

# IDŐJÁRÁS

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## **Weather induced variability of nitrogen exchange between the atmosphere and a grassland in the Hungarian Great Plain**

**Attila Machon<sup>\*1,2,4</sup>, László Horváth<sup>1</sup>, Tamás Weidinger<sup>3</sup>,  
Krisztina Pintér<sup>4,5</sup>, Balázs Grosz<sup>3</sup>, Zoltán Nagy<sup>4,5</sup>, and Ernő Führer<sup>6</sup>**

<sup>1</sup>*Hungarian Meteorological Service,  
Gilice tér 39, H-1181 Budapest, Hungary*

<sup>2</sup>*Center for Environmental Science, Eötvös Loránd University,  
Pázmány P. sétány 1/A, H-1117 Budapest, Hungary*

<sup>3</sup>*Department of Meteorology, Eötvös Loránd University,  
Pázmány P. sétány 1/A, H-1117 Budapest, Hungary*

<sup>4</sup>*Institute of Botany and Ecophysiology, Szent István University,  
Páter Károly utca 1, H-2103 Gödöllő, Hungary*

<sup>5</sup>*Plant Ecology Research Group of Hungarian Academy of Sciences,  
Páter Károly utca 1, H-2103, Gödöllő, Hungary*

<sup>6</sup>*Hungarian Forest Research Institute,  
Papréti 17, H-9400 Sopron, Hungary*

*\*Corresponding author; E-mail: machon@caesar.elte.hu*

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**Abstract**—The paper describes some of the preliminary results of the ecological research on nitrogen exchange in a grassland in central Hungary (Kiskunság National Park, Bugacpuszta). The changes in different climate parameters evidently affect not only N-deposition but also N-exchange and N-gas emissions through the processes of soil and plant metabolism. Measurements of nitrogen fluxes and basic meteorological parameters have been started above a semi-natural grassland ecosystem in 2002. Seasonal and long-term nitrogen exchange (both emission and deposition) is under climatic control. In the years of 2006 and 2007, the amount of the deposited N markedly decreased (by 27% and 15%, respectively) compared to the average of the earlier (2002–2004) years. The main source of the deposited N is NH<sub>3</sub>. The ratio of dry to wet deposition varies between 1.5 and 2.3. In the dry year of 2007, emissions of N<sub>2</sub>O were four times lower compared to the

average ( $90 \text{ mg N m}^{-2} \text{ yr}^{-1}$ ) of the earlier years caused by the changes in weather conditions including lower precipitation and  $1 \text{ }^\circ\text{C}$  higher annual average temperature. In the year with higher precipitation (2010),  $\text{N}_2\text{O}$  emissions increased again and reached  $180 \text{ mg N m}^{-2} \text{ yr}^{-1}$  when the annual rainfall was twice the normal rate. It should be noted that soil water filled pore space (WFPS) cannot explain all of the variations in  $\text{N}_2\text{O}$  fluxes. With increasing soil temperature,  $\text{NO}$  flux grows faster than  $\text{N}_2\text{O}$  up to  $20 \text{ }^\circ\text{C}$  until the role of other factors (e.g., water stress, nutrient supply, and other complex processes linked to heat stress) will determine the magnitude of metabolism. The relatively high soil  $\text{N}_2\text{O}$  flux under  $5 \text{ }^\circ\text{C}$  could come from the thawing period 2–3 months after wintertime which could result in high emission peaks for a few days with low soil temperature. It seems to be the case that soil temperature usually generates short term variability of trace gas exchange, whereas the magnitude of the biogenic emission is dominantly controlled by soil wetness, pH, and other site specific factors. The net N flux – excluding grazing, manure, farm management, etc. – ranged between  $9.5$  and  $13 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ . Reduced  $\text{N}_2\text{O}$  emission presents a potential negative feedback to emission; on the other hand, vegetation can become a net  $\text{CO}_2$  source in extremely dry years such as 2003 and 2007 as a positive feedback to climate change.

*Key-words:* N-exchange, N-emission, N-deposition, climate perturbation, grassland, denitrification

## ***1. Introduction***

The climate in middle and southern Europe will likely be warmer, with drier summers; in some parts with shorter or mild and wetter winters, and more variable patterns of rainfall and temperature in the 21st century as predicted by *EEA* (2004). Grasslands are one of the most widespread vegetation types in Europe and, especially, in the Hungarian Great Plain, which also appear to be drifting to desertification. This is an important environmental problem, because grassland ecosystems are important in the cycle of nitrogen (N) and carbon (C) between the land surface and the atmosphere. Sensitivity of N balance of grasslands to climatic perturbations such as drought remains uncertain (*Schimel et al.*, 1996; *Reich et al.*, 2006).

