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N_2O emission and nitrogen and carbon leaching from the soil in relation to long-term and current mineral and organic fertilization – a laboratory study

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ABSTRACT

Sosulski T., Szara E., Szymańska M., Stępień W. (2017): N_2O emission and nitrogen and carbon leaching from the soil in relation to long-term and current mineral and organic fertilization – a laboratory study. Plant Soil Environ., 63: 97–104.

The paper presents the results of a laboratory experiment aimed at the assessment of N_2O emissions, NO_3^-, NH_4^+ and carbon (C) leaching from agricultural soils subjected to long-term mineral and organic fertilization. Our results show that long-term treatment impacts the N_2O emissions from loamy-sand Luvisols to a greater extent than the recent single application of mineral or organic fertilizers. The N_2O fluxes from soils with higher C_{org} content that results from long-term organic fertilization exceed those from soils with lower C_{org} content subsequent to long-term mineral fertilization. Our research confirms previous reports that the intensity of N_2O emission is related to soil moisture. The NO_3^- leaching depended on the recent application of fertilizers with a stronger influence of single application of NH_4NO_3 than farmyard manure. Long-term fertilization did not impact the NO_3^- leaching.

Keywords: short-term fertilization; gas emission; macronutrient, denitrification, nitrification, nitrate

Among multifactorial impacts, the soil content of nitrogen (N)-mineral, organic carbon (C) and oxygen with the pH and temperature of the soil act as the proximal regulators of the N_2O production in the soil (de Klein et al. 2001). The content of organic C, total N and mineral N forms in the soil are determined by the long-term and current/recent fertilization (Sosulski et al. 2015, Stehlíková et al. 2016). Our earlier research on long-term experiments in Central Poland showed no differences in N_2O emissions from manured and mineral-treated soils (Sosulski et al. 2014). Based on the earlier results it was concluded that the ability to distinguish between the effects of long-term fertilization and those of current application of fertilizers as part of long-term fertilization was insufficient.

Denitrification is one of the most effective ways of removing NO_3^- from the groundwater (Greenan et al. 2006). The C:NO_3^--N ratio in the leachates can serve as an indicator of the potential capacity of the waters to remove excess NO_3^- through denitrification (Brye et al. 2001). An analysis of the content of dissolved organic carbon and nitrates in the leachates should help understand the question of the water pollution with nitrates in Poland (Andersen et al. 2016).

The aim of this laboratory study was to examine the long- and short-term effects of mineral and organic fertilization on the direct N_2O soil emissions alongside the N and C leaching from the soil.

MATERIAL AND METHODS

This study was conducted at the Warsaw University of Life Sciences. The soil was collected up to 1 m depth in an natural horizons arrange-

