (Supplementary Figures S4 and S5). In

contrast, there was a negative deviation from the long-term average of the long-term series from April to October.

Mean N leaching (median values of all years and averaged for all lysimeters with the same land use) showed significantly different levels after 38 study years for grassland and arable land, at 12.1 kg ha⁻¹ and 20.5 kg ha⁻¹, respectively (Supplementary Table S3).

For grassland, 2.5 kg ha⁻¹ was the lowest mean annual N leaching loss measured at the fertilization treatment of 50% on the soil type loam. In contrast, the highest yearly N leaching losses occurred in the amount of 68.1 kg ha⁻¹ on the soil type loess with grassland use and a fertilization treatment of 150%. The N leaching load in grassland use corresponded to the applied mineral N fertilization for the four different soil types studied. Accordingly, the lowest N leaching losses occurred at the 50% fertilization treatment. On the other hand, the fertilization treatment of 150% showed the highest N loads. The differences between the N-loads of the fertilization treatments, 50% and 150%, were highly significant for all soil types. There were also significant differences between the 100% and 150% fertilization treatment for soil type loess (Figure 3).

In the case of arable land use, the lowest median annual N leaching loss was measured at 12.1 kg ha⁻¹ with a fertilization treatment of 50% on the soil type loess. The fertilization treatment of 150% on sand soil type resulted in the highest N load of 36.6 kg ha⁻¹ with arable land use. Leaching corresponds to the application of mineral fertilization for soil types: loamy sand, sand, and loess. Accordingly, the lowest N leaching was recorded here for the 50% fertilization treatment and the highest for the 150% fertilization treatment. In the case of soil type loess, the fertilization treatment

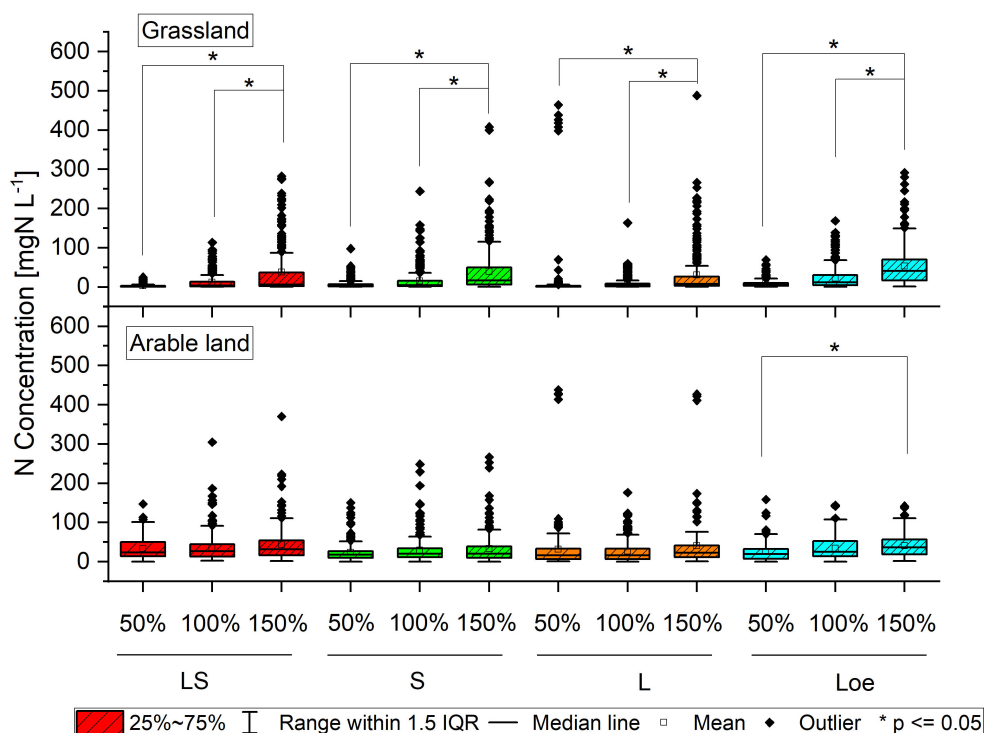


FIGURE 2

Boxplot of N concentrations measured in lysimeters filled with the soil types loamy sand, sand, loam, and loess under three fertilization treatments (50%, 100%, and 150%) for arable and grassland management (asterisk with bracket as significance mark) for the 38 years observation period 1985–2023.

TABLE 3 Median of monthly N concentrations (mgN L⁻¹) (period May 1984–April 2023).