Interactions between climate perturbations and changing dynamics of N-cycle of grasslands have received much less attention than the C-cycle and corresponding interactions in forests. Some studies were carried out for Hungarian grassland sites on how net ecosystem exchange (NEE) of carbon dioxide, respiration processes (*Balogh et al.*, 2005, 2008), or the greenhouse gas (GHG) flux will change as a consequence of climate change (*Nagy et al.*, 2005, 2007). The markers such as heat and drought extremes – which will become more frequent as a result of the climate perturbations – can also have an impact the nitrogen budget through trace gas soil fluxes as well as the carbon and water balance (*Pintér et al.*, 2008) due to soil functioning, and they may lead to reduced plant growth or changes in grassland species composition. Some European studies pointed out the effects of soil condition on  $\text{NO}$ ,  $\text{NO}_2$ , and  $\text{N}_2\text{O}$  emission from different soils (*Schindlbacher et al.*, 2004) or on  $\text{CH}_4$  uptake by sandy grassland

ecosystems (*van den Pol-van Dasselaar et al.*, 1998). Many aspects of the relation between N and C cycles (e.g., *Soussana and Hartwig*, 1996), estimation of the nitrogen and carbon fluxes (*Levy et al.*, 2007) and budgets (*Ammann et al.*, 2009; *Skiba et al.*, 2009) in grassland were also studied.

The changed weather condition – through the changes of soil conditions (e.g., soil temperature, humidity etc.) – greatly affects the soil processes like mineralization, decomposition, nitrification, and denitrification as the significant sources and controllers of atmospheric C and N trace gases (*Conrad*, 1996) and the sink of  $\text{NH}_4^+$  and  $\text{NO}_3^-$  substrates. Due to the processes above, the organic/inorganic N pool of soil or soil emission of NO,  $\text{N}_2\text{O}$ , as well as N uptake of plants are subject to change. These processes are linked with other stressors (e.g., heat stress, water deficiency, etc.) and may easily influence plant physiology, productivity or plant species composition.

The objective of this work is to investigate the nitrogen exchange between atmosphere and grassland and its dependence on climatic conditions, and focusing on links with reactive trace gas emissions and N-deposition, and on the possible feedbacks to soil-vegetation dynamics.

## 2. Materials and methods

Investigations were conducted at the *Bugacpuszta* nature reserve research site (46.69 °N, 19.60 °E, h = 110 m asl) in the Hungarian Great Plain. Measurements of nitrogen fluxes and basic micrometeorological parameters above a semi-natural ecosystem were started in 2002. This station also provides input data for modeling, validation, and calibration of models. The climate is continental with annual average temperature of 10.5 °C, and the average precipitation is about 530 mm year<sup>-1</sup>. The vegetation is semi-arid sandy grassland (*Cynodonti Festucetum pseudovinae*) where *Festuca pseudovina*, *Carex stenophylla*, *Salvia pratensis*, and *Cynodon dactylon* are the dominant plant species. The landscape is flat. The soil is a Chernozem-type sandy soil (*Machon et al.*, 2010) (sand : silt : clay = 82 : 7 : 11% is the average composition of the top 20 cm soil).

Soil emission of NO has been determined by dynamic chamber method as described in *Horváth et al.* (2006). Soil  $\text{N}_2\text{O}$  flux measurements were carried out bi-weekly (2002–2004), later weekly (2006–2010) by 8 parallel static soil chambers (*Clayton et al.*, 1994; *Christensen et al.*, 1996). Samples were taken at  $t = 0, 15,$  and 30 minutes after closure of chambers. Concentration changes (within 30 min) were determined by gas chromatography-mass spectrometry (GC-MS). According to statistical analysis, the non-systematic bulk error (CV: coefficient of variation) of sampling and analysis, estimated using parallel sampling, was always below 10%. In 2007, the detector was changed to electron capture (GC-ECD). Three-stage filter pack method was used for daily sampling of particles and gases followed by ion-chromatography (nitrate, nitric acid) and

spectrophotometry (indophenol-blue method for ammonium and ammonia) (EMEP, 1996). The minimum detection limit (MDL) is  $0.1 \mu\text{g N m}^{-3}$  for all components. The precision (bulk relative error) of sampling and measurements was around 10%. Dry deposition of nitrogen dioxide, ammonia, nitric acid vapour, nitrate, and ammonium particles were inferred using dry deposition velocities (Horváth *et al.*, 2005) measured above surfaces with the same characteristics as those of Bugacpuszta station. Fluxes of  $\text{NH}_3$ , within the atmosphere and the canopy (excluding soil emission) were also inferred this way. Wet depositions of nitrate and ammonium ions were calculated on the basis of daily precipitation sampling and concentration measurements of ammonium and nitrate ions by the analytical methods described in Machon *et al.* (2010). The calculated bulk error of precipitation sampling and concentration measurements was around 10%. Minimum detection limit was  $0.05 \text{ mg NL}^{-1}$  for both ions. Meteorological parameters like precipitation, air and soil temperature, and soil moisture were also observed.