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ment from three of five replications of long-term exclusive mineral (Hist-CaNPK) and organic (Hist-FYM (farmyard manure)) treatments forming part of a long-term fertilization experiment with rye-monoculture carried out since 1923 in Skierniewice. In the Hist-CaNPK treatment mineral fertilizers were applied annually at the following annual rates: 90 kg N/ha (ammonium nitrate); 26 kg P/ha (triple superphosphate); 91 kg K/ha (potassium chloride 50%). In the Hist-FYM treatment manure was applied annually at the dose of 20 t/ha. Lime was applied to both investigated treatments every four years at the dose of 1.43 t Ca/ha. The soil is Luvisols (FAO 2006) of the type of loamy sand with the following fractions in the 0–25 cm layer (Ap horizon): 7% < 0.002 mm; 5% 0.002 to 0.05 mm; 87% > 0.05 mm, in the 26–50 cm layer (Eet horizon): 5% < 0.002 mm; 5% 0.002 to 0.05 mm; 90% > 0.05 mm, below 50 cm of depth (Bt/C horizon) 14% < 0.002 mm; 8% 0.002 to 0.05 mm; 78% > 0.05 mm. The content of soil organic carbon and total nitrogen reached in the soil treated with mineral fertilizers (Hist-CaNPK) was: 5.70 g C/kg ± 0.26 and 0.56 g N/kg ± 0.03 in the 0–25 cm layer, 1.49 g C/kg ± 0.18 and 0.179 g N/kg ± 0.01 in the 26–50 cm layer and 1.92 g C/kg ± 0.09 and 0.195 g N/kg ± 0.01 below 50 cm of depth. The content of soil organic carbon and total nitrogen reached in the soil treated with manure (Hist-FYM): 10.47 g C/kg ± 0.92 and 0.96 g N/kg ± 0.02 in the 0–25 cm layer, 2.00 g C/kg ± 0.16 and 0.192 g N/kg ± 0.01 in the 26–45 cm layer and 2.17 g C/kg ± 0.12 and 0.221 g N/kg ± 0.01 below 50 cm of depth. In the spring 2012, after air-drying and sieving through a mesh with 5 mm openings (Ap soil horizon 7.8 kg 0–30 cm, 7.2 kg Eet horizon 30–55 cm and 12.3 kg Bt/C horizon 55–100 cm) the soil was placed in PVC (polyvinyl chloride) pipe (ø = 155 mm, length 1 m). Mineral, organic and NIL (non-treated) treatments were applied to the soils Hist-CaNPK and Hist-FYM. Nitrogen was applied at a dose of 2 g per pipe (106 g N/m^2); ammonium nitrate (5.88 g/pipe) was scattered on the soil surface while fresh FYM (416.7 g/pipe) was placed at the depth of 15 cm. Each treatment had 3 replications. No plants were cultivated. The experiment was carried out in an air-conditioned laboratory at the temperature of 22°C. On 15-May-2012, the soils were irrigated with distilled water until obtaining a minimum leachates (excluded from the analysis). Subsequently, six irrigations with 1 L of distilled water were applied at approximately one-month intervals starting on 25-May-2012. Leaching was observed on Day 1 to Day 3 following the irrigation. On each day, the leachates volume was measured and samples were collected. The content of NH_4^+-N, NO_3^--N and C in the leachates was measured using segmented flow analyzer model San Plus Analyzer (Skalar Analytical BV, Breda, the Netherlands) and Thermo Electron-C analyzer model TOC-500 (Shimaidzu, Kyoto, Japan), respectively.

The N_2O emission from the soil was measured by infrared spectroscopy using the portable FTIR spectrometer model Alpha (Bruker, Ettlingen, Germany). The N_2O-N flux from the soil was calculated as an increase in the N_2O concentration in the chamber (ø = 16 cm, h = 18.5 cm) after a 10-min exposure to the soil surface. The results were extrapolated to 24 h and 1 m^2. The first measurement was taken on 15 May 2012 before the soil rewetting. Statistical analyses were performed with the SPSS software 21.0 (IBM, Chicago, USA). The Kruskal-Wallis ($P < 0.05$) and the Mann-Whitney U test and Bonferroni correction for multiple comparisons were used ($P < 0.017$), as the data were not distributed normally and the variance differed in a statistically significant way.

RESULTS

Irrespective of the experimental fertilizer, N_2O-N fluxes from the soil historically treated with FYM (Hist-FYM) exceeded those from the soil historically treated with mineral fertilizers (Hist-CaNPK) and the distributions of N_2O-N emissions from those soils differed in a statistically significant way ($P < 0.001$) (Table 1). On the Hist-CaNPK soil, higher N_2O-N soil emissions were observed following an experimental application of NH_4NO_3 compared with FYM. In contrast, on the Hist-FYM soil, higher N_2O-N emissions were noted upon an experimental application of FYM compared with NH_4NO_3. Application of different experimental treatments to the soil under given historical fertilization resulted in no significant differences between the distributions of N_2O-N emissions ($P = 0.087$). The fluctuations in N_2O-N emission were similar across different historical and experimental treatments. Irrigation-related soil moisture determined the dynamics of N_2O-N emissions (Figure 1).