| Soil type | Use | Fert. level | Month | | | | | | | | | | | |
|-----------|-----|-------------|-------|------|------|------|------|------|------|------|------|------|------|------|
| | | | May | Jun | Jul | Aug | Sept | Oct | Nov | Dec | Jan | Feb | Mar | Apr |
| LS | GL | 50% | 1.0 | 0.9 | 1.9 | 1.5 | 1.0 | 1.0 | 0.9 | 1.2 | 1.8 | 1.1 | 1.0 | 1.2 |
| | GL | 100% | 2.5 | 1.8 | 2.8 | 5.6 | 1.4 | 0.9 | 2.1 | 5.0 | 4.0 | 3.9 | 5.0 | 6.3 |
| | GL | 150% | 4.3 | 5.1 | 2.3 | 3.7 | 3.0 | 2.7 | 4.3 | 5.4 | 6.1 | 8.4 | 10.3 | 12.2 |
| | AL | 50% | 23.2 | 24.9 | 11.2 | 7.8 | 16.3 | 45.6 | 67.2 | 35.3 | 27.8 | 19.4 | 23.6 | 19.6 |
| | AL | 100% | 27.9 | 19.2 | 14.5 | 17.2 | 39.1 | 50.6 | 48.5 | 34.0 | 33.7 | 21.2 | 24.4 | 20.4 |
| | AL | 150% | 27.4 | 17.1 | 8.3 | 20.3 | 35.7 | 76.7 | 48.9 | 53.4 | 41.3 | 23.2 | 27.7 | 22.5 |
| S | GL | 50% | 2.4 | 1.8 | 1.9 | 2.1 | 1.4 | 2.0 | 3.9 | 3.5 | 5.4 | 5.0 | 3.5 | 2.8 |
| | GL | 100% | 2.2 | 1.9 | 7.1 | 3.2 | 2.4 | 2.5 | 8.1 | 7.3 | 9.5 | 6.8 | 4.9 | 3.9 |
| | GL | 150% | 6.0 | 6.0 | 33.6 | 12.5 | 10.5 | 20.4 | 18.6 | 22.5 | 24.8 | 23.7 | 14.5 | 10.2 |
| | AL | 50% | 17.5 | 4.7 | 9.1 | 16.5 | 13.5 | 17.3 | 19.9 | 19.6 | 21.5 | 18.2 | 18.3 | 15.7 |
| | AL | 100% | 23.9 | 9.5 | 9.4 | 24.7 | 27.7 | 40.0 | 21.4 | 25.1 | 21.4 | 21.5 | 15.4 | 14.2 |
| | AL | 150% | 18.7 | 15.2 | 9.6 | 39.4 | 35.1 | 20.4 | 21.0 | 25.1 | 32.5 | 20.6 | 13.9 | 12.8 |
| L | GL | 50% | 0.9 | 1.7 | 1.8 | 1.3 | 0.6 | 1.0 | 1.0 | 1.0 | 1.3 | 1.0 | 1.0 | 1.3 |
| | GL | 100% | 3.2 | 1.9 | 3.1 | 2.9 | 2.4 | 2.7 | 1.7 | 3.8 | 3.9 | 5.4 | 4.1 | 3.9 |
| | GL | 150% | 4.7 | 3.8 | 4.8 | 5.5 | 5.0 | 8.2 | 8.1 | 5.8 | 7.3 | 13.2 | 9.1 | 13.9 |
| | AL | 50% | 33.4 | 17.1 | 6.6 | 3.6 | 16.3 | 29.1 | 20.9 | 18.0 | 12.9 | 15.7 | 14.8 | 11.1 |
| | AL | 100% | 24.1 | 2.0 | 4.7 | 3.1 | 12.9 | 23.3 | 34.2 | 13.9 | 14.8 | 16.9 | 15.6 | 15.9 |
| | AL | 150% | 22.4 | – | 12.5 | 10.9 | 24.1 | 35.3 | 38.8 | 25.1 | 28.0 | 18.1 | 18.4 | 21.9 |
| Loe | GL | 50% | 4.8 | 3.6 | 3.4 | 3.6 | 5.4 | 5.0 | 4.8 | 5.1 | 7.0 | 6.7 | 6.6 | 6.1 |
| | GL | 100% | 8.2 | 3.6 | 9.6 | 7.7 | 7.7 | 9.2 | 12.0 | 11.1 | 14.8 | 18.1 | 13.1 | 15.1 |
| | GL | 150% | 44.6 | 37.6 | 35.1 | 24.0 | 44.1 | 31.2 | 36.7 | 40.0 | 46.7 | 42.6 | 39.2 | 46.7 |
| | AL | 50% | 21.8 | 13.8 | 2.9 | 22.7 | 53.8 | 36.5 | 26.6 | 23.0 | 17.5 | 18.5 | 13.2 | 10.7 |
| | AL | 100% | 24.0 | 17.3 | 14.6 | 33.4 | 64.1 | 54.3 | 52.6 | 27.1 | 31.2 | 26.8 | 25.8 | 19.6 |
| | AL | 150% | 41.6 | – | – | – | 26.9 | 9.4 | 63.4 | 47.9 | 34.1 | 38.9 | 38.2 | 31.8 |

of 50% showed the lowest N-load with 12.1 kg ha⁻¹. In the case of arable land use, a significant differentiation of the N-load between the three investigated fertilization treatments comparable to grassland use could not be proven for the studied soil types (Figure 3).

3.4 Dry matter yield

In the result of the Kruskal–Wallace tests of the dry matter (DM) yields (sum of main crop and catch crop), significant effects of fertilization treatment and land use as grassland or arable land could be proven. In particular, the median values of the DM yields for the soil types loam differed significantly from sand. Based on the 38-year study results, a significant influence of the mineral fertilization treatment could be demonstrated for both types of use, grassland and arable land, with the highest yields in the 150% fertilizer treatments (Supplementary Table S4).

For grassland, there were some highly significant differences between fertilizer treatments on all four soil types, with the lowest

DM yield for each of the lysimeter at 50% fertilizer treatment and the highest DM yield at 150% fertilizer treatment. Thus, the differentiation of mean DM yields among the fertilizer levels studied aligned with expectations.

Regarding arable land use, the DM yields significantly differed between the mineral fertilization levels 50% and 150% for the soil types: sand, sandy loam, loam, and loess. Significant differences between the 100% and 150% fertilization treatments were also found for the Loe soil type (Figure 4).

3.5 Nitrogen uptake

Long-term mean N uptake values significantly differed concerning mineral fertilizer treatment and land use. In contrast, soil type had no significant effect on N uptake (Supplementary Table S5 and Figure 5). The highest N uptakes were observed in each of the lysimeters with the highest fertilizer treatment of 150%. The N uptake was lowest in the 50% fertilization treatment

(Supplementary Table S5). For grassland use, long-term mean N uptake values significantly differed among the three fertilization treatments studied, showing a trend similar to DM yields. For arable land use, N uptake showed a comparable tendency. However, significant differences between the 50% and 150% fertilization treatments were only found for the sand soil type (Figure 5).