### 3. Results and discussion

The studied sandy grassland soil has poor water supply with deep water table (6 m). In summer months, high ground surface temperatures can be observed ( $>30^\circ\text{C}$ ) and the upper layer of soil dries out. Nitrate leaching is estimated to be negligible. Suction cups applied below rooting zone did not collect measurable quantity of soil water. The soil N trace gas production depends strongly on soil biology, chemistry, and physics (Smith *et al.*, 2003). Denitrification is stimulated by hypoxia. In dry soils (down to water filled pore space (WFPS) of about 30%, see Fig. 1) this process is limited due to the unfavorable soil conditions for the anaerobic microbial communities (nitrous oxide production and subsequent emission rate are maximized at WFPS range of 40–50% (Horváth *et al.*, 2010)). In drier soils, in the well aerated zone, the nitrification is the dominant process producing NO.

During the summer, microbial productivity is elevated (mineralization, nitrification, immobilization, decomposition, etc.), but denitrification ( $\text{N}_2\text{O}$  production) is suppressed when the WFPS is low. In dormant period, despite the higher WFPS, the activity of microbial community decreases in parallel with soil temperature decrease, resulting in low  $\text{N}_2\text{O}$  production.

Changes in seasonality, distribution, and frequency of precipitation and the total amount of rainfall may have an impact greatly on the nitrogen exchange of this ecosystem, resulting in a switch between the  $\text{N}_2\text{O}$  and NO production (determined by soil processes). Both seasonal and long-term nitrogen exchanges of this ecosystem is, therefore, linked to the soil water content (due to rainfall regime) and soil temperature (Machon *et al.*, 2010). Fig. 1 shows that higher soil NO emissions were observed in comparison to  $\text{N}_2\text{O}$  in the dry soil in summer

seasons, especially in 2007. During summers, the WFPS ranged between 20–30% (with exception of 2010). In 2007, the average soil wetness was 19%, close to the optimum for NO production by nitrification processes. Nitrification processes were dominating this year and NO emission ( $70 \text{ mg N m}^{-2} \text{ yr}^{-1}$ ) was higher than N<sub>2</sub>O soil flux ( $15 \text{ mg N m}^{-2} \text{ yr}^{-1}$ ); later it significantly decreased compared to the earlier (2002–2004) years when average emission was  $90 \text{ mg N m}^{-2} \text{ yr}^{-1}$  (Flechard *et al.*, 2007). In recent years, similar significantly lower emissions of N<sub>2</sub>O were observed at different types of grasslands in Hungary (Horváth *et al.*, 2008), in comparison to 2002–2004, on the basis of weekly sampling. In 2010, N<sub>2</sub>O emissions ( $180 \text{ mg N m}^{-2} \text{ yr}^{-1}$ ) exceeded NO emissions ( $78 \text{ mg N m}^{-2} \text{ yr}^{-1}$ ) due to the unusually high precipitation and WFPS in summer months, favoring denitrification metabolisms.