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Table 1. The soil N_2O-N emission and the NO_3^--N, NH_4^+-N and carbon (C)-content in the leachates from the soils long-term treated with mineral and farmyard manure (FYM) fertilizers upon experimental application of NH_4NO_3, FYM and NIL (non-treated) treatment

Fertilization		Statistics	N_2O (mg N/m^2/day)	NO_3^- (mg N/L)	NH_4^+ (mg N/L)	C (mg C/L)
Long-term	experimental					
CaNPK	NIL	median	0.88	76.2	0.14	5.14
		fractals 25%, 75%	0.53, 1.19	47.0, 156.0	0.10, 0.24	4.00, 6.90
	NH_4NO_3	median	1.00	436.8	0.30	5.41
		fractals 25%, 75%	0.59, 1.25	251.3, 818.0	0.23, 0.37	3.89, 7.27
	FYM	median	0.93	195.9	0.22	6.02
		fractals 25%, 75%	0.59, 1.27	84.1, 305.0	0.16, 0.28	4.51, 7.33
	any treatment	median	0.93^A	184.4^A	0.22^A	5.46^A
		fractals 25%, 75%	0.58, 1.23	69.4, 404.4	0.14, 0.30	4.03, 7.20
FYM	NIL	median	1.06	103.5	0.21	6.38
		fractals 25%, 75%	0.70, 1.32	78.9, 153.6	0.14, 0.34	5.31, 9.30
	NH_4NO_3	median	1.13	462.0	0.43	6.31
		fractals 25%, 75%	0.75, 1.49	245.8, 695.0	0.36, 0.59	4.58, 9.15
	FYM	median	1.29	233.5	0.33	7.29
		fractals 25%, 75%	0.80, 1.53	108.4, 430.0	0.26, 0.46	5.72, 9.31
	any treatment	median	1.14^B	192.0^A	0.33^B	6.62^B
		fractals 25%, 75%	0.76, 1.43	95.3, 449.5	0.20, 0.51	5.25, 9.18
Any treatment	NIL	median	0.95^{ac}	$96,3^{ac}$	0.18^{ac}	5.99^{ac}
		fractals 25%, 75%	0.64, 1.28	62.1, 153.6	0.12, 0.27	4.93, 7.57
	NH_4NO_3	median	1.05^{ae}	449.5^{be}	0.37^{be}	5.87^{ae}
		fractals 25%, 75%	0.72, 1.38	250.2, 734.6	0.27, 0.46	4.13, 8.32
	FYM	median	1.12^{ce}	214.0^{df}	0.27^{de}	6.40^{fd}
		fractals 25%, 75%	0.73, 1.41	91.3, 355. 5	0.17, 0.36	5.09, 8.13

$^{A, B}$indicate significant distribution differences in the soil N_2O-N emission and the NO_3^--N, NH_4^+-N and C content in the leachates from the soils long-term treated with mineral and FYM fertilizers irrespective of the experimental fertilization ($P < 0.05$, Kruskal-Wallis test). Lowercase letters ($^{a, b, c, d, e, f}$) indicate significant distribution differences between the experimental treatments irrespective of the long-term treatment: ^{a,b}between experimental NH_4NO_3 and NIL; ^{c,d}between experimental FYM and NIL; ^{e,f}between experimental NH_4NO_3 and FYM ($P < 0.017$, Mann-Whitney U-test and Bonferroni correction)

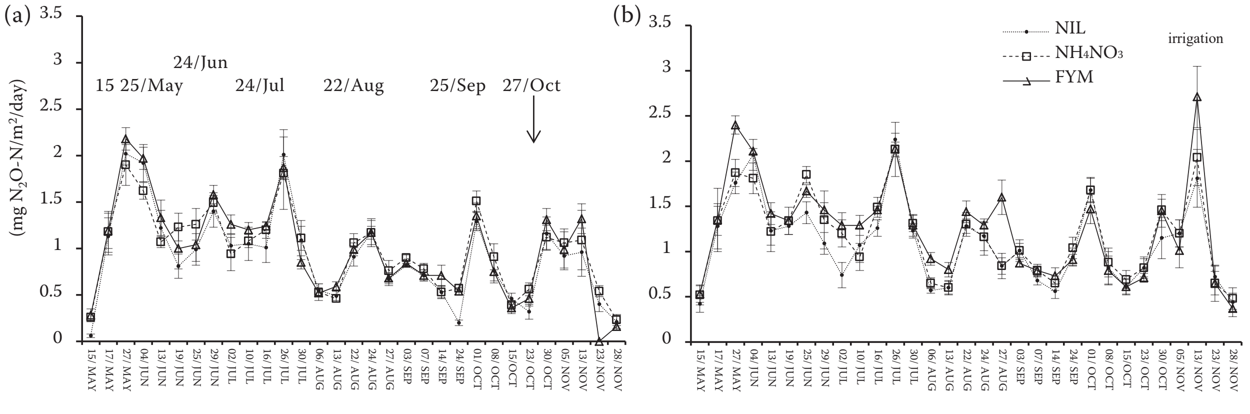

Figure 1. N_2O-N emissions from the soils long-term treated with (a) mineral and (b) manure fertilizers (FYM) upon experimental application of ammonium nitrate and manure and under NIL (non-treated) treatment