3.6 N-budget

For N balancing, the mean atmospheric (wet and dry) deposition over the experimental period of 8.3 kg ha⁻¹ was considered. The crop rotation of the arable land lysimeters included oats with a clover-grass undersow and, in the following year, clover-grass as the main crop. Own investigations on leguminous N-fixation were not available. The literature describes an annual legume N-fixation of about 400 kg ha⁻¹ (73). In the collection of fertilizer law guidelines of the German federal state of Saxony-Anhalt, a symbiotic N fixation of 230 kg ha⁻¹ was set as binding for the cultivation of clover grass (74). This amount of nitrogen had to be considered in the N balance. Clover grass was cultivated in a rotation of 4 years. Thus, an annual balance input of 57.5 kg N ha⁻¹ was included in the yearly balancing. The balance of N inputs (fertilization, legume N-Fixation, and atmospheric input) and N outputs (plant uptake and leaching) showed a negative N

balance in all soil types in the 50% fertilization treatment at grassland use. In contrast, the 100% and 150% fertilization treatments showed balance surpluses of up to 113.1 kg ha⁻¹ for all soil types (fertilization treatment 100% for the loam soil type). Thus, balance surpluses of 21.4 kg ha⁻¹ and 64.4 kg ha⁻¹ for soil type loamy sand were determined for the 100% and 150% fertilization treatments, respectively. For sandy soil, N balance surpluses of 20.5 and 49.4 kg ha⁻¹ and 113.1 and 71.0 kg ha⁻¹ for the loamy soil were calculated. For soil type loess, the balance surplus of the fertilization treatment 150% was 26.6 kg ha⁻¹.

For arable land use, the N budget was negative for the fertilization treatments 50% and 100% for all soil types, with the lowest values occurring in the 50% fertilization treatments with values of -41.1 and -37.1 kg ha⁻¹ for the loamy sand and loam soil types. In contrast, the highest budgets of 8.2 and 14.4 kg ha⁻¹ were calculated for the soil type's sand and loam with the fertilization treatment of 150% (Supplementary Table S6 and Figure 6).

4 Discussion

4.1 Seepage

The observed variation in seepage can be attributed to the different soil physical properties of the lysimeter soil types used

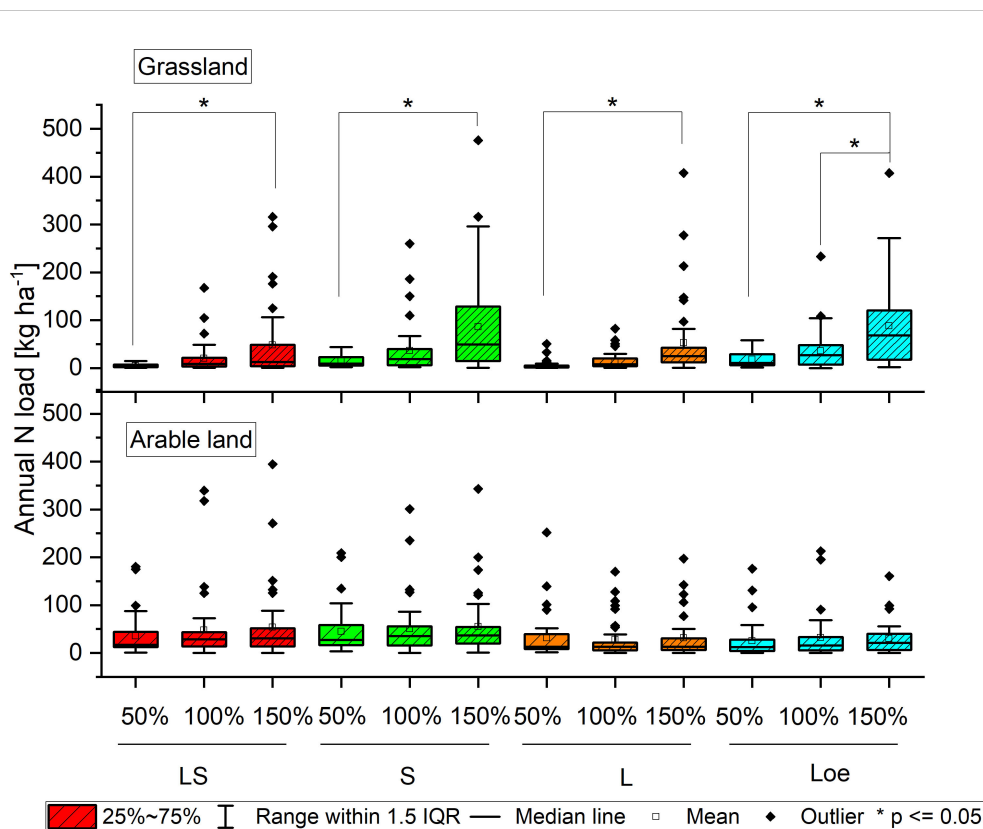
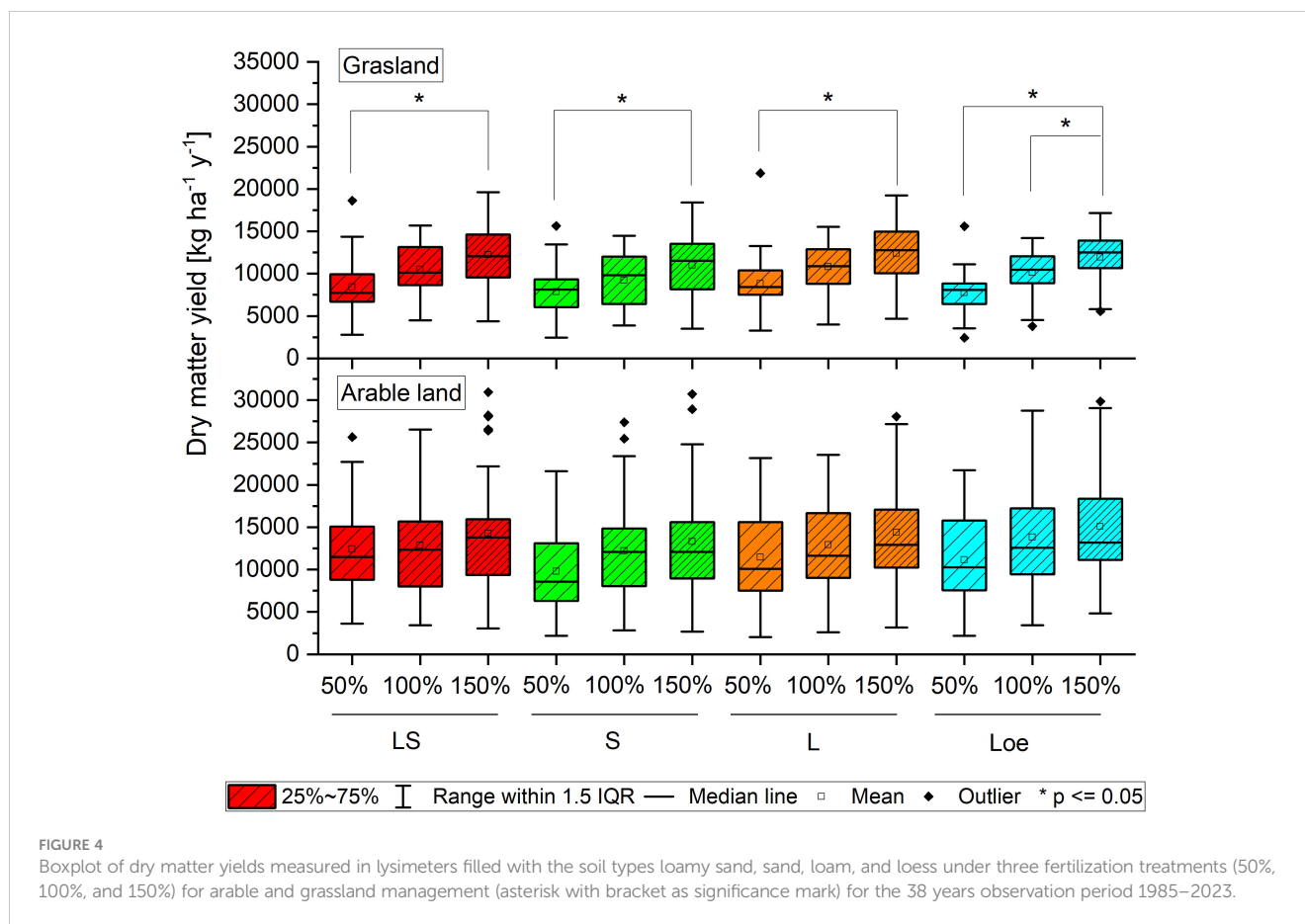