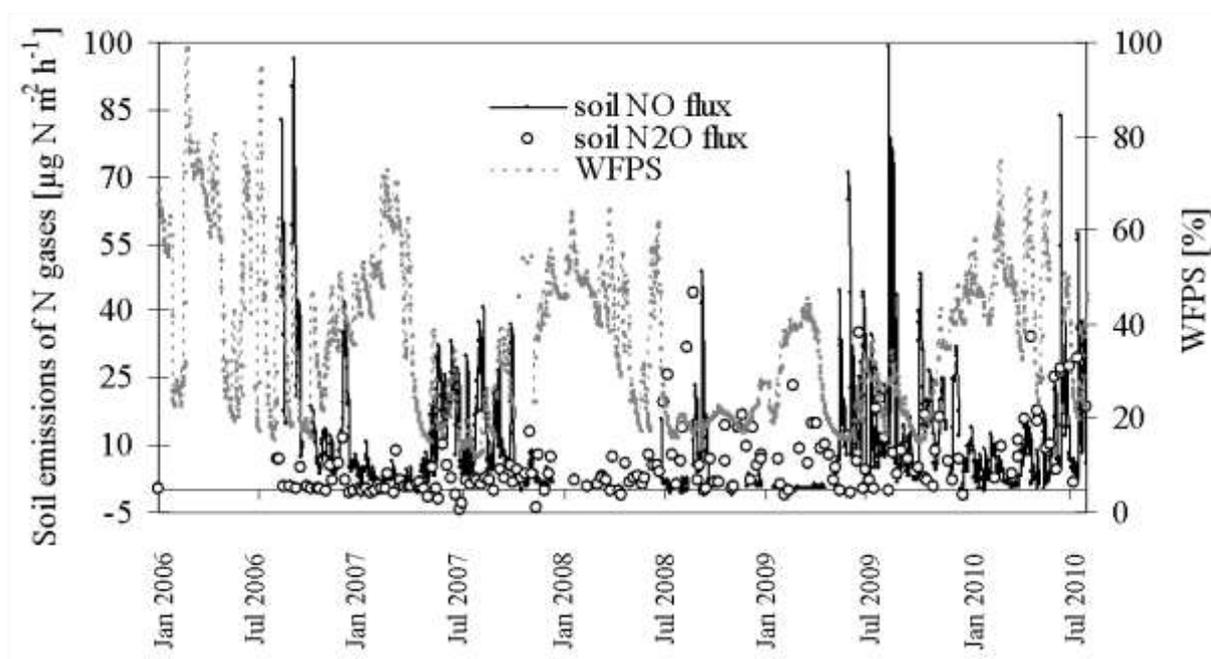


Fig. 1. Soil N gases emissions and linkage to soil humidity (WFPS) at Bugacpuszta station. (Due to instrument failure, there are only a few data points for soil NO flux in 2008.)

Trace gas emissions and N-deposition can be highly variable from season-to-season or year-to-year, but available parameters such as air and soil temperature, precipitation, and soil moisture more or less can explain this variability. Some uncertainty may arise from the complexity of soil and atmospheric processes and/or accuracy of measurements.

It can clearly be seen in *Table 1*, that emission of N<sub>2</sub>O decreases by a factor of 4 in 2006 and 2007 and increases by a factor of two in 2010 compared to the earlier (2002–2004) measurements at the same place. This phenomenon is explainable by soil microbial processes, as the rate of denitrification, etc., strongly depends on the varying weather conditions (e.g., water or heat stress). In 2009, the annual mean temperature was the same as in 2008, but this year was

drier. In contrast, substantial decrease was not observed in the measured flux. It can be explained partly by the fact that the sandy soil dried out within a short time after the rain events, and the effective time for denitrifying bacteria, preferring anaerobic condition, was shorter than in wet soils with higher water retaining capacity. On the other hand, in extremely wet soils emission of nitrous oxide is limited, and reduction leads to production of di-nitrogen (N<sub>2</sub>). The weather conditions (e.g., abiotic stress) prevent optimum soil condition for nitrous oxide production to be sustained for long periods during the study.

*Table 1.* Annual sum of climatic conditions and N-exchange [mg N m<sup>-2</sup> yr<sup>-1</sup>] between the surface and atmosphere at Bugacpuszta station

Measured parameters	2002–2004	2006	2007	2008	2009	2010
Precipitation [mm]	545	524	446	567	486	967
WFPS mean [%] (SD)		34.6 ± 11.7	33.6 ± 16.6	30.2 ± 13.9	27.9 ± 9.67	43 ± 12
T <sub>air</sub> , mean [°C]	10.1	10.1	11.1	11.0	11.2	11
T <sub>soil (5cm)</sub> , mean [°C] (SD)		10.2 ± 6.6	11.5 ± 4.5	11.1 ± 3.7	11.4 ± 3.7	10.9
Wet (NO <sub>3</sub> <sup>-</sup> and NH <sub>4</sub> <sup>+</sup> ) deposition (SD)	-470 ± 23 <sup>a</sup>	-338 ± 17	-420 ± 21	-520 ± 26	-450 ± 23	-578 ± 29
Dry HNO <sub>3</sub> deposition (SD)	-320 ± 32 <sup>a</sup>	-157 ± 16	-172 ± 17	-264 ± 26	-235 ± 24	-295 ± 30
Dry NH <sub>3</sub> deposition (SD)	-460 ± 23 <sup>a</sup>	-418 ± 21	-454 ± 23	-532 ± 27	-425 ± 21	-379 ± 19
Dry NO <sub>3</sub> <sup>-</sup> and NH <sub>4</sub> <sup>+</sup> deposition (SD)	-130 ± 7 <sup>a</sup>	-136 ± 7	-85 ± 4	-109 ± 5	-116 ± 6	-104 ± 5
Dry NO <sub>2</sub> deposition (SD)	-75 <sup>b</sup>	-75 ± 4	-45 ± 2	-80 ± 4	-79 ± 4	-95 ± 5
Soil N <sub>2</sub> O emission	80 <sup>c</sup>	18	15	56	63	180
Soil NO emission	119 <sup>d</sup>	160	79	119 <sup>d</sup>	118	78
Total deposition	-1.455	-1.124	-1.176	-1.505	-1.305	-1.451
<b>Sum of net N flux</b>	<b>-1.256</b>	<b>-946</b>	<b>-1.082</b>	<b>-1.330</b>	<b>-1.124</b>	<b>-1.193</b>