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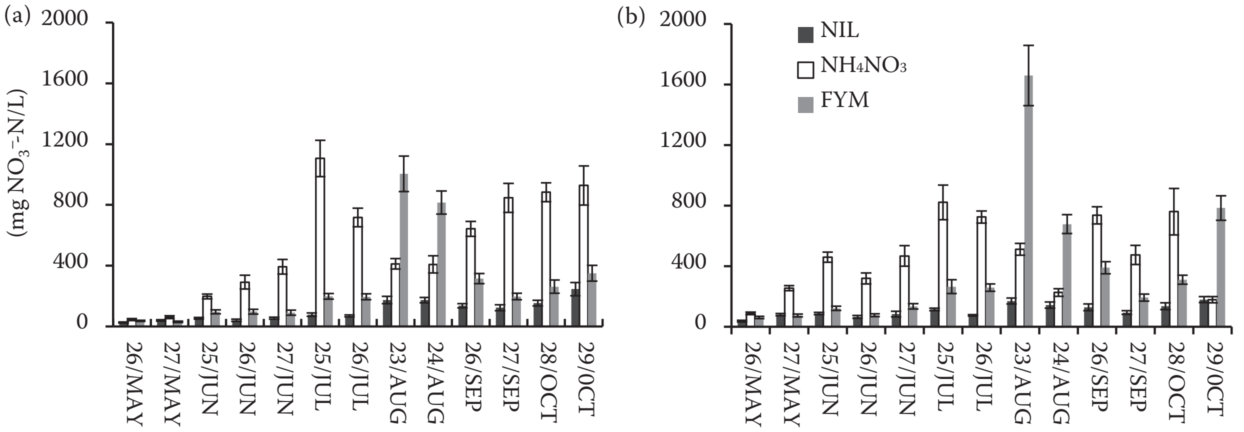

Figure 2. NO_3^--N content in the leachates from the soils long-term treated with (a) mineral and (b) farmyard manure (FYM) fertilizers upon experimental application of NH_4NO_3, FYM and under NIL (non-treated) treatment

NO_3^--N constituted the most important component of the leachates (Table 1). The median NO_3-N content in the leachates from Hist-FYM soil was higher than the median NO_3-N content in the leachates from Hist-CaNPK soil. The content of NO_3^--N in the leachates from experimental NH_4NO_3 treatments were higher than from experimental FYM treatments and higher than from NIL-treatments. The distributions of the content of NO_3^--N in the leachates from those experimental treatments differed in a statistically significant way ($P < 0.001$). Experimental application of NH_4NO_3 and FYM to both soils resulted in a gradual increase in the NO_3^--N leachates content (Figure 2). Peaks under experimental FYM were observed in August, a month later than under experimental NH_4NO_3 treatment. Overall, the content of NO_3^--N in the leachates was 441.9–1441.6 times higher than that of NH_4^+-N as a function of the historical and experimental treatment (Table 1, Figure 3).

The mineral-N (NO_3^--N + NH_4^+-N) losses were higher from Hist-FYM than from Hist-CaNPK soil (Table 2). The mineral-N losses over the study period ranged between 34.9 and 37.9 g N/m^2 upon experimental NH_4NO_3 treatment and between 20.6 and 26.5 g N/m^2 upon FYM application on Hist-CaNPK and Hist-FYM soil, respectively (Figure 4).

The C-content in the leachates from manured soils (both long-term and experimental treatment) was higher than from mineral-treated soils. The C-content in the leachates depended more on the historical FYM treatment than on experimental FYM application (Table 1). Fluctuations in the C leachates content were similar under all experimental treatments (Figure 5).