FIGURE 3
Boxplot of N loads measured in lysimeters filled with the soil types loamy sand, sand, loam, and loess under three fertilization treatments (50%, 100%, and 150%) for arable and grassland management (asterisk with bracket as significance mark) for the 38 years observation period 1985–2023.



(75), and to the improved water utilization of fertilized plant stands according to demand (76). The increased seepage under grassland use (increased by approximately 66.4 mm) is seemingly in contrast to other studies (77, 78). On the one hand, it may result from the high supplemental water supply (until 2004), typical for intensively used grass stands under the former management conditions. On the other hand, higher seepage could result from the lack of soil tillage at the grassland management, which is known to favor the formation of macropores and thus might cause preferential flow with higher seepage amounts (48, 50, 51). In addition, a reduction in infiltration was seen as a result of the cultivation of catch crops, which was integrated into the crop rotation of the lysimeters cultivated as arable land. When catch crops are grown, leachate significantly reduces (79–81).

4.2 N leaching and N concentrations of seepage

Despite increased seepage, the N leaching determined for grassland use was significantly lower than for arable land use except for the 150% fertilization treatment. These results align with other studies reporting a high N efficiency and, consequently, very low NO_3^- leaching of grasslands managed by cutting at moderate N fertilization (44, 82). It is known from the literature that soils under permanent grassland contain more

organic C and organic N than arable soils with solid coupling between the C and N cycles (43, 44, 83, 84). Therefore, converting arable land to grassland was assumed to be a solution for improving soil organic C contents and microbial C and N biomass and associated enzyme activities of degraded arable land (84). Otherwise, there is vast evidence that NO_3^- leaching from grasslands also can be significant, in particular, at intensive grassland management with high N fertilization rates and combined with the more frequent occurrence of drought events due to climate change, which might increase NO_3^- leaching risk (85, 86). Apostolakis et al. (87) found that grassland management intensity was positively associated with NO_3^- leaching risk. In contrast, the intensification effect might be directly by changing nutrient cycling or indirectly by changing plant communities and altering interactions between soils, plants, and soil microorganisms (88, 89). In particular, N fertilization is known to increase plant biomass productivity and nutrient concentrations of plants, resulting in a reduction of the C:N ratio of grassland soils with faster mineralization as well as a reduction of plant diversity (90–92). Other studies showed that high N-surplus fertilizer rates could increase NO_3^- leaching from grassland soils (46, 47). We also observed a relatively high N leaching of all soil types with grassland use in the fertilization treatment of 150%, which was increased by more than 100% compared to the reference fertilization treatment (100%). In the grassland lysimeters with a fertilizer treatment of 150%, the N loads with median values of