<sup>a</sup> Kugler *et al.*, 2008

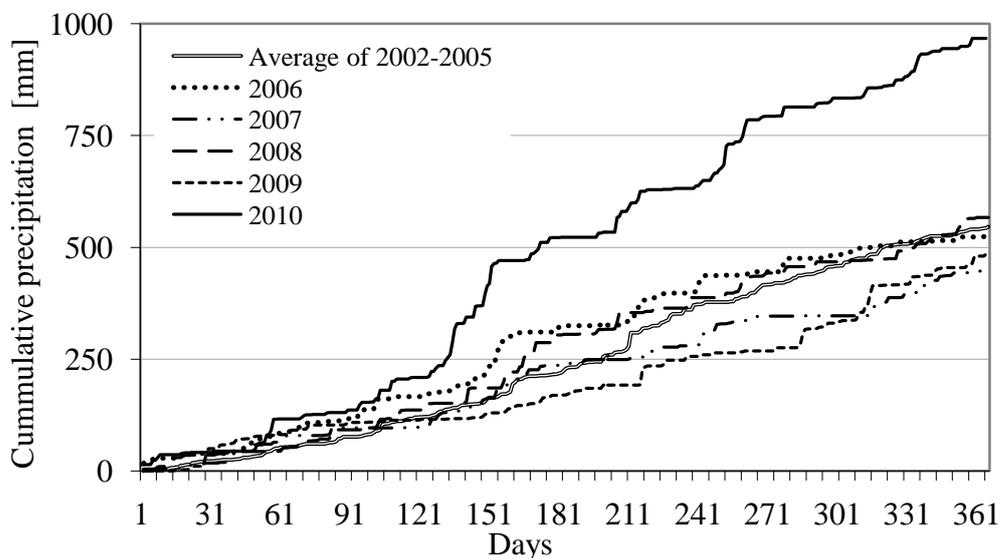
<sup>b</sup> Estimated from the average of 2006–2010

<sup>c</sup> Horváth *et al.*, 2010

<sup>d</sup> No data for technical reason, calculated from the mean NO emission for 2006–2007 and 2009

Wet deposition of nitrate and ammonium together with the dry flux of ammonia and nitric acid vapor are responsible for the majority (80%) of the net N budget.

The sum of the atmospheric N-deposition (excluding deposited organic N) varied between 1.1 and 1.5 mg N m<sup>-2</sup> yr<sup>-1</sup>. It is noteworthy, that the precipitation amount nearly doubled in 2010 (see in *Table 1*), nevertheless, the wet deposition does not follow this pattern. Ratio of dry to wet deposition varied in the range of 1.5–2.3. According to the results (*Fig. 2*), in 2007 and 2009 the annual precipitation amount was significantly lower, and in 2010 it was two times higher than the long term mean. The deficit in the yearly precipitation mainly occurred in July in the vegetation period of 2006–2009 (see *Fig. 3*) resulting in less biomass production, because the drought and the main growing period coincided. At the same time, the lack of precipitation affects the N-cycle through missing wet deposited N, plant uptake, water stress, and other soil processes. In 2007, the annual mean temperature has increased by 1 °C associated with mild winter (soil frost occurred rarely) and less number of rain events were observed. In 2008, the amount of precipitation reached the regular level, but the annual mean temperature remained 1 °C higher as in 2007. The year of 2010 had extra amount of precipitation (it was one of the most wet of the last 100 years; <http://www.met.hu>) and differs from the long-term statistical averages (see in *Figs. 2 and 3*). For this reason, all the circumstances and conditions are different from the previous years.



*Fig. 2.* Cumulative precipitation for the last decade at Bugacpuszta station.

N-compounds can easily be leached into the soil by rain, while canopy can retain dry deposited nitrogen through the N-uptake by stomatae or cuticles, which also depends on the meteorological conditions (*Horváth et al., 1999*). For this reason, in drier years less amount of deposited, reaches the topsoil layers

nitrogen and it can limit the mineral N-conversion to gases (through denitrification and nitrification processes). Reduced availability of soil mineral nitrogen may limit the plant uptake and probably impacts on the plant physiology and biomass production of different species; nevertheless, changes of plant species composition or coverage of plant functional group are not observed. Further research of N-linkage to the coenological experiment of grassland association is needed in the future.