The C:NO_3^--N ratio in the leachates was extremely low under all experimental treatments (Figure 6). Application of both, NH_4NO_3 and FYM resulted in a decreased in the C:NO_3^--N ratio as compared with NIL. FYM treatment was the ma-

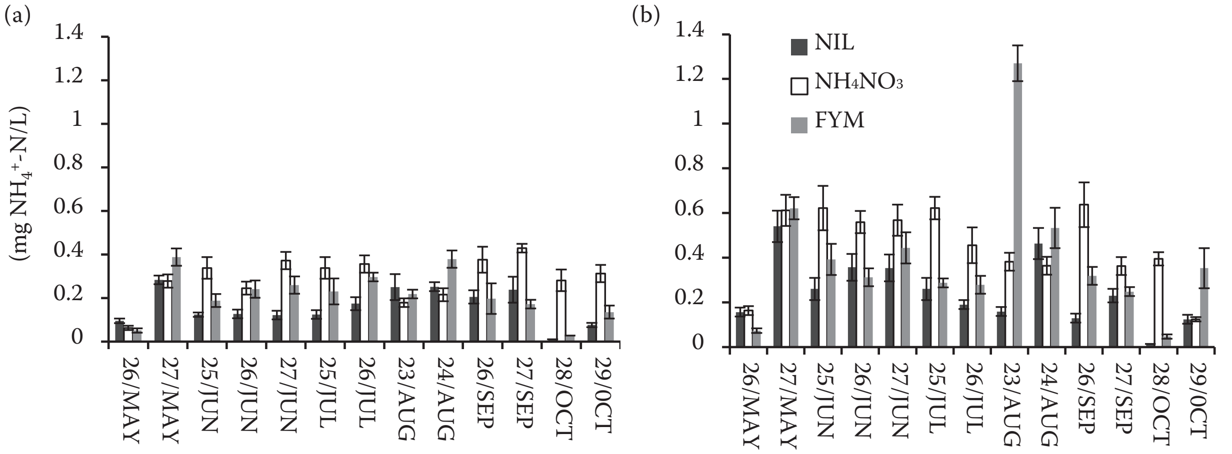

Figure 3. NH_4^+-N content in the leachates from the soils long-term treated with (a) mineral and (b) farmyard manure (FYM) fertilizers upon experimental application of NH_4NO_3, FYM and under NIL (non-treated) treatment

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Table 2. The carbon (C), NH_4^+-N and NO_3^--N outflow from the soils long-term treated with mineral fertilizers and farmyard manure (FYM) upon experimental application of NH_4NO_3, organic and NIL (non-treated) treatment

Fertilization Long-term	 experimental	Statistics	Outflow C (mg C/m^2)	 NH_4^+-N (mg N/m^2)	 NO_3^--N (g N/m^2)
CaNPK	NIL	total	493.91	11.13	6.69
		median	28.62	0.53	0.50
	NH_4NO_3	total	484.90	20.67	34.82
		median	29.15	1.59	2.20
	FYM	total	638.05	23.85	20.59
		median	30.74	1.06	0.99
	any treatment	median	29.15^A	1.06^A	1.03^A
		min–max	3.18–182.84	0.00–7.95	0.02–6.66
FYM	NIL	total	712.77	21.73	8.27
		median	39.22	1.06	0.50
	NH_4NO_3	total	711.71	42.40	37.83
		median	40.28	2.65	2.56
	FYM	total	952.31	42.40	26.50
		median	45.05	2.12	1.58
	any treatment	median	43.46^B	2.12^B	1.26^A
		min–max	9.01–363.03	0.00–13.78	0.08–7.32
Any treatment	NIL	median	30.21^{ac}	1.06^{ac}	0.54^{ac}
	NH_4NO_3	median	33.92^{ae}	2.12^{be}	2.56^{be}
	FYM	median	42.40^{ce}	1.59^{de}	1.59^{df}

^{A,B}indicate significant distribution differences in the C, NH_4^+-N and NO_3^--N outflow from the soils long-term treated with mineral and FYM fertilizers irrespective of the experimental fertilization ($P < 0.05$, Kruskal-Wallis test). Lowercase letters ($^{a, b, c, d, e, f}$) indicate significant distribution differences between the experimental treatments irrespective of the long-term treatment: ^{a,b}between experimental NH_4NO_3 and NIL; ^{c,d}between experimental FYM and NIL; ^{e,f}between experimental NH_4NO_3 and FYM ($P < 0.017$, Mann-Whitney U test and Bonferroni correction)

jor determinant of the total C-outflow (Table 2). C-outflows from Hist-FYM soil were higher than those from the Hist-CaNPK soil. All experimental treatments resulted in C-outflow peaks in June (Figure 7).