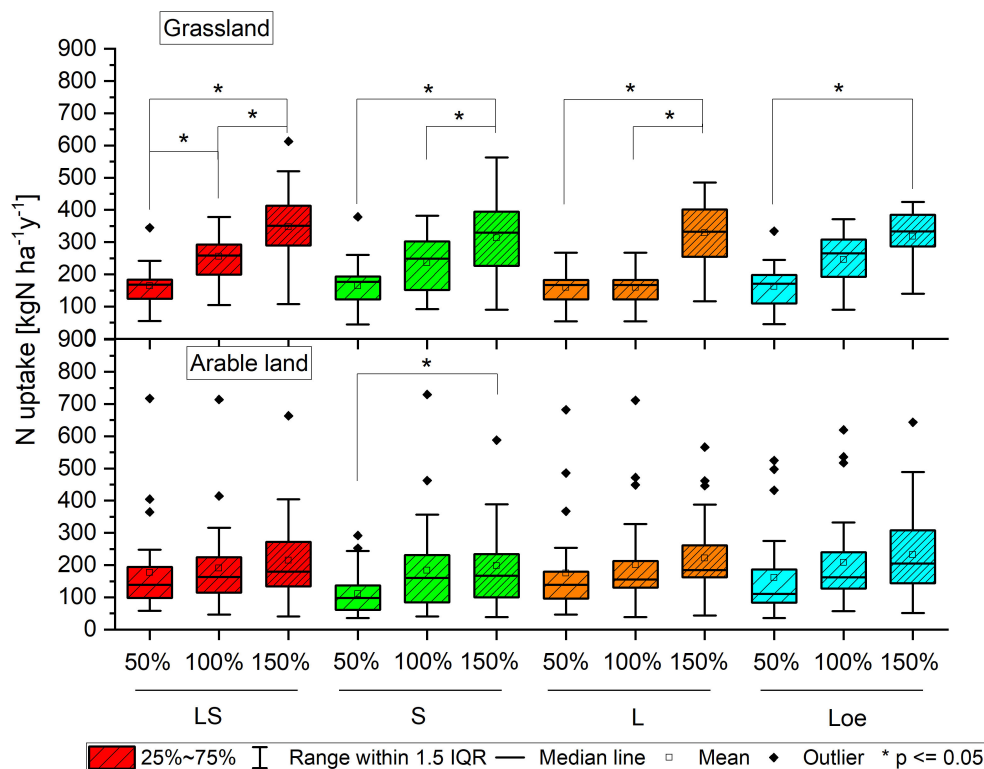


FIGURE 5

Boxplot of N uptake measured in lysimeters filled with the soil types loamy sand, sand, loam, and loess under three fertilization treatments (50%, 100%, and 150%) for arable and grassland management (asterisk with bracket as significance mark) for the 38 years observation period 1985–2023.

12.9 kg ha⁻¹ (loamy sand), 49.4 kg ha⁻¹ (sand), 25.0 kg ha⁻¹ (loam), and 68.1 kg ha⁻¹ (loess) were higher than in the arable lysimeters with 30.7 kg ha⁻¹ (loamy sand), 36.6 kg ha⁻¹ (sand), 13.0 kg N ha⁻¹ (loam), and 21.0 kg ha⁻¹ (loess), except for the loamy sand soil type. However, differences were only significant for the loamy soil. N loads of grassland lysimeters at the 150% fertilizer treatment were higher than those reported in the literature (44, 87, 93) and presumably caused by the intensive management (fertilization and irrigation).

The determined median annual N leaching showed a level of 2.5 to 68.1 kg ha⁻¹ depending on the soil type, fertilization treatment, and land use corresponded to the level described in the literature (94–96). The lowest leaching losses were measured with 50% fertilization in grassland use. Compared to the 100% fertilizer treatment, the N loads were only 35.8% (loamy sand), 50.0% (sand), 30.6% (loam), and 39.2% (loess) of the loads measured with the 100% fertilization treatment. When averaging over all investigated soil types and fertilization treatments, the grassland or arable land use resulted in almost identical annual N leaching of 20.2 kg ha⁻¹ (grassland) and 21.9 kg ha⁻¹ (arable land). These minor differences in the mean N loads between both management systems were only caused by the high N leaching at the 150% fertilization treatment in the grassland lysimeters (Supplementary Table S3). In the other fertilizer treatments (50%, 100%), N leaching of grassland use was lower than arable land use except for the 100% treatment of loess soil. These results align

with previous studies reporting reduced NO₃⁻ leaching of grassland soils (44, 82, 83).

The determined N leaching is consistent with the level of N leaching under arable land use published in the literature using lysimeter and field experiments (95, 97–102). The annual N leaching found in this experiment for the fertilization treatments 100% and 150% on soil types loamy sand and sand in arable land was slightly below the level of >50 kg N ha⁻¹ given by Asmus (103) for sandy soils in the state of Brandenburg, Germany. Unexpectedly, increased N fertilization rates did not significantly increase N leaching in all soil types of arable land lysimeters as observed for grassland use (see Figure 3). Although the trend of decreasing N loads with a 50% reduction of N fertilization and increasing N leaching with increased N fertilization rates (150%) was observed for almost all soil types, no significant differences between the fertilization treatments could be proven. For grassland use, as expected, an increase of N leaching with increased N fertilization was observed, but the arable land lysimeters showed no significant reaction to increased N fertilization. For example, the loam soil had an equal level of N leaching at the fertilization treatments, 50% and 150% (13 kg ha⁻¹). In contrast, the 50% and 100% fertilization levels showed slightly increased N leaching compared to the 150% treatment. These results contrast previous studies demonstrating a close relationship between amounts of N fertilizer and N leaching losses (104, 105). Earlier studies estimated that 19%–24% of the N applied was leached (104, 106). For our