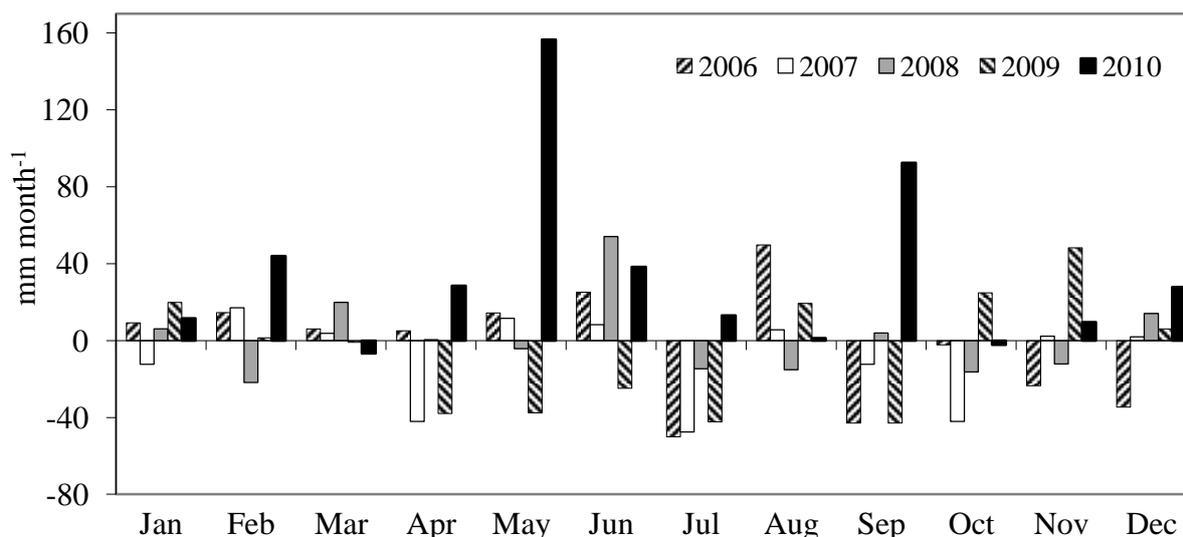
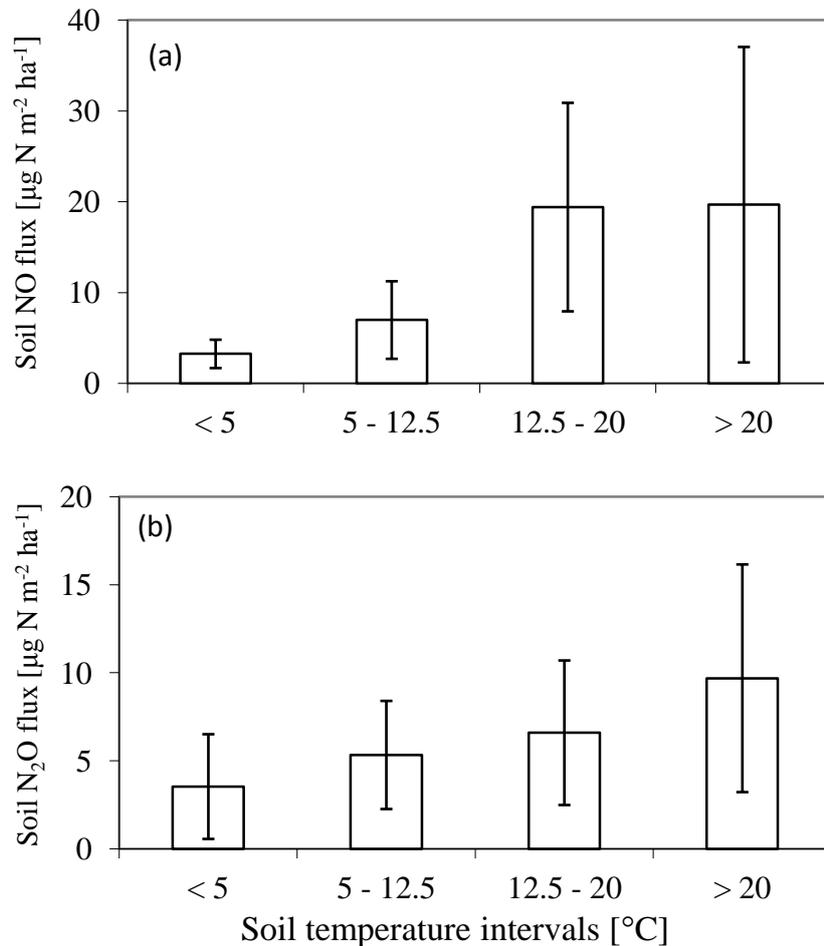


Fig. 3. Deviation of monthly precipitation of the studied years from the average of long term period (1986–2006) at Bugacpuszta station.