DISCUSSION

The available organic C is a critical factor that controls the denitrification, heterotrophic nitrification and N_2O emission from the soil (Wang et

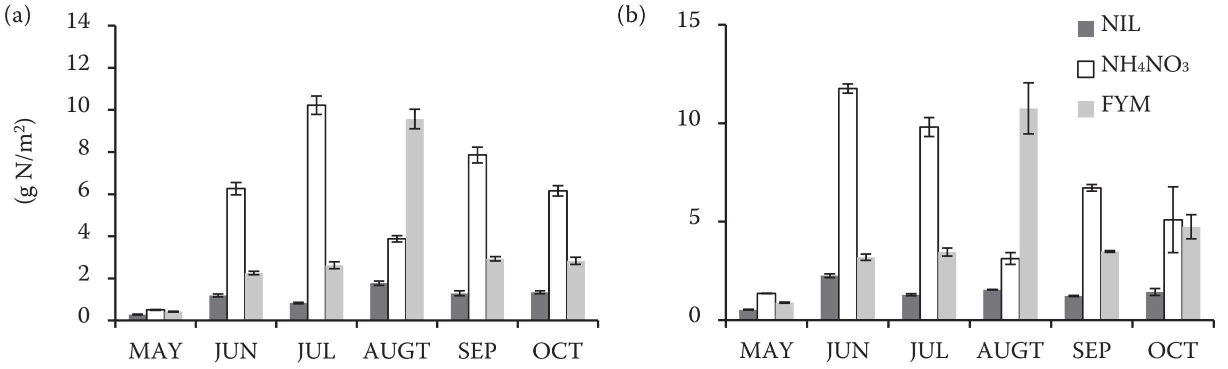

Figure 4. Mineral-N soil leaching from the soils long-term treated with (a) mineral and (b) farmyard manure (FYM) fertilizers upon experimental application of NH_4NO_3, FYM and under NIL (non-treated) treatment

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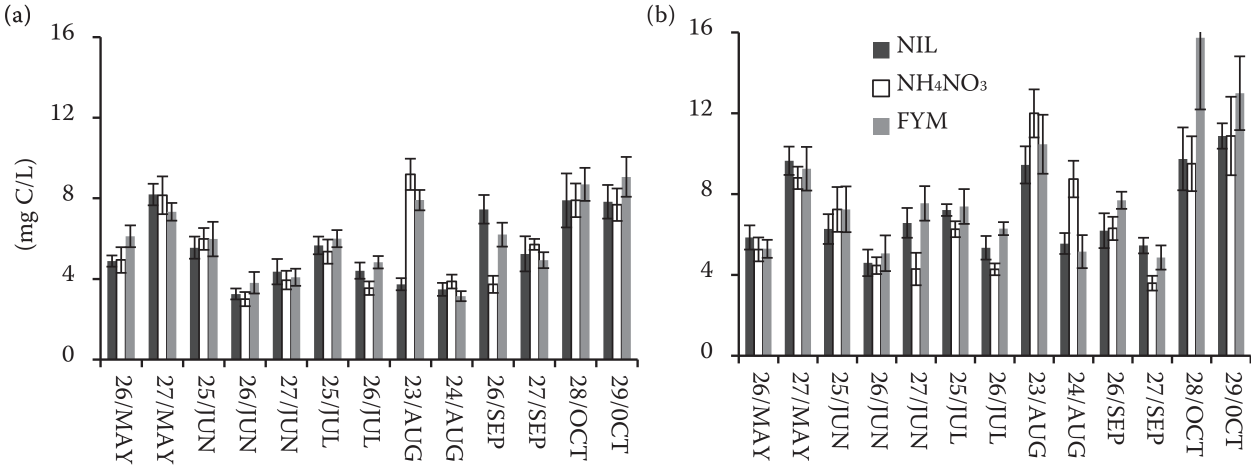