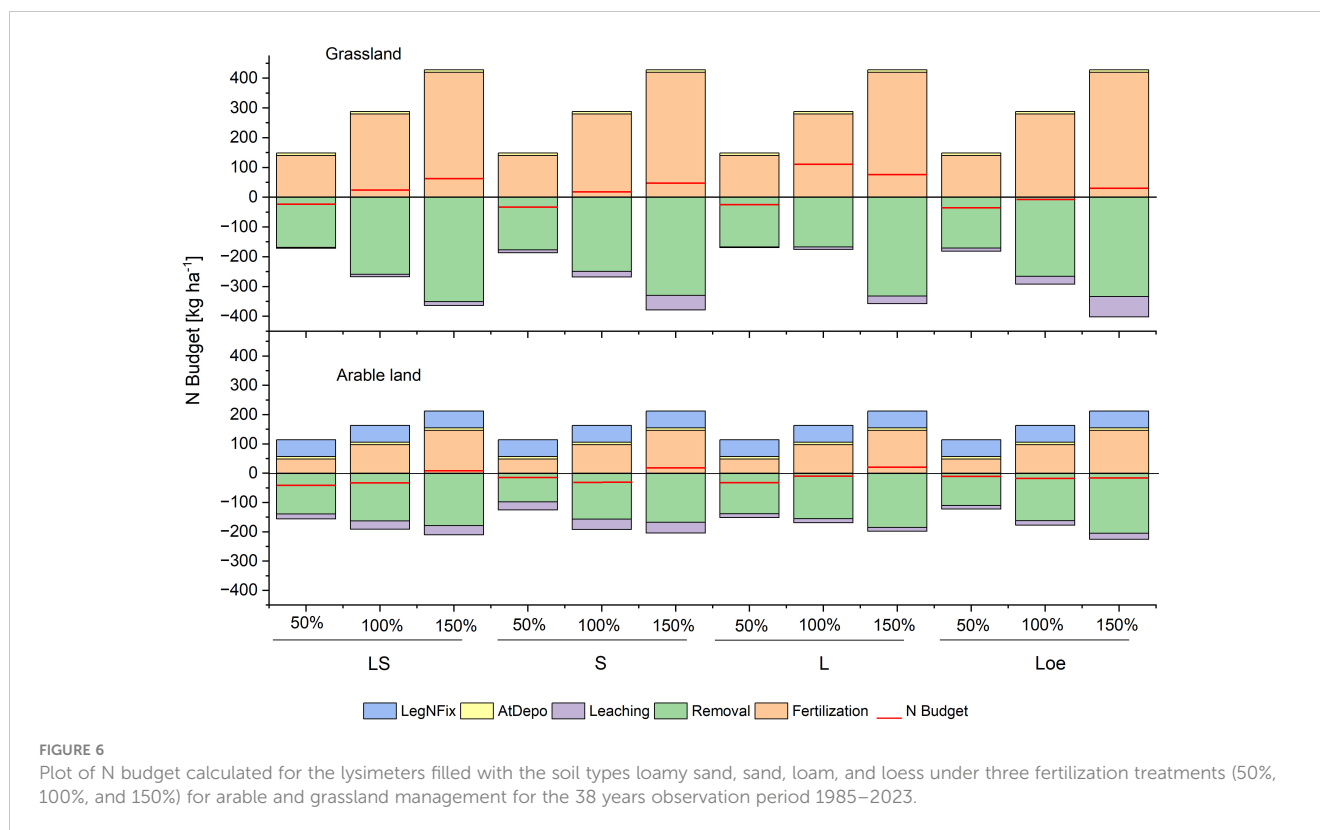


FIGURE 6

Plot of N budget calculated for the lysimeters filled with the soil types loamy sand, sand, loam, and loess under three fertilization treatments (50%, 100%, and 150%) for arable and grassland management for the 38 years observation period 1985–2023.

study, it can be assumed that soil- and management-related factors (soil type, texture, soil tillage, crop rotation, and others) mask the effect of increased N fertilization rates, as also shown by Dieser et al. (41). Different studies reported that the prehistory of cropping (carry-over effect) may affect soil N mineralization and, hence, NO_3^- leaching (21). The largest share of the excess fertilizer N remained in the soil in the organic N pool, and soil microbial biomass built up pools of degradable soil organic N that may subsequently mineralize (31, 32). The soil organic C management is crucial for maximizing the long-term benefit of crop fertilizer applications and minimizing NO_3^- leaching (35). Thus, management-related factors that favor mineralization processes (soil tillage, N-rich crop residues) may considerably affect NO_3^- leaching. In this context, the impact of soil tillage and crop residues on mineralization processes and, consequently, N losses might cover the effects of increased N fertilization in the arable lysimeters. These management-related factors are not as pronounced at grassland use.

We observed differences in N leaching between studied soil types at arable land use, with the highest N loads from the sand and loamy sand soils and significantly lower N loads from the loam and loess soils. Dieser et al. (41) also reported that soil type and texture are among the decisive factors for NO_3^- leaching potential, in addition to the main crop and precipitation in October. Above this, the management strategy in the arable land lysimeters with summer catch crops was very N efficiently, as shown by the high N uptake of excess fertilizer N and consequently low N balances (see Figure 6). It can be assumed that, due to the integration of summer catch crops in the cereal-fodder crop rotation, N surpluses especially in the

150% treatment were absorbed by the catch crops and converted into biomass. This could be one reason for the small and non-significant differences between the different fertilization treatments in arable lysimeters. Numerous studies identified the cultivation of catch crops as the most effective measure for reducing N leaching (100, 101, 107).