NO and N<sub>2</sub>O soil emissions as consequences of nitrification and denitrification are, like all biological processes, influenced by temperature and correlate with the soil temperature as many studies discussed before (*Smith et al.*, 1998; *Gödde and Conrad*, 1999; *Horváth et al.*, 2010), but sometimes the correlation remains poor or not readily understandable (*Clayton et al.*, 1997). With increasing soil temperature, the NO flux increases faster than N<sub>2</sub>O up to 20 °C; until the role of other factors (e.g., water stress, nutrient supply, and other complex processes linked to heat stress) will determine the rate of metabolism of soil microorganisms (see *Fig. 4*). The relatively high soil N<sub>2</sub>O flux under 5 °C could be the reason of thawing period after 2-3 month wintertime which resulting in high emission peaks for a few days even at low soil temperature (*Priemé and Christensen*, 2001; *Müller et al.*, 2002).

The optimal soil wetness for NO and N<sub>2</sub>O is ranged around 20–30% and 40–50%, respectively (see *Fig. 5*) for this sandy soil. The optimal WFPS content is reached only in 2010 for N<sub>2</sub>O. High water content (WFPS > 70%) was rarely observed, so the dependence of N-gas production on soil humidity is incomplete, but we expect (according to earlier studies) that emissions of both gases are continuously suppressed with higher WFPS content (*Horváth et al.*, 2010).

It should be noted that the soil water filled pore space can not explain all of the variations of N<sub>2</sub>O fluxes (see *Fig. 4b*). It seems to be more likely, that soil temperature usually generate short-term variations of the trace gas exchange, whereas the magnitude of the biogenic emission is predominantly influenced by soil wetness, and other factors (*Meixner and Yang, 2004*).



*Fig. 4.* Variation of nitric oxide flux (a) and nitrous oxide flux (b) as a function of soil temperature at Bugacpuszta station.

On the other hand, *Kool et al. (2007, 2009a)* studied our soils by incubation technique using isotope tracers of oxygen and nitrogen, when O-exchange between water and intermediate forms of the N-transformations were measured. By this novel approach they showed that nitrifier denitrification (nitrite reduction by ammonia oxidizers) can be a contributor to the majority of N<sub>2</sub>O production at Bugacpuszta site, thus, N<sub>2</sub>O can be produced at lower water content – this phenomenon may explain the secondary higher emission on low (20–30%) WFPS – see in *Fig. 5b*. This biochemical pathway also demonstrates that pH may be the major factor determining nitrifier-induced N<sub>2</sub>O production,

and the community of microorganisms may not be the key driver in different pathways of  $N_2O$  formation (Kool *et al.*, 2010). By this methodology it was also observed that water is effectively the main oxygen source (instead of  $O_2$  as it was assumed earlier) in  $N_2O$  formation and possibly in the formation of other nitrogen oxides in some European soil samples including Bugacpuszta. The oxygen isotopic measurement of  $N_2O$  showed that in Bugacpuszta the soil  $NH_4^+$  can be the nitrogen source (and it does not necessarily reflect that  $NO_3^-$  is functioning as a substrate) in  $N_2O$  formation though the nitrifier denitrification as an alternative pathway of metabolism of microorganisms (Kool *et al.*, 2009b). Further investigations are needed for better understanding the metabolism processes.