Figure 5. Carbon (C)-content in the leachates from the soils long-term treated with (a) mineral and (b) farmyard manure (FYM) fertilizers upon experimental application of NH_4NO_3, FYM and under NIL (non-treated) treatment

al. 2005). According to Jäger et al. (2011), long-term FYM treatment increases the C_{org} content, which leads to an increase in the N_2O emissions from the soil; the study has shown, however, that the effect of fertilization history on N_2O emission is small compared with short-term effects of the current fertilization. In our study, the N_2O-N fluxes from the soil treated long-term with FYM exceeded those from the soil treated long-term with mineral fertilizers irrespective of the applied experimental fertilizer. Of the experimental treatments, FYM resulted in higher N_2O-N emission from the soil historically treated with FYM but not from the soil historically treated with mineral fertilizers. N_2O-N emission was impacted more by the historical FYM treatment than by experimental FYM application. Low C-content in the soil may hinder denitrification (Rivett et al. 2007) through a departure of C:NO_3^--N ratio in the soil from the value optimal for denitrification (Her and Huang 1995). Sosulski et al. (2013) found higher content of labile carbon in the soil under organic fertilization than under the mineral one. In our study, experimental application of NH_4NO_3 to the Hist-CaNPK soil resulted in higher N_2O-N emission compared with the application of FYM to the Hist-CaNPK soil. Jäger et al. (2011) obtained similar results in a laboratory experiment with soils from three long-term experiments in Germany. Ball et al. (2004) mitigated the N_2O emission from the soil by approximately 90% via substituting a mineral fertilizer with manure. Velthof et al. (2003) have shown that the N_2O soil emission upon an application of manure can exceed the emission upon application of a mineral fertilizer if the content of mineral-N and easily decomposable N and C in the manure is

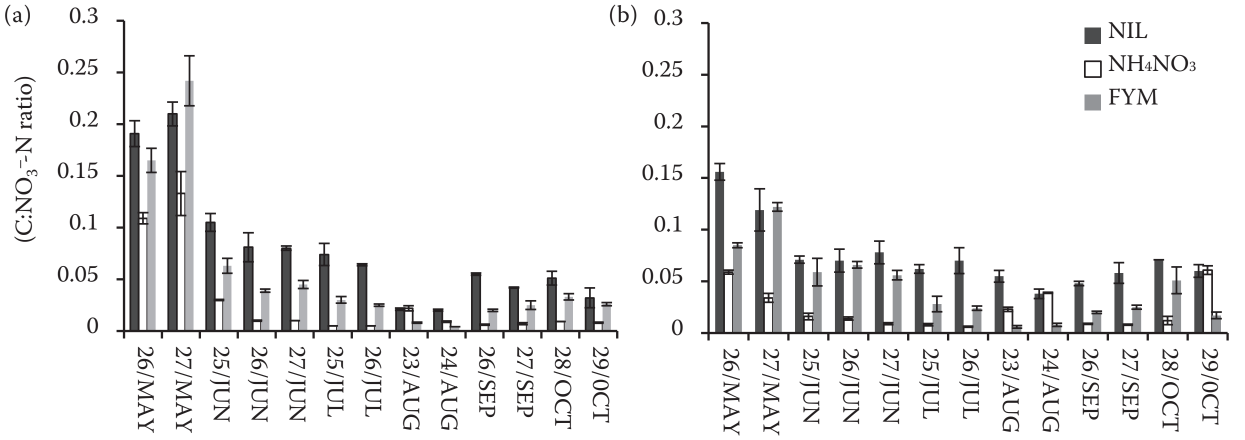

Figure 6. C:NO_3^--N ratio in the leachates from the soils long-term treated with (a) mineral and (b) farmyard manure (FYM) fertilizers upon experimental application of NH_4NO_3, FYM and under NIL (non-treated) treatment