Nevertheless, the results presented here indicate an effect of a 50% reduction in mineral fertilization, which resulted in significantly reduced N leaching for grassland use. For arable land use, the trend of reduced N-leaching with a reduced N-fertilization of 50% (excluding soil type loam) was observed, with no significant differences between fertilization treatments. This statement contradicts the results of our first evaluation after 7 years of study (62, 108). Here, the lowest mean annual NO_3^- leaching of 6 kg N ha⁻¹ was found on the loamy sand soil with grassland use and the highest of 24 kg N ha⁻¹ on sand with arable land use. It was concluded that the fertilization had a noticeable influence on only seepage and DM yield. The more intensive vegetation growth due to higher mineral fertilization treatment also increased water demand. This became apparent from the significant reduction observed in seepage with increasing fertilization and the increased dry matter DM yield. However, the situation was reversed if the plants were sub-optimally fertilized: the plants were not able to convert sub-optimally applied fertilizers into biomass. Consequently, the leaching rates were increased because of enlarged seepage water volume. Therefore, the expected reduction in N loads with a 50% reduction in mineral fertilization could not be statistically proven in the studied application spectrum.

In the case of grassland use, the limit value of $50 \text{ mgNO}_3 \text{ L}^{-1}$ (11) could be achieved in the fertilization treatment of 50% and 100% based on the long-term monthly median values of the N concentration for the soil types: loamy sand, sand, and loam. In contrast, the limit value was frequently exceeded in the 150% fertilization treatment of all soil types and the 100% treatment of the loess soil. The NO_3^- N concentrations determined in the lysimeter study correspond to the range of values that is also known from other studies at a comparable level of mineral fertilization and comparable seepage amounts from lysimeter studies (15–17, 19). A continuous exceedance of the EU drinking water limit was also found by Biernat et al. (109) by comparing organic and conventional land management systems. In contrast, even the low N intensity management systems led to increased NO_3^- concentrations in seepage water. Our lysimeter study showed that NO_3^- leaching at arable land use was affected by the amount of applied N fertilizer and soil mineralization, including all factors controlling mineralization processes (soil type, texture, crop rotation, soil tillage, and others). These results align with other studies that showed only a tiny contribution of applied fertilizer to N leaching (110). Accordingly, Kühling et al. (111) reported that significant N leaching from soil mineralization was also observed without N fertilization.

It was shown in our study that, even at a drastic reduction of N fertilization level by 50%, which exceeded the required reduction in the German Fertilizer Ordinance (112) of 20% markedly, no significant effect on NO_3^- concentration in seepage and N leaching amounts could be achieved for arable land use. Comparable results with a tiny contribution of a reduction in N fertilization levels below the level stated by the Fertilizer Ordinance to reducing N leaching was also documented by Kage et al. (113) for winter wheat and winter oilseed rape.

Our lysimeter trial showed that a reduction of N fertilizer as the only measure was insufficient to reduce NO_3^- leaching for arable land use. Thus, other strategies in addition to a reduction of N input are required. Several studies show that managing soil organic matter, including management-related factors controlling mineralization processes, could be critical in reducing NO_3^- leaching on arable land (35, 114). In this context, management measures such as the reduction of the intensity of soil tillage, crop rotations with the reduction of N-intensive crops (e.g., winter wheat), and reduction of crops leaving high soil mineral N in autumn (e.g., rape, maize) could be effective measures to reduce the risk of NO_3^- leaching (41). Above this, mineral and organic N fertilization affect the decomposability of the organic N pool and control the N release from mineralization processes. In this context, C:N ratio of organic fertilizers was assumed as a reliable predictor for native soil N mineralization (115–117). Masunga et al. (117) reported that organic fertilizers with lower C:N ratios stimulate more significant N mineralization and microbial biomass. In contrast, the meta-analysis of Liu et al. (118) suggested only a minor influence of C:N ratios of organic fertilizers on the direction of native N soil mineralization. Nevertheless, it is known from different authors that the application of organic fertilizers with high

C:N ratios might significantly increase soil organic C contents and, thus, contribute to the effective retention of excess fertilizer-derived N in the organic N pool in the long term (119, 120). In general, the avoidance of N inputs beyond the economic optimum, a high-use efficiency of N fertilization with a better synchronization of N inputs and crop demands, N-efficient application technologies, and N stabilization of fertilizers by the use of nitrification inhibitors are essential to sustain high crop productivity with low N losses (26, 31).

Because the fertilizer-derived N retained in the organic pool can be mineralized during the whole year, it is critical for reducing N losses that the soil is also covered with plants in autumn (and winter) and vegetation-free fallow should be avoided in general (31). Furthermore, catch crops are known to be an effective measure of accumulating excess N and might contribute significantly to reducing the risk of NO_3^- leaching (31, 100, 121, 122). Nevertheless, it must be considered that the mitigation potential of catch crops depends on a successful stand establishment, which underlies a sizeable yearly variation (123). In this context, the optimal sowing date is decisive for sufficient biomass development for efficient uptake of excess N before the beginning of the leaching period (81). It was assumed that the integration of legumes in the crop rotation of the arable lysimeters in our study also might impact NO_3^- leaching by supplying N-rich crop residues favoring mineralization processes in times without plant demand. It is known from other studies that grass-clover leys might have a high risk of NO_3^- leaching due to a high N supply following mineralization of the crop residues during a low N demand in the subsequent crop in autumn (124–126).

4.3 Dry matter yields, N uptake, and N balances

For grassland use, DM yields have differed significantly between the fertilization treatments, with the lowest yields in the 50% fertilization treatment and the highest in the 150% fertilization treatment. A reduction of N fertilization by 50% resulted in a decrease in yields of maximal 23.6% in the loamy sand (see [Supplementary Table S4](#)). In contrast, the sand showed a minor reduction of yields of 17.0% in the 50% fertilization treatment compared to the reference level (100%). An increase of N fertilization by 50% (150% fertilization treatment) increased the DM yields by 17.4% (sand and loam) to 19.5% (loess) compared to the reference (100%). Thus, compared to the steep increase of N leaching losses in the 150% fertilization treatment, the N fertilization above the reference treatment only showed little effect on DM yields in the grassland lysimeters.