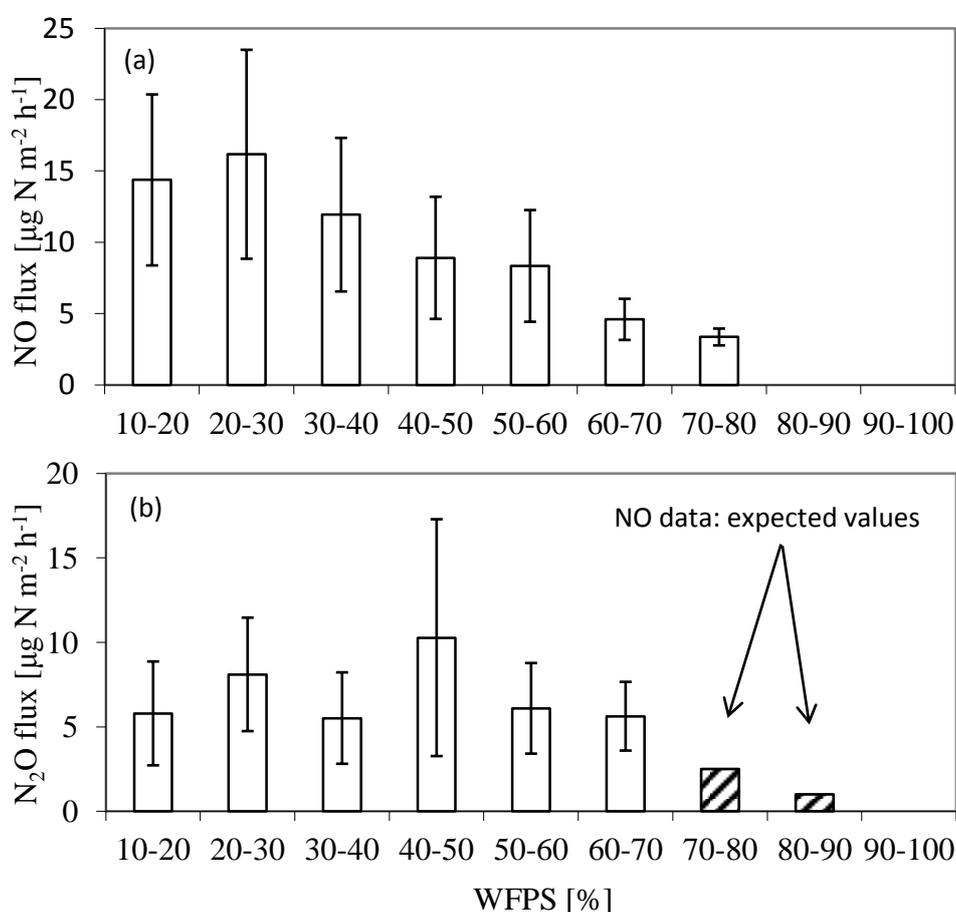


Fig. 5. Variation of nitric oxide flux (a) and nitrous oxide flux (b) as a function of water filled pore space at Bugacpuszta station.

The changes in N-exchange may impact on the carbon cycle as well as on plant uptake leading to opening up and desertification of the grassland, in spite of the fact that the studied ecosystem is evolved and more adapted to drought (in terms of NEE, etc.) than other grasslands (Pintér *et al.*, 2008).

#### 4. Conclusions

Weather perturbations can substantially modify both timing and magnitude of N-deposition and soil N-gas emission. In summertime, parallel with the precipitation deficit, less easily available nitrogen is deposited to the surface leaching to the rooting zone; thus, it can limit the mineral N uptake by plants or may affect the soil emission (through suppress of microbial processes) of N-gases during the main vegetation period.

Summarizing, many soil process (e.g., nitrification, denitrification, N-leaching) are strongly depend on soil temperature and precipitation as ecological drivers. The changes in these parameters influence directly the soil nitrogen gas emission rates, though the complex system of the relationships makes it difficult to explain all changes of NO and N<sub>2</sub>O formation. Though NO flux was higher than N<sub>2</sub>O flux, the soil emission was an order of magnitude lower than the atmospheric deposition (see *Table 1*).

The seasonal fluctuation of N<sub>2</sub>O and NO emission has been mainly influenced by precipitation. N<sub>2</sub>O emissions were not significant in the N-budget at this site in the last years. Reduced N<sub>2</sub>O emission (occurring through desertification due to perturbed climate conditions) means a potential negative feedback to the greenhouse effect. On the other hand, the vegetation can turn into being a net CO<sub>2</sub> source in extremely dry years like 2003 and 2007 (*Pintér et al.*, 2008; *Barcza et al.*, 2009) as a positive feedback for climate change. The ratio and strength of this two phenomena can not be neglected, since the area of temperate grass covered surface is large and will be increasing with increasing aridity of climate (and/or agricultural policy of Hungary). Extended periods of soil water deficit and high air and soil temperatures can affect a wide range of plant physiological functions. The plant communities will be frequently exposed to naturally induced droughts and should become open grassland in accordance with the value of the changing weather conditions.

Due to the forecasted potentially drying Hungarian climate, more frequent natural fires, as ecosystem function distractions, will occur in the dry sandy grassland (Hungarian Great Plain). The estimated nitrogen loss by fires equals to or even exceeds the amount of nitrogen from atmospheric deposition.

Based on the observed phenomena, it can be concluded that climate extremes are significant factors in soil organism functioning and dynamics of N-exchange and emissions. As a result, the N-content of the soil is continuously changing with the climatic anomalies (*Czóbel et al.*, 2008) due to the pool of NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> – which depends on the N-consumption and exchange of soil microbial community – affecting the plant N-uptake (demand), plant growth, etc. The living roots and bacteria are competitors for the same nutrients, so plants also induce effect on soil N-transformations. Further research relating to soil biochemistry in natural grasslands is also needed. Compared to the average of earlier years (13.8 kg N ha<sup>-1</sup>; *Kugler et al.*, 2008), less nitrogen deposited onto

the surface (11.2 and 11.7 kg N ha<sup>-1</sup>) in the driest period (2006 and 2007) and more nitrogen deposited (15.0 and 14.5 kg N ha<sup>-1</sup>) in the wetter years (2008 and 2010) during the study period. Wet deposition of nitrate and ammonium together with the dry flux of ammonia and nitric acid vapor is responsible for the majority (80%) of the net N-deposition flux. Seasonal and annual variation of N-gas emissions and N-deposition can be considerable, but precipitation, soil moisture, air and soil temperature, as easily measured parameters, can more or less explain this variability.

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