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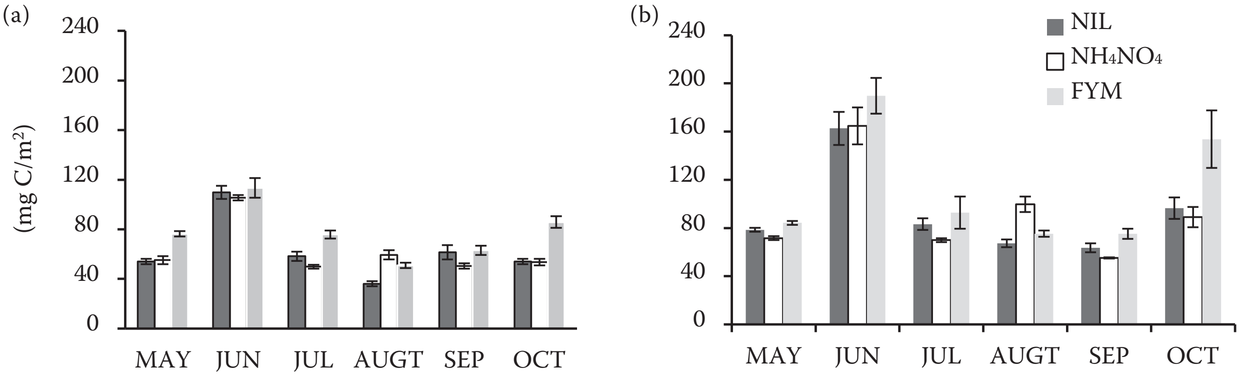

Figure 7. Carbon (C)-soil leaching from soils long-term treated with (a) mineral and (b) farmyard manure (FYM) fertilizers upon experimental application of NH_4NO_3, FYM and under NIL (non-treated) treatment

high, such as in liquid manure. Amha and Bohne (2011) demonstrated an increase in the N_2O flux from the soil with increasing soil moisture. In our study, the most pronounced increase in the N_2O soil emission was observed after the rewetting of the soil, whereas subsequent irrigations applied at monthly intervals had a smaller yet noticeable effect on the N_2O-N fluxes. This observation was consistent with the results by Flessa et al. (2012), who reported the highest N_2O emissions after heavy precipitation and Ruser et al. (2006) who observed an increase in the N_2O emission after rewetting of the soil above 70% WFPS (water-filled pore space). However, Yanai et al. (2007) detected no detectable increase in N_2O emissions following rewetting of the soil at 64% WFPS.

Based on the examination of drainage water collected at drain outlets and from drainage ditches carried out in Poland over the years 1998–2002 in a net of 722 farms, Igras (2004) described the NO_3-N content in soil leachates as ranging from 0.01 to 166 mg N/L. In our study, comparable NO_3^--N concentrations in the leachates were observed only upon NIL-treatments. Experimental application of NH_4NO_3 and FYM to both soils resulted in a much higher NO_3 content in the leachates. Application of NH_4NO_3 increased the NO_3-N content in the leachates by over 2–4 times in the function of the historical treatment. The influence of manuring on the NO_3 content in the leachates was much less pronounced. Our data are contradictory to the research by Kücke and Kleeberg (1997), who determined that groundwater N-contamination occurred as a result of organic fertilization due to mineralization of organic compounds. On the other hand, Elmi et al. (2005) obtained similar NO_3^--N content in drainage water from mineral and organic-fertilized soils. Gradual increases in NO_3^--N content were observed in the leachates with maximum values reached approximately 2 and 3 months after the application of NH_4NO_3 and FYM, respectively. The differences in the dynamics of NO_3^--N leaching between the soils subjected to different experimental treatments can be explained by the mineralization and nitrification required to produce NO_3^- after the application of FYM in contrast to an immediate availability of NO_3^- following the application of NH_4NO_3.

The C-content in the leachates depended more on the Hist-FYM treatment than on the experimental FYM application. The C-content in the leachates from the soils experimentally and/or historically treated with FYM was greater than from those under mineral fertilization. These data can be explained in the context of the research by Sosulski et al. (2013), who detected a higher content of extractable organic carbon in the arable soils treated organically than in those under conventional mineral fertilization. As already discussed, a low C-content (< 5 mg C/L) inhibits denitrification (Rivett et al. 2007). In our study, the median C content in the leachates was 5.14 mg C/L or higher; however the C:NO_3^--N ratio (0.004–0.242:1) was far from the value considered optimal (0.9:1 to 3:1) for complete denitrification (Her and Huang 1995). An inadequate C:NO_3^--N ratio with subsequent inhibition of denitrification is supposed to be the main reason of permanent pollution of the groundwater with nitrates.

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