The arable lysimeters showed no significant differences between the fertilization treatments for the loamy sand, sand, loam, and loess soil (see [Figure 4](#)). The reduction of N fertilization by 50% in the lysimeters used as arable land did not lead to a significant decrease in DM yields. For the sand soil, reduced N fertilization (50% treatment) reduced DM yields by 29.0%, whereas no significant

decrease in N leaching could be proved. An increase of N fertilization by 50% compared to the reference level (100% treatment) increased DM yields by 11.6%, leading to a significant decrease in N leaching. These results are in contrast to other studies reporting a close relationship between N leaching losses and N fertilization rates (5, 105, 106). Kühling et al. (111) observed that, for a karst aquifer with loamy sand in northwestern Germany, high precipitation amounts decreased N leaching losses by 10%–20%, reduced N fertilization rates, and no significant reductions in crop yields.

Several studies described the relationship between N leaching and applied N fertilizer rates by linear, exponential, or quadratic models (5, 22, 104–106, 127). Some authors reported considerable yield reductions and only small reductions of N leaching below the economic optimal N rate, whereas leaching losses increased exponentially above the economic optimum (82, 128, 129). The economic optimal N rate is calculated based on N incrementality testing (113). In our lysimeter study, an economic optimal N rate was not determined to define levels of different fertilization treatments. However, it can be assumed that the economic optimal N rate was not exceeded excessively in our study for arable land lysimeters because the N balances of the 150% fertilizer treatment were relatively low with 2.1 kg ha⁻¹, 8.3 kg ha⁻¹, 14.4 kg ha⁻¹, and -13.6 kg ha⁻¹ for the soil types loamy sand, sand, loam, and loess, respectively (see Figure 6), indicating a high N efficiency by uptake of excessive fertilizer N by main crops and catch crops. Overall, it must be noted that the calculation of the N budgets was fraught with uncertainties. On the one hand, the symbiotic N fixation for arable land use was estimated at 230 kg ha⁻¹ based on the guideline values for the cultivation of clover grass valid in the follow-up documents of the German Fertilizer Ordinance (74). Another reference gives this as 400 kg ha⁻¹ under comparable conditions (73). On the other hand, an atmospheric N deposition of 10 kg ha⁻¹ to 41 kg ha⁻¹ was determined in container tests using the ¹⁵N dilution method (130–132).

In contrast to the arable lysimeters, intensive grassland use with irrigation and high N fertilization (150%) can be assumed to have a significantly lower N efficiency compared to arable land use in this study, which was made clear by the high N balance of up to 113.1 kg ha⁻¹ in the grassland lysimeters (see Figure 6). This could be the reason for the substantial increase in N leaching at the 150% fertilizer treatment in the lysimeters managed as grassland, which is consistent with the strong N leaching at N application above the economically optimal rate described in the literature (82, 128, 129).

5 Conclusions

The present long-term study showed that increased N fertilization rates significantly enhanced N leaching losses and NO₃⁻ concentrations in seepage for grassland use. Thus, a reduction of N fertilization levels by 50% was appropriate to reduce NO₃⁻ leaching and to comply with the current limit value

for NO₃⁻ in groundwater of 50 mg NO₃⁻ L⁻¹ (11.3 mg N L⁻¹) at simultaneously reduced yields by maximal 29%. In contrast, no significant effects of reduced N fertilization levels on N leaching were proved for arable land use, even at a reduction of N fertilization by 50%. This is because excess nitrogen from fertilizers is retained in the organic nitrogen pool and later released through mineralization processes. For agriculture, this suggests that reducing N fertilization alone is not sufficient to reduce NO₃⁻ leaching and comply with the European limit value for NO₃⁻. Rather, the management of organic carbon in the soil appears to be decisive for N losses. Consequently, for arable land use, in addition to avoiding N fertilization levels beyond the economic N optimum, management measures should be targeted to reduce N losses of mineralization processes in times with small plant demand. Other efficient measures to reduce N leaching are the N conservation of excess N in the biomass of catch crops and the synchronization of crop rotations without vegetation-free fallows in N leaching sensible times (autumn, winter). In addition, appropriate management measures to reduce N leaching can be proposed, such as reduced tillage, increasing the N efficiency of fertilization (technical and chemical solutions), crop rotations avoiding crops known to leave high mineral N levels in autumn, such as crops with high N requirements, crops with a need for intensive tillage during harvest, and crops with a high proportion of N-rich crop residues after harvest. Furthermore, we assume that management measures contributing to an increase in the soil's organic carbon (SOC) content are essential to keeping the excess nitrogen in the SOC pool long term. In this context, the integration of SOC-increasing crops (e.g., legumes), organic fertilization (manure with higher C:N ratio), and retention of agricultural by-products (straw, sugar beet leaves) on the field could reduce N leaching.

In addition to taking into account the specific site conditions, the design of the crop rotation must be an integral part of strategies to minimize N losses. The reduction of mineral N fertilizer use should be combined with crop cultivation and crop rotations adapted to the specific site conditions. Within the present lysimeter study scope, it was impossible to investigate the processes relevant to the dynamics of N compounds in the soil. The contribution of essential processes, such as symbiotic N fixation by legumes, to the N budget, could only be estimated and should be investigated in further studies. In addition, the effects of the quantity and quality of SOC on the contribution of mineralization processes on the one hand and the retention of excess N on the other hand should be investigated in more detail. Therefore, process studies are essential for a comprehensive understanding of the complex soil processes that can lead to the retention and release of N compounds.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material. Further inquiries can be directed to the corresponding author.

Author contributions

HR: Conceptualization, Data curation, Software, Writing – original draft, Writing – review & editing. NT: Conceptualization, Writing – original draft, Writing – review & editing. RM: Supervision, Writing – review & editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fsoil.2024.1345073/full#supplementary-material>

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